PROCEEDINGS
OF THE
SPECIALISTS’ MEETING
ON
THE SAFETY PROBLEMS
ASSOCIATED WITH
THE HANDLING AND STORAGE
OF UF6

Boekelo, The Netherlands, 27 - 29 June 1978

Hosted by the
URENCO NEDERLAND v.o.f.
with the support of the
MINISTRY OF SOCIAL AFFAIRS

COMMITTEE ON THE SAFETY OF NUCLEAR INSTALLATIONS
OECD NUCLEAR ENERGY AGENCY
38, boulevard Suchet, 75016 Paris, France
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FOREWORD

Uraniumhexafluoride (UF₆) is the process material used in UF₆ manufacturing and conversion plants, diffusion and centrifuge plants for uranium enrichment and fuel element fabrication plants. A specialists' meeting on the safety problems associated with the handling and storage of UF₆ promoted by the Committee on the Safety of Nuclear Installations of the OECD Nuclear Energy Agency, was held in Boekelo, The Netherlands, June 27 - 29, 1978.

The primary objective of the meeting was to exchange the experience gained over many years in those countries dealing with this process material in the field of design, construction and operation of UF₆ facilities, including transport and storage.

The papers in this report have been reproduced directly from the contributions of the authors.

For each session, the Session Chairmen drafted summaries, based on the papers and the discussions; these summaries are taken up at the end of the papers of each session. Also the panel discussion is summarized in this way.

Many thanks are due to all participants, to the Session Chairmen and to the members of the Program Group for their contribution to the scientific value of the meeting, to Urenco Netherlands Operations B.V. for all the practical arrangements and to the Ministry of Social Affairs of The Netherlands for the timely publication of the Proceedings of the Meeting.

C.J. van Daatselaar
Chairman of the Meeting
The Organisation for Economic Co-operation and Development (OECD) was set up under a Convention signed in Paris on 14th December, 1960, which provides that the OECD shall promote policies designed:

- to achieve the highest sustainable economic growth and employment and a rising standard of living in Member countries, while maintaining financial stability, and thus to contribute to the development of the world economy;

- to contribute to sound economic expansion in Member as well as non-member countries in the process of economic development;

- to contribute to the expansion of world trade on a multilateral, non-discriminatory basis in accordance with international obligations.

The Members of OECD are Australia, Austria, Belgium, Canada, Denmark, Finland, France, the Federal Republic of Germany, Greece, Iceland, Ireland, Italy, Japan, Luxembourg, the Netherlands, New Zealand, Norway, Portugal, Spain, Sweden, Switzerland, Turkey, the United Kingdom and the United States.

The OECD Nuclear Energy Agency (NEA) was established on 20th April 1972, replacing OECD's European Nuclear Energy Agency (ENEA) on the adhesion of Japan as a full Member. NEA now groups eighteen European Member countries of OECD and Australia, Canada and Japan, with the United States as an Associated country. The Commission of the European Communities - takes part in the work of the Agency.

The objectives of NEA remain substantially those of ENEA, namely the orderly development of the uses of nuclear energy for peaceful purposes. This is achieved by:

- assessing the future role of nuclear energy as a contributor to economic progress, and encouraging co-operation between governments towards its optimum development;
- encouraging harmonisation of governments' regulatory policies and practices in the nuclear field, with particular reference to health and safety, radioactive waste management and nuclear third party liability and insurance;

- forecasts of uranium resources, production and demand;

- operation of common services and encouragement of co-operation in the field of nuclear energy information;

- sponsorship of research and development undertakings jointly organised and operated by OECD countries.

In these tasks NEA works in close collaboration with the International Atomic Energy Agency, with which it has concluded a Co-operation Agreement, as well as with other international organisations in the nuclear field.

OECD, 1976.

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Director of Information OECD
2, rue André-Pascal, 75775 PARIS CEDEX 16, France.
The NEA Committee on the Safety of Nuclear Installations (CSNI) is an international committee made up of scientists and engineers who have responsibilities for nuclear safety research and nuclear licensing. The Committee was set up in 1973 to develop and co-ordinate the Nuclear Energy Agency's work in nuclear safety matters, replacing the former Committee on Reactor Safety Technology (CREST) with its more limited scope.

The Committee's purpose is to foster international co-operation in nuclear safety amongst the OECD member countries. This is done essentially by:

(i) exchanging information about progress in safety research and regulatory matters in the different countries, and maintaining banks of specific data; these arrangements are of immediate benefit to the countries concerned;

(ii) setting up working groups or task forces and arranging specialist meetings, in order to implement co-operation on specific subjects, and establishing international projects; the output of the study groups and meetings goes to enrich the data base available to national regulatory authorities and to the scientific community at large. If it reveals substantial gaps in knowledge or differences between national practices, the Committee may recommend that unified approach be adopted to the problems involved. The aim here is to minimise differences and to achieve an international consensus wherever possible.

The technical areas at present covered by these activities are as follows: particular aspects of safety research relative to water reactors, fast reactors and high-temperature gas-cooled reactors; probabilistic assessment and reliability analysis, especially with regard to rare events; siting research as concerns protection against external impacts; fuel cycle safety research; the safety of nuclear ships; various safety aspects of steel components in nuclear installations; licensing of nuclear installations and a number of specific exchanges of information.
The Committee has set up a sub-Committee on Licensing which examines a variety of nuclear regulatory problems, provides a forum for the free discussion of licensing questions and reviews the regulatory impact of the conclusions reached by CSNI.
In publications, reference to the proceedings of this Meeting should be made as follows:

CSNI Report No 33, Proc. OECD (NEA) CSNI Spec. Meet. on the Safety Problems associated with the handling and storage of UF$_6$;

Boekelo, June 1978.

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38 Boulevard Suchet,
75016 PARIS, France.
Tuesday, 27 June 1978

8.30  Registration.

9.30  Opening remarks, welcome addresses.

SESSION I : SAFETY PRINCIPLES FOR AND EXPERIENCE IN UF₆ HANDLING, STORAGE AND TRANSPORT - PART 1

Session Chairman: B.H. van de Wijngaert

10.00  Handling of UF₆ in U.S. gaseous diffusion plants.
       (A.J. Legeay)

10.20  COFFEE

10.40  The safety problems associated with the handling and the storage of UF₆ at EURODIF Tricastin.
       (P. Dabernard, E. de Dorlodot)

- 10.40 -
10.40 Safety aspects of uranium hexafluoride manufacture and reconversion at Springfield Works.  
(A. Price)

UF₆ handling at the URENCO Nederland centrifuge plants, Almelo.  
(F. Stockschläder, J. Christofzik)

11.40 Discussion

12.10 LUNCH

AFTERNOON Visit to URENCO centrifuge plants, Almelo.

Wednesday, 28 June 1978

SESSION I: SAFETY PRINCIPLES FOR AND EXPERIENCE IN UF₆ HANDLING, STORAGE AND TRANSPORT - PART II

Session Chairman: E.S. London

9.00 Design considerations for UF₆ systems.  
(W.K. Walrave, H. de Winter, B. Dekker)

The uranium hexafluoride vaporisation unit in the F.B.F.C. plant at Romans.  
(J.M. Blum)

Yield and storage of depleted uranium in the Federal Republic of Germany.  
(W. Kayser)

Safety considerations involved in the choice of containers for storage of UF₆ tails.  
(D.F. Norsworthy, C. Howarth)
10.20  COFFEE

10.40  Discussion

SESSION II.A: UF₆ BEHAVIOUR. THEORETICAL ASSESSMENTS
Session Chairman: C. Jouannaud

11.10  The safety problems associated with the handling and storage of UF₆.
(W. Thomas)

A quantitative forecast of behaviour of UF₆ release in the air.
(A. Doumenc, R. Gérard)

Atmospheric dispersion and consequences of a UF₆ release caused by valve rupture in a hot 30B cylinder.
(A.M. Ericsson)

12.10  LUNCH

14.00  Safety analysis of UF₆ transportation.
(F. Israël)

14.20  Discussion.

SESSION II.B: UF₆ BEHAVIOUR. EXPERIMENTAL VERIFICATIONS
Session Chairman: Dr. Banaschik

14.50  Fundamentals of UF₆ accidental release.
(H. Bouzigues, M. Mezin)
Studies and results in safety of uranium hexafluoride installations.
(A. Raggenbass, M. Berlin, J.C. Malet)

15.30 COFFEE

15.50 Discussion

SESSION III : INCIDENT EXPERIENCE
Session Chairman : G.T. Hull

16.10 Experience on UF₆ handling in uranium manufacturing processes
(F. Schinzer, R. Hille)

UF₆ release experience and prevention in United States gaseous diffusion plants.
(G.T. Hull)

An experience of accidental release of UF₆
(A. Ducouret)

17.10 Discussion

Thursday, 29 June 1978

9.00 FINAL PANEL : CONCLUSIONS AND RECOMMENDATIONS OF THE SPECIALIST MEETING
Panel Chairman : F.A. Stockschläder

10.30 COFFEE

10.50 FINAL PANEL (continued)

12.00 Concluding remarks

12.30 CLOSURE OF THE MEETING
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OPENING ADDRESSES
Opening remarks by Mr. O. Ilary at the CSNI Specialist Meeting on the Safety Problems associated with the handling and storage of UF6.

Mr. Stockschaeder, Mr. Van Daatselaar, Ladies and Gentlemen,

I am pleased to welcome you on behalf of the Director General of the OECD Nuclear Energy Agency to this CSNI Specialist Meeting on the safety problems associated with the handling and storage of UF6. It also gives me pleasure to transmit to you best wishes from the Chairman of the Committee on the Safety of Nuclear Installations (CSNI) for a successful meeting.

Over the past four years this Committee of NEA has sponsored some 20 Specialist Meetings and various other co-operative activities in the field of nuclear safety research and regulatory matters.

Because not all of you may be familiar with CSNI activities, I think it would be perhaps appropriate to speak a few words about CSNI and its role in international co-operation.

The merits of the international co-operation in nuclear safety and regulatory matters are today, I believe, undisputed. Let me just recall briefly the three fundamental reasons for this:

1. There is a clear need for the different countries to compare experience and share the effort involved in reducing the risks associated with nuclear power and in maintaining its excellent safety record.

2. Safety and regulatory questions are especially well suited for intergovernmental co-operation - such as is carried out in the framework of NEA - as these questions are the primary responsibility of public bodies in our Member countries.

3. Visible and vigorous international co-operation in nuclear safety leading to a consensus on important safety questions is likely to ease
ease public acceptance of nuclear power.

In the light of these basic incentives for international co-operation, the first concerted effort by the NEA in the field of nuclear safety developed within the then CREST, or Committee for Reactor Safety Technology, as from 1965. As it was appropriate at that time, this Committee was concerned with those basic aspects of nuclear technology affecting reactor safety. In 1973 the present Committee on the Safety of Nuclear Installations (CSNI) was set up, in order to expand the co-operation between Member countries in nuclear safety research and also to provide a forum for a free exchange of information and views between national regulatory and licensing authorities.

CSNI brings together the most influential voices in nuclear safety research and licensing in the OECD area, and its working objectives are essentially two:

- to enrich and broaden, by means of technical exchanges, the fund of data available to designers and to regulatory authorities for taking their decisions in the matter of safety;
- to achieve consensus of experts and responsible people on key safety issues.

With these aims in mind, a flexible working arrangement has been adopted as follows:
- CSNI meets in plenary session, usually once a year, primarily for setting up direct co-operative ventures in safety research, and to co-ordinate relevant national work;
- under CSNI direction, special studies are carried out by a number of groups of experts, working groups and task forces, or through specialist meetings;
- results and conclusions are subsequently reported to and evaluated by CSNI and, through its national delegates, they are diffused to national safety authorities.

-It-
It is probably appropriate to recall and point out that this effort to CSNI, by its very nature and its objectives, is focussed on the solution of selected advanced problems and in no way can it be confounded with the development of safety codes or guides, which is a specific responsibility of the IAEA. We can even say that, contributing to the broadening of the safety data base, the CSNI programme is complementary with that of IAEA and in fact, we strongly support the IAEA activities in this field.

I need not remind you of the spectacular expansion of nuclear safety activities in the OECD area in recent years. This development has been accompanied by an even stronger increase in international contacts and collaboration.

Let me just give you a few figures to illustrate the importance of nuclear safety in terms of money in the OECD area. Our Member countries spent last year some 15-20 billion dollars to construct and operate nuclear power plants and the associated fuel cycle facilities. At the same time expenditure of OECD governments on safety research amounted to about 800 million dollars a year, and this sum is divided among some 1100 different research projects; these projects are listed and described in the CSNI Nuclear Safety Research Index, which is published annually.

Obviously, CSNI's contribution to such a complex programme is necessarily restricted to those questions judged most important and urgent by a sufficient number of Member countries. While it is important to be as comprehensive as possible in the area of general information exchange - such as the Nuclear Safety Research Index - the limitation of resources, both nationally and internationally, dictates that operational co-operation on specific topics such as pursued by CSNI ancillary bodies (working groups, task forces, etc.) must be limited to those which have been accorded highest priority. In this respect CSNI produced in 1976 a list of priority topics on which to base its future programme. This list includes some 70 specific questions grouped in a few broad areas covering safety research and regulatory and licensing matters. The areas concerning safety research were, until recently -
recently, the following:

- Thermal reactor safety research, including such topics as emergency core cooling problems, international comparisons of computer codes for the analysis of loss of coolant accidents, fuel behaviour in accident conditions, etc.
- Fast reactor safety, covering subjects such as sodium/fuel interactions, aerosol behaviour in accident conditions, etc.
- General safety, including the safety of nuclear ships, the definition and treatment of rare events in the probabilistic safety assessments, the mechanical and material problems of steel components of power reactors, etc.

CSNI, as I said, has for several years been traditionally active in the field of reactor safety, but, considering widespread interest in fuel cycle studies and growing public and governments reluctance to accept nuclear power without a clear demonstration of the feasibility and safety of the entire fuel cycle - particularly of spent fuel storage and reprocessing, and radioactive wast management - it decided in 1976 to initiate joint international activities in this field. Another major consideration justifying this decision was the need for assuring that a coherent safety approach would be applied to all types of nuclear installations, throughout the entire fuel cycle.

For this purpose a Working Group was set up in 1977, under the Chairmanship of Mr. Jouannaud, who is here with us and will chair one of the sessions of this meeting. The task of the group was to review the safety status of the fuel cycle and to assess for each stage the potential risks and safety problems involved and the prevention systems adapted, identifying critical safety issues and problem areas where further research or analysis was needed.

The Working Group so far reviewed these problems, reaching a consensus on a number of key safety issues, and a preliminary status report on the safety of the fuel cycle has been prepared.

This report briefly discusses the potential risks and safety problems presented by each stage of the fuel cycle excluding nuclear reactors,
as well as the present state of knowledge and research in this field. This first report is not intended to address such a complex matter in a deep and exhaustive manner, but rather to be an overview containing concise expert statements on the various problems involved. However, it already includes a list of the most critical safety issues identified by the Working Group and the problem areas requiring further research or analysis, and is concluded by a series of proposals and recommendations to CSNI for future work.

One of these areas identified by the group is represented by the safety problems of handling, storage and transport of UF6. The group had, in fact, recognised that the risk of accidental release of UF6, although not significantly high, was not sufficiently known and studied and required a deeper insight. In fact, a large release of UF6 into the atmosphere can occur due to errors of operation in UF6 handling and storage operations or following a sudden loss of confinement barriers due to external impacts, such as earthquakes, aircraft crashes, explosions, fire, etc. It was, in particular, recognised that little was known about the behaviour of large masses of liquid UF6 when released into the atmosphere and that more information was needed on the reaction mechanisms of UF6 with air moisture, its evaporation rate, the atmospheric dispersion and ground deposition processes of the solid and vapour components, its reactions with hydrocarbons in case of an airplane crash, etc.

On the basis of these indications of the Working Group, CSNI deemed it appropriate to stimulate an exchange of information in this field through the organisation of this specialist meeting.

Allow me to remind you that CSNI expects that your conclusions from this meeting will state the present situation of knowledge and experience in the field of UF6 safety, and will point out what further practical action of international co-operation in this field you recommend for implementation by CSNI and NEA.

Before making room for the experts, I should like to thank the URENCO Company and the Ministry of Social Affairs of The Netherlands for having so kindly offered to host this meeting. Particular thanks are also due - to -
to Mr. Kell of Urenco and his collaborators who have worked so hard to lay the necessary foundation for a successful meeting.

I wish you success in your deliberations.

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Ladies and Gentlemen,

First of all it is a pleasure for me to welcome you and I hope you will have a pleasant and fruitfull stay in the Netherlands. With respect to this expert meeting on the safe handling of UF₆ I would like to say a few words.

In the framework of safety evaluations of the different steps in the nuclear fuel cycle, main emphasis has always been given to the nuclear reactors. It is in the recent years that the tail end of the fuel cycle, in particular the ultimate disposal of high level radioactive waste is getting the required attention. In many studies, carried out in different countries during the last years, it has been shown that the first steps in the fuel cycle (excepted mining) as conversion, enrichment and fuel element fabrication, do not contribute in any significant way to the risk of nuclear energy production. To my opinion this does not mean that the safety aspects of the handling and transport of uraniumhexafluoride can be disregarded.

There has been, and there still is, quite a lot of information exchange with respect to the safety of nuclear power reactors. Therefor I think we may welcome the proposal of the Committee on the Safety of Nuclear Installations to have an expert meeting on the safety problems associated with the handling and storage of UF₆. In the next three days we will have an excellent opportunity to hear about the experience gathered in different countries with the handling of UF₆. And, more than just listening, as we do not have an overloaded programme, there will be ample time for discussions.

With respect to the programme itself, this morning and a part of tomorrow will be devoted to the safety principles for and experience in handling, storage and transport of UF₆. This morning in particular will be focussed on design principles, general safety aspects and operating experience in the larger installations in the United States,
United Kingdom and the Netherlands.
Tomorrow some more specific details will be highlighted.
In the next session of tomorrow consideration will be given to the behaviour of UF₆; papers on theoretical views as well as experimental verifications will be presented.
Then in this meeting's presentations we will hear about incidents. In fact I think it will proof the statement that UF₆ can be handled without any unacceptable risk to the workers or to the public.
Finally on Thursday morning there will be a panel session. More details will be communicated later, but in any case it will be a perfect opportunity to give your views or suggestions on the matters discussed, and to indicate the areas where further development might be useful for a safe handling of UF₆.
Ladies and Gentlemen, I wish you a very successful meeting and I declare the meeting open.
PAPERS, INCLUDING SUMMARY AND PROGRAMME GROUP'S CONCLUSIONS
SESSION I
SAFETY PRINCIPLES FOR AND EXPERIENCE IN UF₆ HANDLING, STORAGE AND TRANSPORT
PART I
Handling of UF₆ in U.S. gaseous diffusion plants

by

A.J. Legeay

Operations Division
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ABSTRACT

A comprehensive systems analysis of UF₆ handling has been made in the three U.S. gaseous diffusion plants and has resulted in a significant impact on the equipment design and the operating procedures of these facilities. The equipment, facilities, and industrial practices in UF₆ handling operations as they existed in the early 1970's are reviewed with particular emphasis placed on the changes which have been implemented. The changes were applied to the systems and operating methods which evolved from the design, startup, and operation of the Oak Ridge Gaseous Diffusion Plant in 1945.

INTRODUCTION

Uranium enrichment in the United States is accomplished in three gaseous diffusion plants operated for the U.S. Department of Energy. The oldest plant, the Oak Ridge (Tennessee) Gaseous Diffusion Plant, has been in continuous operation for more than 30 years. Figure 1 is an aerial view of the plant. Figure 2 shows the Paducah (Kentucky) Gaseous Diffusion Plant, which was started in 1952. Both of these plants are operated by the Nuclear Division of Union Carbide Corporation. The third plant, shown in Figure 3, is the Portsmouth (Ohio) Plant, operated by Goodyear Atomic Corporation, which also began operating in the mid-1950's. The three plants are located in rural areas, 15 to 30 kilometers away from high-density population centers.

Since the beginning of operations, emphasis has been placed on personnel safety and avoidance of UF₆ releases and insults to the environment. In 1973 this effort was expanded when a three-plant committee was established to make a comprehensive and systematic analysis of the UF₆ handling in gaseous diffusion plants. The need for this action arose from (1) an expanding enrichment industry which would more than double plant capacity; (2) an ever-increasing sensitivity to personnel safety and environmental insult; and (3) the potential for various types of UF₆ releases due to equipment failure. The analysis was divided into 15 areas of UF₆ handling operations, as shown in Table I, and a thorough evaluation of appropriate safety criteria as applied to UF₆ systems, equipment, and procedures led to more than 100 recommendations.

This report will briefly describe the UF₆ handling operations in the gaseous diffusion plants, emphasizing the precautions taken to prevent the release of UF₆. The UF₆ handling operations are those involved in the sampling, transferring, feeding, and withdrawing UF₆ in the gaseous diffusion plants and do not include the operation of the diffusion cascade separating equipment. The committee's investigation and resultant recommendations which have major impact on the equipment design and operating procedures of the UF₆ handling facilities will be discussed in some detail. Some of the more significant areas of concern and the resultant actions taken follow:

1. The design of the liquefaction and vaporization system which includes an autoclave has been modified to assure that over-pressurization of the cylinder does not occur while heating UF₆. It is further designed to provide containment for releases of uranium hexafluoride and its reaction products if the primary containment provided by the cylinder should be lost.

2. Connections used for the transfer of UF₆ gas or liquid in sampling, transferring, or feeding operations are of special design and are tested and inspected prior to use to assure their integrity.
3. Administrative procedures and controls have been adopted to prevent the introduction of reactive organics and other contaminants into UF₆ containers, connecting lines, and sample volumes which could cause violent reactions, rupture of the system, and release of UF₆.

4. A degree of confinement of UF₆ releases will be provided by modifications to the buildings where UF₆ handling operations are done. UF₆ detectors will be provided to activate the shutdown of building exhaust systems and closure of doors (with option to override). A scrubber system on the building exhaust would facilitate emergency operations for safe equipment shutdown, leak isolation, minimizing environmental insult, and simplifying the decontamination of the area.

5. Handling of cylinders which contain liquid UF₆ is minimized in the gaseous diffusion plants, since the potential for a major release is greatest when the UF₆ is in a liquid state. Containers of liquid are elevated to minimum heights and moved shortest possible distances in processing.

The committee analysis did not consider the potential for a nuclear criticality event. Extensive criticality studies have been performed in the past on these systems, and design and operating considerations assure subcritical conditions during all foreseeable, credible circumstances.

OVERVIEW

An overview of all UF₆ handling operations is shown in Figure 4. All facilities shown are similar at the three plants and are ideally suited to a combined systems analysis. UF₆ feed cylinders enter the plant by either truck or railcar. They are removed by crane and if sampling is required they are heated and homogenized to the liquid state in steam-heated autoclaves or atmospheric steam hoods. The samples are analyzed for chemical purity and isotopic concentration. After sampling the UF₆ is cooled to a solid before being transported to the feed station. Feed vaporization is also accomplished by autoclaves. After separation in the gaseous diffusion equipment, both the enriched and depleted material are withdrawn by a compression-liquefaction system into cylinders. The product cylinders are again cooled before being transported to the sampling and transfer autoclaves. Cooled waste cylinders, commonly called "tails," are moved to permanent storage areas. When the three diffusion plants are being operated at full uprated capacity, 3,750 tails cylinders and about 13,000 square meters of cylinder storage area will be needed each year.
The integrity of UF₆ containers (cylinders) and their valves is the front-line of defense for avoiding UF₆ releases. The largest size steel cylinders used for UF₆ handling have an internal volume of 4.05 cubic meters and a capacity of 12,700 kg of UF₆. The risk of releasing large quantities of UF₆ is greatest when the cylinder contents are liquid and the cylinder integrity is violated by accidental conditions. For instance, nearly all of the 12,700 kg of UF₆ could be discharged from the cylinder in 15 minutes if the 2.5 cm cylinder valve breaks off when the valve is in the 6 o'clock position. With the valve located in the gas phase of the cylinder, an initial leak rate of 34 kg/min UF₆ could be experienced with as much as 7,300 kg of UF₆ released in 6 hours under adiabatic conditions.

Types of Cylinders

Four types of cylinders are most commonly used in the diffusion plants. Three of the cylinders and valves meet the ANSI Standard 1/ for "Packaging of Uranium Hexafluoride for Transport." The fourth type, 48G cylinder, is not contained in the ANSI Standard and is used exclusively for UF₆ tails storage. Table II shows the model numbers and other pertinent information for these cylinders.

More detailed information on cylinder types and UF₆ handling is given in ORO-651, Rev. 4, "Uranium Hexafluoride: Handling Procedures and Container Criteria." 2/

Inspection, Cleaning, Testing, and Weighing

All cylinders are coded pressure vessels and added precautions are taken to inspect them externally, to check their cold pressure, and to weigh them at the time of delivery to the diffusion plants. Any dent, visible crack, or significant removal of metal renders the cylinders unacceptable. While external inspection might sound like a rather simple procedure, it is critical to preventing a release while transporting or heating the cylinders.


Another inspection requirement is a cold pressure check on full feed and product cylinders and all empty cylinders before filling with UF₆. The purpose is to assure the absence of volatile contamination. The presence of foreign materials could violate the system design pressure by their vapor pressure contributions or by the reaction with UF₆. The cold pressure check may not detect the presence of organic compounds such as vacuum pump oil, so operational certainty must be attained that oil is not introduced into cylinders during cylinder evacuation. It is the current practice at Oak Ridge to evacuate UF₆ cylinders by the use of air ejectors. If cylinders are evacuated with equipment which uses oil-lubricated pumps or compressors, a trap, filters, and check valves should be installed in the system to prevent inadvertent transfer of oil into the evacuated cylinder.

Each cylinder is weighed before heating or filling as a safety precaution since the physical properties of UF₆ lead to the possibility of rupturing an overfilled UF₆ cylinder by heating. To avoid this occurrence, the cylinders are fabricated with a known volume and a maximum fill weight is established for each cylinder type. An operational control is to weigh each cylinder before it is heated to assure the cylinder is not overfilled.

Cylinder Valves

The cylinder valves must be compatible with UF₆ and resistant to stress corrosion cracking in UF₆, HF, and industrial environments. Valves of 2.5 cm diameter are used on cylinder models 30B, 48X, 48Y, and 48G. The body, packing nut, packing ring, and packing follower are aluminum-silicon-bronze alloy C636. The stem is Monel; the packing and cap gasket are virgin Teflon. This valve is shown in Figure 9. The valve is specifically designed for UF₆ cylinder application and is built to rigidly controlled specifications. The aluminum-silicon-bronze is a single-phase, stress-corrosion-resistant alloy. Only virgin Teflon is specified to preclude the possibility of contaminated Teflon reacting with UF₆. The cylinder valve threads are tinned with

50-50 tin-lead alloy to provide a leak-tight fit into the cylinder.

In the cylinder fabrication a half coupling is used to provide the engaging threads for the valve. Cylinders with only tapped holes have been found to increase the incidence of small UF₆ releases around the threaded portion of the valve. A half coupling is used to receive the plug in the other end of the cylinder to avoid similar release potentials.

When a valve replacement is made, it is a policy to use a new valve and not to rebuild valves.

All full cylinders being transported must be equipped with either a valve protector (Figure 10) or be enclosed in a protective package (Figures 11 and 12).

CYLINDER HANDLING EQUIPMENT

The equipment for transporting cylinders containing liquid and/or solid uranium hexafluoride consists of cranes, straddle carriers, forklifts, and Tourn-a-haulers (Figures 13, 14, 15, and 16). The cranes have limit controls, emergency stop switches, and adequately designed cable and lifting fixtures to assure safe operation. The hydraulic or mechanical lift systems, brakes, and steering must function properly for safe operation of the straddle carriers and forklifts. Straddle carriers must be well designed and maintained in proper condition. The hydraulic system is improved by the installation of hold valves on the hydraulic cylinders which actuate the lift mechanism for the straddle carriers. The hold valves contain the hydraulic fluid once it is in the cylinder. Thus, the cylinder mechanism could not fail as a result of a break in the hydraulic system.

Added protection from dropping a cylinder can be gained by providing a positive connection between the cylinder and carrier. There are a variety of methods to pin the cylinder to the carrier through a hole in the cylinder lifting lug. Tourn-a-haulers, unlike cranes, straddle carriers, and forklifts, do not transport UF₆ cylinders suspended from the ground. The cylinder is lifted by a crane-like mechanism and set down on cradles on a truck bed.

The UF₆ handling systems analysis revealed that the most critical aspect is handling UF₆ in the liquid phase. The principal recommendations were geared toward minimizing liquid handling, since it poses the greatest potential for a large release. Many recommendations that resulted from the analysis have been implemented and some are shown below.

1. Training, testing, and licensing of heavy equipment operators is done by an independent group. A continued strong degree of reliance must be placed upon well-trained human operators and their adherence to procedures.
2. A gas mask and escape rope are provided in cab-operated cranes where a UF₆ release may occur. Cab-operated cranes are typically 10 meters above ground level and provide no easy means of escape for the operator should a mishap occur.

3. All straddle carriers are equipped with an alarm system to alert the driver if the cylinder is not in the fully raised position.

4. Steps have been taken to minimize or eliminate the movement of partially full liquid UF₆ cylinders. The heavy liquid poses a threat of sloshing, which causes the cylinder to become unbalanced while being hoisted. If partially full liquid cylinders must be moved, a double-hook crane is recommended.

AUTOCLAVES AND STEAM HOODS

The heating of UF₆ in cylinders is an operation which increases the potential for a UF₆ release. The autoclaves and steam hoods are used for the three cylinder heating operations: feeding to the diffusion plant, sampling incoming cylinders, and sampling and transferring product to customer-owned cylinders.

Feeding

All future feed stations will consist of pressurized steam-heated cylinder enclosures called autoclaves (Figure 17). These units have a design pressure of about 11 bars and provide a positive secondary containment vessel. The heating medium is 1.6 bars steam and the operating temperature is 113°C. Recommendations of the UF₆ Handling Committee have been implemented in these units as follows:

1. Any cylinder should have the cylinder valve opened prior to being heated for feeding. The opened valve is connected to a low-pressure alarm associated with a timer and a high pressure alarm. If the rate of pressure rise is less than prescribed, a plugged line or valve malfunction may be the cause and an alarm is actuated.

If contaminants are present and the cylinder overpressurizes, the high-pressure alarm would sound. The source of heat is cut off at either alarm point; the alarms are audible and the areas are continually manned.
In the Paducah Plant, rupture disks are used in the cylinder manifold and provide pressure relief into surge drums if a high-pressure condition is encountered.

2. Each feed autoclave is monitored internally with a UF₆ detection device. If a UF₆ leak occurs, the material reacts with the steam and increases the conductance of the condensate. Conductivity cells are used to monitor the condensed steam. An audible alarm is provided, along with automatic cutoff of heat supply and automatic closure of the condensate drain valve.

3. A liquid level indicator in the autoclave will cut off the heat supply and sound an alarm if the condensate is not properly draining. This level gauge controls the water in the autoclave to a maximum of 23 kg and is required to control the pressures which may be generated by a UF₆ and water reaction.

4. Feeding areas are monitored with UF₆ detection devices. At the alarm point automatic isolation of the cylinders from the manifolds occurs. The devices in 2, 3, and 4 above are tested monthly.

5. Respiratory protection equipment is required during the opening and closing of a UF₆ system.

Sampling

The autoclaves used for UF₆ sampling differ from those used for feeding in that they are capable of tilting and rotating the cylinder to provide for liquid sampling and transfer. Steam hoods (Figure 18) are also used for sampling, although they are an older technology and will eventually be replaced by autoclaves. Prior to heating for sampling, a cold pressure check is made to assure the absence of volatile contaminants. The cylinder is heated with the valve in the 12 o'clock position, which is above the liquid level of the UF₆. The valve is opened and attached to a pressure monitor throughout the required heating period to assure that the pressure does not exceed 5.2 bars at 93°C or 8.6 bars at 113°C. A thermocouple is placed on the cylinder and shuts off the steam if the temperature is higher than 113°C. Heating time is approximately 6 hours for 10- and 14-ton cylinders. All cylinders are sampled with the valve below the liquid level in the 3-5 o'clock or the 7-9 o'clock position.
The cylinder is connected to the fixed sampling volume with a length of Monel, nickel, or copper tubing. The tubing, called a "pigtail," is certified and is inspected prior to each usage. The connection is pressure tested prior to introduction of UF₆. Prior to opening the sampling system to atmosphere, the system is purged with air, evacuated to 34.5 millibars and back-filled with dry air or nitrogen to atmospheric pressure. The purging and evacuating operations are designed to minimize the possibility of environmental insult when the lines are opened to the atmosphere.

Transfer

Figure 19 is a schematic diagram of a typical transfer operation. An autoclave is used to heat the cylinder and contains the cylinder during the transfer operation. This system is vulnerable to UF₆ releases at the time the cylinder is being connected to the transfer manifold since the autoclave is open and mechanical connections are being made. Proper operation techniques will avoid UF₆ releases.

Once the cylinder is connected to the manifold and the autoclave closed, liquid UF₆ is drained by gravity through heated lines and valves into evacuated cylinders. The evacuated cylinder is positioned on a scale and is continually monitored to prevent overfilling. The systems are designed to be equipped with automatic cutoff devices which are actuated either by the scales or indication of ambient UF₆.

Several actions have been taken as a result of the system analysis, particularly concerning the pigtail, or length of tubing connecting the cylinder to a manifold or sampling volume.

1. The pigtail is an engineered piece of equipment. Each individual pigtail is tested, coded, and provided with a unique serial number. Flexible hoses are not approved for pigtails.

2. The design of the pigtail heater is such that the resistance and the operating voltage of the heater limit the temperature rather than depending on a control device.

3. All pigtails are dimensionally checked and tested to 2 times the specified design pressure.

4. Each pigtail is visually inspected prior to
each usage to determine that there is no damage to the pigtail fitting, threads, or any deforming, kinking, or flattening of the tubing. The pigtail is not a disposable material that wears out, but a reusable piece of equipment. Pigtails are decontaminated after each use. Cleanliness of the pigtails is mandatory.

5. Only virgin Teflon gaskets are used. Used Teflon may have absorbed contaminants that will react with UF₆. Gaskets are used only once.

6. All connections are leak tested prior to introduction of UF₆.

7. Either an emergency cutoff valve should be incorporated in the pigtail or crimping tools should be available to isolate the pigtail contain the UF₆ in a hot cylinder when the valve will not close.

UF₆ WITHDRAWAL

Both tails and product are withdrawn from the diffusion plants by compression systems which compress the UF₆ above the triple point pressure (see phase diagram in Figure 20) and then cool it to condense it to a liquid. The UF₆ flows as a liquid into an evacuated UF₆ cylinder positioned on a scale. The uranium hexafluoride pressure associated with the withdrawal operation is a potential source of release.

The product and tails withdrawal facilities are monitored by a UF₆ release detection system with strategically located sensing heads which, if activated, would automatically close a valve between the withdrawal system and the cylinder being filled. Further isolation, compressor shutdown, and venting of the UF₆ pressure could be accomplished in a relatively short period of time from the control room. Existing steam distribution headers could be used to "knock down" UF₆ vapors if a significant release were to occur.

The emergency closure valves provided in the liquid UF₆ drain lines are actuated when the cylinder weight reaches its fill limit. This action prevents overfilling of the cylinder. The cylinder scales and/or load cells are routinely calibrated. If a cylinder is inadvertently overfilled, it is not removed from the withdrawal facility until the overfilled condition has been resolved. The resolution is usually low temperature evacuation to the cascade through vent return lines.

Most of the safety concerns expressed about pigtails in the transfer section also apply to withdrawal.
CONTAINMENT

Additions and modifications have been made and are planned for facilities within the diffusion plants where the greatest potential for large UF₆ releases exist. These modifications are aimed at reducing the likelihood of a release, limiting the size of a release, minimizing personnel exposure in a release, containing a release within a small building area, and facilitating decontamination of buildings and recovery of uranium compounds. The following are examples of these modifications:

1. Steam lines and valves are provided to permit steam down and hydrolysis of UF₆ in areas where a release has occurred.

2. Emergency exhaust systems are provided to vent minor localized releases to an alumina trap before venting to the atmosphere. A UF₆ release will activate the shutdown of the building ventilation system and closure of doors.

3. Positive secondary containment is provided when heating a UF₆ cylinder to the liquid state.

4. Control rooms are operated under a positive pressure compared to process areas where UF₆ releases are possible.

5. Valves and expansion joints in high-pressure portions of UF₆ piping are buffered.

SUMMARY AND CONCLUSIONS

The three U.S. gaseous diffusion plants have operated a combined 80 years and have handled large quantities of UF₆ during that time. While UF₆ releases have occurred, there have been no lasting discernible effects to either the environment or personnel.

A detailed analysis has been recently performed which further reduces the potential for releases at the diffusion plants. Some of the most significant results of the analysis include:

1. Minimize handling cylinders which contain liquid UF₆.

2. Positive secondary containment during heat-up of UF₆.

3. Certified connections between UF₆ cylinders and other process equipment.

4. Coded UF₆ cylinders and rigidly controlled UF₆ cylinder valves.

5. Provide positive building containment.

6. Install facility ventilation systems with a gas scrubber.

7. Segregate UF₆ handling areas from the control room areas.
TABLE I

FIFTEEN AREAS OF UF$_6$ HANDLING RECOMMENDATIONS

<table>
<thead>
<tr>
<th>Cylinder</th>
</tr>
</thead>
<tbody>
<tr>
<td>Inspection and Testing</td>
</tr>
<tr>
<td>Integrity</td>
</tr>
<tr>
<td>Handling</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>UF$_6$ Operations</th>
</tr>
</thead>
<tbody>
<tr>
<td>Feeding</td>
</tr>
<tr>
<td>Sampling</td>
</tr>
<tr>
<td>Transfer and Assay Blending</td>
</tr>
<tr>
<td>Withdrawal</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Handling of UF$_6$ Releases</th>
</tr>
</thead>
<tbody>
<tr>
<td>Containment</td>
</tr>
<tr>
<td>Emergency Personnel and Equipment</td>
</tr>
<tr>
<td>Securing the Release</td>
</tr>
<tr>
<td>Communications</td>
</tr>
<tr>
<td>Decontamination</td>
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</table>

<table>
<thead>
<tr>
<th>Valves</th>
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</thead>
<tbody>
<tr>
<td>Inspection, Testing, and Replacement</td>
</tr>
<tr>
<td>Rebuilding</td>
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</tbody>
</table>

<table>
<thead>
<tr>
<th>Flexible Cylinder Connectors (&quot;Pigtails&quot;)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Design and Testing</td>
</tr>
</tbody>
</table>
**TABLE II**

**UF₆ CYLINDER DESCRIPTION**

<table>
<thead>
<tr>
<th>Cylinder Model No.</th>
<th>Diameter (cm)</th>
<th>Use</th>
<th>Capacity</th>
<th>Shown in Figure</th>
</tr>
</thead>
<tbody>
<tr>
<td>30B</td>
<td>76</td>
<td>Product Shipment</td>
<td>2,277 kg</td>
<td>5</td>
</tr>
<tr>
<td>48X</td>
<td>120</td>
<td>Feed or Product</td>
<td>9,539 kg</td>
<td>6</td>
</tr>
<tr>
<td>48Y</td>
<td>120</td>
<td>Feed or Product</td>
<td>12,501 kg</td>
<td>7</td>
</tr>
<tr>
<td>48G</td>
<td>120</td>
<td>Tails (Waste)</td>
<td>12,701 kg</td>
<td>8</td>
</tr>
</tbody>
</table>
Figure 1
AERIAL VIEW OF OAK RIDGE PLANT

Figure 2
AERIAL VIEW OF PADUCAH PLANT
Figure 3
AERIAL VIEW OF PORTSMOUTH PLANT

Figure 4
UF₆ HANDLING OPERATIONS
Figure 5
CYLINDER MODEL NO. 30B

DIAMETER: 76 cm
CAPACITY: 2,277 kg

Figure 6
UF₆ CYLINDER MODEL 48X

DIAMETER: 120 cm
CAPACITY: 9,539 kg
**UF₆ CYLINDER MODEL 48Y**

Diameter: 120 cm  
Capacity: 12,501 kg

Figure 7  
Cylinder Model No. 48Y

**UF₆ CYLINDER MODEL 48G**

Diameter: 120 cm  
Capacity: 12,701 kg

Figure 8  
Cylinder Model No. 48G
Figure 9

2 1/2 CENTIMETER UF₆ CYLINDER VALVE

Figure 10

CYLINDER VALVE PROCTOR
Figure 11

PROTECTIVE PACKAGE FOR MODEL 30B CYLINDERS

Figure 12

PROTECTIVE PACKAGE FOR MODEL 48X AND 48Y CYLINDERS
Figure 13
CRANE

Figure 14
STRADDLE CARRIER
Figure 15
FORKLIFT

Figure 16
TOURN-A-HAULER
Figure 17
AUTOCLAVE

Figure 18
STEAM HOODS
Figure 20

UF₆ PHASE DIAGRAM
The safety problems associated with the handling and the storage of UF₆ at Eurodif Tricastin by
P. Daberard, E. de Dorlodot
THE SAFETY PROBLEMS ASSOCIATED WITH THE HANDLING AND THE STORAGE OF UF6 AT EURODIF TRICASTIN

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Designer of the Reception and Feeding Installations of EURODIF

Edouard-Michel de Dorlodot
Chef de Service à EURODIF PRODUCTION, Pierrelatte

After describing the methods of handling uranium hexafluoride in EURODIF, the authors explain the safety criteria which guided the design of the Tricastin installations.

With the help of slides, they show how these criteria were applied in the realisation of both the reception and despatch area and of the installations directly connected to the cascade.

Finally, they refer to the training of personnel.
Before discussing the safety aspects of the problems, we shall describe
the EURODIF factory at TRICASTIN, and the main operations necessary
from the reception of the customers uranium at the entrance to the
delivery of the enriched product.

The EURODIF factory is a gaseous diffusion plant of 10,8 million
SWU capacity, designed for low enrichment of less than 5%. That means
that big quantities of UF6 have to be handled every year, even as
enriched uranium.

1. As you know a diffusion process is a continuous one in a leak proof
installation, so the handling problems are mainly localised in the
feeding and extraction installations. We intend to present these
problems to-day.

1.1. The feed material

The natural uranium arrives as UF6 in standard 48Y containers.
At the entrance, the containers are inspected for integrity,
contamination and activity. After weighing and sampling for
analysis, the material is accepted if all data are within the
norms set out in the specifications of the contracts. If not,
special action has to be taken with the customer.

The material received is stored for some time pending utili-
sation. Then the container is heated in an oven so as to pump
the UF6 in gaseous form in the process installations. During
this operation the material flow is continuously measured.

The empty container is brought back to the reception building
where a second weighing permits exact measurement of the
amount of material taken by EURODIF.

After a leak test, and checking integrity and possible conta-
mination, the container is sent back to the customer. If neces-
sary for long storage, the container can be pressurised with
dry nitrogen.

1.2. The product

The customer supplies the containers for the enriched uranium,
in general standard 30B. The customer's cylinder is not filled
directly from the withdrawal position in the enriching plant,
and special cylinders are used between the process and the
transfer installations.

Both types of cylinders are inspected before and after use for
mechanical integrity, contamination and possible activity.
They are also weighed for quantity when full and when empty.

When all is in order, the 30B cylinder is sent to the customer
with a protective overpack.

1.3. The tails material

The tails material is withdrawn from the enriching plant in
48Y cylinders. These containers are subjected to the same control:
and weighing operations as the cylinders of the other types
of material.

The containers with UF6 tails are stored in anticipation of
transformation into a better form for long storage, probably
uranium oxide.
1.4. Filling up of the installations and evacuation

Special operations are sometimes necessary, mainly for maintenance and repair, and naturally for the initial start-up.

These operations are the filling up or evacuation of parts of the factory. They are done in special installations but the same types of containers and material are used as for the above uranium handling routines.

2. SAFETY CRITERIA

After this description of the EURODIF factory and the routines for handling uranium hexafluoride in containers, we shall now see what safety criteria guided the design of the installations to prevent accidents and to give protection, above all to people, against the dangers of UF6.

2.1. Double fence criterion

In general, all handling cycles or reactors take place in cells which are closed during normal operation. Persons can only have access to these rooms by an intervening door.

This situation involves remote control of the process and so reinforces the safety of personnel.

It was not possible to maintain this criterion in every case for the connecting of containers, but where not, the container valve is always in "twelve o'clock" i.e. in the top position, and such operations are conducted in closed buildings.

2.2. Chemical risk

To avoid the risk of contact between water or moisture and uranium hexafluoride, the containers in the oven are heated by electrically heated air.

This method had a second advantage - it introduces no criticality risk.

2.3. Movement of containers

Particular attention has been given to container manipulations. There are so many movements every day that a strict organisation is necessary to avoid difficulties. The design of the building has been studied so as to eliminate the crossing of the different cycles of the containers. Special systems have been elaborated: overhead cranes, trolleys, gripping mechanisms mainly for the reception building where most of the movements of containers occur.

2.4. Control of containers

For control, we have seen earlier that there are systematic checks for integrity and contamination at the entrance to EURODIF and just before leaving, but there are also contamination checks after most operations where the valve has to be opened.
There are also torque wrenches to measure the opening or closing torque of valves. Where torque is too high, the valve is changed.

There are also pressure tests on full containers (in fact under pressure) to detect any volatile product and avoid problems when connecting them to the plant.

The transport of hot containers with liquid hexafluoride is limited to a strict minimum. For enriched uranium, special cooling rooms have been installed in the buildings, so that removal of liquid enriched uranium hexafluoride outside the buildings is forbidden.

At the connecting stations to the process plant or other equipment, an interlocking system has been devised which blocks the trolley while the connecting pipe is fixed to the container.

2.5. Elimination of human error

Most of the installations are operated by computers and they give alarms on many measures:
- too high temperature
- too high pressure
- detection of a leak of UF6 or hydrofluoric acid and so on ...

2.6. Evacuation ways and alarm

The evacuation ways have been studied so that staff are able to leave any working post and get out in a short time.

There are horn and buzzer alarms and arrows to give the direction of evacuation.

3. REALISATION

By means of slides, we shall show how the criteria described above have been applied to the construction of the EURODIF factory.

Firstly, in regard of the security and safety problems we came to the design of a complex which complements the diffusion cascades. In this design, the main objective is to concentrate here all in and out movements of uranium hexafluoride and to control all active materials which have to pass through the EURODIF installations.

Slides (Figures)

In Figure 1 you see the general drawing of this complex. The "A" building is the Reception and Despatch building. There the containers are weighed and sampled, the empty ones are also leak tested, and the enriched uranium is transferred to the customers cylinders.

In "A", there is a laboratory to check the quality of the process material.

Round buildings A and A' are two hectares of cylinder storage parks, and to the north you see the railway sidings and the installations for unloading containers.

The A and A' zones form the reception area which is open to customers to follow all the checks and analyses of the uranium material.

In the B building we find all the functions directly related to the cascade: the feeding, the extraction of the enriched or tails products, and also the head purification of the process.
From this building, a gallery gives access to the four process buildings and to the control room (in D) from where it is possible to operate all process and auxiliary installations with the help of two computers.

This design has as objective the easy circulation and handling of containers. All activities outside the factory such as deliveries, checks and storage are placed on one side, while on the other side are those related to the process itself.

Figures 2 and 3 give the present progress on the construction of these installations on which we are now running the initial tests.

Figures 4 and 5 show the overhead crane for loading or unloading if cylinders. You will notice the special rocking lever design to improve safety where there is unbalance due to bad distribution of the material in the cylinder.

The next figure (No. 6) gives the type of vehicles used for movements on the parks and to the buildings. You may notice the special hydraulic gripping system. It gives two advantages: the driver can operate it on his own from his seat and there are four fixing points which give good stability during lifting and movement.

The seventh figure is of the empty cylinders test stand. You see the crane installation.

In the Reception building, where quite a lot of container movements occur, handling is by overhead rail cranes (two in each gallery) for movements along the length of the building, and by rail trolleys for movements across. The trailers go into the oven or on to the weighing platforms.

The next figure (No. 8) shows the entrance to the weighing rooms and sampling installations. You see both rail cranes in parallel and the rail trolley which gives access to the working post.

In the ninth figure there is a weighing installation. This room is air conditioned and the platform balances are very accurate, the precision being about 1 kg in 20 tonnes. The picture shows calibrating tests going on; standards of mass were on the platform.

In slide 10 we see the mass metrology laboratory which will be used to verify the accuracy of the mass measures.

Figure 11, 12, 13 and 14 give different views of the oven in which the uranium hexafluoride is heated. When in use, these ovens are closed and present a barrier to any possible escape of vapour in the event of an incident.

Before opening the doors, the atmospheric content is checked, and special cleaning is possible.

The inside is made of all-welded stainless steel, and all the connecting devices are inside the cell.

Round the cells and in the galleries, the floor is covered by a special epoxy resin which gives a leak proof film with a good chemical and mechanical resistance; it is easy to wash.

Figures 15 and 16 show the cells of the process installations. These rooms are gas tight, and are protected by concrete walls calculated to resist the maximum credible accident.
The atmospheric content is monitored to detect quickly the presence of hydrofluoric acid gas.

In Figure 17 you see the intervention zone which gives access to the cells. Checks are very strict on these, and access is only permitted after the shut down of the installations.

The next figure (18) shows the observation window of one of these cells. In this connection, we invite your attention on the problem presented by very large amounts of apparatus and in particular of valves.

A special reference numbering plan has been developed and each valve, for example, carries a plate with its registration number. Many valves, even hand operated ones, have a position indicator with signal to the control room.

The 19th figure gives a view of the control room. The operators run all the installations with the aid of a few terminals; they are helped by two computers.

On the last figure, 20, you see the special computer installation for uranium administration. It will give all the necessary documents, in particular for the EURATOM and IAEA inspections.

CONCLUSION

We have presented some aspects of the handling of uranium hexafluoride in the EURODIF installations, and have tried to show you aspects in regard to safety. In a short time, it was not possible to go into the details nor to show every point, but our goal was to present to you that safety was one of the prime objectives of the designers of EURODIF.

We are also conscious that even with all the safety measures provided, there are still many risks, for a part of the safety comes from human activities.

In this respect, EURODIF gives particular attention to personnel recruitment. All staff have experience in the nuclear energy field, most of them have worked in the PIERRELATTE diffusion plant.

Everybody coming to work in EURODIF undergoes various training schedules, general presentation of the installations, process information, and safety preparation. This occupies about two months on first joining EURODIF.

There is a special section to follow the preparation and the training of personnel.

Finally, EURODIF keeps in touch with other uranium users to get experience on safety, and is receptive to all ideas and suggestions for its improvement.
Safety aspects of Uranium Hexafluoride
Manufacture and Reconversion at Springfield Works
by
A. Price
SAFETY ASPECTS OF URANIUM HEXAFLUORIDE
MANUFACTURE AND RECONVERSION AT SPRINGFIELDS WORKS

A Price
British Nuclear Fuels Limited
Fuel Division, Salwick, Preston, England

Manufacture of UF₆ and its reconversion into solid products has been carried out at Springfields Works for over 25 years. During this period the processes have changed from simple batchwise operations into more sophisticated higher capacity processes. In recognition of the hazardous nature of UF₆, safety has been an important factor at both the design and operational stages. The control system adopted has proved quite successful in limiting the number of incidents. Nevertheless recent doubts as to the adequacy of the procedures adopted has culminated in a safety assessment of the situation and the risks presented in the potentially most hazardous areas have been quantified.
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1. Introduction
2. Uranium Hexafluoride Manufacturing Experience at Springfields Works.
5. Hazards.
8. Conclusion.
1. INTRODUCTION

Uranium Hexafluoride (UF₆) has been manufactured at Springfields Works since 1952 and product and waste UF₆ from the Isotope Enrichment Plants has been converted into uranium dioxide (UO₂) or uranium tetrafluoride (UF₄) for almost as long. To date over 40,000 toles UF₆ has been handled in these processing units.

During this time the processes have changed from simple batch wise operations to the more sophisticated high capacity continuous processes at present in use. Safety has been of prime importance requiring equally intensive attention at the design and operational stages.

The paper describes the processes used at the Works, the type of hazards encountered, safety attitudes adopted, incidents experienced and current safety policy.

2. URANIUM HEXAFLUORIDE MANUFACTURING EXPERIENCE AT SPRINGFIELDS WORKS

Uranium hexafluoride has been produced at Springfields Works since 1952. Until 1964 a batchwise-process was used in which uranium tetrafluoride was fluorinated with chlorine trifluoride, provided by Imperial Chemical Industries Limited, in a stirred reactor equipped with a condensing system for collection and purification of the product.

Two experimental units were constructed in 1951 and production of UF₆ commenced in 1952. Operations continued until these units were shut down permanently, one in 1953 and the other in 1954.

The construction of these two experimental units was followed in 1953 by the installation of a further four units. The new units were of improved design and the condensing system was considerably simplified. A further major improvement was the installation of a fluorocarbon heating and cooling system in place of brine and steam. Production from these units commenced in 1953 and continued until 1955, when they were taken out of service for modification.

All the UF₆ produced during this period was collected and liquified in the condensers and bottled in the liquid state and this method of collection had posed many difficulties. In 1954 a further three units were built. These units differed in several respects from the other units, the most notable feature being the method of collecting the UF₆. This involved the transfer of the gaseous product from the units, across a steam heated pipe bridge to a water cooled cylinder situated in a separate UF₆ filling building. The gaseous method of UF₆ filling proved so successful that the other units were modified to accommodate gaseous filling of cylinders.

Operation of these units continued satisfactorily until 1964, when the process was replaced by a continuous system using fluorine in place of
It was realised at an early date that the direct reaction between fluorine and uranium tetrafluoride in a fluidised solids reactor, although technically more difficult offered the possibility of appreciable economic gain.

A pilot plant constructed in the early 1960's consisted essentially of a monel fluidised solids reactor provided with a filter and condenser system and a fluorine station where liquid fluorine supplied from Imperial Chemical Industries Limited could be gassed into the plant. Following successful operation of the plant it was modified on the grounds of monetary saving to incorporate a fluorine cell installation and so avoid the necessity of transporting fluorine liquid from the fluorine plant operated by Imperial Chemical Industries Limited, at Rocksavage Works, Runcorn.

The main modification was the replacement of the existing liquid fluorine supply system by electrolytic fluorine cells provided with a system for periodically replenishing the cells with anhydrous hydrofluoric acid. Primary regenerative and secondary clean-up absorbers were incorporated in the fluorine feed to the reactor to remove hydrofluoric acid. A recycle compressor was installed in the line to the scrubbers to control the process gas flow through the plant. The latter had the advantage of permitting waste gas recycle to the reactor and was installed as an experimental facility. The steam/brine heating and cooling system was replaced by a fluorocarbon system.

This plant, designated the Prototype Fluidised Solids Uranium Hexafluoride Plant was commissioned in 1964. Its design throughput was 150 tes U/annum, but further additions of fluorine cells later doubled in capacity. A total of 1,100 tes U was produced in the plant before it was closed down.

The plant was used as the basis for design of the large production units now in operation on the Works. These were installed in the building originally used for the production of UF₆ by the uranium tetrafluoride-chlorine trifluoride process. The first commissioned in 1968 has a capacity of 3,000 tes U/annum and the second commissioned in 1974 has a capacity of 6,000 tes U/annum. Liquid cylinder filling incorporating new types of valves and jointing materials was adopted to meet the increased filling rates required.

A total of well over 20,000 tes UF₆ has been produced in these units to date.

3. **URANIUM HEXAFLUORIDE RECONVERSION EXPERIENCE AT SPRINGFIELDS WORKS**

Since the mid 1950's reconversion of both enriched and waste UF₆ into
solid uranic products has been carried out at Springfields Works

On a relatively small scale, two processes housed in the same building reconvert waste material. In one process full UF₆ cylinders (2 tonne capacity) are heated in a steam bath to melt the UF₆ and the UF₆ is allowed to pass in the liquid form via a dip leg into a dissolver containing water to form a uranyl fluoride/hydrofluoric acid solution. The dissolver is fed continuously with water.

In the second process the UF₆ is gassed off from the cylinders at low pressure (0.6 kgs/cm²) using hot water sprays controlled by the UF₆ line pressure, then metered into the feed end of a kiln, where it reacts with steam in the gas phase to give a very active UO₂F₂. The UO₂F₂ produced is converted to ceramic UO₂ in a countercurrent stream of hydrogen and steam as it passes down the kiln.

Several such systems installed in a purpose built building adjacent to the kilns are used in the ceramic uranium oxide powder production plant which provides the oxide for CAGR fuel and export programmes.

The type of cylinders handled are the type 0236 cylinder for deliveries of waste enrichment UF₆ from the UK Diffusion Plant and types 30A and 30B from other sources. A type 0007 container originally used for gaseous filling in the manufacturing plant is used for enriched UF₆ in addition to the standard types 30A and 30B.

4. PRESENT URANIUM HEXAFLUORIDE MANUFACTURING PROCESS

Production of uranium hexafluoride is carried out by reacting uranium tetrafluoride (UF₄) with gaseous fluorine in nitrogen in a fluidised bed of inert calcium fluoride (CaF₂) at around 475°C. External electrical heaters arranged in four zones on the outside of the reactor wall automatically control the bed temperature. As the reaction is exothermic, the heat input requirements to the reactor steadily decrease as the throughput increases until eventually the heat evolved by the reaction can exceed the heat losses. A cooling system is provided which is automatically actuated at this stage. The UF₄ is pneumatically transported from the UF₄ Production Plant, to a large storage hopper which is used as an accounting point for the uranium received. From here the UF₄ is pneumatically transported as required to a feed hopper sited alongside the reactor. A screw feeder at the base of this hopper meters the UF₄ into a larger screw feeder which is designed to feed both UF₄ and CaF₂ and is attached to the reactor.

Additions of small quantities of classified CaF₂ are periodically made to the reactor from the CaF₂ feed hopper also sited adjacent to the reactor.

Solids carried over from the reactor in the main gas stream are retained on one of two primary filters; a backing filter is provided
to cover the eventuality of failure of the primary filter. These filters are sited external to the reactor. The gas stream then passes to a bank of four condensers, one of which is in line as a primary condenser and another as a backing condenser, where the product condenses out. Excess gas passes to atmosphere via cold traps and a wet scrubber system.

The reactor bed is fluidised with a \( \text{F}_2/\text{N}_2 \) gas mixture at sub-atmospheric pressure and the gases are extracted through the reactor filter and condensers by an oil free reciprocating compressor which recycles the gases to the reactor to improve the fluorine efficiency. Two such units are available both of which are supplied with fluorine from the adjacent electrolytic fluorine manufacturing plant.

The uranium hexafluoride which collects in the condensers is transferred in the liquid form either to type 0236 cylinders for the Capenhurst Enrichment Plant or to types 48F or 30A cylinders for the US or USSR Diffusion Plants. Two separate filling stations are provided for this purpose one for the type 0236 cylinders and the other for the types 48F and 30A cylinders. A sampling system for type 48F cylinders is provided in the filling station; systems to enable the hex to be sampled from the transfer lines are also available.

Cooling and heating of the condensers is achieved using a fluorocarbon refrigeration and heating system.

The reactors and filters are installed in one cell and the condensers in another. The oil free reciprocating compressor is installed in a separate cubicle under the condenser cell.

5. HAZARDS

5.1 Basic Properties

At room temperature UF\(_6\) is a colourless, crystalline solid with a significant but low vapour pressure. When heated at atmospheric pressure the crystals sublime without melting and the vapour pressure reaches 760 mm Hg at a temperature of about 56°C. At higher pressures the crystals will melt, at a temperature of about 64°C and this melting is accompanied by a very substantial increase in specific volume.

Uranium hexafluoride is a highly reactive substance. It reacts chemically with water forming soluble reaction products. It reacts with most organic compounds and with many metals. Its reactivity with most saturated fluorocarbons is very low. It does not react with oxygen, nitrogen, or dry air. It is sufficiently inert to aluminium, copper, monel, nickel and aluminium bronze that they can be exposed to UF\(_6\) without excessive corrosion.
5.2 Prime Hazard

The prime hazard in the event of a failure of containment arises from the reaction between UF\textsubscript{6} and moisture, normally present in the atmosphere, according to the equation:

\[ \text{UF}_6 + 2\text{H}_2\text{O} \rightarrow \text{UO}_2\text{F}_2 + \frac{1}{2} \text{HF} \]

With gaseous UF\textsubscript{6}, this reaction proceeds rapidly liberating some heat and generating gaseous hydrogen fluoride (HF) together with a fine aerosol of uranyl fluoride (UO\textsubscript{2}F\textsubscript{2}). Hydrogen fluoride has a much higher vapour pressure than UF\textsubscript{6} and due to the formation of four molecules of HF for each molecule of UF\textsubscript{6}, the reaction is accompanied by a substantial volume increase at atmospheric pressure. The HF is highly corrosive in aqueous solution and has a Threshold Limit Value (TLV) for airborne concentration of 3 ppm or 2.5 mg/m\textsuperscript{3}.

No formal agreement exists regarding the emergency reference level (ERL) to be used for hydrogen fluoride (HF) when analysing the possible effect on the general public of a major release of UF\textsubscript{6}. The figure currently employed for the purpose of defining a hazard range for emergency planning is 5 ppm (4.5 mg/m\textsuperscript{3}).

The UO\textsubscript{2}F\textsubscript{2} produced by the reaction is also very toxic. At high airborne concentrations the fluoride component can produce effects broadly similar to HF but, for a major release, the limiting UO\textsubscript{2}F\textsubscript{2} airborne hazard range is more likely to be set by the cumulative effect of uranium inhalation. At low levels of enrichment the chemical toxicity of uranium exceeds its radio-toxicity; the TLV for soluble uranium compounds is 0.2 mg/m\textsuperscript{3}.

For occupational short period exposures the following inhalation dose limits are recommended:

<table>
<thead>
<tr>
<th>Duration</th>
<th>Uranium concentration (mg/m\textsuperscript{3})</th>
<th>Total Dose (mgm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>10 days</td>
<td>0.2</td>
<td>20</td>
</tr>
<tr>
<td>10 hours</td>
<td>1.0</td>
<td>10</td>
</tr>
<tr>
<td>1 hour</td>
<td>4.0</td>
<td>5</td>
</tr>
<tr>
<td>0.1 hour</td>
<td>6.0</td>
<td>1</td>
</tr>
</tbody>
</table>
No formal agreement exists regarding the ENL to be used for UO$_2$F$_2$. An inhalation dose limit of 4 mg/m$^3$ of uranium is used as a basis for estimating potential hazard ranges.

Deposition of UO$_2$F$_2$ can result in the contamination of all surfaces, including agricultural crops and grassland. The rate at which deposition will occur and hence the contamination contours will be very dependent on atmospheric conditions at the time of a release.

5.3 **Magnitude of Hazard**

It is clear that the sudden release of large quantities of UF$_6$ if windborne could conceivably cause large numbers of casualties. In theory, such an occurrence could in certain weather conditions produce lethal concentrations in places twenty miles from the point of release. The actual number of casualties would depend on the emergency procedures available and the population density.

5.4 **UF$_6$ Manufacturing Plant Hazards**

5.4.1 **UF$_6$ Release due to Containment Failure**

Releases of UF$_6$ are possible in the areas listed below for the reasons specified.

i. **Reactor Rooms**

Failure of vessels, joints, valves, instruments or lines giving vapour release. It is also possible for UF$_6$ to be released in the absorber room of the fluorine plant under non-standard plant conditions but only in the gaseous form. Such releases are unlikely to be other than minor.

ii. **Condenser Rooms**

Failure of vessels, joints, valves, lines or instruments can give either liquid or gaseous release. Overloading of the structure could lead to a condenser falling from its location and producing a liquid or gaseous UF$_6$ leak.

Maloperation of the system could lead to both gaseous and liquid UF$_6$ leaks.

iii. **Transfer Lines**

Failure of lines may take place due to corrosion or mechanical damage. Joints, valves or instruments may fail. These occurrences may lead to gaseous or liquid release. Hydraulic bursting may occur due to trapped UF$_6$. Localised heating of a manifold system to remove a UF$_6$ plug is very hazardous. Liquifying a quantity of UF$_6$ in a restricted space, such as between two solid UF$_6$ sections or between two valves, can
readily result in hydraulic rupture of the system and UF$_6$ release.

iv. Cylinder Filling Operations

Failure of flexible connections and valves, instruments or joints can lead to liquid or gaseous UF$_6$ release. This can be brought about by mechanical impact. Opening of condensers to disconnected filling lines or removal of cylinders whilst still connected, by maloperation, can produce gaseous or liquid UF$_6$ release.

The fracture or removal of a valve on a cylinder by impact or mishandling could lead to gaseous release. Maintenance operations on filling lines whilst filling is in progress or disconnection of a cylinder when not shut-off due to maloperation or valve misfunction could lead to gaseous or liquid release.

A cylinder may be overfilled by completely filling with liquid. If the cylinder is heated after overfilling, it may be hydraulically ruptured. The unscrewing of the Superior valve on type 48F and type 30A cylinders could lead to gas release.

The movement of cylinders in particular the type 48F containing liquid using a single point lift could lead to instability and the cylinder falling, rupturing and discharging its contents.

Insufficient cooling before maintenance of a cylinder or line may cause problems. If the valve is removed too soon after filling type 0236 cylinders prior to fitting of the blank, a gaseous hex release may occur.

The use of a damaged cylinder may produce problems during filling operations.

v. Liquid Sampling of Type 48F Cylinders

Liquid hex is hot, pressurised and very mobile. Movements of partially filled cylinders results in surges of the dense liquid which can upset handling equipment and cause loss of control.

The fill limits for cylinders are established for a maximum temperature of 121°C. Heating cylinders above this temperature may result in liquid expansion to a volume exceeding that of the cylinder, which could result in hydraulic rupture. An overfilled cylinder could rupture on reheating.
The removal of, or damage to a cylinder valve, the failure of connections to the pressure gauge or sampling rig due to component failure or mechanical damage or the breaking of connections whilst still open to the cylinder, could all lead to gaseous or liquid UF₆ release.

The rotation of a cylinder whilst still connected to the sampling rig with the cylinder valve open or damage to the cylinder or connections from other cylinder movements could lead to discharge.

The magnitude of these releases can vary according to the conditions applying at the time.

5.4.2 UF₆ Release due to Fire or Explosion

UF₆ is not of itself inflammable but if a container of the material were present in a fire, the container would explode by virtue of the internal stresses built up in the container and deluge its contents over a wide area.

The presence of oil or other impurities in a cylinder could lead to release of UF₆ by explosion.

5.5 UF₆ Reconversion Plants Hazards

Releases of UF₆ could occur due to the occurrences specified below:-

i. Failure of a cylinder on heating due to hydraulic burst if the cylinder were overfilled.

ii. Failure of joints on the liquid or gas lines.

iii. Failure of the liquid or gas lines through corrosion or mechanical damage.

iv. Local overheating of the liquid or gas lines by electric heating systems.

v. Failure of valves on the liquid or gas lines.

vi. Failure of instruments, notably pressure gauges.

vii. Leaks from the Type 0236 cylinder whilst in transit within the building.

viii. Leaks at the gun or at the dissolver, the latter especially if the dissolver were empty.

ix. A UF₆ cylinder being involved in a fire.
6. DESIGN AND OPERATIONAL SAFETY PRINCIPLES AND FEATURES

6.1 Construction Philosophy

The plant is constructed in an area of low seismic activity. The possibility of missiles both natural and man-made and of aircraft impinging upon the structure has been rejected as being too remote. Hazards to other workers on site associated with the processing of chemical and radioactive material are reduced by housing the plant in a standard brick/steelwork/concrete building. Process effluent gases are monitored and discharged to atmosphere via a 60' stack.

The inhalation and ingestion hazard presented by uranyl fluoride and HF is controlled by containing the materials within vessels which for the most part are located within cells. Back diffusion of gas is prevented by establishing pressure gradients between the cells and the working areas with the extraction and plenum systems. As far as practical all major penetrations through the cells are sealed but where this is impossible a linear air flow of 200 ft/min is aimed at through the gap into the cell.

The concrete cells therefore provide secondary containment against the spread of airborne contamination from the vessels. The floors of the cells are in concrete, no drainage channels are provided in the floors policy being to retain material within the cell.

The two UF$_6$ filling stations are constructed external to the main plant; one for type 48F cylinders and the other for type 0236 cylinders. The type 48F filling station has two filling points and a steam heating bath for sampling these cylinders. The UF$_6$ feed pipes and associated ancillary lines pass through the main wall of the plant into the type 0236 cylinder filling station and via a lagged line to the type 48F cylinder filling station. The two major hazard areas are therefore isolated making the problem of introducing more rigid operational safeguards easier. Containment from the outside atmosphere is by a roller door or concertina doors.

The necessity of adopting the design safety principle of double primary containment of UF$_6$ lines in the Springfields Plants has been debated at some length. Where the lines pass through cell areas and ducts it was not considered necessary. In other areas namely,

i. Where liquid UF$_6$ lines run outside the confines of a cell,

ii. The coupling lines to the cylinders in the two filling stations,

iii. The sampling line,
iv. The section of line between the condensers and the liquid runoff valve,
either this principle has been adopted or people working in the area have been provided with protective equipment and closer supervision maintained.

Mild steel is used for vessels and pipework where the temperature of the process gaseous is below 100°C. Above this, monel is used. More exotic materials are used for valves and instruments in some instances. Special consideration has been given to the mechanical design of process equipment to ensure leakproof construction. Vessels are of all welded construction with all containment welds fully radiographed. Argon arc welding is used throughout and weld details are to the approved standards. The vessels and heat exchangers have been mechanically tested to a suitable pressure. Instrumentation is designed to offer high standards of containment to the UF₆.

Valves have in general been designed specifically for their compatibility and operational resilience in the reaction gas environment. One exception is the superior valve whose use on type 30 and 48 cylinders is mandatory for the US enrichment plants.

The Superior cylinder valve is considered to be a somewhat unpredictable valve operationally; it does not back seal, it can be screwed out of the cylinder and the spindle can be screwed out of the body and special operational safeguards are necessary in its use.

In general, the attitude has been to construct plant to high standards with little degree of duplication and to accept a low probability of failure which would require maintenance after decontamination.

6.2 Input and Extraction Systems

A plenum system delivers air to the control corridor at a rate of approximately 3000 cfm. This is intended to maintain the area at a slightly higher pressure than the cell areas and reduce the likelihood of UF₆ penetrating the cell walls into the working areas.

Extraction is provided from the reactor room, condenser rooms and compressor rooms and the gases are discharged to atmosphere via a 45' stack. Separate extraction systems serve the two filling stations and cylinder cleaning facilities. Duplication and notification of failure is provided.

6.3 UF₆ Cylinders

i. Types 48 and 30

These cylinders are manufactured to the specified drawing,
the safe working pressure is 200 psig. The vessels are ASME code approved regarding material of construction weld fabrication details etc. Only one valve and one plug are installed to minimise points of leakage.

The actual water capacity of the cylinder is measured following manufacture and is stamped on the cylinder. Cylinders are provided with a plate giving the volume details. Examinations carried out on the cylinders comprise a four-yearly hydraulic test and intermediate examinations; the hydraulic test pressure is 500 psig.

A UF₆ cylinder is removed from service (for repair or replacement) when it is found to have leaks, excessive corrosion, cracks, bulges, dents, gouges, defective valves, damaged stiffening rings or skirts, or other conditions which, in the judgement of the examiner, render it unsafe or unserviceable in its existing condition.

Repairs or alterations to parts subject to pressure are followed by a hydrostatic strength test and plug or valve replacements are followed by air-leak tests, when possible. Repairs to structural attachments are not followed by pressure or leak tests of the cylinder unless repair of torn or deformed areas of pressure-containing materials are involved. The cleanliness of UF₆ cylinders is of serious concern since the reaction of UF₆ with hydrocarbon oils and some other impurities is quite vigorous and can result in serious explosions. The inside of new cylinders is therefore thoroughly cleaned of all grease, oil, scale, slag, oxides, dirt, moisture and other foreign matter. The surfaces are left clean, dry and free of all contamination but not chemically passivated.

ii. Type 0236 Cylinders

The manufacturing drawing was first issued in April 1953. A standard specification AESS(E)34/22700/7 was first issued in 1968 and this has subsequently been replaced by AESS(E)34/39900/7.

The specification calls for thorough internal drying and degreasing, after hydrotest and before vacuum testing, but no special steps are taken to passivate the internal surfaces. A testing and examination procedure is carried out. The cylinder volume has been checked in a number of cases and found to be 636 ± 30 litres.

6.4 UF₆ Manufacturing Cycle

The reactor, gas filtering, condensing and waste gas recycling system relies on a partial vacuum in the system to draw fluorine
and recycle gases through the reactor bed. If a major leak occurred in the system air would enter, the pressure would rise and the process stop. Under these conditions it is not possible to postulate a significant UF₆ release.

The maximum theoretical temperature of the fluorocarbon heating medium used to heat the condenser is 141°C assuming the relief valve on the boiler fails to operate at its setting of 10 kg/cm². If the UF₆ in the condenser contains 5% HF, the pressure could be 300 psi, i.e. in excess of the design figure for the condenser.

An instrumentation system, separate from the process control instrumentation, automatically shutting off the steam supply to the fluorocarbon boiler in the eventuality of a high condenser pressure has therefore been provided.

Calculations carried out on the possible reaction between the fluorocarbon heating and cooling medium and liquid UF₆ according to the equation

\[ 2 \text{CFC}_3 + \text{UF}_6 \rightarrow 2 \text{CF}_2\text{Cl}_2 + \text{UF}_4 + \text{Cl}_2 \]

indicate that should a leak occur during the heating cycle the pressure release system should prevent an overdesign pressure developing in the condenser.

A weighing system on the UF₆ feed hopper provides a check on the UF₄ fed to the reactor and hence the UF₆ charged to the condenser and this was considered to be sufficient to prevent condenser overloading.

The problem of operator error in perhaps switching a hot condenser on line has been recognised and an interlock provided.

6.5 UF₆ Filling and Sampling

The major hazard areas are those of uranium hexafluoride filling and sampling. The type 48F cylinder filling station was originally built to accommodate two filling points for these cylinders. The station was later extended to provide for a sampling system for type 48F cylinders. The hazardous nature of the hex sampling operation and the filling operation was recognised and whilst the necessity to provide sampling equipment quickly meant that a system ideal from the design safety stand point was not possible it was felt that close operational control could compensate for a lack of design sophistication.

Filling connections to type 48F cylinders are through stainless
steel flexible hoses. The wall thickness of these hoses is 0.015" and although they are protected by overbraiding, they are susceptible to mechanical damage. The flexibles are checked or tested before putting into service and during service. An interlock system is provided which prevents UF$_6$ filling commencing unless a key switch is activated. The key is kept in the possession of the Plant Supervisor. Interlocks have been fitted to the sampling system so that power to the rotation motor is not available unless all the correct trap doors are in closed position. The system is rigidly supported so that the sampling line cannot break due to movement.

6.6 **Operational Techniques**

The variations of density, vapour pressure and physical state with temperature of UF$_6$ require the development and use of safe handling procedures. Pressure testing of equipment is always carried out following maintenance or installation.

Overfilling of the condenser is prevented by closely monitoring the UF$_4$ feed and relying on the fact that the pressure drop across a condenser will increase to a level too high to permit the plant to be operated before it becomes filled to an unsafe level.

Overfilling of cylinders is prevented by adhering to weight limitations and an alarm sounds at a preset figure. If slight overfilling occurs, the cylinder is connected to an evacuation system and the excess UF$_6$ removed, without heating the cylinder, until the correct fill limit is obtained.

It was realised that leaks in a cylinder containing liquid UF$_6$ would be difficult to control. Caution is exercised in handling cylinders until the contents have solidified.

Any hex cylinder which has been subjected to heating is not opened to the atmosphere until it has been stored for a period sufficient to allow the whole of the contents to reach ambient temperature.

The following, more specific, operational precautions are taken to minimise the chances of hex release:

i. A 48F cylinder is only lifted to a height of < 1' directly after filling when moving off the weighscales. 36 hours are allowed to elapse before the cylinder is lifted out of the filling station.

ii. A type 48F cylinder is left for 8 hours after removing the top of the sampling steam bath before it is moved and then it only moves 2' above floor level.
iii. Liquid sampling is carried out by the Analytical Services Department. The connection of the sampling rigs to the liquifier and the rotation of the cylinder is carried out by Engineering Department and Production Department under plant clearance.

The fuses for the drive motor are only put in when the cylinder requires rotation and is under the control of the Plant Supervisor.

Supervision of the sampling operations is always carried out by the Plant Supervisor.

iv. The cylinder filling operation is only commenced after inspection of the system by the Plant Supervisor who checks that the system has been assembled correctly and pressure tested. He then inserts the key in the interlock switch to enable the filling operation to be activated.

v. Personnel wearing appropriate protective clothing, breaking flanges on vessels containing UF₆ first loosen the nuts and bolts and "crack" the joint. If a persistent substantial leak occurs, the bolts and nuts are re-tightened at once and cooling or purging is carried out before continuing.

6.7 Cylinder Cooling Time prior to Devalving or Maintenance

The temperature inside type 48F and type 0236 or 30A cylinders during filling with liquid UF₆ can be as high as 105°C. During sampling of the type 48F cylinders, the temperature is controlled at 95 - 100°C.

Operations which rely for their safety on cooling these cylinders adequately are:-

i. Removal of the valve adaptor from a type 0236 cylinder to fit the blank flange for transit.

ii. Valve removal, due to some malfunction, from types 48F and 30A cylinders after filling or heating for sampling.

It has been calculated that it takes 5½ days for 95% of the UF₆ in a type 0236 cylinder to solidify. The behaviour of the mixed phase in a cylinder cannot be predicted with certainty. It is possible that the behaviour of two cylinders cooled for the same time in this situation may be quite different on opening to the atmosphere. Adhesive notices labelled "not to be devalved until ......." are fitted to the cylinder by the Plant Supervisor immediately after filling. The alternative of fitting a castel lock system was rejected as it was considered unsuitable.

It has been calculated that the time taken for the contents of a type 48F cylinder to solidify is 10.5 days with natural cooling with no containment and 8.5 days with water cooling. An isolated
cooling curve plotted on a type 48F cylinder after a minor incident elsewhere showed that the theoretical times are not optimistic. Only one type 48F cylinder has so far been subjected to valve replacement at Springfields and this was carried out after 5 days in accordance with accepted US practice. No significant UF₆ release was experienced. This however may well have been fortuitous in view of the unknown mixed phase UF₆ behaviour. A time of 10 days natural cooling is now adopted before maintenance.

6.8 UF₆ Reconversion

Design and operational safety principles and features concerning containment and materials of construction are similar to these for the UF₆ Manufacturing Plant. Thus the hex liquification and vaporisation equipment is installed in a purpose built extracted area. Containment comprises the pipework system and a heating box for the line. A leak in the baths would be detected by a conductivity meter in the condensate outlet line. A positive reading on the meter diverts the condensate to recovery, the steam supply to the bath is replaced by cold water and the operator is informed by an audible alarm.

Where liquid UF₆ is transferred from a cylinder to a dissolver:

i. The UF₆ cylinder feed lines and dissolver are segregated from the rest of the plant in a fully contained and extracted compartment.

ii. The UF₆ cylinder pressure is monitored by an alarm. Should the pressure rise above 3 kgs/cm² the steam supply is switched off and cooling water put on the cylinder.

6.9 Fire and Explosion

The possibility of a UF₆ cylinder becoming involved in a fire in the plants appears remote.

There is not a great deal of combustible material in the areas and the gas driven truck used for movement of type 0236 cylinders does not contain enough fuel to reach the temperature necessary for a UF₆ cylinder burst. It is true that the oil cooled transformer/rectifiers in the fluorine plant present a hazard (there is a 10⁻³ probability of an oil-cooled/rectifier catching fire) but UF₆ cylinders are not stored near this facility. Standard fire alarm systems and evacuation procedures are provided for and the Works Fire Brigade would be on hand before a fire of any magnitude could develop.

The danger of an oil or petrol tanker crashing into a type 48F cylinder at the loading point on the east side of the plant or
during transit to a storage area, and getting involved in a fire is significant. However the loading road is closed to other traffic, and transport of inflammmable materials within the factory only takes place along well defined routes specified by the Transport Manager.

Should a fire occur appropriate action is specified to ensure that the plant is shutdown safely. The fire alarm circuitry has a battery backing system should the mains electricity fail and this should be adequate to give the necessary warning so that a UF$_6$ release due to a hurried shutdown does not occur.

6.10 **Alarm Evacuation and Emergency Systems**

Portable equipment is installed in specified high risk areas to detect high atmospheric contamination. A fire alarm system and manual fire fighting equipment afford protection in the event of fire or explosion. In the UF$_6$ manufacturing plant at ground floor level several exit doors are available and there are two at first floor level. Fire doors are installed to contain fires. The reconversion plants are in single storey areas with adequate exit facilities. A separate system of emergency lighting for safe evacuation of personnel is installed, which comes on automatically should the mains supply fail. Batteries are designed to maintain the supply for one hour. 24 hours are needed to recharge the batteries.

Separate emergency electrical supplies are also provided for the fire alarms. An emergency procedure is available to cover the eventuality of a UF$_6$ release. The system provides for:

i. remote safe shutdown of the process by manual action from outside the area.

ii. safe means of evacuation by the personnel.

iii. warnings at the entrances to the affected areas.

6.11 **Lifting Equipment**

The surging of liquid in partially filled cylinders and the eccentric centre of gravity of cooled cylinders has been recognised. The lifting equipment used in the type 48F filling station has been specially adapted for this type of work; a special adaptor for the type 48F cylinders designed to enable the cylinder to be lifted safely with connections at four points, has been provided. An interlock system is provided so that a cylinder cannot be moved out of the plant without the door being open. A fork lift truck and stillages are used for the type 0236 cylinders. Recent studies have indicated that it would be better
to use a two-point lifting arrangement with the lifting points spanning the centre of gravity at all times when handling liquid filled type 48F cylinders and steps are in hand to implement this.

6.12 Plant Operational Philosophy

Plant rules, operating instructions and a plant manual are provided. Emergency instructions to cover the eventuality of leaks of UF$_6$ are available. Reliance is based primarily on local training for the plant operators.

6.13 Failure of Services

Clear specification of what is required should a steam or electricity failure occur, is made since both these have obvious safety connotations. In the event of a steam failure, particular attention is necessary to traced lines to ensure that draining of liquid UF$_6$ is complete. Twin supplies have been provided from separate steam and electric mains.

7. ADEQUACY OF DESIGN AND OPERATIONAL SAFETY PRINCIPLES AND FEATURES.

7.1 Incidents Experienced

Judged solely by the number of incidents which have been experienced on site the design and operational safety principles and features have been satisfactory. The incidents which have occurred have been of a minor nature. In the early days of UF$_6$ manufacture during the 1950's leaks were commonplace but the small scale of operations (batches were only approximately 130 kgs U) ruled out major leakage. Leaks from thermocouple points in the vapour phase were fairly commonplace but these were contained in the cell. Since these early days there have been only six releases deemed of sufficient importance to warrant formal local investigation and these are detailed below:

i. A release of approximately 20 kg UF$_6$ occurred in the Experimental Fluid Bed UF$_6$ Plant, due to the failure of two bursting discs in a vent line from a condenser to a scrubber system when approximately 200 kg UF$_6$ passed to scrubber. The incident was caused by the uncontrolled application of steam heating to the condenser in direct contravention of operating instructions. The scrubber system became over-loaded and 20 kgs unabsorbed UF$_6$ passed to atmosphere via the scrubber stack. The incident caused no injury to personnel or equipment. (1962)

ii. A release of approximately 13 kg UF$_6$ occurred in the Low Enriched Reconversion Plant when a leak from a UF$_6$
cylinder occurred in a copper line connecting joint. The leak was contained by cooling the cylinder with solid carbon dioxide and no fumes entered the plant area. There was no injury to personnel but an abnormal white plume was observed to be emitted from the Building exhaust stack causing temporary contamination in the vicinity of the outside area. (1963)

iii. A release involving 50 kgs (maximum) of $^{235}$U enriched UF$_6$ occurred in the Enriched Oxide Powder Production Plant. The cause was the breaking of a flange joint, prior to replacement of a faulty pressure gauge, on a line used to feed UF$_6$ from a heated cylinder to the Pyrohydrolysis Reactor. Unfortunately neither the cylinder valve nor the line valves had been closed. The leak lasted for approximately 20 minutes but there was no injury to personnel or damage to equipment. (1969)

iv. During construction of the second UF$_6$ manufacturing unit the steam supply to sections of trace heating on the first unit was inadvertently turned off whilst liquid UF$_6$ was being transformed to a cylinder. The UF$_6$ solidified and a hydraulic pressure split of the line occurred on subsequent reheating releasing approximately 20 kg UF$_6$. There was no injury to personnel (1969)

v. A release of approximately 15 kgs UF$_6$ occurred at the Prototype UF$_6$ Production Plant when a joint failed on the gaseous transfer line to a type 0236 cylinder. There was no injury to personnel (1969)

vi. Two type 0236 cylinders containing 2294 and 2404 kgs UF$_6$ bulged and cracked at a weld whilst being heated in a water bath in the UF$_6$ Production Plant to remove excess material. The release of UF$_6$ was about 5 kgs. There was no injury to personnel. (1969)

7.2 Safety Assessments

As the scale of operations increased doubts were expressed on site regarding the adequacy of the safety precautions and in 1976 on the instruction of the General Manager a Safety Assessment Group of five experienced technologists comprising a chemist, a plant manager, a mechanical engineer, an instrument engineer and a group leader with safety assessment experience were assigned for a period of six weeks to examine critically the UF$_6$ handling procedures on site. Over 150 recommendations for improvement were suggested and these were discussed at senior level by a committee chaired by the Deputy General Manager.
7.3 On Site and Off Site Hazards Associated with the Accidental Release of UF₆

One recommendation of the Safety Assessment team was that a more detailed study of the hazards presented by possible large scale releases of UF₆ to workers on the site and to the public be investigated, the possible consequences of such escapes be examined and the need for a full scale Site Emergency Scheme assessed.

The approach adopted was to identify the worst possible liquid UF₆ discharge and the worst possible gaseous UF₆ discharge accidents. A study of the hazards to personnel arising from an accident involving a site release of uranium hexafluoride requires:

i. A study of conditions that could give rise to a release.

ii. An estimate of the extent and duration of the release.

iii. A determination of the concentration profile with distance in relation to the Emergency Reference Level (ERL) pertaining to the chemicals involved.

It has been noted earlier that an instantaneous release due to earthquake or air crash on cylinders or equipment containing liquid UF₆ at the Springfields Works is so improbable as to be disregarded. In addition the risk of major fire conflagration within the hex manufacturing or reconversion plant was investigated and a similar conclusion reached. The condition that could conceivably give rise to significant UF₆ losses was therefore concentrated on a study related to plant operations in the plants.

In the UF₆ manufacturing plant the typical quantity of liquid UF₆ that could give rise to a hazard (solid hexafluoride does not present a hazard of comparable magnitude) is 12 tonnes, the weight that is charged into type 48F cylinders. In the other plants the uranium is converted into other compounds and exists in liquid form in amounts not exceeding 2 tonnes UF₆ and only in one case is it used in liquid form. For the first stage of the Safety Study the implications of losses from the Hex Manufacturing plant, handling 12 tonnes lots of UF₆ was analysed. The analysis investigated any incident from three standpoints:

i. The radioactive hazards due to particles arising from U₂₃₈, U₂₃⁵, U₂₃⁴.

ii. The toxic hazard due to ingestion of uranyl fluoride.

iii. The toxic hazard due to ingestion of hydrogen fluoride arising from the reaction UF₆ + 2H₂O → UO₂F₂ + 4HF, which takes place when UF₆ meets a moist atmosphere.
7.3.1 The Largest Potential Release of Liquid UF₆

Discounting catastrophic fracture of a type 48F cylinder the worst conceivable accident is the fracture of a line carrying liquid UF₆ or the breaking off of the \( \frac{7}{8''} \) superior valve due to an accidentally applied physical force with the valve open to liquid UF₆. Such an accident is possible at the liquid hex sampling stage. Discharges of similar magnitude could also arise from the condenser system where uranium hexafluoride is held as liquid prior to run down to the cylinder filling area.

A quantitative probability analysis based on the reliability of the equipment involved and the operational procedures followed indicates a probability of such an occurrence of approximately \( 10^{-8} \). - \( 10^{-7} \).

7.3.2 The Largest Potential Release of Gaseous UF₆

During a cylinder filling operation liquid uranium hexafluoride is flowing from condenser to cylinder over a period of about 6 hours (minimum time about 4 hours). Any fracture of the run down line would result in a liquid release from the condenser and a simultaneous gaseous release from the cylinder being filled. The former would approximate to the conditions mentioned above if the fracture were close to the run down valve. A fracture in the cylinder filling station would correspond to a gas release condition since the condenser valve could in this event be shut off. If the incident happened when the cylinder was full, this would represent the worst situation involving a gas release and it is this situation that has been analysed. A similar probability analysis to 7.3.1 indicates a comparably low probability of occurrence (\( 10^{-7} \)).

7.3.3 Analysis of UF₆ Release Rates

Calculations were carried out to ascertain both the quantity of UF₆ that would be released to atmosphere and the rate at which it would be discharged from the cylinder for both the above cases. It was assumed that at the start of the incident the cylinder contained 12 tonnes UF₆ and the contents of the cylinder were at 100°C in conformity with normal plant operation. The calculations deduced the discharge rates for liquid discharge for \( \frac{7}{8''} \) holes. (The assumption that the temperature is 100°C for its vapour release is very pessimistic as the temperature in the cylinder falls during transfer).

It was concluded that in the case of the liquid release the full quantity could be discharged and approximately 7 tes would be converted to vapour at an average release rate of 250 kg/min. In the case of the gas release the release rate
would be much smaller (of the order of 20 kg/min) and the total release would be around 7 to UF₆, the release occurring in three stages viz:-

i. A high pressure discharge due to the superheat of the liquid, with the initial leak rate being approximately 56 kg/min.

ii. A steady release over a period of approximately 3½ hours, while liquid is freezing to solid at around 18 kg/min.

iii. A slow rate of release of 11 kg/min as the solid cools from 64°C to 57°C at which point the vapour pressure reaches atmospheric.

The velocity of discharge has been calculated at 200 miles/hour on initiation of the incident (this is about the sonic velocity).

7.3.4 Hazard Analysis

The toxic, radiation and HF hazards arising from the same situation were estimated for Category F weather conditions associated with a wind speed of 2 M/sec and ground level release using the standard Pasquill curves. These gave ERL fatal and distress ranges of 1 KM, 3 KM and 13 KM respectively for HF, an ERL of 9 KM for toxicity of UO₂F₂ and 800M for radiation due to inhalation. These represent the average type of weather conditions experienced and would be exasperated in unfavourable conditions.

The modifications necessary to existing emergency procedures are currently being reassessed in the light of these results.

8. CONCLUSION

At Springfields Works the safety aspects of toxic gas handling are under constant scrutiny and improvement. By this policy it is hoped to avoid a major release of such material into the environment. Legislation, following the deliberations of the Advisory Committee on Major Hazards, expected within the next year will provide further opportunities for re-examination of accepted procedures.
UF₆ handling at the
URENCO Nederland centrifuge plants, Almelo
by
F. Stockscläder, J. Christofzik
UF6 HANDLING AT THE
URENCO NEDERLAND CENTRIFUGE PLANTS, ALMELO

by F.A. Stockschläder and J. Christofzik
Urenco Nederland Operations B.V., Almelo

Abstract

In each of the three Urenco centrifuge uranium enrichment plants at Almelo, UF6 under pressure is only handled in enclosed areas which are kept slightly below atmospheric pressure. The air in the plants is continuously monitored and, if a UF6 release occurs, is automatically diverted through a scrubber system.

Further, automatic quick closing valves in the pipework are so arranged that, in a maximum credible accident, only 50 kg liquid UF6 could escape in the enclosed area.

Finally, containers used for transporting UF6 can only be disconnected from the plant under safe conditions as verified by a cross-checking procedure.

The anyhow small number of UF6 releases of trivial quantities since operation began could be significantly reduced still further relative to the increasing UF6-throughput in the Almelo plants.
Introduction

Urenco Nederland has three centrifuge enrichment plants. These comprise the two pilot plants, the Dutch SP1 and the German SP2 (SP = separation plant) with a capacity of 20 t SW/a and 24 t SW/a respectively, and the commonly built industrial scale production plant SP3 with a capacity of 200 t SW/a (see Table 1 for details). The two pilot plants have been fully operational since end 1975 and early 1976 respectively, and the industrial plant, now in course of completion, has reached about 60% of its final capacity.

Enrichment is effected by UF6-gas centrifuges arranged in cascades, these being the smallest operational sub-unit of a plant capable of fully completing an enrichment task. Total plant output is accordingly obtained by connecting a sufficient number of cascades in parallel.

UF6 handling process

Natural UF6 (called "Feed") is transported to the plants in 48" or 30" containers and passed direct from these into the process system. Depleted UF6 ("Tails") or enriched UF6 ("Product") is drawn off from the system into 48" or 30" containers conforming to ORO 651.

The principle of the internal UF6 flow scheme in the plants is shown in Figure 1.

In order to introduce UF6 into the process, the feed container is heated to about 70°C in an autoclave. The contents of the container are liquefied and the gas phase above the liquid, which is extracted as feed, reaches a pressure of about 2 bar inside the container. The gas then flows through pressure reduction stations, situated behind the autoclaves, at low pressure into the cascades, where the process gas pressure is then only a few millibars.

The two gas streams leaving the cascades as enriched and depleted material are collected in chilled (-70°C) desublimers as a solid. When a desublimer is full, its contents are liquified by heating (+70°C) and drawn off straight into a transport container.

Except for the loading, transport and storage of containers, all handling of filled containers takes place in enclosed areas of the plant.

Safety aspects of the UF6 systems

UF6 at above atmospheric pressure is present only in the feed system and desublimer/take-off areas. Thus, only in these areas do the conditions exist for a possible UF6 release. Further, only here does container handling (connection and disconnection) take place. In all other areas, UF6 is present as a solid or at very low pressure.

To limit and control possible UF6 releases, the above mentioned systems and working areas have been located together in a special area, (Zone I), in each separation plant. To prevent any UF6 released spreading beyond Zone I accommodation, the area is provided with its own ventilation system. This keeps the air at a pressure which is about 10 Pa lower than that of adjoining areas and effects a frequent change of air (~5/h).

1) ORO 651 Uranium hexafluoride: Handling procedures and container criteria; US Dept. of Commerce, April 1977
Exhaust air passed to atmosphere from here is continuously monitored at several points and, in the event of an incident (e.g. increased uranium activity), automatically diverted to a retaining system which filters out air pollution by uranium and the consequently arising HF.

The limit for the cleanliness of the exhaust air from this area is a yearly average rate of airborne activity of $10^{-13}$ Ci/m$^3$ admissible under the existing licence. For comparison, the ICRP-value for uranium in inhaled air is $\text{mpc} = 3 \times 10^{-11}$ Ci/m$^3$ for an employee working in the radiological field 168 hours a week.

Due to the corrosive properties of UF6 pipework and valves consist of UF6 resistant materials such as stainless steel or aluminium. All connexions are welded - unless the process requires otherwise.

Where connexions have to be undone (flanges), either metal packing or high fluorinated heat resistant soft material packing is used. All welded connexions in the pressure and liquid areas are subjected to 100% X-ray examination, and a system is only passed for operational use after the X-ray photographs have been expertly examined by specialists. A single leak may not exceed a value of 10 Torr 1/sec. That requires the application of strict standards in testing systems for tightness, and the test pressure exceeds the normally expected working pressure by a factor of 4-5. The feed container is located in an autoclave which is subject to routine checks periodically.

Process material pipework systems and container transport and handling systems are located in relation to each other in such a way that accidental damage to pipework cannot occur. For instance, transport containers are automatically locked in position during filling and can only be removed when the valves in the take-off piping have been closed. Process operating conditions (temperature, pressure) are monitored and regulated automatically by multiple controls, and where maximum values are exceeded, actions are again taken automatically (e.g. heating switched off). Important elements of the automatic control and safety systems are the automatic valves.

The UF6 pipework in the pressure and liquid areas is separated into well-defined sections, as regards the volume contained, with automatic quick-closing valves so that in the event of a fault they shut off automatically and thus enclose small quantities. This measure allowed the definition of a maximum credible accident as being the release of 50 kg UF6 in the production plant in the event of a failure.

**UF6 releases**

Since operation on site began in 1972, some 430 t U have been put through the three enrichment plants. This corresponds to 350 to 500 container manipulations and about 1500 discharges of the desublimers.

During the whole of this period of operation, 19 releases of UF6, including all minor incidents, have been recorded, and in every case the amounts released were:

- in the order of tenths of grams
- in gaseous form, and
- fully dealt with by the ventilation system installed, i.e. none spread beyond the area of Zone 1.
A further characteristic was that all releases occurred while operational work was being undertaken: there have been no spontaneous releases.

Of the 19 releases, 18 took place in the pilot plants and only one has occurred in the industrial plant. The causes of the releases were chiefly technical failures. In 13 cases, the release came from small leaks in the valve flange connexions or valve packing of liquid piping. Expecially in the pilot plants, flange connexions had been given preference since lack of experience with valves in the liquid piping suggested that replaceable valves were desirable.

The leaks arose mainly through the failure of two components of the flange connexion:
- the packing material, Teflon, was unfit for the constant effect of surface pressure and temperature;
- in some other cases the tensile strength of the flange bolts had become inadequate. These were replaced and metal packing was introduced.

As a result of these experiences, welded connexions were given widespread preference in the industrial plant.

In five other cases, release of UF6 was the result of improper working methods when disconnecting cold traps or sampling flasks. Such cases are classified as "Maloperation". The release of UF6 almost always occurred because residual gaseous UF6 in the flange connexion was not correctly removed before the flange was unbolted, or because the quantity of the residual UF6 was higher than estimated. The latter was in fact the case for the only release so far experienced in the industrial plant where a desublimer was opened up which was assumed to be completely free of UF6 on the grounds of earlier action.

In 12 of the cases mentioned, incorporation by employees of minor quantities of uranium could not be excluded, and this was confirmed for 5 releases where the notifiable limit of 50 μg U per litre of urine was exceeded; 7 of the staff were affected. In all other cases of suspected incorporation, the quantity was found to have been below 10 μg U/litre.

For comparison, ICRP quotes a maximum inhalation value of 2.5 mg U per day. With the passing of 11 urine a day and 80% of the inhaled quantity, 50 μg U/litre corresponds to 2.5% of the above ICRP value.

Figure 2 gives the distribution of UF6 releases since operation began (in six monthly periods). The lower diagram shows the recorded releases, including all minor incidents; the middle diagram, such cases where a urine sample was taken to check for possible incorporation, and the upper diagram cases of notifiable incorporation. The distribution of incidents over operation time appears to be regular, but a sharp drastic relative decrease of incidents due to increased UF6 throughput could be found (Figs. 3a and 3b).

Operating procedures

Apart from the above mentioned positive measures to reduce the probability of a release, special regulations, works directions, and working procedures have been set out for the safety of container handling and other operations.
All work is carried out against check lists or specific task approvals which, in particular, also detail the safety measures. These details are so drawn up that a task can at any time be passed on to others. All work is undertaken by at least two persons so that it is cross-checked.

Important points on working procedures are given below:

1. Container handling only occurs when the container is at its safest, i.e. transport and connecting and disconnecting are only carried out when the UF6 contents are proved to be solid and there is sub-atmospheric pressure in the container. Containers under pressure are neither handled nor are they readily accessible since they (feed and product containers) are in autoclaves or heating chambers.

2. Before release of connexions, UF6 is removed and the system is several times cleared through with gaseous nitrogen. Before connexions are released or made, tests (leak, pressure etc. tests) are carried out to make sure that the next step is permitted.

3. Handling operations in the above named areas are carried out where the following equipment is on the spot:
   - suction hose directly connected to the air cleaning system; this is used to draw off any gaseous UF6 or HF set free during the release of connexions;
   - a counter to monitor activity;
   - protective breathing apparatus which is kept ready in a safe place nearby (or frog suits); breathing apparatus is worn at the start of work where the release of UF6 is expected;
   - CO₂ fire extinguisher;
   - plastic sheeting or similar material to restrict the area affected.

4. Staff have instructions that should trouble arise, they are to leave the room, put on protective clothing and then take the appropriate action. The radiation protection officer is to be advised immediately.

5. Staff are trained in radiation protection, first aid, and fire fighting, regular refresher courses are held.

Conclusion

Summarizing, the following can be stated:

1) most of the incidents occurred in the pilot plants;
2) after a learning phase, these incidents have been significantly reduced in the production plant.

To achieve this, each and every incident was rigorously analysed. Procedural checks and/or cross-checks were progressively improved and/or modified wherever necessary. And finally the UF6 hardware system was continuously developed on the strength of operational experience with the pilot plants by improving the valve sealing material or by replacing flanges by welded connexions.

Thus, while the chance of a human error still remains, the possibility of it having serious consequences has, due to continued effort, been reduced to a minimum.
<table>
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<th>Pilot Plant 2</th>
<th>Industrial Plant 3</th>
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<td>Dec.76</td>
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<td>Cumulative number of container movements</td>
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</table>

**Table 1**

**Design and Operation Data**
Figure 1

URENCO NEDERLAND OPERATIONS B.V.

Cylinder

Desublimer

Heating

Cooling

Desublimer

Cylinder

Vacuum

Cf-Drive system

Energy

Cascades

F

T

Autoclave

Cf-Cooling

CENTRIFUGE ENRICHMENT PLANT
UF₆ - RELEASES FROM 1972 TO 1978
URANIUM THROUGHPUT AND RELEASES
UF₆ - RELEASES RELATED TO A STANDARD THROUGHPUT
SUMMARY
By the Session Chairman
B.H. van de Wijngaert
SESSION I, PART I

Safety Principles for and Experience in UF₆ Handling; Storage and Transport.

Summary

Under the heading mentioned above, four papers were presented illustrating UF₆ handling as it is performed in practice in the United States (Oak Ridge, Paducah, Porthmouth), France (Tricastin), Great Britain (Springfield) and The Netherlands (Almelo).

Summarizing, it may be concluded that at all locations under review the handling of UF₆ takes place on the basis of apparently generally known and accepted safety principles.

As a result, the UF₆ handling procedures practiced in diffusion plants, UF₆ manufacturing and reconversion plants and centrifuge plants appear to be quite similar.

The avoidance of UF₆ releases can be recognized as the basic safety principle in designing equipment and drafting procedures for handling UF₆.

The handling of liquid UF₆ material is considered the most critical activity which may cause potential UF₆ releases, such as:
- the feeding of UF₆
- the withdrawal of UF₆ and
- the sampling and transfer to customer cylinders.

From a safety point of view, the goal strived for is to minimise the handling of UF₆ in the liquid state.

Handling of UF₆ in the solid state is considered to create a much less dangerous situation as far as UF₆ releases are concerned. In spite of that, sophisticated transport and lifting devices are in use to exclude all possible damage to containers from handling activities.

The number of UF₆ releases and the amounts of UF₆ set free during the operation time of the Springfield Works and the Almelo Centrifuge Plant show -
show that the design and operational safety principles and features are satisfactory.
The releases that occurred never caused hazardous situations or injury to personnel.

During the discussion a number of short questions were put. Asked about the release rate of solid UF$_6$ material via a broken off container valve, Mr. Legeay answered that such an event is of minor importance and can always be handled easily.

As to the heating time of 48" containers by means of electrically heated hot air Mr. De Dorlodot mentioned the figure of 14 hours.

Asked about the inspection procedures for large tails storage areas Mr. Legeay answered that only visual inspection of the containers takes place at irregular intervals.

Finally Mr. De Dorlodot made clear that long term storage of tails material will be done by Eurodif in the form of UO$_2$.

Items tabled in written form and to which attention will be paid during the panel discussion on Thursday 29th June concern:
- criticality considerations
- hydrolysis of UF$_6$ in case of a release
- dose limits for inhalation.
SESSION I
SAFETY PRINCIPLES FOR AND EXPERIENCE IN
UF₆ HANDLING, STORAGE AND TRANSPORT
PART II
Design considerations for centrifuge enrichment plants
by
W.K. Walrave, H. de Winter, B. Dekker
DESIGN CONSIDERATIONS FOR CENTRIFUGE ENRICHMENT PLANTS

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ABSTRACT

The design basis to insure safe handling of UF₆ feed systems for small and large enrichment plants is outlined. Also the recovery of enriched product and depleted tails in cooled desublimers will be discussed, as well as the filling of containers by liquifying the desublimer content. The further development of methods to transfer gaseous UF₆ from the desublimers into the final containers will be explained as a further improvement on safety. In the future the use of compressors to freeze UF₆ directly into the containers by passing the expensive desublimers will be considered. Special precautions will be outlined to control the ventilation air and equipment to remove UF₆ and HF from these air streams.
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1. INTRODUCTION
The design of centrifuge enrichment plants is, due to the fact that uranium hexafluoride (UF₆) is processed, subject to governmental regulations laid down in the Netherlands Nuclear Energy Act and the implementation regulation measures which are in force since 1 January 1970. This paper describes the basic safety principles to be considered when designing plants in order to be in accordance with the regulations resulting from the Nuclear Energy Act.

Furtheron, the design considerations for UF₆ systems, building and ventilation systems are explained in detail.

2. SAFETY PRINCIPLES

In order to ensure safe operation of centrifuge enrichment plants, safety measures related to handling of UF₆ as well as standard safety measures need to be taken into consideration.

The standard safety measures concern the normal industrial precautions related to fires and explosions, electrical safety, interruptions in electric power supply, etc.

The principles, adapted in relation to the safe processing of UF₆, concern:

a. design of UF₆ systems in such a way that the chance on UF₆ releases is minimized,
b. design of UF₆ systems in such a way that in case of a release the maximum amount of UF₆ is limited,
c. design of building lay-out in such a way that potential hazardous systems are located in separate areas,
d. design and installation of cleaning and decontamination systems that can handle the maximum UF₆ release without risks for personnel and environment.
3. DESIGN CONSIDERATIONS

3.1 General

Safety measures related to the handling of UF₆ focus on those parts of the process plant where UF₆ is present at superatmospheric pressures. Parts of the UF₆ process plant where superatmospheric pressure can exist, are:

- **Feed system**
  
  Solid UF₆, arriving at site in 30" or 48" containers, is heated to 70°C, purified and fed to the cascade at reduced pressure.

- **Take-off system**
  
  The product and tails streams, which are respectively enriched and depleted in U⁰²³⁵ percentages, are collected in desublimers cooled to -70°C. When filled to a certain level, these vessels are emptied by heating to +70°C and subsequent draining of liquified UF₆ into containers.

The cascade systems operate at very low pressure of approx. 1000 pascal at room temperature, with very low UF₆ inventory. For those reasons the cascade system is not considered to be a hazardous system.

For those parts of the plants where superatmospheric pressure can exist, the consequences of equipment failure are minimized by special design feature instrumentation and proper containment. Furthermore, an emergency ventilation system, equipped with U and HF monitors, shall remove UF₆ and secondary reaction products, such as fine solids of UO₂F₂ and gaseous HF, from the plant exhaust air.

In the next paragraphs the design feature of the following items will be explained more extensively:
- UF₆ feed system
- UF₆ take-off system
- building lay-out and ventilation
- air-cleaning equipment

3.2 UF₆ feed system

The UF₆ needed in the plant is of an extremely high purity. Gasses with low molecular weight, such as N₂, O₂ and HF, are a disturbing factor in the centrifuge process. To remove these, UF₆ is heated up to melting at 64°C to free any trapped air and HF. This light gas together with some UF₆ is released to a desublimer cooled to -70°C. After the pressure/temperature relationship confirms that the percentage of light gas is sufficiently low that UF₆ gas can be fed to the cascade, the necessary valves and pressure regulating valve are opened.

The UF₆ pressure required in the cascades is well below the equilibrium pressure of 8000 Pa at ambient temperature; in other words it is possible to feed UF₆ by sublimation from the solid phase. This is practised in the pilot plant SP2 with 48" feed containers. However, this mode of operation is not suitable for large capacity streams, due to heat transfer limitations. Therefore, the feed system of the SP3 plant is based upon heating/liquefaction of UF₆.
For safety reasons the containers are heated inside autoclaves with a design pressure of $10^6$ Pa. In case accidental UF$_6$ release would occur - as a consequence of a failure of the flexible connection or leakage of the container valve - all escaped UF$_6$ is contained inside the autoclave and can be recovered together with remaining UF$_6$ in the feed container.

Heating medium in the autoclaves is air which, in turn, is heated by means of condensing steam inside finned tubes. Open steam heating has been considered as an alternative for large plants.

Owing to improved heat transfer, open steam heating results in higher UF$_6$ flow rates. However, special precautions and blow-off systems are necessary to cope with pressure and temperature rise in case of UF$_6$ reaction with steam or condensate. Since it has been demonstrated that the capacity increase from 200 to 1000 tons SWU/year can still be handled with a reasonable number of air-heated autoclaves, open steam heating has not been proposed for the SP4.

The simple approach of UF$_6$ heating with open steam in a non-pressure resistant steam chest in various installations abroad is gradually being replaced by autoclaves.

### 3.3 Take-off system

The UF$_6$ product and tails flows from the cascades are recovered by freezing in desublimers cooled to -70°C. This low temperature is chosen to maintain a minimum pressure in the desublimers. In this way sufficient suction capacity is provided, effecting good mass transfer from the cascades to the desublimers while satisfying rather strict pressure drop demands. Traces of HF and other low-molecular gases are removed by a vacuum pump system, provided with NaF adsorption traps to retain HF and uranium compounds. Discharge lines of the vacuum pumps are passed through a wet scrubber prior to release to the ambient.

To empty the desublimers, the content is heated to approx. 75°C. Liquid UF$_6$ is then drained into containers. In the drain lines a number of automatic valves with interlock system are installed, while an orifice will restrict the flow of liquid UF$_6$ to no more than 50 kg/min.

If inadvertent UF$_6$ release is detected, e.g. caused by valve failure, the remotely operated valves are closed, whereas the orifice controlled flow limitation will result in reduced UF$_6$ release. Containers are moved only after solidification of the UF$_6$ content.

Future take-off systems tend to avoid filling of containers with liquid UF$_6$. In SP4 transfer of UF$_6$ into product and tails containers is planned via the gaseous phase at a pressure of approx. $6 \times 10^4$ Pa. Consequently, situations whereby large quantities of UF$_6$ are kept at superatmospheric pressure are avoided. Experiments in Almelo have confirmed that this way of processing, which already has been practised over many years by B.N.F.L. in Capenhurst, results in sufficiently large sublimation and desublimation rates to justify incorporation in SP4. However, a large number of containers are needed to collect all UF$_6$, due to heat transfer characteristics. Tails containers are cooled to approximately 10°C by a sprinkler system. Product containers are
cooled by cold air to avoid possible criticality problems caused by the use of water.

One stage further, the desublimers can be avoided altogether by compressing UF₆ flows from the cascades to such a pressure that direct desublimation into containers is feasible at moderate cooling temperature. Once a dependable compressor with sufficient compression ratio has been developed, this may result in substantial savings.

3.4 Building lay-out and ventilation

All systems operating with UF₆ at superatmospheric pressure, such as the feed station, desublimers and product and tails container filling systems, are located in a separate part of the building. This area is continuously ventilated by a once-through system, equipped with monitors for U and HF detection and air-cleaning equipment.

To obtain maximum containment, air intake and exhaust have been designed to maintain a slight underpressure in the UF₆ area.

Entry and removal of containers is done through a material sluice, through which container lorries can be driven without upsetting the ventilation system. Personnel access is also by a sluice arrangement equipped with alpha-beta hand and foot monitors. Decontamination facilities are available.

The UF₆ area has only small windows using reinforced glass, to ensure maximum containment.

3.5 Air-cleaning and effluent handling

Exhaust streams which under certain circumstances may contain uranium compounds together with hydrogen fluoride, are:
- vacuum pump outlets,
- exhaust air from local exhausts in decontamination rooms, laboratories, etc.,
- exhaust air from the UF₆ area.

Vacuum pump discharge lines and local exhausts are connected to a continuously operating air-cleaning system, where UF₆, U and fine mist of pump oil are removed.

The ventilation system of the UF₆ area is equipped with an emergency air-cleaning system. This emergency air-cleaning system is located in a bypass of the normal ventilation system of the UF₆ area and comes into operation only after accidental UF₆ release has been detected by means of monitoring instruments or by the operators.

Because of the larger air volumes to be handled in the SP4, the recirculation of the exhaust air to the hotbox area is considered in order to optimize air-cleaning.

The outlet air from the air-cleaning systems is discharged to the exhaust stack. Air contamination trapped in the air-cleaning system is collected in an effluent collection system that is kept completely separated from the public sewer system.

The discharge of contaminated solid and liquid materials takes place conform governmental regulations.
The uranium hexafluoride vaporization facility
at FBFC's plant at Romans, France
by
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THE URANIUM HEXAFLUORIDE VAPORIZATION FACILITY
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Les principes de sûreté qui ont été appliqués lors de l'étude et de la réalisation de la station de vaporisation de l'UF₆ ont été:

1) d'empêcher l'apparition du risque par un confinement permanent de l'UF₆ gazeux en mettant en jeu plusieurs barrières successives;
2) de piéger toute fuite accidentelle, de façon à éviter toute nuisance à l'environnement et d'assurer la protection des travailleurs.

Les particularités de construction de cette installation sont présentées ainsi que les calculs sur les conséquences d'une fuite accidentelle.

The principles of security adhered to at the stages of design and construction of the uranium hexafluoride vaporization facility were:

1. to obviate potential hazards by setting up a sequence of barriers to ensure the permanent containment of gaseous uranium hexafluoride;
2. to trap any accidental escape so as to obviate any pollution of the environment and ensure the protection of the workforce.

The salient features of the installation are described and calculations included showing the consequences of any accidental leakage.
SECURITY

The principles of security adhered to at the stages of the design and construction of FBFC's uranium hexafluoride vaporization facility at Romans were:

1. to obviate potential hazards by setting up a sequence of barriers to ensure the permanent containment of gaseous uranium hexafluoride;

2. to arrest any accidental leakage at the vaporization shop itself so as to obviate any pollution of the environment and, at the same time, ensure the highest standard of operator safety and protection.

DESCRIPTION OF FACILITY

The vaporization facility, the ultimate capacity of which will amount to 1200 tonnes of uranium per annum, forms part of a complex built for the production of fuel elements from enriched uranium hexafluoride.

The Romans fuel fabrication plant comprises three main production shops, engaged respectively in the following process stages:

1. Vaporization of uranium hexafluoride
   Conversion of uranium hexafluoride to powdered uranium dioxide

2. Pelletising of uranium dioxide
   Sintering
   Manufacture of fuel pins
   Fuel assembly and verification

3. Recycling of production rejects
   Effluent treatment.

Containment levels 1 and 2

The uranium hexafluoride arrives at the plant in type 30 A or 30 B containers holding a maximum of 2200 kg of hexafluoride, equivalent to some 1500 kg of uranium.

The containers have to be heated to around 80°C to vaporize the hexafluoride, which is a solid at room temperature.

To this end, each container is lowered into a hot-air vaporizer, where it rests on a cradle.

The reason for using hot-air ovens will be explained in due course.

For reasons of safety and continuity of supply to the conversion kilns, there are two vaporizers per kiln. These are employed alternately to ensure strict compliance with the rule that a cylinder of uranium hexafluoride may only be handled when cold, i.e. when the hexafluoride is in the solid state.
Additionally, the container is emptied via the gas phase, the cylinder being connected to the conversion plant by the top to obviate any escape of liquid hexafluoride.

Connection is by means of a flexible metal connector.

As a further contribution to safety, the flexible connector is inspected and tested before connection in the vaporizer. It is also changed regularly, as necessary.

FBFC opted to employ hot-air vaporizers on safety grounds, because a criticality hazard can arrive if enriched uranium (in this case at up to 5% enrichment) finds itself, in whatever form, mixed with a moderator such as water. Water pipes have been completely banned from the vaporization shop.

As against this, the hot-air vaporizer has the disadvantage of the slower rates of heating/cooling obtainable than in the case of water or steam-heated ovens.

This disadvantage has been overcome by using a double-walled oven and blowing air through the space so formed in order to increase the rate of cooling of the container when necessary.

In the case of vaporizers heated by steam or water, any leakage of uranium hexafluoride is detected simply via a variation in the pH of the water. In the case of the hot-air oven, however, a means of detecting hydrogen fluoride in the oven atmosphere is necessary.

Again, should a major incident arise, the cylinder - and particularly the valve and flexible connection - are cooled with carbon dioxide so as to bring about crystallization of the hexafluoride.

The vaporizer employed is of the shaft type, complete with bolted-down top designed to reduce any escape of hexafluoride to a minimum.

Front-loading ovens cannot easily be made as gas-tight.

The first-level containment system, which will in any case always contain the bulk of the uranium hexafluoride in the course of the process operation, consists of the cylinder itself.

When its mechanical strength is compared to the temperatures and pressures employed in the vaporization operation (80°C/1.7 bar), the likelihood that the cylinder could burst seems so slight that this first barrier to escape is seen as highly reliable.

It is followed up by the cylinder-emptying valve and piping inside the oven, which is connected to the outside line feeding the conversion kiln.
The cylinder outlet valve is screwed to the cylinder and will already have been tested on filling at the enrichment plant. It can be operated from outside the oven.

The feed line carrying the hexafluoride to the conversion reactor also comprises three other valves, two of which are automatically operated.

Containment level No. 2 consists of the vaporizer itself, even if it cannot be considered as perfectly gas-tight. Because the top is tightened down onto the body of the oven by means of handwheels, the oven will itself either contain any escape of hexafluoride, or at least retard its passage into the oven shop.

**Containment level No. 3**

The third barrier to any possible escape of hexafluoride consists of the vaporizer shop itself. This is an isolated room with no openings other than the cylinder admission lock (normally closed), personnel admission lock and a third opening consisting of an emergency exit, which will also normally be kept closed.

The control room is located outside, and does not connect directly with, the vaporizer room.

Temperature, pressure and rate of flow are permanently monitored from the control room, from which lead the means of control/regulation of the various items of equipment, including valves.

The vaporizer room and inner lock are ventilated in a manner which ensures that the pressure in the room is negative with respect to atmosphere and that there is a constant flow of outside air through the opening into the room, amounting to ca. 1 metre/sec. The air in the room is renewed four times per hour. The ventilation system, which is common to the other parts of the building, is connected to the emergency electrical circuit in case of breakdown of the mains.

In the event of escape of uranium hexafluoride, the ventilation system is modified - as described in more detail below - in order to maintain the effectiveness of the containment system.

**Complete emptying of cylinders**

Simply heating the cylinder is not enough to empty it completely.

When the pressure in the cylinder falls below 0.4 bar (gauge) the main system cuts out and a secondary pumping system takes over. This comprises a cold trap for each pair of vaporizers. The cold trap is connected to a mobile pumping unit comprising a vacuum pump (of the dry type) and chemical trap.
The pump effluent is filtered and passes through a scrubbing column before going to the discharge stack.

Once the pressure in the cylinder has fallen to ca. 100 mbar, evacuation is discontinued, the cylinder cooled to approximately room temperature and nitrogen admitted to restore pressure to atmospheric.

The cold trap is then heated externally by means of an electrical heating circuit in order to vaporize any condensed hexafluoride and pass this to the conversion reactor.

The mobile vacuum pump employed is of the dry type in order to obviate any risk of suck-back of oil, which would react explosively with the uranium hexafluoride.

**FAULTY OPERATION**

Since normal operation by definition excludes any possibility of escape of hexafluoride, there is no chemical pollution to affect operating personnel or, even more evidently, the environment. Thus, before being disconnected from the cooled cylinder, the feed lines are flushed with nitrogen to remove any residual hexafluoride and thus obviate any emission, however slight, at this point. The operators concerned will be wearing the necessary protective clothing and safety mask.

Let us now consider what could occur in the event of an operating incident: i.e. possible causes, how the fault is detected, how it is remedied and the potential consequences.

The risk may be considered as virtually nil as long as the full cylinder remains "cold", i.e. at room temperature, at which the hexafluoride will be solid. It will also be nil when the cylinder is empty and has again been cooled. This being so, any potential hazard will exist only as from the time heating of the cylinder commences and until such time as the cylinder has been completely emptied. Throughout these operating stages, the cylinder remains permanently in the vaporizer.

An escape may occur:

a. From the cylinder itself. This would imply a breach of the cylinder wall, which - having regard to the mechanical properties of the metal employed - is highly improbable. Despite this, the eventuality will be considered.

b. At the cylinder valve. It should be emphasized that any true leakage could occur only in the event that the valve became detached from the cylinder (as a result of fracture) or if the valve jammed in the open position while not connected (or only imperfectly connected) to the transfer line. But an imperfect connection would be detected at the time of preliminary tests and nitrogen flushing.
c. From the flexible connector inside the oven or the points at which this connector is itself connected at either end.

d. From piping or valves outside the oven up to the point at which they leave the oven shop.

The most serious leakage would be that of type (a), although its actual consequences would depend on the size of the breach in the cylinder wall.

The type of accident referred to under (b) would give rise only to an escape over a longer period of time since the rate of escape would be limited by the valve opening. The same is true of type (c) or type (d) incidents.

However, even a slow leakage of uranium hexafluoride ($\text{UF}_6$) will give rise to the formation of an opaque and toxic cloud of uranyl fluoride ($\text{UO}_2\text{F}_2$), which would rule out any possibility of local remedial action unless gas-tight safety equipment including self-contained breathing apparatus is available.

The means of preventing any possibility of escape of hexafluoride are as already discussed in connection with the first level of containment.

Quite apart from the precautions taken at the design stage (securing of cylinder in the vaporizer, choice of materials, location of flexible connector within the oven, etc.), the operating procedure provides for gas-tightness testing prior to heating and the vaporization operation is permanently monitored from the control room.

Process equipment is also inspected at regular intervals.

**Means of detection**

Any leakage, however slight, within the oven is detected by a hydrogen fluoride analyser, this substance being the immediate product of any contact between uranium hexafluoride and atmospheric moisture. The detector itself is located outside the oven but samples the air inside the oven, and employs an ionization probe to detect the presence of any fluorine compounds.

The resistivity recorded by the detector is transmitted to the control room, where, should it exceed a certain threshold value, it will activate an audio-visual alarm system. The aggregate quantity of hydrogen fluoride released can thus be monitored and the leak traced to a particular oven.
Provision has also been made for second-level detection in the shop. This employs two separate systems, namely a detector of the previous type fitted to the ventilation circuit and an opacity detector. A concentration of 8 ppm will activate an audio-visual alarm in the control room. This detection system will therefore respond to any sudden appearance of hydrogen fluoride but does not enable the origin of the leak to be pinpointed.

Lastly, the frequent presence of operating personnel in the vaporizer room favours detection by visual observation, given that a concentration of as little as 1 mg of uranyl fluoride per cubic metre is visible, whereupon the alarm can immediately be given (by telephone).

In sum, therefore, there will always be two - and very frequently three - levels at which an escape of hexafluoride can be detected. Any leakage from the piping systems external to the vaporizers will be detected by the analyzer on the ventilation line, always supposing it escapes the attention of operating personnel.

Additionally, any significant leakage from the piping systems would cause the hexafluoride feed pressure to one of the conversion reactors to fall and this would be noted in the control room.

In conclusion, therefore, any leakage would be detected very rapidly, if not immediately, even in the event of malfunctioning of one of the analyzers.

Remedial action

The type of action taken will be determined by the seriousness and location of the leak. Thus:

. A very slight leak inside a vaporizer will be observed in the control room as a gradual increase in the hydrogen fluoride concentration plotted by the pen recorder. Should this eventuality arise, heating of the oven is discontinued and cooling is commenced. Since the leak concerned is, by definition, no more than slight, no further action need be taken until the cylinder has been fully cooled, whereupon shop personnel can take appropriate steps to locate and remedy the fault.

The small amounts of uranium hexafluoride and hydrogen fluoride released will be extracted by the ventilation system (which employs two-stage filtration, a local rate of flow of 10,000 m³/hr and total flow for this building of 200,000 to 390,000 m³/hr).

. A leak of a more substantial, although still limited, nature occurring inside an oven will be detected and dealt with in the same way, possibly with the additional use of emergency cooling. This involves blowing carbon dioxide into the vaporizer and onto the cylinder itself - and particularly
around the valve and flexible connection - so as to bring about the rapid cooling of the container and cause the hexafluoride to crystallize at the point of escape, particularly if this affects the valve or flexible connection.

Again, a limited escape of hexafluoride from the piping systems external to the vaporizers can be reacted to by simply closing the outlet valve, by remote operation from the control room, and switching the oven to cooling. Should it not be possible to pinpoint the leak immediately, the same action may be extended to all the vaporizers on line. Emergency cooling with carbon dioxide can also be used.

Finally, consideration has to be given to the possibility of a massive - and, therefore, sudden - escape of hexafluoride, when the drill described above would be incapable of stemming the release of the gas.

Any such massive escape will inevitably be detected at both, or all three, of the levels already discussed and will bring into action, as described below, measures to:

a. protect operating personnel;
b. contain and trap the escaped gas.

Protection of personnel

This relates in the first instance to the operators present in the oven shop, who will not normally number more than two or three.

All personnel present in the shop carry gas-masks round their neck and wear overalls and gloves. Any substantial escape of hexafluoride will immediately be visually detected in the room, quite apart from the activation of the audible alarm, and the standing instruction is then simply to don masks and evacuate the shop (see Post scriptum, page 12).

The shop has three emergency exits, two of which are located at opposite ends of the room. This being so, an escape of hexafluoride is hardly likely to deny access to all three exits within the first few seconds following the escape, unless there were to be a number of simultaneous leaks at various points in the shop. Nevertheless, the possibility that visibility could be totally obscured by an opaque cloud has been taken into account. In this eventuality, the operators can feel their way quite surely to the exits by means of a handrail running along the walls and leading to the three doors. This emergency exit route is kept absolutely clear.

It should be emphasized that a massive escape of hexafluoride is conceivable only in the event of a type (a) incident involving a cylinder or valve. Also, even in this worst-case scenario, it is not feasible for a toxic cloud to fill the room instantaneously (in view of its volume of 2500 m$^3$) and the cloud formed would in fact tend to rise towards the ceiling because of
the temperature differential. Finally, even though the oven cannot be described as absolutely gas-tight, it will serve to contain the escape momentarily.

This being so, the personnel will have enough time to leave the room, which would in fact require no more than some ten seconds.

The standing instructions are quite clear in this respect: in the event of an escape of hexafluoride, personnel must don masks and evacuate the oven shop.

Special provision has also been made to protect the environment by ensuring the containment and retention of any escape of hexafluoride and the hydrogen fluoride produced by its reaction with atmospheric moisture.

**Containment and trapping of hexafluoride and hydrogen fluoride**

As already indicated, the containment system consists of the oven shop itself, which has only three exits.

These exits are not gas-tight in themselves (inter alia, the inner lock door includes provision for the passage of pipes for the supply of air to breathing apparatus). The ventilation system employed is however such that all movement of air is from outside into the vaporizer shop, thus preventing any emission of toxic gas from the room. The ventilation system could be switched to the emergency electrical circuit and continue to function throughout the trapping procedure described below.

Thus, should an escape of hexafluoride occur, a special extraction system is brought into action to clear the shop and suck the toxic discharge (uranyl fluoride and hydrogen fluoride) into an absorption and filtration facility.

The reason for this is that containment is possible only if normal air extraction is discontinued since, otherwise, the hexafluoride present in the atmosphere of the shop would be discharged to atmosphere via the stack downstream of the conversion facility. While the uranyl fluoride aerosols formed would no doubt be arrested by the outlet filters, the hydrogen fluoride formed on contact with atmospheric moisture would not be arrested and would also be likely to damage and thus reduce the efficiency of the existing filters.

The technique employed therefore consists, in the first instance, of closing a valve in the main ventilation duct so as to discontinue extraction by the normal route. This is effected by remote control from the control room, standing instructions being to close the valve as soon as the hydrogen fluoride alarm goes off. Additionally, provision has been made for blanking off the oven room extraction outlets so as to select the point of extraction most appropriate to the location of the leak. The blanking-off operation relies on a set of sluices
fitted in front of the eight vaporizer shop prefilters and which can be operated manually and simultaneously from outside the building by means of a set of linkages.

The escape of uranium hexafluoride having been thus circumscribed, it can be dealt with as described below. The system employed, which by-passes the main ventilation duct, extracts the polluted air from the shop via an emergency extraction line incorporating a caustic potash scrubber, the gaseous effluent from which then passes to the normal discharge stack upstream of the "absolute" filters. This gaseous effluent, which will now be virtually free of hydrogen fluoride, is then filtered and diluted in the main flow of ventilation air extracted from the conversion building. It should be stressed that the emergency extraction system (with particular reference to the scrubbing tower) is located outside the building so as to obviate any presence of water in the conversion building.

The emergency system is brought into operation manually from the control room, and is of course employed concurrently with the drill already discussed, i.e.:
- the oven concerned is switched from heating to cooling, based on circulation of air between the double walls
- closure of valve upstream of any leak located on a hexafluoride line external to the ovens
- if necessary, switching of all ovens to cooling
- use of carbon dioxide cooling.

The potassium hydroxide scrubber is designed to neutralize and arrest 98% of pollutants.

In view of its large size and unfavourable geometry, it has been poisoned with boron glass rings to ensure against any criticality hazard arising from accumulation of enriched uranium in the scrubbing tower.

WORST-CASE SCENARIO

In what follows, the working assumption is that of the maximum possible accident, i.e. the rupture of a full 30 B cylinder, containing 2200 kg of uranium hexafluoride at a temperature of 80°C.

Thermodynamic calculations show that 60% of the contents of the container can escape rapidly in the event of opening to atmosphere.

As already noted, the hexafluoride will hydrolyse completely and instantaneously on contact with atmospheric moisture to yield uranyl fluoride (UO$_2$F$_2$) and hydrogen fluoride (HF).

The hydrolysis of 1320 kg of uranium hexafluoride yields 300 kg of hydrogen fluoride.
By virtue of its very high density, the bulk of the uranyl fluoride formed will tend to settle in the immediate vicinity of the escape, but this will not be true of the aerosols formed.

After passing through the prefilters, the polluted air is extracted by the emergency system, which is designed for complete renewal of the air over a period of 2.5 hours against the 15 minutes for normal operation.

The choice of the slower rate of renewal is dictated by the need to protect the environment, the reasoning being that if a quantity of hydrogen fluoride has to be released to atmosphere such release should preferably be spread over an extended period of time in order to keep down the outside concentration of hydrogen fluoride.

The polluted air is therefore extracted via the caustic potash scrubber, which has been designed for 98% retention efficiency. We shall however assume an efficiency of only 95%. On this basis, approximately 15 kg of hydrogen fluoride, diluted by the main ventilation air, will be released to atmosphere over a period of 2.5 hours.

As against this, the quantity of uranyl fluoride released will be negligible, thanks to the action of the "absolute" filters on the main ventilation air extraction system.

The environmental pollution assessment is based on the method recommended by A. Doury\(^{(1)}\) (formula 20), according to which the concentration per unit volume \(\chi\) of the pollutant is given by:

\[
\chi = \frac{\lambda z Q}{\left(\frac{u}{2} \frac{1-k_y-k_z}{2} \frac{k_y}{k_y-k_z} \frac{k_z}{k_y+k_z} A_y A_z x y + k_z \right)} \\
\exp \left(-\frac{1}{2} \left[ \left(\frac{u}{A_y} x \right)^{2k_y} \left(y - y_o \right)^2 + \left(\frac{u}{A_z} x \right)^{2k_z} \left(z - z_o \right)^2 \right] \right)
\]

\(^{(1)}\) A. Doury - Une méthode de calcul pratique et générale pour la prévision numérique des pollutions véhiculées par l'atmosphère.

CEA Report R - 4280, Rev. 1
where:

- \( x \) = coordinate of position in the direction of the wind, reckoned from the point of emission
- \( y, z \) = coordinates at right angles to the direction of the wind
- \( y_0, z_0 \) = coordinates of the point of emission and of the axis of the plume (direction of wind)
- \( \lambda_s Q \) = rate of emission of pollutant
- \( \bar{u} \) = mean velocity of carrier fluid (assumed to be uniform)
- \( k_i \) = dimensionless exponent
- \( A_i \) = pseudo-coefficient of diffusion.

The coefficients \( k_i \) and \( A_i \) vary with meteorological conditions (normal or low scatter) and the "transfer time" required for a "burst" of pollutant to travel from the point of emission to the point of measurement.

These coefficients are tabulated in the Appendix.

The formulae can be used to plot sets of curves (2) (cf. Figs. 2 and 3) showing variations in atmospheric transfer coefficient (CTA) with distance from the point of emission and wind speed.

The CTA merely has to be multiplied by the rate of emission of pollutant (per second) to obtain the ground-level concentration immediately below the plume at any point.

The curves shown (Figs. 2 and 3) relate to a stack height (h) of 15 metres since, although the actual height of the stack is 23 metres - to which has to be added the hydrodynamic head - we have, in order to take account of the effect of surrounding buildings, adopted the very conservative figure of 15 metres as the height of the source of emission.

Again, in order to obviate any dependence on favourable meteorological conditions, the envelopes of the graphs dependent on \( \bar{u} \) shown in Figs. 2 and 3 have been plotted for the conservative conditions of normal or low atmospheric scatter.

The superimposition of these two envelopes (Fig. 4) shows that normal scatter is preponderant over short distances (up to 500 metres), giving way to low scatter at greater distances.

(2) A. Doury, R. Gérard and M. Picol - Abaques d'évaluation directe des transferts atmosphériques d'effluents gazeux. CEA Report DSN No. 84, Rev. 1, March 1977
Since the factor of importance in any case of chemical pollution is concentration, allowance has to be made for dilution by main ventilation air (pollution-free).

In the case of the Romans plant, the flow of main ventilation air amounts to 240,000 m$^3$.h$^{-1}$ for the initial construction phase, and will ultimately increase to 390,000 m$^3$.h$^{-1}$.

The worst-case emission concentration (at the stack) will therefore amount to 25 mg.m$^{-3}$ (for 240,000 m$^3$.h$^{-1}$).

Using this figure, concentration has been plotted against distance (Fig. 5) and it will be seen that as from a distance of 20 metres from the stack the hydrogen fluoride concentration (in the worst-case scenario) is less than 2 micrograms per cubic metre, i.e. only one-thousandth of the maximum permissible concentration to which the workforce may be exposed under the regulations applying in most countries (2 mg.m$^{-3}$).

In designing the Romans plant, therefore, every precaution which can reasonably be taken in industry has been taken to ensure that any incidents or improbable accidents which could occur in the course of the vaporization of uranium hexafluoride would have strictly negligible consequences for the surroundings.

---

**Post scriptum**

on page 7. Protection of personnel

"Since the writing of this paper our philosophy has changed. Since the time necessary to don a protective mask is longer than the time to leave the workshop (less than 10 seconds), and rule is to go out of the room as quickly as possible if necessary with the aid of the handrails.

However, it must be mentioned that if a manipulation has to be done on a container the workforce wears protective clothing (cowl) which is supplied by external purified air".
Fig. 2 - Coefficient de transfert atmosphérique maximal au sol en fonction de la distance sous le vent d'une émission ponctuelle de hauteur effective 15 m
[Conditions atmosphériques: diffusion normale - vents de 0 à 30 m.s⁻¹]
Fig. 3 - Coefficient de transfert atmosphérique maximal au sol en fonction de la distance sous le vent d'une émission ponctuelle de hauteur effective 15 m. (Conditions atmosphériques: diffusion faible - vents de 0 à 5 m.s⁻¹)
Fig. 4 - Coefficient de transfert atmosphérique maximal au sol en fonction de la distance sous le vent d'une émission ponctuelle de hauteur effective 15 m.
### ANNEXE

- Conditions météorologiques "normales (troposphériques moyennes)

#### TABLEAU I

Diffusion horizontale

<table>
<thead>
<tr>
<th>Temps de transfert</th>
<th>$k_x = k_y = k_h$</th>
<th>$A_x = A_y = A_h$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$t + \Delta t$</td>
<td>sans dimension</td>
<td>m$^{-1}$/s$^{-1}$</td>
</tr>
<tr>
<td>1</td>
<td>2</td>
<td>3</td>
</tr>
<tr>
<td>0 à 4 mn</td>
<td>0,859</td>
<td>$4,05 \cdot 10^{-1}$</td>
</tr>
<tr>
<td>4 mn à 1 jour</td>
<td>1,130</td>
<td>$1,35 \cdot 10^{-1}$</td>
</tr>
<tr>
<td>1 jour à 6 jours</td>
<td>1,000</td>
<td>$4,63 \cdot 10^{-1}$</td>
</tr>
<tr>
<td>6 jours à 15 jours</td>
<td>0,824</td>
<td>$6,50 \cdot 10^0$</td>
</tr>
<tr>
<td>après 15 jours</td>
<td>0,500</td>
<td>$2,00 \cdot 10^5$</td>
</tr>
</tbody>
</table>

#### TABLEAU II

Diffusion verticale

<table>
<thead>
<tr>
<th>Temps de transfert</th>
<th>$k_z$</th>
<th>$A_z$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$t + \Delta t$</td>
<td>sans dimension</td>
<td>m$^{-1}$/s$^{-1}$</td>
</tr>
<tr>
<td>1</td>
<td>2</td>
<td>3</td>
</tr>
<tr>
<td>0 à 4 mn</td>
<td>0,814</td>
<td>0,42</td>
</tr>
<tr>
<td>4 mn à 49 mn</td>
<td>0,855</td>
<td>1,00</td>
</tr>
<tr>
<td>&gt; 40 mn</td>
<td>0,500</td>
<td>20</td>
</tr>
</tbody>
</table>
- Conditions météorologiques extrêmes de faible diffusion dites "mauvaises" (stratosphériques moyennes)

\[
\begin{align*}
 k_h & = \text{cf Tableau I} \\
 A_h & = \text{cf Tableau I} \\
 k_z & = 0.500 \\
 A_z & = 2.00 \times 10^{-1} \text{ m}^{-1} \text{ s}^{-1} = 1.73 \times 10^{-4} \text{ m}^{-1} \text{ s}^{-1} = 1.73 \times 10^{-2} \text{ km}^{-1} \text{ s}^{-1}
\end{align*}
\]
Yield and storage of depleted uranium in the Federal Republic of Germany by

W. Kayser
Under several aspects, depleted uranium can be regarded as valuable raw material. By the year 2000 an accumulated quantity of about 150,000 te of UF6 will have to be stored in the Federal Republic of Germany. Presently, two storage facilities the capacities of which being 150 and 1,000 te are under operation where the material is stored in 48" transport containers in one layer configuration. This concept seems to be reasonable, too, in the view of protection against environmental impact. Up to now, no incidental or accidental situations occurred.
1. INTRODUCTION

Depleted uranium is produced as tails material in the isotope separation process the main goal of which is increasing the \( \text{U}^{235} \) contents in natural uranium to about 3%. All enrichment processes require the gaseous compound of uranium hexafluoride, UF6. Usually, the enrichment work is performed under a toll enrichment contract between the enrichment company and the utility so that the UF6 is property of the latter. Under normal conditions, the depleted UF6 remains in the area of the enrichment plant although - in terms of enrichment - it could be regarded as waste material. In some cases, for instance according to COCOM requirements, depleted uranium from an enrichment contract with the USSR had to be taken back by the customer.

On the other hand, from the view of long term energy supply, depleted uranium has to be considered as valuable material. Firstly, if uranium prices would increase in future, and, at the same time, enrichment cost would decrease further depletion of the material could be worthwhile. Secondly, in any case and without respect to commercial aspects, depleted uranium represents a strategic reserve for energy supply. Finally, \( \text{U}^{238} \) can be used in fast breeder reactors.

Currently, the application of depleted uranium is limited to shieldings against ionizing radiation and, according to the mechanical properties of the metal and the chemical behaviour, some minor kinds of utilization in the non-nuclear field.

2. YIELD OF DEPLETED URANIUM IN THE FEDERAL REPUBLIC OF GERMANY

The estimates on nuclear power capacity to be installed in the Federal Republic of Germany during the following twenty years have changed since 1976. This mainly results from significant delays in the licensing procedures initiated by public opposition against nuclear energy. Figure 1 shows the predicted values which vary between 30 and 40 GWe for 1985 and between 47 and 72 GWe for 1990. Regarding the progress of licensing procedures just being performed, the lower value seems to be more realistic.

These values have been used as basic data for the calculation of the yield of depleted U up to the year 2000. The calculation was performed by means of a special computer programme which allows the variation of

- assumptions on recycling of uranium from spent fuel
- assumptions on recycling of plutonium from spent fuel
- installed power capacity versus time
- tails assay.

As far as fixed by contracts, the actual tails assay was used. The results of the calculations are shown in figure 2.

\[ \text{COCOM} = \text{Coordinating Committee on East-West Trade} \]
Figure 1: Predicted nuclear power capacity in the Federal Republic of Germany [1,2]

Figure 2: Accumulated quantities of depleted uranium produced by nuclear fuel supply in the Federal Republic of Germany
The upper limit results from the maximum nuclear power increase of fig. 1 under entire renunciation of recycling. The tails assay is assumed as to be 0.25 %. Accordingly the lower limit is based on the minimum increase of fig. 1. Partial reprocessing and recycling of SNM (special nuclear material) and a tails assay of 0.2 % are assumed.

Additionally, a reference case has been calculated which is characterized by a 0.25 % tails assay and recycling conditions being reasonable in the view of current philosophy.

A sensitivity analysis showed that even by extreme assumptions on
- reprocessing and recycling
- increase programmes for alternative reactor types such as high temperature and fast breeder reactors
- reduction of tails assay to 0.15 %

the results will not vary essentially.

From the calculations described a yield of

160,000 te

of depleted uranium up to the year 2000 seems to be a reasonable figure.

The quantities to be stored within the Federal Republic are much smaller because, as already mentioned, the depleted uranium usually is stored in the area of the enrichment plant. So generally no quantities need to be stored in a country which does not operate an enrichment plant. The only exception is given by an enrichment contract between the Federal Government and the USSR. Under these assumptions, all quantities of depleted uranium will originate from two sources:

1. from toll enrichment in the USSR
2. from future enrichment plants being operated in the Federal Republic.

Assumed that the enrichment capacity in Germany would reach 7 mio swu/a', the accumulated quantity to be stored would follow figure 3, i.e. it will increase from about 600 te in 1977 to about 100,000 te in 2000.

If the utilities would make use of their options to keep the depleted uranium as their property this quantity could be enlarged up to 160,000 te.

'swu = separative work units
3. STORAGE OF UF6

At the time being depleted uranium worldwide is being stored as uranium hexafluoride, UF6. In the Federal Republic of Germany about 150,000 te UF6 containing 100,000 te U will have to be stored in the year 2000.

The two types of storage containers presently used in the FRG (48" F and 48" Y container) are steel containers of 48" or 1,219 mm in diameter, 150" or 3,810 mm in length, and a wall thickness of 15.9 mm. They differ in minor details and their net capacities are 12,300 kg and 12,500 kg, resp. The containers are layed out and licensed for transport, too.

The Umschlag & Speditionsgesellschaft "Braunkohle" GmbH, Mannheim, is storing about 1,000 te UF6 which have been taken back from the USSR in Weisweiler near Aachen.

Type F and type Y containers are in use lying on squared timbers on the concrete bottom of a building of light steel construction, the dimensions of which being 97.5 m by 25 m, and 11 m in height. The distances from the walls and between the containers are wide enough to allow convenient visual inspections and monitoring measures (figure 4).
The building is surrounded by a wire fence, 2 m in height. The area between fence and building is lighted up during night and monitored by detectors.

NUKEM GmbH, Hanau, is storing under federal contract about 150 te UF6 in 48" type F containers in an open air storage within the fabrication area (fig. 5).
The ground under the containers consists of prefabricated concrete plates. The containers are lying on squared timbers, too. The distance between them is 1 m. The storage area is surrounded by a wire fence of the same height like that at the USG storage. The storage area is situated within the nuclear park which is protected by another fence and guards. So additional protection measures are not necessary. The containers, especially valves and condition of walls, are inspected in a four weeks turn. The anticorrosion coating is renewed once a year and, if necessary, more frequently.

4. SAFETY ASPECTS

The behaviour of UF6 in case of an incidental or accidental situation has to be understood from its physical and chemical properties. The most important of them are its triple point at $64^\circ C$ [3,4] and 1517 m bar and its chemical reaction under presence of water or water vapor according to the equation

$$UF6 + 2H2O \rightarrow UO2F2 + 4 HF.$$ 

An environmental impact in case of UF6 release is due to its chemical poisonousness and its radioactivity. Criticality problems do not need to be considered as depleted UF6 cannot become critical in any configuration [5].

Of all incidental and accidental situations to be discussed, as there are transport, corrosion, radiant heat of sun, earthquake, hurricane, thunderstorm, theft, explosion, sabotage, and airplane crash, only those ones have to be regarded causing release of big quantities of gaseous UF6 to the air.

These situations have been investigated with the result that there are only two severe situations which, under unfavourable conditions, can cause dangerous impact:

1. theft followed by explosion within a densely populated area and
2. airplane crash followed by a fuel fire of more than 5 minutes duration [6].

In the first case, theft of containers from a storage facility can be excluded because of the protection measures and the need to dispose of special equipment. In case of theft during transport, the UF6 quantity is limited. In the second case, the calculations were based on the specification of 48" type G containers with a wall thickness of 8 mm especially being used for storage purposes. Even in this case, calculations performed for the distance of 1 km show that the HF concentration would reach only a small fraction of the value mortal for human.

Additional measures which could be provided in order to limit the extent of environmental impact have been considered theoretically. The conversion of UF6 into other chemical
uranium compounds such as UF4, UO2, or U3O8 with reconversion into UF6 before further enrichment or fuel fabrication seems to be possible but would increase the storage cost by the factor 3 to 4. In order to reduce the duration of fuel fire after an airplane crash, several provisions can be made. So a special shape of the ground under the containers could ensure that the fire would be located in a favourable position, for instance.

5. CONCLUSIONS

At the time being, by reasons of energy supply the storage of depleted uranium cannot be renounced. Storage in the chemical form of UF6 is a well approved technique and most economic. Especially in storage areas in one layer container configuration, additional measures in order to reduce the possible consequences of a fuel fire after airplane crash by relatively simple means seem to be technically feasible if necessary. It seems to be worthwhile to gain better knowledge of the behaviour of UF6 in that case of accident by additional R&D work.

As a final statement, up to now no UF6 release has occurred in the German storage facilities for depleted UF6.

6. REFERENCES


Safety considerations involved in the choice of containers for storage of UF₆ tails by
D.F. Norsworthy, C. Howarth
SAFETY CONSIDERATIONS INVOLVED IN THE CHOICE OF CONTAINERS FOR STORAGE OF UF₆ TAILS

by

D F Norsworthy, Safety and Reliability Directorate, U.K.A.E.A. Culcheth

C Howarth, British Nuclear Fuels Limited, Risley

The paper summarises the outcome of a safety study to decide which of two types of container to recommend for long term storage of depleted uranium hexafluoride.

Subjects discussed include:-

(a) Operational safety of the Type 0236 2 Tonne Vertical container in use at present.

(b) Quantification of the ability of both the Type 0236 and the Type 48Y to withstand exposure to a major fire.

(c) Estimates of hazard ranges for HF and uranium in adverse weather given a 'quasi-instantaneous' release of UF₆ contents.

(d) A discussion of other safety factors of relevance when making a choice between the containers.
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1. INTRODUCTION

In 1975 British Nuclear Fuels Ltd (BNFL) were already storing several thousands of tonnes of depleted Uranium Hexafluoride (UF$_6$) at Capenhurst. This material had built up, over a period of 15 to 20 years, as 'tails' from the Diffusion Plant; the large number of 2 tonne containers involved was distributed around the site in buildings and on concrete rafts in the open. It was recognised that future arisings from the Diffusion Plant and from the Centrifuge Plants now coming on stream will continue to augment the total holding.

To ensure that proper account was taken of all safety aspects when choosing the type of container to be used for depleted material from the new plant, the Safety and Reliability Directorate of the U.K. Atomic Energy Authority (SRD) were asked to undertake an independent safety assessment aimed at identifying:–

(a) The safety implications of existing storage and handling arrangements at Capenhurst.

(b) The relative merits, from a safety viewpoint, of the 2 tonne vertical container and an alternative 12 tonne horizontal container.

(c) Any other safety considerations which might be of relevance to the long term storage/handling of UF$_6$ 'tails'.

This paper summarises the outcome of the SRD study and some of the actions which resulted from it.

2. NATURE OF HAZARD

Gaseous UF$_6$ is used at Capenhurst to obtain Uranium which has been enriched in the fissile isotope (U$_{235}$). The product so generated contains more fissile material than natural Uranium and care must be exercised to avoid any risks of a "critical assembly" being formed.

However, as a natural consequence of this enrichment process, both the Diffusion Plant and Centrifuge Plants generate a large quantity of 'tails' material which has lost a fraction of its fissile content. Although this depleted UF$_6$ still contains U$_{235}$ (it may even be economic to recycle it through the plant) the proportion present is never sufficient to sustain a chain reaction even under the most favourable conditions. As a result, there is no possibility of a critical assembly being formed even if a large quantity of 'tails' hex is mixed in gaseous, liquid or solid form with an optimum amount of hydrocarbon moderator material.

At room temperature UF$_6$ is a colourless, crystalline solid which sub-tends a significant but low vapour pressure (Fig. 1). When heated at atmospheric pressure the crystals sublime without melting and the vapour pressure reaches 760 mm Hg at a temperature of about 56°C. At higher pressures the crystals will melt, at a temperature of about 64°C and this melting is accompanied by a very substantial increase in specific volume (Fig. 2). Although the material reacts chemically with most hydrocarbons, many metals, alcohol, ether and water it does not react with moisture free air. The primary hazard to personnel in the event of a boundary failure arises from the reaction between UF$_6$ and the moisture normally present in the atmosphere.
Reaction \[ UF_6 + 2H_2O \rightarrow UO_2F_2 + 4HF \]

Molecular weights: \( UF_6 \) 352, \( H_2O \) 36, \( UO_2F_2 \) 308, \( HF \) 80

With gaseous \( UF_6 \), this reaction proceeds rapidly liberating some heat and generating gaseous hydrogen fluoride (HF) together with a fine aerosol of uranyl fluoride (\( UO_2F_2 \)). Hydrogen fluoride subtends a much higher vapour pressure than \( UF_6 \) (Fig. 1) and due to the formation of four molecules of HF for each molecule of \( UF_6 \) the reaction is accompanied by a substantial volume increase at atmospheric pressure. The HF is highly corrosive in aqueous solution and has a Threshold Limit Value (TLV) for airborne concentration of 3 ppm or 2.5 mgm/m³ (Ref. 1). The following extract from (Ref. 2) summarises its toxicology:

"It is extremely irritating and corrosive to the skin and mucous membranes. Inhalation of the vapour may cause ulcers of the upper respiratory tract. Concentrations of 50-250 ppm are dangerous even for brief exposures. Hydrofluoric acid produces severe skin burns which are slow in healing. The subcutaneous tissues may be affected becoming blanched and bloodless. Gangrene of the affected areas may follow".

No formal agreement exists in the U.K. regarding the emergency reference level (ERL) to be used for hydrogen fluoride when analysing the possible effect on the general public of a major release of \( UF_6 \). The figure currently accepted by the Alkali Inspectorate for the purpose of defining a hazard range for emergency planning at Capenhurst is 5 ppm (4.5 mgm/m³).

The \( UO_2F_2 \) produced by the reaction is also very toxic. At high airborne concentrations the fluoride component can produce effects broadly similar to HF but, for a major \( UF_6 \) release, the limiting \( UO_2F_2 \) airborne hazard range is more likely to be set by the cumulative effect of uranium inhalation. At the level of enrichment appropriate to 'tails' material the chemical toxicity of uranium exceeds its radio-toxicity - the TLV for soluble uranium compounds is 0.2 mgm/m³ (Ref 1).

For occupational short period exposures Table VI of Ref 3 recommends the following inhalation dose limits:

<table>
<thead>
<tr>
<th>Duration</th>
<th>Uranium Concentration (mgm/m³)</th>
<th>Total Dose (mgm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>10 days</td>
<td>0.2</td>
<td>20</td>
</tr>
<tr>
<td>10 hours</td>
<td>1.0</td>
<td>10</td>
</tr>
<tr>
<td>1 hour</td>
<td>4.0</td>
<td>5</td>
</tr>
<tr>
<td>0.1 hour</td>
<td>6.0</td>
<td>1</td>
</tr>
</tbody>
</table>

The time of passage of the toxic cloud, at distances of a few kilometres is likely to be between 0.1 and 1 hour. Hence, since no formal
agreement exists regarding the MNL to be used for UO$_2$F$_2$, an inhalation dose limit of 4 mg/m$^3$ of uranium is used in this report as a basis for estimating potential hazard ranges.

Deposition of UO$_2$F$_2$ can result in the contamination of all surfaces, including agricultural crops and grassland. The rate at which deposition will occur and hence contamination contours will be very dependent on atmosphere conditions at the time of a release.

3. TWO TONNE VERTICAL CONTAINER (TYPE 0236)

The design and operating intent for these containers had never been collated into a formal document, hence the first task was to identify and comment on the adequacy of the steps taken to minimise both the probability and potential consequences of an accident during filling, handling, storing and emptying the existing containers. Possible causes of a UF$_6$ release were identified as follows:

3.1 Handling Accident

Due to the large number of containers being handled the chance of a handling accident was seen as substantial. A drop test performed at Springfields in 1968 showed that dropping a loaded container onto a flat surface from a height of 6 ft is unlikely to produce a gross failure but there is clearly some risk of a container being "spiked" during handling and this could result in penetration, with partial spillage of contents and/or ingress of moisture. Additionally, if local weakening has occurred due to corrosion e.g. in the crevice formed by the skirt and the lower dished end, there is an increased chance of a failure if the container is dropped or lands heavily. With regard to external corrosion of containers stored in the open a specialist consulted by SRD suggested that:

EITHER - Arrangements should be made to periodically inspect all containers at an interval to be agreed. Any paint damage or corrosion should be rectified to an approved procedure

OR - A method of 'coating' exposed surfaces of the containers on a routine basis should be developed, tested and specified. The agreed specification should include:

(i) The choice of anti-corrosion material
(ii) The method and frequency of application
(iii) The confirmatory programme of sample inspections

The second approach of developing a method which is suitable for coating the exposed surface on a routine basis has been followed. Samples of anti-corrosion materials have been applied and the specialist's report of the examination of treated UF$_6$ containers after an 11 months trial, concluded that these anti-corrosion compounds should greatly extend the outdoor life of the containers.
Available evidence from U.S.A. (Ref 4) suggested that internal corrosion should not present a problem, but, to check that differences in U.K. conditions of use pointed out by SRD did not invalidate this conclusion, two containers which had been used for 'tails' storage for 17 years were withdrawn from service and examined in detail. Internal corrosion was found to be negligible.

At ambient temperature the UF\textsubscript{6} in the container will be a crystalline solid but, if a spill occurs, reaction with environmental moisture will produce HF vapour and a fine aerosol of UO\textsubscript{2}F\textsubscript{2}. If surfaces were wet, with pools of water around, the airborne hazard would be greater than if the moisture available was limited to that in the surrounding air. One kg of water will react with approximately 10 kg of UF\textsubscript{6} to form approx. 2 kg of HF and a release of this size would present a serious hazard to site personnel in the vicinity. Hence it was concluded that, with sufficient moisture present a handling accident leading to a spill of UF\textsubscript{6} could produce a serious local hazard with some risk of exceeding the emergency reference level at the site boundary. However, the off-site consequences would be small compared with the potential of a container failure due to overheating.

The only handling accident which has the potential for releasing a substantial quantity of UF\textsubscript{6} vapour is the dropping of a container whilst in transit over the feed stations. The handling of containers within the Diffusion Plant building has been reviewed to minimise movements over on-line containers but the layout is such that there are a limited number of cases where feed containers must be moved over other feed positions. Special precautions are taken for these movements and the problem has been avoided in the design of new plants.

3.2 Spontaneous Failure during Storage

The material stored in these containers is in solid form and subtends a low vapour pressure at normal atmospheric temperatures. The maximum internal pressure without artificial heating is unlikely to exceed (say) 5 psig hence a gross failure is highly unlikely. However, pitting attack, if allowed to proceed unchecked, could result in minor leakage of air carrying UF\textsubscript{6} vapour and this would tend to show as a "festering" of UO\textsubscript{2}F\textsubscript{2} in the vicinity of the leaks.

3.3 External Missiles

The consequence of a missile hitting and penetrating a container would be comparable with 'spiking' during handling i.e. partial spillage of contents and/or moisture ingress.

A case of special importance would be a crashing aircraft since this could result in the contents of one or more containers becoming exposed to a severe fire. An early SRD
assessment examined whether the addition of UF$_6$ to a hydrocarbon/air fire could drastically alter its severity; this is reproduced as Appendix 1, from which it will be seen that, whilst the total heat generated is unlikely to increase significantly, the rate of combustion may possibly be increased. Appendix 8 outlines a suggested basis for estimating the chance of an aircraft crash affecting a container storage area, from this it will be seen that, even for the storage of a large quantity of material, the chance of such an incident is low.

3.4 Loss of voidage during refeeding

The only operating sequence which involves deliberate heating of a tails container and hence the potential for releasing a large quantity of UF$_6$ vapour is 'refeeding'. Two types of accident might occur here:-

(a) A conventional leak – the amount of UF$_6$ vapour released would be unlikely to produce a serious off-site hazard.

(b) Overpressurisation of the container and associated filling line by expansion, whilst isolated from the plant during warm up to feed temperature – this could result in a disruptive failure of the container and an uncontrolled release of a large quantity of UF$_6$ vapour, into the Plant Area with some possibility of a serious off site hazard. Since completion of the SRD study:-

(i) the fill limit, applicable to the selection of existing Type 0236 containers for refeeding, has been reduced from 2150 Kg to 2050 Kg.

(ii) the manufacturers now measure and certify the volumetric capacity of all new containers of this type.

(iii) all containers are examined for signs of corrosion when taken from the stockpile for refeed.

3.5 Failure due to fire

When heated in a closed container UF$_6$ has a melting point of about 64°C and this change of state is accompanied by a substantial increase in volume. In addition the coefficient of expansion of the liquid is much higher than that of the mild steel container and if heating continues it is inevitable that the container will be overpressurised and eventually fail.

Such a failure would result in a sudden release of the pressure which was keeping the UF$_6$ in liquid form and as a result it would change state rapidly. The proportions of vapour and solid formed would be dependent on the amount of heat stored in the liquid. If the total contents of the container were liquid when a disruptive failure occurred the vapour cloud produced would be likely to carry the solid UF$_6$ with it into the atmosphere.
BNFL are satisfied that, with the precautions taken, the chance of a container becoming involved in a major fire whilst in storage at Capenhurst is negligible. Nevertheless there is a possibility that a container of this type could become involved in a transport fire, especially if it becomes necessary to move large numbers of containers to and from the site by road. The potentially serious consequences of this type of failure were sufficient to justify closer examination. (Sections 4 and 5 below).

4. THE EFFECT OF FIRE ON A 2 TONNE CONTAINER

Appendix 2 analyses the effect of fire on a loaded 2 Tonne container using the method of calculation recommended in the IAEA Regulations for the safe transport of radioactive materials. Although total immersion in an 800°C fire is a severe case it should be borne in mind that flame temperatures higher than this could occur and also that if 'free' UF₆ is available this may possibly increase the rate of burning.

The first section of the Appendix calculates the internal pressure for a container loaded to its nominal maximum (2150 Kg), heated to a temperature of 80°C. The remaining sections outline the basis for reaching the following general conclusions:

(a) For a starting temperature of 15°C all heat entering the container after the first 2.5 minutes would be available to produce UF₆ vapour on failure.

(b) Pressure build up depends on mass content and volume, a mass content of 2.3 Tonnes could produce failure before the contents are fully liquefied.

(c) With a mass content of 2 Tonnes the time to liquefy is about 12 minutes.

(d) If the fire is still burning after the completion of melting the UF₆ temperature rises at 10-12°C per minute until failure occurs. With a mass content of 2 Tonnes failure would probably occur at a temperature of 120-150°C (i.e. 5 to 8 minutes after completion of melting).

(e) For a failure 20 minutes after the start of the fire approximately 1600 Kg of UF₆ would vaporise when the pressure was released and the remainder would be expected to become airborne in particulate form.

Note: As a consequence of the study 0236 containers are no longer transported without protection. To date a specially adapted vehicle is used. Later this year a number of double metal skinned insulated protective packages will be available.
5. VAPOUR CLOUD BEHAVIOUR

The results of a large scale test reported by Van Ulden (Ref 5), which involved the release of 1 Tonne of the heavy gas Freon 12 (4 x air density) showed the initial expansion to atmospheric pressure to be accompanied by intensive mixing which reduced the mixture density to about 1.25 (i.e. a volume ratio Freon/Air of 1/11.5). No details are given in the reference of the way the cloud was generated but it is understood this was achieved by pouring liquid Freon onto a water pool - a very different mechanism from the disruptive failure of a vessel containing a liquid which 'flashes' at atmospheric pressure.

If it was feasible for a 1.6 Tonne cloud of UF₆ vapour to be generated without mixing, the volume - at atmospheric pressure, 56.5°C - would be about 125m³. If the UF₆ mixed with ten times its own volume of dry air during initial expansion:

* Cloud Volume = 125 + 1250 = 1375m³
Cloud Weight = 1.6 + 1.6 = 3.2 Tonnes
Mean Density = 2.3 Kg/m³

* This ignores vaporisation of particulate as vapour pressure reduces.

It will be seen that, ignoring any reaction with moisture a UF₆ vapour/air cloud will always be heavier than the surrounding air.

Once the initial mixing phase was complete Van Ulden's test showed (for the Freon/Air mixture) that the heavy cloud subsided under the influence of gravity, to a height of less than a metre, spreading out to cover an area far larger than would be predicted by using a 'neutral buoyancy' model. Subsequently as the cloud moved away from the source, carried by the wind, the height increased only slowly and did not exceed about 10 metres at a distance of 1 Km. No attempt has been made to use this result, or the associated theory, to predict the behaviour of a UF₆ vapour cloud since a number of important differences must be recognised:

1. The initial mixing phase is unlikely to be comparable and may result in a smaller cloud of much higher density than in the experiment.

2. Conversion to HF will occur continuously as the cloud size increases at a rate dependent upon the available moisture, e.g. =

   50% humidity at 20–25°C = 10 gm/m³ H₂O approx

   Total volume of air needed = 2 x 10⁴ m³
   to convert 2 Tonnes of UF₆

3. Turbulence effects at the cloud periphery may be influenced by this HF generation since, as well as the heat of reaction, each molecule of UF₆ will produce 4 molecules of HF and a corresponding volume increase.
4. If the ground is dry deposition of solid UF₆ due to heat transfer, may deplete the cloud.

The main conclusion of the Van Ulden report is seen as relevant and important, viz: "The results of the experiment together with the model clearly show that the gaussian model cannot be used for heavy gases, without a proper correction for density spread. As a result of density spread higher concentrations over larger areas should be expected."

A fuller discussion of the factors which could influence density, as the cloud is diluted with moist air, is given in Appendix 3. For simplicity the assessment of hazard ranges which follows has been based on an assumption of neutral buoyancy both prior to and after HF formation.

6. HYDROGEN FLUORIDE HAZARD RANGES

Appendix 4 explains the basis used to calculate HF concentration at various distances downwind for both 'quasi-instantaneous' (puff) releases and continuous releases at constant rate.

From Table 1 of the Appendix it will be seen that, for adverse weather (Pasquill Category 'F') the average airborne concentration of HF in the cloud at a distance of 3 Km downwind is estimated to be 150 mg/m³. With a cloud passage time of 3.5 minutes this would present a very serious health hazard. Appendix 5 reviews available data on the toxicity of HF vapour and compares this with chlorine. In an attempt to crystallise thinking, the curves for 'fatalities' and 'distress' given in Fig 3 have been derived from information relevant to chlorine (Ref 6) using a 'weight for weight' equivalence between the hazards. When considering the estimated hazard ranges subsequently derived it should be noted that:

(a) There is no rigorous data to support the assumption made.

(b) The precise meaning of the 'fatal' and 'distress' curves is not brought out in Ref 6, eg it may well be that some fatalities could occur in the band between the two lines and that serious distress might be caused to susceptible members of the public, outside the hazard range predicted from the 'distress' curve.

(c) A 'weight for weight' equivalence corresponds to an HF/Chlorine ppm ratio of 3.5/1, ie for a given time of exposure, HF is assumed less dangerous than chlorine by a factor of 3.5 when considering concentrations expressed in ppm.

(Note:- TLVs are in the ratio 3:1 ppm)

With the above points in mind it is possible to use the derived curves for HF to estimate relevant hazard ranges for a cloud release of UF₆.

Estimated HF concentrations and cloud passage times at various distances downwind (taken from Appendix 4, Table 1) have been used to derive the Category 'F' line shown on Fig 3 for a 2 Tonne release.
In addition, again using data presented in Ref. 7 cloud concentrations and passage times have been calculated to plot the Category 'D' line shown on the Figure.

NB. No attempt has been made to correct for "gravity slumping", hence the results obtained may well be optimistic, particularly at the shorter ranges.

The assessed hazard ranges of the hydrogen fluoride (HF) from a 'quasi-instantaneous' vapour cloud release of 2 tonnes of UF₆ are:

<table>
<thead>
<tr>
<th>Category 'F' Weather</th>
<th>'Fatal' Hazard Range</th>
<th>1.8 Km</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>'Distress' Hazard Range</td>
<td>5.0 Km</td>
</tr>
<tr>
<td></td>
<td>'ERL' Hazard Range</td>
<td>14.0 Km</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Category 'D' Weather</th>
<th>'Fatal' Hazard Range</th>
<th>0.5 Km*</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>'Distress' Hazard Range</td>
<td>1.3 Km*</td>
</tr>
<tr>
<td></td>
<td>'ERL' Hazard Range</td>
<td>4.5 Km</td>
</tr>
</tbody>
</table>

*These ranges may be significantly increased by "Gravity Slumping".

7. URANYL FLUORIDE HAZARD RANGE

When UF₆ reacts with water two highly toxic compounds are formed:

\[ UF₆ + 2H₂O \rightarrow UO₂F₂ + 4HF \]

\[ 352 \quad 36 \quad 308 \quad 80 \]

In Section 6 we have concentrated attention on the possible implications of a cloud release of UF₆ in relation to the hazard from Hydrofluoric Acid (HF), however, the other product of the reaction Uranyl Fluoride (UO₂F₂) is also highly toxic since, in addition to taking one third of the total fluorine it also carries all the uranium. This compound would be generated as a fine aerosol in the cloud as it travelled and consideration must be given to both inhalation hazard and ground contamination.

No attempt has been made, in this 'overall' assessment, to analyse the problems in depth but it seems reasonable to take account of the toxicity of the material as a fluoride by applying a simple factor (1.5) to the HF concentrations already derived. This would not significantly affect the estimated hazard ranges and Appendix 6 concentrates attention on the chemical toxicity of the UO₂F₂ as a soluble uranium compound (the radiotoxicity of depleted 'tails' material will be less limiting). From the Appendix it will be seen that:-
In Class 'F' weather, if no account is taken of cloud depletion due to surface deposition, the assessed 'ERL' hazard range for uranium inhalation (25 km) is greater than the ERL hazard range for HF inhalation.

In view of the shortage of relevant information regarding deposition velocity for UO$_2$F$_2$ no attempt has been made to assess the consequences of the postulated release in terms of ground contamination.

8. FUTURE STORAGE - THE CHOICE OF CONTAINER

The alternative container, used at present for some UF$_6$ shipments to and from Springfields, and used as the feed container for the Centrifuge plants is the Type 48Y (nominal capacity 12 tonnes UF$_6$). Appendix 7 examines the effect of a fire on this type of container and concludes that it should be capable of withstanding a 800°C fire for the 30 minute period necessary to meet the IAEA Transport Regulations for a Type A package (Ref 8). However, if the fire continues beyond that time, or if its intensity is such as to produce a higher flame temperature, the risk of a catastrophic failure is high.

Such a failure (caused by expansion of the UF$_6$) would release a substantially larger vapour cloud in this case and the concentration of toxic vapour at any given point in the path of the cloud would increase approximately in proportion to the mass content. The potential consequences of a serious fire for both types of container are summarised in the following table.

<table>
<thead>
<tr>
<th>Type</th>
<th>Total heat required</th>
<th>Time to failure</th>
<th>Maximum release</th>
<th>'Fatal' hazard range*</th>
<th>'Distress' hazard range*</th>
<th>'ERL' hazard range*</th>
</tr>
</thead>
<tbody>
<tr>
<td>0236</td>
<td>$2 \times 10^5$ BTU</td>
<td>15-20 mins</td>
<td>2 Tonnes</td>
<td>1.8 Km</td>
<td>5 Km</td>
<td>14 Km</td>
</tr>
<tr>
<td>48F</td>
<td>$1.35 \times 10^6$ BTU</td>
<td>30-35 mins</td>
<td>12 Tonnes</td>
<td>4 Km</td>
<td>12 Km</td>
<td>30 Km</td>
</tr>
</tbody>
</table>

* Based on total generation of HF within the cloud, Category 'F' weather and no allowance for "gravity slumping" or buoyancy due to heat of reaction (see Section 5).

When considering the choice of container for the storage of the very large quantities of depleted material expected to arise from the Enrichment Plants in the future, account must be taken of all factors which could influence safety through to final disposal. At some future time stockpiling at Capenhurst will have reached its maximum and, unless a conversion plant is built on the site, it will then be necessary to:

- either introduce an additional hex. transfer operation immediately prior to shipment,
- or use the selected type of container to transport depleted material to another site for storage or reprocessing.

If the second alternative is chosen the suitability of the storage container for routine transport in quantity becomes an important consideration.

Clearly the 12 tonnes container has a distinct advantage from this point of view since:
(a) It is internationally recognised as being satisfactory for transporting hex on a routine basis.

(b) It meets the IAEA Regulations for Type A packaging with regard to a transport fire.

(c) Handling problems involved in both despatch and receipt are likely to be reduced since the load carried in a single container is the same as that in six Type 0236 containers.

The smaller number of 12 Tonne containers required for storage also gives important operational safety advantages during handling at Capenhurst (or elsewhere) since, if the chance of an accident involving a hex spill due to "spiking" is assumed equal for both types, the overall risk of such an accident is reduced by a factor of 6. (In practice, with the smaller numbers involved, increased attention is possible to the safety of individual handling operations and the reduction in risk of an accident is likely to be a factor of at least 10.) However, since the 2 Tonne container has its valve removed at an early stage before transporting, the chance of a spill is somewhat less.

One important disadvantage of the 12 Tonne container, from a safety viewpoint, must be taken into account:-

Since the rate of direct desublimation is slow the UF6 is normally introduced into these containers in liquid form and solidification takes several days. If the number of filling stations is limited it is necessary to disconnect the full container and move it before the contents have solidified. This sequence of operations offers the potential for a vapour cloud release without heating of the container.

Even with strict operational safeguards applied, the large number of containers to be handled on a routine basis in the future suggests that the chance of an accidental leak during handling must be high in relation to the chance of a catastrophic failure due to fire. However, the potential hazard range of such a leak will be governed by the release rate and the containment provided. Thus, despite the higher chance of a release, with careful design it should be possible to demonstrate a negligible hazard to the public.

To summarise, SRD concluded that:-

(a) Direct desublimation into a 2 Tonne vertical container of the type currently in use for 'tails' storage avoids the possibility of a substantial release of vapour during or immediately after filling. However, given appropriate attention to detail, it should be possible to ensure that a 12 Tonne container does not have the potential to generate a large UF6 vapour cloud during this phase of operation.
(b) As and when it becomes necessary to transport the total quantity of depleted UF$_6$ being generated by the Centrifuge Plants away from Copenhurst, the 12 Tonne container will have the major safety advantage that it is better able to withstand a serious transport fire.

Based primarily on these conclusions, recognising the potentially serious consequences of overpressurisation due to fire, it was recommended that long term arrangements for the storage and transport of depleted material should be based on use of the 12 Tonne horizontal container.
References


VAPOUR PRESSURE CURVES FOR HF AND UF₆

FIG 1
ASSUMED DENSITIES OF SOLID AND LIQUID UF₆

FIG 2
VARIATION OF CLOUD PASSAGE TIME AND AVERAGE CONCENTRATION WITH DISTANCE DOWNWIND

CONCENTRATION EXPOSURE PERIOD CURVES

DISTRESS RANGES

FATAL HAZARD RANGES

E.R.L. HAZARD RANGES

DERIVATION OF HAZARD RANGES

FIG 3
APPENDIX 1 - THE CHEMICAL EFFECTS OF UF₆ ON A HYDROCARBON FIRE

by F Abbey

1. There seem to be four possible routes by which the UF₆ might affect the fire chemically:

(a) by itself burning in air;
(b) by reacting with the hydrocarbon;
(c) by reacting with water vapour produced by hydrocarbon combustion;
(d) by catalysing the reaction between the hydrocarbon and air.

2. The statement occurs in a number of places in the literature that UF₆ does not react with dry oxygen, nitrogen or air and this is what one might expect from the large free energy of formation of this material (ΔF°₂₉₈ = -486.3 k cals/mol as against, for UO₂, for example, ΔF°₂₉₈ = -246.55 k cals/mol)*. Thus, UF₆ will not contribute to the fire by itself burning in the normal sense.

3. On the other hand it is well known that UF₆ reacts with many organic materials to give UF₄ and a mixture of HF and fluorinated carbon compounds. Consider for example, the standard free energy associated with the following possible reaction with methane, a hydrocarbon for which the necessary thermodynamic data (at 298°K) are available:

4 UF₆ (gas) + CH₄ → 4 UF₄ + CF₄ + 4 HF

The free energy change for this reaction can be calculated as follows:

ΔF°₂₉₈ (k cals)

4 UF₆ (gas) → 4 U + 12 F₂

4 U + 8 F₂ → 4 UF₄

CH₄ → C + 2 H₂

C + 2 F₂ → CF₄

2 H₂ + 2 F₂ → 4 HF

Hence, the reaction is favourable on free energy grounds and at flame temperatures might well proceed rapidly.
Since reactions of this kind are possible, one must consider the extent to which they might contribute heat to the fire. Two extreme cases will be examined, one where the fluorine released from the hex reacts entirely with hydrogen and one where it reacts entirely with carbon. By comparing the difference between the heat generated in these reactions and the heat which would have been generated had the hydrogen or carbon burned to H$_2$O and CO$_2$ respectively one can gain an idea of the effects on the fire.

* All thermodynamic data are taken from the following references:
  US Bureau of Mines Circular 500

Case 1 - Fluorine Reacting with Hydrogen

\[
{\text{UF}_6} + {\text{H}_2} \rightarrow {\text{UF}_4} + 2\text{HF}
\]

The heat of reaction $\Delta H^\circ$ for this reaction can be calculated as follows (the two temperatures chosen are the extremes of the range for which data is readily available):

<table>
<thead>
<tr>
<th>$\Delta H^\circ$ (k cals)</th>
</tr>
</thead>
<tbody>
<tr>
<td>298$^\circ$K</td>
</tr>
<tr>
<td>$\text{UF}_6$ (gas) $\rightarrow$ U + 3F$_2$</td>
</tr>
<tr>
<td>2F$_2$ + U $\rightarrow$ UF$_4$</td>
</tr>
<tr>
<td>H$_2$ + F$_2$ $\rightarrow$ 2HF</td>
</tr>
</tbody>
</table>

This is to be compared with the reaction

\[
\text{H}_2 + \frac{3}{2}\text{O}_2 \rightarrow \text{H}_2\text{O} \text{ (gas)}
\]

for which the heats of reaction are -58.547 k cals at 298$^\circ$K and -49.480 k cals at 1000$^\circ$K respectively. Hence, the net increase in the amount of heat generated if the reaction with UF$_6$ takes place is 65.7 - 58.547 = 7.2 k cals per mol of UF$_6$ at 298$^\circ$K and 61.2 - 49.480 = 11.7 k cals per mol of UF$_6$ at 1000$^\circ$K. These are very small amounts of heat and for a 100 tonne shipment of UF$_6$, for example, would be equivalent to the addition of about 3 tonnes of additional hydrocarbon to the fire.
Case 2 - Fluorine Reacting with Carbon

The most conservative result will be obtained when each carbon reacts with four fluorines, since the fewest number of carbons will then be prevented from burning to CO$_2$, i.e. when the reaction is:

$$\text{UF}_6 \text{ (gas)} + \frac{3}{2} \text{C} \rightarrow \text{UF}_4 + \frac{3}{2} \text{CF}_4$$

The heats of reaction are then as follows:

<table>
<thead>
<tr>
<th>Reaction</th>
<th>$\Delta H^0$ (k cal)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\text{UF}_6 \text{ (gas)} \rightarrow \text{U} + 3 \text{F}_2$</td>
<td>$+505.7$ $+504.1$</td>
</tr>
<tr>
<td>$2 \text{F}_2 + \text{U} \rightarrow \text{UF}_4$</td>
<td>$-443$ $-436$</td>
</tr>
<tr>
<td>$\frac{3}{2} \text{C} + \text{F}_2 \rightarrow \frac{3}{2} \text{CF}_4$</td>
<td>$-81.25$ $-81.35$</td>
</tr>
<tr>
<td>$\text{UF}_6 \text{ (gas)} + \frac{3}{2} \text{C} \rightarrow \text{UF}_4 + \frac{3}{2} \text{CF}_4$</td>
<td>$-18.55$ $-13.25$</td>
</tr>
</tbody>
</table>

These are to be compared with the heats of reaction for

$$\frac{3}{2} \text{C} + \text{O}_2 \rightarrow \text{CO}_2$$

which are $-47.025$ k cal at $298^0K$ and $-47.2$ k cal at $1000^0K$. Hence, in this case if the reaction with UF$_6$ takes place it will actually result in the net removal of heat from the fire.

In summary, therefore, it is clear from these calculations that in terms of the total heat generated the UF$_6$ can no more than marginally increase the severity of fire, if at all, by reacting with the hydrocarbon. In principle the possibility exists that the reaction between hex and the hydrocarbon may be faster than the reaction between the hydrocarbon and air and there is the related physical effect that the UF$_6$ is released at the centre of the fire whereas air must diffuse in from outside. Either of these mechanisms could only increase the rate of combustion, however, and not the total heat generated.

4. As an alternative to reacting directly with the hydrocarbon, it is possible for the UF$_6$ to react with water produced by the combustion of the hydrocarbon. The most exothermic hydrolysis would occur if both reactants were in the gas phase and both products were formed as aqueous aerosols.

$$\text{UF}_6 (g) + 2 \text{H}_2\text{O} (g) \rightarrow \text{UO}_2\text{F}_2 (aq) + 4 \text{HF} (aq) \Delta H = -33.5 \text{ k cal/mole}$$

This heat of reaction is almost certainly an overestimate since it is probable that the hydrolysis products would be in an anhydrous form in the vicinity of a fire. However, taking this value for the heat of reaction the complete hydrolysis of 100 Tonnes of UF$_6$ would be approximately equivalent, in terms of additional heat generation, to burning a further 2 Tonnes of hydrocarbon fuel.
It should be remembered that this represents a pessimistic maximum value, and in reality the additional heat liberated by hydrolysis is likely to be significantly smaller due to the removal of UF₆ by direct reaction with the hydrocarbon, by evaporation and dispersion of unreacted UF₆ and by the formation of anhydrous hydrolysis products.

5. The remaining method by which the hex might affect the fire is by catalysing the reaction between the hydrocarbon and air. A priori this seems very unlikely and, in any case, would again increase only the rate of combustion and not the total heat generated.
APPENDIX 2 – THE EFFECT OF A FIRE ON A 2 TONNE CONTAINER (TYPE 0236)

The containers are initially filled and sealed at a temperature of about 3°C and a volume of air is trapped above the UF₆. As the temperature is increased the partial pressure of the trapped air will rise due to (a) the compression ratio resulting from UF₆ expansion reducing the free volume, and (b) the ratio of final to initial absolute temperatures. The present limit for normal filling is set at 2150 Kgs but the manufacturing drawings do not specify a container volume.

Consider two alternatives, each of which is thought to be well within the possible manufacturing range:— (1) 23.5 ft³ (2) 22.0 ft³

1. To calculate internal pressure @ 80°C

<table>
<thead>
<tr>
<th>Container Volume</th>
<th>UF₆ Vol</th>
<th>Air Vol</th>
<th>Compression Ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td>Case 1</td>
<td>665 litres</td>
<td>416</td>
<td>610</td>
</tr>
<tr>
<td>Case 2</td>
<td>623 litres</td>
<td>416</td>
<td>610</td>
</tr>
</tbody>
</table>

These figures are based on UF₆ density

5.18 gm/cc @ 3°C
3.53 gm/cc @ 80°C

Case 1
Partial pressure (Air) = 14.7 x 4.5 x 353₉⁰K
= 83.5 psia
Partial pressure UF₆ = 35.5 psia
TOTAL = 119 psia

Case 2
Partial pressure (Air) = 14.7 x 15.9 x 353₉⁰K
= 295.0 psia
Partial pressure UF₆ = 35.5 psia
TOTAL = 330.5 psia

The high compression ratio in Case 2 is due to the very limited free space remaining when the UF₆ expands to 80°C. More generally, if UF₆ expansion leads to a reduction of free volume to less than (say) 25 litres, compression ratios in excess of 10 are likely to result.
In view of the complex dependence of failure temperature on container volume, mass content and strength there is little to be gained by considering a series of specific cases but the conclusion can be drawn that failure will inevitably occur if the UF$_6$ expands to fill the container.

2. To calculate the rate at which heat is absorbed

Using assumptions taken from Ref. 9, para. 720 and listed below, the rate of heat absorption by the container is approximately 200 BTU/sec or 3000 Kcal/min.

- Flame emissivity = 0.9
- Absorptivity = 0.8
- Flame temperature = 800°C = 1932°R
- Container temperature = 150°C = 762°R
- Surface Area = 40.5 ft$^2$

\[ Q \]
\[ \frac{t}{Q} = 0.9 \times 0.8 \times 40.5 \times 1.73 \times 10^{-9} (1932^4 - 762^4) \]
\[ = 5.05 \times 10^{-8} (1.4 \times 10^{13} - 3.2 \times 10^{11}) \]
\[ = 7.06 \times 10^5 \text{ Btu/hr} \]
\[ = \frac{7.06 \times 252}{60} \times 10^2 \approx 3000 \text{ Kcal/min} \]

3. Time to reach atmospheric sublimation temperature (330°K)

- Assume Initial temperature = 15°C = 298°K
- UF$_6$ content = 2000 Kg/m (nominal)
- Container heat capacity - negligible
- Enthalpy @ 330°K = 8.860
- @ 298°K = 7.545

\[ \text{Difference} = 1.315 \text{ K cal/mole} \]

\[ \frac{2 \times 10^6 \text{ gm}}{352 \text{ gm/mole}} = 5.7 \times 10^3 \text{ moles} \]

\[ \therefore \text{Total heat needed} = 7.5 \times 10^3 \text{ K.cal} \]

\[ \therefore \text{Time} = 2.5 \text{ minutes} \]

From this point, all extra heat will be available to generate vapour at atmospheric pressure.
4. **Time to melt total contents**

Liquid enthalpy @ 337.2°K = 13.760

\[
\begin{align*}
298 & = 7.545 \\
& \quad 6.215 \text{ K cal/mole}
\end{align*}
\]

Total heat needed = 3,54 x 10^4 K cal

\[\text{Time 12 minutes}\]

5. **Rate of temperature rise of liquid**

This will depend directly on the specific heat of the UF₆ which varies with temperature.

Assume an approximate value of 46 cal/mole °C

Heat to raise temperature 1°C = 46 x 5.7 = 262 K cal

\[\text{Rate of temperature rise} = \frac{3000}{262} = 11.5 \text{°C/min}\]

6. **Mass of UF₆ vaporised (12 minutes)**

If the expansion on change of state causes overpressurisation the fraction of UF₆ vaporised can be calculated by equating the heat gain liquid → vapour to the heat loss liquid → solid.

Liquid enthalpy = 13760

Solid enthalpy (330°K) = 8860

Heat Loss = 4900 cal/mole

Sublimation heat (330°K) = 11500 cal/mole

\[\text{Fraction Vaporised} = \frac{4900}{11500} = 42.5\%\]

\[\text{Mass Vaporised} = 850 \text{ Kgs}\]

7. **Mass of UF₆ vaporised (20 minutes)**

If we arbitrarily assume the container fails after heating for 20 minutes the fraction of the UF₆ which will vaporise can be calculated as follows:
Time to reach sublimation temperature = 2.5 mins

Heat available for sublimation = $3000 \times (20 - 2.5) \times K \cdot cal$

= 5250 K cal

Heat of sublimation ($330^\circ K$) = 11.5 K cal/mole

"" Quantity of vapour = $\frac{5250}{11.5}$ moles

= $\left( \frac{5250}{11.5} \times \frac{352}{1000} \right)$ K.gms

Mass Vaporised = 1600 K.gms
APPENDIX 3 - FACTORS AFFECTING THE DENSITY OF A UF₆ CLOUD
by S R Haddock

Pure UF₆ vapour released at the sublimation temperature (56.5°C) corresponding to 1 atm. pressure is approximately 11 times denser than air and clearly would exhibit gravity slumping similar to that observed in the Van Ulden experiment. Very large volumes of air would have to be entrained in the UF₆ vapour cloud as it was formed if its density was to be reduced to a value at which it might be expected to exhibit neutral buoyancy. It might be argued that mixing with air at ambient temperature would cool the UF₆ sufficiently to cause condensation; precipitation of UF₆ particulate could then reduce the overall density of the cloud. However, although the temperature of the UF₆ will fall quite rapidly as it mixes with air because of its low heat capacity relative to that of air, the partial pressure of UF₆ will also decrease as the cloud is diluted. As this reduction in partial pressure should be sufficient to ensure that the UF₆ remains in vapour phase, unless the air is very cold (< 0°C), it seems unlikely that this is a possible mechanism for significantly reducing the cloud density.

Another possible mechanism for reducing the cloud density is the hydrolysis of UF₆ by atmospheric water which will convert it to uranyl fluoride and hydrogen fluoride.

\[
UF₆ (g) + 2 H₂O (g) \rightarrow UO₂F₂ (aq) + 4 HF (aq) \quad \Delta H = -349.4 \text{ kJ/mol}
\]

It is known that HF may exist in a highly associated form in the vapour phase but under the condition of partial pressure and temperature likely to prevail in the cloud it is probable that the HF will be formed as the monomer (i.e. less dense than the surrounding air). It should be noted, however, that although the hydrolysis of one molecule of UF₆ will produce four molecules of HF, the density decrease associated with this will be offset to some extent by the removal of two molecules of water vapour which is even less dense than HF monomer.

The other hydrolysis product, UO₂F₂, will appear as a fine aerosol and will precipitate from the cloud at a rate dependent on particle size and the cloud dimensions. Although the conversion of UF₆ to UO₂F₂ is accompanied by a modest decrease in mass it also involves a large reduction in volume. Thus, unless it can be demonstrated that the UO₂F₂ precipitates rapidly from the cloud, it seems that its formation is more likely to contribute an increase to the cloud density rather than a reduction and further offset the density decrease associated with HF formation.

It can be seen that the hydrolysis of UF₆ is strongly exothermic which will reduce the cloud density. However, by the time enough air has been mixed with the cloud to cause significant quantities of UF₆ to be hydrolysed, the thermal capacity of the cloud will probably be sufficiently large to ensure that any temperature increase will be small. The complete hydrolysis of 2 Tonne of UF₆, neglecting any deposition or reaction with ground moisture, would require approximately 200 kg of water which, even at high relative humidities, is unlikely to be immediately available. Moreover, as soon as hydrolysis products appear within the cloud there will be other species competing with the UF₆ for the available water. Under a wide range of conditions of temperature and pressure mixtures of gaseous HF and water vapour are capable of condensation to form aqueous aerosols. Similarly
$\text{UO}_2\text{F}_2$ is very hygroscopic and will rapidly absorb water from its surroundings to form an aqueous aerosol. Thus, the hydrolysis of UF₆ may not be as complete as the total available mass of water vapour might suggest.

On balance it appears that although hydrolysis of UF₆ will be accompanied by a decrease in cloud density due to HF formation, heat generation and enhanced precipitation of uranium as $\text{UO}_2\text{F}_2$, it seems improbable that this will effect the large reduction necessary to render the cloud buoyancy neutral in the early stages of dispersion.
APPENDIX 4 - DOWNWIND CONCENTRATIONS OF HF - 2 TONNE UF₆ RELEASE

UF₆ reacts with moisture as follows:

\[
\text{UF}_6 + 2\text{H}_2\text{O} \rightarrow \text{UO}_2\text{F}_2 + 4\text{HF}
\]

For a 2 tonne release of UF₆, the total generation of HF is therefore 
\[
2000 \times 60 = 456 \text{ Kg}
\]
illustrated by considering two alternative cases:

**Case 1 Quasi-instantaneous release (Cat. F weather)**

If a UF₆ vapour cloud is formed by the disruptive failure of an overheated container, the HF will be generated within the cloud as it travels and, if density slumping is ignored, this is equivalent to an instantaneous HF release. Fig. 6 of Ref. 8 can now be used to estimate the Total Integrated Dose (TID) from the cloud, at various distances, for a unit release (Table 1, column 2). Multiplication by the total release then gives TID from the cloud for our case (column 3).

For toxic substances it is also important to know the concentration and to obtain this we determine the time of passage of the cloud, using the following formula originally proposed by Pasquill

\[
t = 1.6 \times 10^{-2} \varphi \frac{d}{u}
\]

\(t\) (seconds), \(\varphi\) (degrees), \(d\) (metres), \(u\) (metres/sec).

**Note:** This assumes an along-wind spread equal to "arc width" and the cloud centre moving at the full wind speed.

The average concentration can now be obtained by dividing the TID by cloud passage time and peak concentration is roughly twice the average.

On this basis we obtain:

<table>
<thead>
<tr>
<th>Distance (metres)</th>
<th>Fig 6 (gm sec/m³ per gm)</th>
<th>TID (gm sec/m³)</th>
<th>Cloud passage (sec)</th>
<th>Av. Conc. (mgm/m²)</th>
<th>Peaks (mgm/m³)</th>
</tr>
</thead>
<tbody>
<tr>
<td>(10^3)</td>
<td>(3.4 \times 10^{-4})</td>
<td>157</td>
<td>75</td>
<td>2100</td>
<td>4200</td>
</tr>
<tr>
<td>(3 \times 10^3)</td>
<td>(6.6 \times 10^{-5})</td>
<td>30</td>
<td>203</td>
<td>148</td>
<td>300</td>
</tr>
<tr>
<td>(10^4)</td>
<td>(1.25 \times 10^{-5})</td>
<td>5.7</td>
<td>603</td>
<td>9.5</td>
<td>19</td>
</tr>
<tr>
<td>(3 \times 10^4)</td>
<td>(3.3 \times 10^{-6})</td>
<td>1.5</td>
<td>1566</td>
<td>0.96</td>
<td>2</td>
</tr>
</tbody>
</table>

It will be seen from this table that the peak concentration is approximately four times the "acceptable" level of 4.5 mgm/m³ at a distance of 10 Km downwind.
Case 2 - Generation at "Uniform" Rate for 30 Minutes (Category F weather)

Again using Fig. 6 of Ref. 8 but now determining the cloud concentration direct from a uniform release rate of \( 456 \times 1000 \text{ gm/sec} = 253 \text{ gm/sec} \) we obtain the results presented in columns 2 and 3 of Table 2. Alternatively if it is assumed that wind direction changes sufficiently during the period of the release one can use Fig. 8 of Ref. 8 to obtain the results presented in columns 4 and 5 of the Table.

**TABLE 2**

<table>
<thead>
<tr>
<th>Distance (metres)</th>
<th><strong>Fig. 6</strong> (gm/m(^3) per gm/sec)</th>
<th>Concentration (mgm/m(^3))</th>
<th><strong>Fig. 8</strong> (gm/m(^3) per gm/sec)</th>
<th>Concentration (mgm/m(^3))</th>
</tr>
</thead>
<tbody>
<tr>
<td>( 10^3 )</td>
<td>( 3.4 \times 10^{-4} )</td>
<td>86.0</td>
<td>( 1.1 \times 10^{-4} )</td>
<td>28.0</td>
</tr>
<tr>
<td>( 3 \times 10^3 )</td>
<td>( 6.6 \times 10^{-5} )</td>
<td>16.7</td>
<td>( 2.2 \times 10^{-5} )</td>
<td>5.57</td>
</tr>
<tr>
<td>( 10^4 )</td>
<td>( 1.25 \times 10^{-5} )</td>
<td>3.17</td>
<td>( 4.2 \times 10^{-6} )</td>
<td>1.06</td>
</tr>
<tr>
<td>( 3 \times 10^4 )</td>
<td>( 3.3 \times 10^{-6} )</td>
<td>0.83</td>
<td>( 1.1 \times 10^{-6} )</td>
<td>0.28</td>
</tr>
</tbody>
</table>

Note: The concentrations derived from Fig. 8 for a "Prolonged" release are **average** figures for the total release period and rely on the assumption that the wind will swing sufficiently, during the period of the release, to miss any selected point for two thirds of the total release time.

**Conclusion**

Comparing the results in Tables 1 and 2 it can be seen that, if a rapid generation of HF is assumed, the peak concentration at a distance of 3 Km (approximately 2 miles) downwind in Category 'F' weather can be over fifty times the mean concentration derived for a prolonged (30 minute) release at uniform rate. This factor is made up as follows:

\[
\text{Ratio of Cloud Passage times} \frac{1800}{200} = 9 \\
\text{Peak to Average Factor (Rapid Release)} = 2 \\
\text{Wind Spread Factor (Prolonged Release)} = 3 \\
\text{Overall Factor} = 54
\]

Both these calculations ignore the possibility of a substantial UF\(_6\) vapour cloud being carried downwind, but also ignore any advantage to be gained from buoyancy effects due to heat generation and/or the lower density of the HF. When considering the 'potential' hazard to the surrounding population from HF following a UF\(_6\) release, it is important to examine all factors which may influence the maximum rate at which HF can be generated.
APPENDIX 5 - THE SIMILARITIES BETWEEN THE TOXICITIES OF HYDROGEN FLUORIDE VAPOUR AND CHLORINE GAS by S R Haddock

The following physiological effects of HF vapour have been observed. (Ref. 10)

a. Effects on Rabbits and Guinea Pigs

<table>
<thead>
<tr>
<th>CONC. mg/m³</th>
<th>EFFECT</th>
</tr>
</thead>
<tbody>
<tr>
<td>15</td>
<td>'Long term' exposure tolerable</td>
</tr>
<tr>
<td>25</td>
<td>No deaths after 41 hours, but subsequent weight loss</td>
</tr>
<tr>
<td>&lt; 50</td>
<td>Mild irritation, coughing and sneezing</td>
</tr>
<tr>
<td>&gt; 50</td>
<td>Severe irritation, paroxysms of coughing, copious discharge from eyes and nose</td>
</tr>
<tr>
<td>&lt; 100</td>
<td>No fatality after 5 hour exposure, but severe irritation</td>
</tr>
<tr>
<td>500</td>
<td>Not fatal in 15 minutes, but animals severely affected</td>
</tr>
<tr>
<td>1000</td>
<td>Not fatal in 30 minutes, but tissue damaged</td>
</tr>
<tr>
<td>1500</td>
<td>Fatal in 5 minutes</td>
</tr>
</tbody>
</table>

b. Effects on Man

<table>
<thead>
<tr>
<th>CONC. mg/m³</th>
<th>EFFECT</th>
</tr>
</thead>
<tbody>
<tr>
<td>25</td>
<td>Tolerable for several minutes. Mild irritation of eyes and nose</td>
</tr>
<tr>
<td>50</td>
<td>Immediate sour taste. Irritation of eyes and nose</td>
</tr>
<tr>
<td>100</td>
<td>'Maximum tolerable exposure' 1 minute. Sour taste, irritation of eyes and nose, smarting of the skin</td>
</tr>
</tbody>
</table>

Considerably more information is available on the toxicology of chlorine, particularly on its physiological effect as a function of concentration and exposure time. Comparison with the data on HF seems to indicate that the acute toxicities of these two gases are quite similar. For example, the responses to a 1 minute exposure to relatively low concentrations of chlorine are described as follows:-

<table>
<thead>
<tr>
<th>CONC. mg/m³</th>
<th>EFFECT</th>
</tr>
</thead>
<tbody>
<tr>
<td>15</td>
<td>Irritation</td>
</tr>
<tr>
<td>50</td>
<td>Coughing and mild distress</td>
</tr>
<tr>
<td>150</td>
<td>Distress</td>
</tr>
</tbody>
</table>
In view of the limited nature of the quantitative data available for HF it seems reasonable to assume that in terms of mass its toxicity is equivalent to that of chlorine for the present purposes of assessing the consequences of an accidental release.

It should be noted, however, that there are no rigorous data to support this assumption. Moreover, postmortem examinations of animals repeatedly exposed to relatively low levels of HF have given evidence of damage to lung, liver and kidney of a type that suggested that some process was involved beyond that usually associated with a purely irritant gas. Furthermore, it has been demonstrated that storage of fluoride in the bone occurs as a result of repeated exposures to concentrations that are only slightly above the irritant level. In view of this it may be necessary to exercise some caution in comparing HF with chlorine for exposures other than those which are rapidly fatal. For example, exposure to a large but sub-lethal dose of HF may have some long term effect on the health of an individual that would not be produced by a similar exposure to chlorine because of this bone storage mechanism.
APPENDIX 6 - INHALATION HAZARD RANGE OF URANYL FLUORIDE (UO₂F₂)

For a 2 Tonne release of UF₆ complete reaction with moisture will generate

\[
\frac{2000 \times 308}{352} = 1750 \text{ Kgm of } UO₂F₂,
\]

containing \[
\frac{2000 \times 238}{352} = 1350 \text{ Kgm of depleted uranium}
\]

If the release is in the form of a UF₆ vapour cloud this UO₂F₂ will be generated, as a fine aerosol, within the cloud as it travels downwind. Some ground deposition will inevitably occur, resulting in both cloud depletion and surface contamination. No authoritative data exists for UO₂F₂ to permit an accurate assessment of the rate at which the cloud is likely to be depleted in specified weather conditions but two cases are considered below to illustrate the importance of "deposition velocity".

Case 1 No deposition

For the purpose of this report the Emergency Reference Level (ERL) for inhalation of HF has been taken as 4.5 mg/m³ and the 'ERL' hazard ranges assessed are based on this figure. It will be seen from Section 2 that the 1 hour limit recommended in Ref. 3 for inhalation of uranium in soluble form, is 4 mg/m³. Clearly therefore, if ground deposition is neglected, the 'ERL' hazard range for UO₂F₂ will be greater than that for HF.

Using the basis outlined in Appendix 4 we obtain:

For Class F weather (Wind speed 2 m/s)

<table>
<thead>
<tr>
<th>Distance (metres)</th>
<th>gm sec/m³ per gm</th>
<th>TID (Uranium) gm sec/m³</th>
<th>Cloud passage time (secs)</th>
<th>Average Concentration mg/m³ (Uranium)</th>
</tr>
</thead>
<tbody>
<tr>
<td>10⁴</td>
<td>1.25 x 10⁻⁵</td>
<td>16.9</td>
<td>603</td>
<td>28.0</td>
</tr>
<tr>
<td>3 x 10⁴</td>
<td>3.3 x 10⁻⁶</td>
<td>4.47</td>
<td>1566</td>
<td>2.85</td>
</tr>
</tbody>
</table>

Hence, interpolating between these values, we conclude that the concentration in the cloud will be 4 mg/m³ (the chosen ERL) at a distance of approximately 25 Km.

For Class D weather (Wind speed 5 m/s)

The hazard range is estimated to be approx 8 Km.

Case 2 With deposition (vg = 10⁻² m/s)

Ref 21 suggests a method of calculating ground deposition and cloud depletion for a fission product release, making use of the concept of a deposition velocity (vg) where:

\[ vg = \text{deposited activity per unit area} \div \text{cloud dosage}. \]
Class F Weather (Wind speed 2 m/s)

Ref 10 Fig. 2 provides a series of curves of axial cloud dosage against distance downwind for a short release at ground level in Class F weather, taking account of cloud depletion.

These curves are derived assuming a fixed source at the origin whereas the UO₂F₂ would be generated within the cloud as it travelled.

If a deposition velocity of 10⁻² m/s is arbitrarily assumed (for illustration) cloud dosages can be read directly from Curve 1 (Fig 7) and the following results are obtained:

<table>
<thead>
<tr>
<th>Distance (metres)</th>
<th>gm sec/m³ per gm</th>
<th>TID (Uranium) gm sec/m²</th>
<th>Cloud passage time (secs)</th>
<th>Average Concentration (Uranium) mgm/m³</th>
</tr>
</thead>
<tbody>
<tr>
<td>3 x 10³</td>
<td>2.2 x 10⁻⁵</td>
<td>29.7</td>
<td>203</td>
<td>146</td>
</tr>
<tr>
<td>10⁴</td>
<td>2 x 10⁻⁶</td>
<td>2.7</td>
<td>603</td>
<td>4.6</td>
</tr>
<tr>
<td>3 x 10⁴</td>
<td>10⁻⁷</td>
<td>0.135</td>
<td>1566</td>
<td>0.086</td>
</tr>
</tbody>
</table>

Hence it can be concluded that if a deposition velocity of 10⁻² m/s is used the estimated hazard range for inhalation of UO₂F₂ is reduced from 25 km to approx 10 km i.e. less than the hazard range for HF (14 km).

Class D Weather (Wind speed 5 m/s)

Using Ref 10 Fig. 1 the following results are obtained:

<table>
<thead>
<tr>
<th>Distance (metres)</th>
<th>gm sec/m³ per gm</th>
<th>TID (Uranium) gm sec/m²</th>
<th>Cloud passage time (secs)</th>
<th>Average Concentration (Uranium) mgm/m³</th>
</tr>
</thead>
<tbody>
<tr>
<td>3 x 10³</td>
<td>2.5 x 10⁻⁶</td>
<td>3.38</td>
<td>162</td>
<td>20.8</td>
</tr>
<tr>
<td>10⁴</td>
<td>2.9 x 10⁻⁷</td>
<td>0.398</td>
<td>610</td>
<td>0.65</td>
</tr>
</tbody>
</table>

By interpolation, continuing to use 4 mgm/m³ as the inhalation limit for consistency (although the cloud passage time is now less than 0.1 hours) the hazard range is estimated to be approx. 5.5 km.

In Class D weather the cloud depletion due to deposition is less marked than in Class 'F' weather and the 'ERL' hazard range of the UO₂F₂ is likely to be comparable with or greater than the 'ERL' hazard range for HF.
APPENDIX 7 - THE EFFECT OF A FIRE ON A 12 TONNE CONTAINER

Basic Assumptions:-

- Volume = 142.7 ft³
- Weight = 5200 lb
- Surface Area = 155 ft²
- UF₆ Content = 12 Tonnes
- HF content = 1% mole (600 ppm by wt)
- Air content Case 1 NIL
  Case 2

Initial Ambient Temperature = 15°C (60°F)

Rate of heat input, 800°C flame temperature

Using the notation of Appendix 2

\[
\frac{Q}{t} = 0.9 \times 0.8 \times 155 \times 1.73 \times 10^{-9} \quad (1932^4 \text{ - negligible})
\]

\[
= 1.93 \times 10^{-7} \quad (1.4 \times 10^{13} \text{ - neg})
\]

\[
= 2.7 \times 10^6 \text{ BTU/hour}
\]

\[
= \frac{2.7 \times 252}{60} \times 10^3 = 1.13 \times 10^4 \text{ Kcal/min}
\]

Heat capacity of container

Assume specific heat = 0.117 Kcal/Kgm°C

\[
5200 \times 0.454 \times 0.117 = 277 \text{ Kcal/°C}
\]

Time to melt total contents

| Liquid Enthalpy @ 337.2°C | 13.760 |
| Solid Enthalpy @ 298°C | 7.545 |

\[
12 \text{ Tonnes} = \frac{1.2 \times 10^7}{352} = 3.41 \times 10^4 \text{ moles}
\]

Heat to melt UF₆ = 2.13 \times 10^5 \text{ Kcal}

Heat to raise container temp. = 1.08 \times 10^4 \text{ Kcal}
Total heat uptake = $2.24 \times 10^5$ Kcal

.'. Time taken = $\frac{2.24 \times 10^5}{1.13 \times 10^4}$ = 19.8 minutes

Subsequent rate of temperature rise

Assume 46 cal/mole°C as Sp. Ht. of UF₆
Heat to raise UF₆ by 1°C = 1570 Kcal
Heat to raise container 1°C = 277 Kcal
= 1847 Kcal/°C

'. Rate of temp rise = \( \frac{1.13 \times 10^4}{1.85 \times 10^3} \) = 6.13°C/minute

'. Temperature at 30 minutes = \((64.2 + 6.13 \times 10.2)^°C\) = 130°C

To calculate failure temperature

Case 1 No air present

Under these conditions the container will fail when all voidage is taken up unless the vapour pressure reaches an unacceptable value prior to this time.

Density to fill void = \( \frac{12 \text{ Te}}{4.04 \text{m}^3} \) = 2.97 gm/cc

'. From Fig 2 Failure temp = 150°C

At this temperature Vapour Pressure UF₆ = 178 psia
UF₆ + 1% HF = 286 psia

Hence the container will fail due to loss of all voidage at approx. 150°C.

Time to failure = 19.8 + \( \frac{150 - 64.2}{6.13} \)
= 34 minutes

Case 2 Container at atmospheric pressure due to leakage

Initial pressure of air 760-50 mm = 13.7 psia

Initial volume of air \((4.04 - \frac{12}{5.1})\text{m}^3 = 1.68 \text{ m}^3\)
Try 130°C

\[ UF_6 \text{ volume} \quad = \frac{12}{3.18} = 3.57 \text{ m}^3 \]

\[ \therefore \text{ Air volume} \quad = 4.04 - 3.57 = 0.47 \text{ m}^3 \]

\[ \text{Air pressure} \quad = 13.7 \times \frac{1.67 \times 403}{0.47 \times 298} = 66 \text{ psia} \]

Even with 1% HF present this should not fail the container.

Try 140°C

\[ UF_6 \text{ volume} \quad = \frac{12}{5.10} = 3.37 \text{ m}^3 \]

\[ \therefore \text{ Air volume} \quad = 0.17 \text{ m}^3 \]

\[ \text{Air pressure} \quad = 13.7 \times \frac{1.67 \times 413}{0.17 \times 298} = 186 \text{ psia} \]

\[ \therefore \text{ Total pressure} \quad = 186 + 178 = 364 \text{ psia \ (No \ HF)} \]

\[ 186 + 286 = 472 \text{ psia \ (1\% \ HF)} \]

Although the pressure is above the test value, failure is unlikely to occur unless the container has been weakened, e.g. due to corrosion in service. From these two cases it can be seen that the presence of air in the container does not significantly reduce its ability to resist fire.

To calculate size of Vapour Cloud (150°C failure)

\[ \text{Total heat input} \quad = 1.13 \times 10^4 \times 33.8 \quad = 3.85 \times 10^5 \text{ Kcal} \]

\[ \text{Heat to container} \quad = 277 \times 135 \quad = 3.74 \times 10^4 \text{ Kcal} \]

\[ \therefore \text{ Heat to } UF_6 \quad = 3.48 \times 10^5 \text{ Kcal} \]

\[ \therefore \text{ Heat per mole } UF_6 \quad = \frac{3.48 \times 10^5}{5.41 \times 10^4} = 10.2 \text{ Kcal/mole} \]

\[ \text{Heat to reach sublimation temp. (app 3)} \quad = 1.315 \text{ Kcal/mole} \]

\[ \text{Heat available for sublimation} \quad = 8.88 \text{ Kcal/mole} \]

\[ \text{Total } UF_6 \text{ vapourised} \quad = \frac{12 \times 8.88}{11.5} \quad = 9.25 \text{ Tonnes} \]

If the failure is 'catastrophic' the 2.75 tonnes of solid UF$_6$ is likely to be carried out of the container in the vapour cloud.
APPENDIX 9 - THE CHANCE OF AN AIRCRAFT CRASH AFFECTING A UF₆ STORAGE AREA

For the purpose of illustration we will assume that 15,000 Tonnes of depleted uranium may eventually be stored as UF₆ Tails in 2 Tonne vertical containers.

Size of Storage Area

With 1" spacing between the containers and a "square pitch" array the ground area occupied is:

\[ 32" \times 32" = 0.8 \text{ sq yards per container} \]

If provision for access is made, this area will need to be increased by a factor estimated as 9/4.

<table>
<thead>
<tr>
<th>Effective Area per Container</th>
<th>Quantity of depleted uranium</th>
<th>Quantity of UF₆</th>
<th>Number of containers</th>
<th>Storage Area required</th>
</tr>
</thead>
<tbody>
<tr>
<td>( = 1.8 \text{ sq yards} )</td>
<td>( = 15,000 \text{ Te} )</td>
<td>( = 22,000 \text{ Te} )</td>
<td>( = 11,000 )</td>
<td>( = 6.5 \times 10^{-3} \text{ sq miles} )</td>
</tr>
</tbody>
</table>

NOTE: If the 12 Te Horizontal container is used the ground area required will depend on layout but could well be larger than with the 2 Te vertical container.

Light Civil Aircraft

In this case the 'crash zone' area is likely to be small and it is appropriate to consider direct impact on the storage area only. If crashes which have occurred within a 5 nautical mile radius of the airfield of landing or departure are ignored, available data suggests an average of 15 per annum on a UK land area of about \( 10^5 \) square miles. Hence assuming a random distribution:

\[ P = \frac{15 \times 6.5 \times 10^{-3}}{10^5} = 1 \times 10^{-6} \text{ per year} \]

Heavy Aircraft (\( > 5000 \text{ Kg} \))

The limited data available suggests that for our present purpose it is reasonable to assume an average of 7 per annum for this category. In this case the plane and its fuel load are much heavier and a 'crash zone' of 100 yards diameter will be assumed.

To take account of this both the length and width of each storage area should be increased by 100 yards. The extent to which this will increase the target area will depend on the geometry of the storage array(s).
Case 1  Single storage area (200 yds x 100 yds)

The target area now becomes:

\[
300 \text{ yds} \times 200 \text{ yds} = 6 \times 10^4 \text{ sq yds} \\
= 2 \times 10^{-2} \text{ sq miles}
\]

\[
\therefore P = \frac{7 \times 2 \times 10^{-2}}{10^5} = 1.4 \times 10^{-6} / \text{Yr}
\]

Case 2  Five storage areas (100 yds x 40 yds)

The target area now becomes:

\[
5 \times 200 \text{ yds} \times 140 \text{ yds} = 1.4 \times 10^5 \text{ sq yards} \\
= 4.5 \times 10^{-2} \text{ sq miles}
\]

\[
\therefore P = \frac{7 \times 4.5 \times 10^{-2}}{10^5} = 3.2 \times 10^{-6} / \text{Yr}
\]

Hence the combined probabilities are:

Case 1 \[2.4 \times 10^{-6} \text{ crashes/yr}\]
Case 2 \[4.2 \times 10^{-6} \text{ crashes/yr}\]

The calculations presented here are purely illustrative and no account has been taken of the probability that a crash may occur which does not produce a significant fire.
SUMMARY
By the Session Chairman
E.S. London
SESSION I, PART II

Summary

Two authors stressed a basic principle that containers should be moved only after solidification of the UF₆ content. It may be profitable to discuss what this means in terms of time.

One author expressed a preference for avoiding take-off systems which fill containers with liquid hex and stated that the transfer of product and tails into containers via the gaseous phase is a desirable aim.

Where there may be criticality problems the avoidance of moderators for cooling and heating systems is to be preferred.

With regard to the protection of operators in the event of in-plant incidents one author emphasized the importance of aids to maintaining the means of escape.

For the storage of hex tails, either indoors or outside, suitable anti-corrosion coatings are available for the external surfaces of the containers. One author referred to two measures which would limit the environmental impact of the low probability event of fire in a tails storage area following an aircraft crash; these are i) the conversion of UF₆ into another chemical form and ii) the shaping of the ground underneath the containers in a favourable way. One paper compared the suitability from the public safety standpoint of 2 tonne and 12 tonne containers for hex tails and concluded that the larger container is preferable since it is better able to withstand a serious transport fire. There was some criticism from the floor of the model taken by the author, it being felt that the approach was too simplistic and led to a pessimistic conclusion.

This topic warrants further discussion.
SESSION II A

UF₆ BEHAVIOUR

THEORETICAL ASSESSMENTS
Environmental impact of a potential UF₆ - release resulting from an accident in a UO₂ fuel fabrication plant by W. Thomas
ENVIRONMENTAL IMPACT OF A POTENTIAL UF$_6$-RELEASE RESULTING FROM AN ACCIDENT IN A UO$_2$ FUEL FABRICATION PLANT

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Abstract

An analysis has been performed of the possible environmental impact of accidental UF$_6$ releases from a fuel fabrication plant. Radiological and chemical consequences due to soluble uranium and hydrofluoric acid have been quantified in model calculations. The results of the study indicate that only a small chemical or radiological impact to the environment is to be expected from in-plant accidents. Doses of only a few rem should occur outside the facility during a criticality accident. An airplane crash followed by fire constitutes a severe risk for a relatively small area in the vicinity of the plant.
1. INTRODUCTION

Uranium hexafluoride (UF₆) constitutes the main feed material for producing uranium dioxide (UO₂) powder or pellets in a fuel fabrication plant. Currently the plant operations for fuel fabrication in Germany are based on the wet conversion processes using ammonium diuranate (ADU) or ammonium hydrogen carbonate (AUC) as intermediate conversion products. The only existing full scale plant in operation is Reaktor-Brennelemente-Union (RBU) at Hanau which has a capacity of about 1000 tons/year. A second plant, EXXON Nuclear at Lingen will contain only a small UF₆ conversion facility in project step II.

Uranium hexafluoride is a very reactive substance reacting readily with water, many metals and most organic compounds. A reaction of UF₆ with water produces uranium oxifluoride (UO₂F₂) and the highly corrosive and toxic volatile chemical, hydrogen fluoride (HF). The uranium particles from this reaction constitute a fine, white material, which easily can be dispersed to the environment. If inhaled the soluble uranium acts as a chemical toxin especially to the kidneys. For Light Water Reactor (LWR) fuel element fabrication, typical enrichments are in the order of 2 to 4 % U-235. Bone is the critical organ in respect to radiological effects of the uranium. With enriched material an accidental criticality is possible if UF₆ is converted to a well-moderated solution of UO₂F₂ in water.

Several potential accident scenarios were investigated to quantify possible consequences of small or remarkable releases of UF₆ to the environment [1].

2. CHEMICAL AND RADIOLOGICAL TOXICITY OF UF₆

A basis for the assessment of the hazard to humans in case of an accidental release of UF₆ was developed by compiling data of toxic concentrations in air and the related existing limits. Only a few extrapolated data exist for UF₆ itself and the data for hydrogen fluoride are not fully consistent, especially for short exposures to high concentrations. The collected sources agree that no lethal effects on humans should be anticipated for HF-concentration less than 40 mg/m³. The correlation of lethal concentration and time of exposure is rather difficult to be quantified. Therefore, the data given in Table I should be considered as the most probable values derived from a rather limited data base [2, 3, 4, 5].
Table I: Toxicity Data for Hydrogen Fluoride

<table>
<thead>
<tr>
<th>Detection by smell</th>
<th>Concentration [mg HF/m³]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Maximum permissible concentration for 8 h-exposure</td>
<td>2.5</td>
</tr>
<tr>
<td>Respiratory discomfort</td>
<td>25</td>
</tr>
<tr>
<td>Lethal concentration</td>
<td></td>
</tr>
<tr>
<td>Exposure &gt; 1 h</td>
<td>40</td>
</tr>
<tr>
<td>Exposure 5 min</td>
<td>400</td>
</tr>
<tr>
<td>Exposure 1 min</td>
<td>1500</td>
</tr>
</tbody>
</table>

Concentration of about 1 mg/m³ of finely distributed soluble uranium are visible. Based on the ICRP-lung model, 150 mg of soluble uranium may be lethal if inhaled [6]. The radio-toxicity of low enriched uranium is of less concern. In the design of plants in the FRG the following limits have to be kept in case of accidents at the boundary of the facility area [7]:

Table II: FRG - Radiation Limits in Case of Accidents

<table>
<thead>
<tr>
<th>Organ</th>
<th>Maximum dose limit, rem</th>
</tr>
</thead>
<tbody>
<tr>
<td>Whole body</td>
<td>5</td>
</tr>
<tr>
<td>Bone</td>
<td>30</td>
</tr>
<tr>
<td>Lung, Liver</td>
<td>15</td>
</tr>
</tbody>
</table>

3. LAYOUT OF THE CONVERSION FACILITY

Enriched UF₆ is shipped and stored in type 30 A or 30 B steel cylinders. Each cylinder contains up to 2200 kg of UF₆. Part of these cylinders are stored outdoors and part are stored indoors of a common industrial building. There is a small storage area where ten cylinders are stored at the headend of the conversion process. The cylinder type 30 B has a diameter of 76 cm and a length of 205 cm. The wall material is steel of a thickness of 12.7 mm. The container is tested to 28 kg/cm² (2.7×10⁵ Pa) of external pressure which is twice the service pressure of 14 kg/cm². The vessel is designed to withstand a service temperature up to 120 °C. A maximum enrichment fo 5 % U-235 is permitted for UF₆ with a maximum impurity of 0.5 % HF.

At the headend of the conversion facility, the cylinders are
heated by steam in specially designed pressure containers up to 100 °C so that the UF₆ is vaporized. The vaporization procedure is supervised by control of temperature and pressure. The steam condensate is controlled by a pH-meter. The valves for steam and UF₆ are automatically closed in case of an alarm from the pH-meter. The UF₆ is transferred in heated pipes to a precipitation vessel where chemical conversion to AUC occurs. A second pH-meter in the precipitator ascertains the right conversion to AUC. A deviation of the given pH-value causes the UF₆ feed line to be closed automatically. After precipitation and filtration the AUC is converted to UO₂ in a fluidized bed calciner.

4. ACCIDENT SCENARIOS AND CONSEQUENCES

Experience of accidental releases of UF₆ in the existing German conversion plant is limited only to smaller events, e.g. small releases in case of repair of piping or removing plugs of UF₆. In no case have the operations represented hazardous situations to the personnel or the environment. In 1968 a release of approximately 160 kg uranium as UF₆ occurred in a blending operation of different enriched material. The release from the heated container was stopped after 3 minutes and the released material was dispersed by the wind. The accident occurred outdoors. An adjacent parking lot and the walls of the building were contaminated by uranium. No damage to the environment by the visible HF-cloud has been reported.

The analysis of accidents for the storage and conversion of UF₆ at a conversion facility includes the following postulated events:

- Damage to unheated containers
- Breakage of a plugged UF₆ connection pipe
- Sudden release of UF₆ from a heated container
- Criticality
- Release of UF₆ in case of an airplane crash

No direct experience is available for the more severe accident scenarios. Calculations of scenarios include necessary assumptions which must be stated arbitrarily as best estimates.

Uncertainty exists in the assumptions for heating and possible rupture of UF₆-containers in case of a fire. Theoretical investigations of this problem are based on rather conservative assumptions. No quantitative data are available for settling mechanisms of released uranium or hydrofluoric acid in form of small droplets. Usually the calculation of concentration of uranium and HF are performed using a Gaussian dispersion model, which seems to overestimate the concentrations at greater distances. It is rather doubtful that the strong chemical reactivity and buoyancy of released uranium oxifluoride and HF can be neglected. Therefore, the following calculations and results should be regarded only as model calculations including a great deal of conservatism.
4.1 Damage unheated containers

Mechanical damage to unheated containers seems possible during a severe impact of a truck or as a result of a transportation accident in the storage area. Without heating the UF$_6$ in the damaged container is in the solid state because the vapor pressure is low, only a slow release of very small quantities of UF$_6$ of HF are to be expected. No chemical or radiological effects outside the confinement of the facility are anticipated. Because a release of UF$_6$ is marked by white UO$_2$F$_2$ the release point easily can be detected.

4.2 Breakage of a plugged UF$_6$ connection pipe

If heating of a UF$_6$ connection pipe fails solidified UF$_6$ may plug the pipe. Due to the expansion during solidification breakage of the plugged pipe is possible. Experience indicates that the released quantities in this type of event are in the order of 10 - 20 g UF$_6$. Similarly, releases from damaged or plugged valves are possible. Release of UF$_6$ from a damaged valve can occur via the steam condensate outlet. The design of the pH-meter and the feed-back to the steam valve have been improved to prevent this type of accident. The concentrations of UF$_6$, UO$_2$F$_2$ and HF in the vicinity of the damaged or plugged valve are in the order of few milligrams/m$^3$. Damage to the filters of the container ventilation is prevented by a washer. The filters of the room ventilation should not be affected, so that a release to the environment should be possible only in minor quantities.

4.3 Sudden rupture of a heated container

Heated UF$_6$-cylinders are only present in the evaporation container. In no case are heated cylinders moved or transported. A release of hot UF$_6$ therefore, could be possible only if the valve or the UF$_6$-container and the outer evaporation vessel are seriously damaged by external forces. Calculations and accident experience indicate that about 50% of the UF$_6$ could be released in about 30 minutes. The rest of the UF$_6$ remains as crystallized solid material in the damaged container.

A scenario for this kind of accident has been set up. The main features of the model are:

- 1100 kg of UF$_6$ are released at a constant rate in 30 minutes into the building.
- The filters of room ventilation fail after 15 minutes.
- HF and finely dispersed uranium are released to the environment. The duration of total release with a ventilation rate of 20 m$^3$/s is about 30 minutes.

Investigations of maximum possible concentrations of heavy metals in air indicate, that only concentrations up to few grams/m$^3$ of air can persist for the anticipated release time. A maximum concentration of 5 g U/m$^3$ is assumed in the scenario leading to a steady release of 100 g U/s. 50% of generated HF is anticipated to react chemically with material...
and equipment inside the facility, the maximum release rate is 45 g HF/s. No direct release of UF₆ without chemical reaction is assumed. Total quantities released to the environment are 190 kg uranium in soluble form and 73 kg HF.

The ventilated air containing noxious HF and uranium is emitted at a height of 10 m. The dispersion model is based on Gaussian diffusion. No settling of uranium or HF-droplets is considered. Assuming unfavourable weather conditions (Class E), the concentrations calculated as function of the distance from the release point are likely to overestimate the environmental impact. The diffusion parameters used are those recently published by a group of German experts [10]. These data mainly are based on field experiments performed at Jülich and Karlsruhe which take account of the roughness of the environment.

The calculated concentrations of HF and uranium versus distance from the release point are shown in Fig. 1. At a distance of 100 m, the HF-concentration reaches the value of 25 mg/m³ which causes lung and eye irritation. The concentration at 1000 m distance corresponds to the maximum permissible concentration for a 8 h-exposure. The released cloud containing HF and uranium lasts for about half an hour with a HF-concentration in the order of 20 mg/m³. People should withdraw from this region, no intake of dangerous or lethal quantities of soluble uranium is to be expected.

For the unlikely case that no evacuation from regions with higher HF-concentrations takes place, integrated doses from total released quantities were calculated for the lung, bone and liver. The calculations are based on soluble uranium (Class D) with an enrichment of 3.5 % and a specific activity of $2.25 \times 10^{-6}$ Ci/g. The dose commitments are from [11], calculated for an activity median aerodynamic diameter of 0.3 µ and a breathing rate of 0.5 l/s. The doses are in the order of a few rem and in no case exceed the limits for accidents. The lung doses are below 0.1 rem for all distances.

The concentration of HF inside the building (volume 40,000 m³) reaches a maximum value of 2200 mg/m³. The lethal concentration for short exposure (1 min) is reached after 15 minutes of release. Since the HF-concentration already causes respiratory irritation close to the point of release in very short time the personnel should evacuate immediately. The risk from inhaled soluble uranium is of less concern in comparison to the inhalation of HF.

4.4 Criticality

A criticality accident with enriched UF₆ is only possible if large quantities are moderated to UO₂F₂-solutions. Only small amounts of UO₂F₂ can be obtained with procedures used for vaporization of residues or cleaning operations. Therefore, this kind of accident is relatively remote. Experiments with high enriched uranium solutions [12] allow a close estimation of the possible maximum number of total fissions. Whereas a typical criticality incident for a UO₂F₂-solution should result in less than $10^{18}$ fissions, an upper limit of $5 \times 10^{18}$ total fissions can
be derived from existing experimental data [12]. A criticality accident results in emission of penetrating neutron and gamma radiation for a short time. Pulsating critical excursions may also be possible. The doses from direct radiation without any shielding are lethal only in a small region of about 25 m distance from the point of criticality. Outside the building the radiation doses decrease very rapidly with increasing distance. Including the shielding effect of a concrete wall of 15 cm thickness, doses from direct radiation have been calculated. At a distance of 150 m from the event, the whole body dose is 5 rem.

A great variety of volatile and nonvolatile fission products is generated by the critical excursion. Part of these are released to the environment. The main contribution to resulting doses in the environment are from relatively short lived halogens and noble gases. At a distance of 100 m the calculated doses under severe weather conditions (stability class E, release height 10 m) are in the order of a few rem for β- and γ-submersion and thyroid (Fig. 2).

4.5 Release of UF₆ in case of an airplane crash

The probability for an airplane crash on a UF₆ conversion facility and the adjacent storage area has been estimated to be in the order of 10⁻⁶/y. Although this is very improbable, severe consequences can be expected from this accident and a subsequent fire. Two different kinds of release have to be investigated:

- The release from damaged or destroyed UF₆-cylinders
- Possible breaks of undamaged containers by subsequent heating with burning kerosene.

Since the building will be destroyed or strongly damaged most of the released UF₆ will be dispersed with the smoky plume. Part of the UF₆ or UO₂F₂ may be converted to nearly insoluble U₃O₈-powder, but it is not possible to quantify this effect.

For the analysis the following scenario has been postulated:

- 10 UF₆-cylinders of type 30 B are stored close to the headend.
- 2 UF₆-cylinders of type 30 B are heated in the evaporization equipment.

Leakage of 50 % of the UF₆ in a time of 5 minutes is anticipated from the heated containers. The cold UF₆-cylinders in storage position are heated by burning kerosene. Calculations of heating time necessary to break a 30 B-cylinder are in the order of 10 - 20 minutes [13]. The kerosene fire is assumed to last about 30 minutes. The calculations of the heating times up to rupture pressure (about 85 kg/cm² = 8.3·10⁶ Pa) seem to be rather conservative due to the assumption of heating a black body without loss of heat so there remains some doubt as to how many containers would fail in this time. For the analysis, a continous leakage of 50 % of the UF₆ from 10 containers in 30 minutes is anticipated. Total released quantities over a distance greater than 100 m are 892 kg uranium and 3000 kg HF.
For the calculation of doses in the environment, only HF and UO₂F₂ are regarded to be present in the plume. Part of the UF₆ may remain unreacted close to the point of impact. 100% of released HF is set free for dispersion, but only 10% of UO₂F₂ is assumed to be transported by the plume over a distance greater than 100 m. The rest of the uranium will remain as UO₂F₂ or U₃O₈ in or near the destroyed facility. As experience indicates, the height of the resultant plume is very dependent from prevailing weather conditions. A typical plume height is in the order of 100 m, but smaller heights have been observed under special weather conditions. Using the diffusion parameters from [10], resulting HF-concentrations for maximum release rate of 1.67 kg/s are shown in Fig. 3 for stability class A. For a plume height of 100 m, the calculated maximum concentrations are in the order of 25 - 40 mg/m³. This causes lung irritation but no lethal HF-burns. If very unfavourable weather conditions exist, plume heights of only 30 - 50 m are anticipated. Calculated concentrations then exceed the minimum lethal concentration of 40 mg/m³ for 1-h-exposure. A maximum value of 270 mg/m³ has been calculated for a plume height of 30 m. Similar curves for uranium concentrations for different stability classes have been calculated.

The calculated concentrations remain because a continuous release is assumed for about 30 minutes. For distances greater than 1 km, the time for inhalation of a lethal quantity of HF or soluble uranium, plus the migration time of the plume, are so long, that evacuation and emergency measures should be effective.

The calculation of the radiological impact, due to dispersion of soluble uranium using dose commitments from [11], shows that the limit for bone dose from an accidental release is reached only under extreme weather conditions (30 rem [7]). For a typical release height of 100 m the calculated maxima of bone dose are in the order of few rem integrated over the total duration of the release.

5. CONCLUSION

The results of the model calculations for in-plant accidents indicate that outside the plant boundary no severe or lethal effects by chemical or radiological impact are to be expected. Inside the building, HF-concentrations in the order of 2000 mg/m³ represent a lethal hazard in case of a great release of hot UF₆ even for short time exposure. Penetrating neutron and gamma radiation emitted from an improbable criticality accident causes lethal whole body doses up to a distance of 25 m if a conservative number of 5·10¹⁸ total fissions is assumed. The environmental impact of an airplane crash results mainly from corrosive HF. Lethal effects from HF should be possible only for unfavourable dispersion conditions in a region up to 1 km from the release point.

Further activities and investigations are needed for a better understanding of the heating mechanism, behaviour of UF₆ during a large release, chemical reactions, and deposition of HF and uranium during dispersion. Experimental results in these fields
are of high importance. More experimental data will help to reduce some conservatism in the calculations and to permit a more accurate analysis of UF₆-releases.

Literature

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Fig. 1:

UF$_6$-RELEASE FROM A HEATED CONTAINER. CONCENTRATIONS VERSUS DISTANCE. MAXIMUM RELEASE RATES: 100 g U/s, 45 g HF/s
Fig. 2:

DOSES FROM A CRITICALITY ACCIDENT

UO$_2$F$_2$-Solution, $5 \cdot 10^{18}$ Fissions in 15 min.
Release of Volatile Fission Products

RElease Height: 10 m
Stability Class E

Dose, rem

10

1

10$^{-1}$

10$^{-2}$

$10^2$ $10^3$ $10^4$ Distance, m

$\beta$ $\gamma$

THYROID CHILD 1y

SUBMERISON
Fig. 3:

HF-CONCENTRATION VERSUS DISTANCE FOR UF₆-RELEASE. RELEASE RATE 1.67 kg HF/s.
PARAMETER: PLUME HEIGHT
A quantitative forecast of behaviour of UF₆ release in the air
by
A. Doumenc, R. Gérard
La prévision quantitative des pollutions consécutives à un rejet d'UF₆ dans l'air peut être effectuée à partir d'un modèle général de transfert atmosphérique.

Le modèle utilisé pour cette prévision est un modèle à bouffées développé au Commissariat à l'Energie Atomique. Les différents types d'accidents possibles sont schématisés par deux types d'émission à la source : une émission ponctuelle prolongée et une émission volumique instantanée. Plusieurs codes de calculs adaptés à ces différentes émissions et aux présentations recherchées par la sûreté ont été développés. Des exemples d'applications pratiques sont donnés et commentés.

A quantitative forecast of behaviour of uranium hexafluoride release in the air can be estimated by a general atmospheric model. The atmospheric dispersion model used for is a puff model made in "Commissariat à l'Energie Atomique". Different kinds of potential accidents are simply represented by two kinds of sources : a continuous point source and an instantaneous voluminous source. Several calculation codes have been developed for these different releases and forms wanted by safety. Some practical examples are given with comments.
1 - PURPOSE AND SCOPE

This study describes the methods actually used to forecast the atmospheric pollutants after an uranium hexafluoride release in air and shows problems encountered in practice.

In a first time, term source has been simplified and stylised, yet the used models are convenient enough in their conception to be adapted according to knowledge evolution and experiments work on hand. For source term definition, a first difficulty is to determine the release conditions, because different causes must be taken into account such as a sudden lost of confinement in an enrichment facility or a release during a container handling. On the other hand, it is allowed that a complete or partial hydrolysis follows the release but the resulting components and the kinematic reaction seem very dependent on water "supply" conditions (air moisture, spattering, simple contact with a surface ...).

In actual knowledge, it is difficult to provide for the exact composition of hydrolysis products, even if the reaction conditions are well known.

It seems that many components are produced during the reaction and last a more or less long time. So after an atmospheric release, it seems possible to find: \( UF_6 ; HF ; x \text{UO}_2\text{F}_2, y \text{H}_2\text{O} ; x \text{UO}_2\text{F}_2, y \text{HF} ; x \text{UO}_2\text{F}_2, y \text{HF}, z \text{H}_2\text{O} ; ... \)

For this different products a general atmospheric transfer model can be used to forecast their dispersion if:

- pollutant products can be considered as minor atmospheric constituent following the same general transfer law of carrying fluid

- pollutant products are gaseous or composed with small size aerosol to be indifferent to gravity (practically, if \( \Phi < 20 \mu m \)).

The atmospheric dispersion model used must take any release into account after the thermodynamic equilibrium with the atmosphere. Therefore it is necessary to be able to consider a point source as well as a voluminous source, an instantaneous or a long period release.
2 - **ATMOSPHERIC MODEL USED**

2.1 General purpose to forecast

The atmospheric dispersion model used is a puff model made in Commissariat à l'Énergie Atomique / 1 /, which gives concentration in air and ground deposition for a pollutant in any spatial point, from any known reference trajectory, near the source as well as very far from it (several hundred kilometers). Model advantages consist especially in separation of the dispersion process in two fundamental components:
- transport by mean wind
- diffusion by turbulence.

This model allows to use, without any subjective interpretation, meteorological parameters measured in siting stations (by example with a meteorological tower outfit):
- wind speed and wind direction function of time (transport)
- vertical stability of the atmosphere (diffusion) determined as possible by measurement of the vertical temperature gradient.

This model has been exposed in previous publications. / 1 / / 2 / It uses a gaussian solution for the diffusion equation and the main difference and originality with other similar models is to consider the standard spatial deviations (σ) of the concentration distributions as a function of transfer duration.

In this model, a continuous release is simply represented as a succession of instantaneous puffs; the diffusion of any of them is function of transfer deviation.

This method allows to take into account not only the transport direction but also the release duration and the exposure duration. From this, three reference models have been developed, for three kinds of release:
- instantaneous point source
- instantaneous voluminous source
- continuous point source.
2.2. Instantaneous point source

This source does not correspond to practice problem, but this type is the most simple and the model for it is the basis for build up models.

The volumic concentration of pollutant from an instantaneous point source is given by

\[
X = \frac{O \cdot E}{(2\pi)^{3/2} \sigma_x \sigma_y \sigma_z} \exp \left( -\frac{1}{2} \left[ \frac{(x-x_0-\bar{u}t)^2}{\sigma_x^2} + \frac{(y-y_0)^2}{\sigma_y^2} + \frac{(z-z_0)^2}{\sigma_z^2} \right] \right)
\]

\(X\) = Concentration of pollutant in air
\(O\) = Total quantity of the pollutant source
\(E\) = Evolution law of pollutant (chemical evolution, radioactive decay ...)
\(\sigma_x, \sigma_y, \sigma_z\) = Standard deviations of the concentration distributions along the three coordinate directions at the centroid of the cloud at time \(t\):

\[
\sigma_x = \sigma_y = \sigma_z = (A_h t)^{k_h}
\]

\[
\sigma_z = (A_z t)^{k_z}
\]

\(A_h, A_z\) = Horizontal and vertical diffusion parameters \((L^{1/k_{-1}})\)
\(K_h, K_z\) = Horizontal and vertical diffusion exponents
\(x\) = Coordinates on the trajectory direction
\(y, z\) = Coordinates orthogonal to the trajectory direction
\(x_0, y_0, z_0\) = Coordinates of the source
\(\bar{u}\) = Mean wind speed
\(t\) = Time after release

Parameters and exponents \(A_h, A_z, k_h, k_z\) come from real diffusion coefficients which are function of time transfer \(x\), and known by experimental various and numerous distributions of pollutants in air from instantaneous releases /1/.

\[
\begin{align*}
\sigma_h^2 &= 2K_h t \\
\sigma_z^2 &= 2K_z t
\end{align*}
\]

Kh, Kz are real horizontal and vertical diffusion coefficients.
Diffusion and parameters exponents, as a function of time after release are given below.

### Normal diffusion

<table>
<thead>
<tr>
<th>Travel time t (seconds)</th>
<th>( A_h ) ((m^1/k\cdot s^{-1}))</th>
<th>( A_z ) ((m^1/k\cdot s^{-1}))</th>
<th>( k_h )</th>
<th>( k_z )</th>
</tr>
</thead>
<tbody>
<tr>
<td>0 à 2,40(\times)10²</td>
<td>4,05(\times)10⁻¹</td>
<td>0,42</td>
<td>0,859</td>
<td>0,814</td>
</tr>
<tr>
<td>2,40(\times)10² à 3,28(\times)10³</td>
<td>1,35(\times)10⁻¹</td>
<td>1,00</td>
<td>1,130</td>
<td>0,685</td>
</tr>
<tr>
<td>3,28(\times)10³ à 9,70(\times)10⁵</td>
<td>1,35(\times)10⁻¹</td>
<td>20</td>
<td>1,130</td>
<td>0,500</td>
</tr>
<tr>
<td>9,70(\times)10⁵ à 5,08(\times)10⁷</td>
<td>4,63(\times)10⁻¹</td>
<td>20</td>
<td>1,000</td>
<td>0,500</td>
</tr>
<tr>
<td>5,08(\times)10⁷ à 1,30(\times)10⁹</td>
<td>6,50</td>
<td>20</td>
<td>0,824</td>
<td>0,500</td>
</tr>
<tr>
<td>&gt; 1,30(\times)10⁹</td>
<td>2,00(\times)10⁵</td>
<td>20</td>
<td>0,500</td>
<td>0,500</td>
</tr>
</tbody>
</table>

### Weak diffusion

<table>
<thead>
<tr>
<th>Travel time t (seconds)</th>
<th>( A_h ) ((m^1/k\cdot s^{-1}))</th>
<th>( A_z ) ((m^1/k\cdot s^{-1}))</th>
<th>( k_h )</th>
<th>( k_z )</th>
</tr>
</thead>
<tbody>
<tr>
<td>0 à 2,40(\times)10²</td>
<td>4,05(\times)10⁻¹</td>
<td>0,20</td>
<td>0,859</td>
<td>0,500</td>
</tr>
<tr>
<td>2,40(\times)10² à 9,70(\times)10⁶</td>
<td>1,35(\times)10⁻¹</td>
<td>0,20</td>
<td>1,130</td>
<td>0,500</td>
</tr>
<tr>
<td>9,70(\times)10⁶ à 5,08(\times)10⁸</td>
<td>4,63(\times)10⁻¹</td>
<td>0,20</td>
<td>1,000</td>
<td>0,500</td>
</tr>
<tr>
<td>5,08(\times)10⁸ à 1,30(\times)10¹⁰</td>
<td>6,50</td>
<td>0,20</td>
<td>0,824</td>
<td>0,500</td>
</tr>
<tr>
<td>&gt; 1,30(\times)10¹⁰</td>
<td>2,00(\times)10⁵</td>
<td>0,20</td>
<td>0,500</td>
<td>0,500</td>
</tr>
</tbody>
</table>

Actually, two atmospheric vertical stability conditions are considered.

The correspondance with atmospheric vertical thermal gradient \((Y)\) is conventionally:

- normal diffusion : \(Y < -0,5 ^\circ C/100 \ m\) (unstable)
- weak diffusion : \(Y \geq -0,5 ^\circ C/100 \ m\) (stable)
2.3. Instantaneous voluminous source

In that release, the sudden important quantity of delivered pollutant makes an horizontal and vertical expanded volume for a so short time (some seconds to some minutes) that it can be looked to as instantaneous. When the thermodynamic equilibrium with atmosphere is reached, the cloud dispersion goes on with the normal atmospheric transfer process.

The cloud, whose volume is a function of energy release, can be considered as a cylinder.

That volume can be brought back to a virtual point source and then it is an instantaneous point source (figure 1).

If $\Delta t$ is a theoretical time necessary for a virtual source to reach the real source dimensions, concentration function of time can be computed with the expression for instantaneous point source, but the time origin must be taken $\Delta t$ back:

$$
\sigma_x = \sigma_y = \sigma_h = \left[ A_h \left( t + \Delta t_h \right) \right]^{k_h}
$$

$$
\sigma_z = \left[ A_z \left( t + \Delta t_z \right) \right]^{k_z}
$$

with:

$$
\Delta t_h = \frac{1}{A_h} \left[ \frac{R_o}{V^2} \right]^{1/k_h}
$$

$$
\Delta t_z = \frac{1}{A'_z} \left[ \frac{\Delta z_o}{V^2 \pi} \right]^{1/k'_z}
$$

$R_o, \Delta z_o =$ radius and height of cylinder source

$A'_h, A'_z, k'_h, k'_z =$ parameters function of source initial dimensions

The other terms of basis expression are unchanged.
2.4. Continuous point source

The method used for a continuous point source is to model the release as a succession of instantaneous puffs (explained in 2.1.). Then the concentration may be calculated by adding the contribution of each puff to the concentration at the point of interest at a given instant. The contribution from puff "i" at point of coordinates x, y, z can be written from the equation:

$$X_i = \frac{Q_i e^{-rac{1}{2} \left[ \left( \frac{x-x_0-\theta_i}{\sigma_x} \right)^2 + \left( \frac{y-y_0}{\sigma_y} \right)^2 + \left( \frac{z-z_0}{\sigma_z} \right)^2 \right]}}{(2\pi)^{3/2} \sigma_x \sigma_y \sigma_z}$$

Where:

- \(Q_i\) = the quantity of pollutant released in puff i
- \(\theta_i\) = travel time of the puff \(i = t-t_i\)
- \(t\) = observation time
- \(t_i\) = birth time of the puff

Other symbols have been explained in 2.2.

The total quantity released \(Q\) is the sum of contributions \(Q_i\):

$$Q = \sum_i Q_i$$

The net concentration at the point of interest can be represented by the summation: \(X = \sum_i X_i\)

This method allows a large use. So it is possible to take into account:

- any duration release from instantaneous source (one puff)
  to continuous source (numerous successive puffs)
- variable discharge release
- sinuous trajectory
3. **EXAMPLES OF PRACTICAL USE**

From this model several computing codes have been developed for different releases and presentation wanted by safety.

A first serie of codes (ECYIN, TRAIR III) computes concentration evolution of pollutants function of time in any spatial point from instantaneous voluminous sources (figure 3).

A second serie of codes (EPOP, TRAIR II) computes the same result from continuous point sources (figure 3).

A third serie of codes (ICAIR) more elaborated draws isoconcentration curves at fixed time and can be superposed upon map (figure 4).

For concrete examples, the use of this codes has been in a good concordance with real measures after experiments over large distances with tracers and interpretation of a past recent incident.

HF concentrations computed by the model for a 500 kg UF₆ release in atmosphere and for different kinds of releases and presentations are shown on figures 3 to 6.

To simplify, it has been supposed a simple, quick and complete hydrolysis of UF₆ giving practically 113,5 kg HF release in all cases.

For small size aerosols of UO₂F₂ or other chemical materials (Ø<20 μm) the model can be used but it is necessary to know the quantity of released small size material.
4. ENCOUNTERED PROBLEMS

4.1. Meteorological conditions

The combined effects of meteorological conditions and real height of release can be seen on figure 5.

This model can take up common meteorological states, but others can be treated:

- by arrangement of diffusion parameters (by example: inversion of temperature limited or no in altitude)
- by addition of a complementary model (by example: precipitations).

When having meteorological measures normally made on a nuclear site:

- wind speed and wind direction
- vertical temperature gradient

it is easy to define mean, extreme or typical site meteorological conditions to evaluate atmospheric transfer.

4.2. Source definition

Source parameters (height of release, volume, ..., physical and chemical composition ...) are very important and actually they are the biggest reason of incertitude in forecasting especially at short distance.

The figure 6 shows the maximum concentration for four various sources:

- point source with instantaneous release (greatest hypothetical case) (A curve)
- voluminous source with instantaneous release
  Limited expansion to the minimum volume necessary to hydrolyse UF₆ by atmospheric humidity (about 5 000 m³ in this example) (B curve).
- voluminous source with instantaneous release, corresponding to the volume get from the heat of the complete hydrolysis and from the initial heat of the surrounding environment (C curve).
- continuous point source corresponding to a release of constant discharge during 10 mn (D curve).
All these sources are released from a same height (40 m) and involved a same quantity of UF$_6$. The maximum concentration curves show a great difference according to the various source especially for short distances. By these results, the strong necessity to have a good knowledge of source parameters is shown. The codes will be completed according to the results of experimental works on hand take into better account of the real UF$_6$ kinematic release and hydrolysis.
REFERENCES


FIGURE 1 - INSTANTANEOUS VOLUMINOUS SOURCE LOOKED TO AS A VIRTUAL POINT SOURCE.
Figure 2 - CONTINUOUS POINT RELEASE MODELLED AS A SUCCESSION OF INSTANTANEOUS PUFFS.
FIGURE 3 - HF CONCENTRATION EVOLUTION IN THE AIR FROM A 500 kg HF RELEASE AT 4000 m DOWNWIND.
NORMAL DIFFUSION, 5m.s⁻¹ WINDSPEED
**FIGURE 4** - 500 kg UF₆ RELEASE POINT SOURCE WITH CONSTANT DISCHARGE. UF₆ ISOCONCENTRATION CURVES IN AIR (g.m⁻³) AT GROUND LEVEL.
FIGURE 5 - 500 kg $^{6}$UF₆ POINT SOURCE RELEASE WITH CONSTANT DISCHARGE DURING 10 mn.

HF MAXIMUM CONCENTRATION IN AIR AT GROUND LEVEL.
METEOROLOGICAL CONDITIONS

wind speed 5 m.s\(^{-1}\)

normal diffusion

SOURCE
A et D point source
B small volume
C large volume

RELEASE
A, B, C instantaneous
D prolonged

FIGURE 6 - 500 kg UF\(_5\) RELEASE BY SEVERAL SOURCES CONFIGURATIONS
Atmospheric dispersion and consequences of a UF₆ release caused by valve rupture on a hot 30B cylinder by A.M. Ericsson
ATMOSPHERIC DISPERSION AND CONSEQUENCES OF A UF₆ RELEASE CAUSED BY VALVE RUPTURE ON A HOT 30B CYLINDER

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Bertil Grundfelt
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The accident analysed is that of valve rupture on a 30B cylinder heated to 100°C and containing UF₆, thus causing gaseous UF₆ to escape.

Most of the UF₆ content of the cylinder is initially in liquid form. This liquid phase will start to boil and the gas will flow out. The contents of the cylinder will then be cooled due to the dissipation of the latent heat of evaporation.
The calculations are based on the following simplifications:

1) The behaviour of the cylinder contents is considered to be adiabatic, i.e. no cooling is considered to take place through the walls.

2) The material transport resistance during boiling is ignored.

3) The pressure-volume work in the gaseous phase can be considered as negligible as compared to the heat dissipation in the form of latent heat of evaporation.

From the risk aspect, all of these assumptions are pessimistic. If free convection in air is assumed to occur on the outside of the cylinder, assumption 1) will give an accessively low cooling rate.

Assumption 2) gives a somewhat high discharge rate, since the pressure in the cylinder at any instant will be equal to the equilibrium pressure at the prevailing temperature. However, the deviation should be very small. But it is difficult to specify how small this deviation is, since an analysis of the material transport resistance during boiling is complicated and is outside the scope of this study.

Assumption 3) gives a cooling rate which is about 0.1 - 0.2% too low.

When the temperature in the cylinder has fallen to the triple point, crystallisation will start. This is assumed to take place at a constant temperature, i.e. the crystallisation rate is assumed to be determined by the dissipation of the latent heat of evaporation.

When crystallisation has been completed, it is assumed that discharge will cease. The triple point is assumed to be about 64°C and the temperature at which the steam pressure above solid UF₆ is 1 atm is approx. 54°C. The specific heat capacity of solid UF₆ within this temperature range is approx. 120 cal/kg, K. The heat of sublimation is 33 000 cal/kg, K. The quantity of UF₆ which would flow out of the cylinder after completed crystallisation (if the cylinder is assumed to contain 500 kg of solid phase) would thus be

\[
\frac{120(64-54)}{33 000} \cdot 500 = 20 \text{ kg}
\]

1.1 Discharge relationships.

The following is based on the Chemical Engineers' Handbook, Perry et al. (5).
The maximum mass velocity on isothermal expansion can be calculated from:

\[ G_{ci} = P_0 \sqrt{\frac{M}{2.718 \cdot R \cdot T_0}} \, (kg/m^2 \cdot s) \]  

(1)

where \( P_0 \) = pressure in the container (Pa)
\( M \) = molecular weight of UF\(_6\) = 355 (kg/kmol)
\( T_0 \) = temperature in the container (K)
\( R_0 \) = universal gas constant = 8314 (J/kmol, K)

\( G_{ci} \) is multiplied by a factor \( F \) which is a function of the ratio of the pressure in the container and the ambient pressure \( e \), the discharge resistance \( N \) and the isentropic index \( k \). The value of \( F \) is specified graphically in the Chemical Engineers' Handbook.

The discharge resistance consists of two components, i.e. sudden restriction with \( N = 0.5 \) and discharge into a large recipient with \( N = 1.0 \).

UF\(_6\) has a very low isentropic index \( k = 1.06 \), i.e. it behaves almost isothermally on expansion. With these values of \( N \) and \( k \), the graph in the Chemical Engineers' Handbook is described reasonably well by the following equations:

\[ F = 0.76 \quad \epsilon < 0.45 \]  

(2a)

\[ F = -1.18381 \cdot \sqrt{\epsilon -1.463735(\epsilon -0.75)^2} +1.68141 \quad 0.45 \leq \epsilon \leq 0.75 \]  

(2b)

\[ F = 1.18381 \cdot \sqrt{\epsilon -4.464022(\epsilon -0.75)^2} +1.689192 \quad 0.75 < \epsilon < 0.95 \]  

(2c)

\[ F = -7 \cdot \epsilon +7 \quad \epsilon \geq 0.95 \]  

(2d)

The relative error in relation to the graph is below 2%, which is well within the reading accuracy in the graph.

The total flow out of the cylinder will be:

\[ G = G_{ci} \cdot F \cdot A_{\text{hole}} \]  

(3)

where \( A_{\text{hole}} \) = cross-sectional area (m\(^2\)) of the discharge hole.

From equations (1) and (3), and at a 1 inch diameter discharge hole, the discharge flow will be:

\[ G = 6.43 \cdot F \cdot P_1 \cdot T_0^{-1/2} \]  

(4)

where \( P_1 \) = pressure in the cylinder (atm).
If the discharge hole is instead assumed to have the same diameter as the major diameter of the valve thread, i.e. 1 11/32 in, the discharge flow will instead be:

\[ G = 11.62 \cdot F \cdot P_1 \cdot T_0^{-1/2} \] (5)

This clearly illustrates the uncertainty of the discharge calculation.

1.2 Material-energy balances.

A material balance on the container gives:

\[ \frac{dm}{dt} = -G \] (6)

where \( m \) = mass of UF\(_6\) in the cylinder (kg).

If the pressure volume work is ignored and the standard condition is set for the liquid at the prevailing temperature, an energy balance gives:

\[ m \cdot C_{p}^{\text{tot}} \cdot \frac{dT}{dt} = -G \cdot \Delta H_{\text{vap}} \] (7)

where \( \Delta H_{\text{vap}} \) = latent heat of evaporation (J/kg)

\[ C_{p}^{\text{tot}} = \text{the weighted mean value of the specific heat capacities of the gaseous and liquid phases (J/kg K)} \]

\[ C_{p}^{\text{tot}} = m_g \cdot C_p^g + m_L \cdot C_p^L \] (8)

where \( m_g \) = mass of the gaseous phase in the cylinder (kg)

\( m_L \) = mass of the liquid phase in the cylinder (kg)

From (7) and (8):

\[ \frac{dT}{dt} = -G \cdot \Delta H_{\text{vap}} / (m_g \cdot C_p^g + m_L \cdot C_p^L) \] (9)

The masses of the gaseous and liquid phases can be obtained by knowledge of the total volume of the cylinder, the total mass of UF\(_6\) in the cylinder and the density of the liquid phase. From the universal gas law:

\[ m_g = \frac{M \cdot P_1 \cdot V_g}{R \cdot T_0} \] (10)

where \( V_g \) = volume of the gas in the cylinder.
But \( V_g = V_{tot} - V_L = V_{tot} - \frac{m_L}{\rho_L} \), which gives:

\[
m_L = \left( m - \frac{M \cdot P_1}{R \cdot T_0} \cdot \frac{1}{\rho_L} \right) \cdot \frac{V_{tot}}{1 - \frac{M \cdot P_1}{R \cdot T_0} \cdot \frac{1}{\rho_L}} \tag{11a}
\]

\[
m_g = m - m_L \tag{11b}
\]

Differential equations (6) and (9) are solved simultaneously by the Runge-Kutta method.

To solve equations (11a) and (9), expressions are necessary for \( C_P, C_L, H_{vap}, \rho_L \) and \( P_1 \) as a function of the temperature. These expressions have been taken from (3):

\[
C_P^g = 382.8 + 0.09366 \cdot T - 3.785 \cdot 10^6/T^2 \tag{12}
\]

\[
C_P^L = 211.907 + 0.76751 \cdot T + 7.87236 \cdot 10^6/T^2 \tag{13}
\]

\[
\Delta H_{vap} = 29193.09 + 170.86 \cdot T - 0.33692 \cdot T^2 + 1.1657 \cdot 10^7/T \tag{14}
\]

\[
\rho_L = 4041 + 3.368 \cdot T - 0.0136 \cdot T^2 \tag{15}
\]

\[
P_1 = \exp \left( 7.2876 \cdot \ln T - 0.014371 \cdot T - 1245.2 \cdot T^{-1} - 248608 \cdot T^{-2} - 31.2935 \right) \tag{16}
\]

The mass of the crystal phase is obtained from

\[
\frac{dm_s}{dt} = G \cdot \Delta H_{vap}/\Delta H_{fus}
\]

where \( \Delta H_{fus} \) = latent heat of fusion (kJ/kg)

\( m_s \) = mass of the crystal phase.

2. Results of the discharge calculations.

A computer program known as "HEXRISK" has been written for the discharge calculation. This program which calculates the mass of UF₆ in the cylinder, the temperature, pressure, flow and the proportion discharged as a function of time following valve rupture, has been run a total of four times, assuming three different starting temperatures and two diameters of the discharge hole:
Run No. 1: $T_0 = 100^\circ C$, $d = 1 11/32"$
Run No. 2: $T_0 = 90^\circ C$, $d = 1"$
Run No. 3: $T_0 = 100^\circ C$, $d = 1"$
Run No. 4: $T_0 = 110^\circ C$, $d = 1"$

The temperature drop as a function of time in run No.3 is plotted in Fig. 2. The proportion discharged from the cylinder varies between 49% (run No.2) and 56% (run No.4). This corresponds to a total UF₆ discharge of between 1 120 kg nad 1 270 kg. About 20 kg must be added to these figures as the quantity which may be discharged after the uranium hexafluoride has crystallised. These 20 kg can be entirely ignored from the risk aspect, since they represent a very small addition to the exposure in the surroundings and since it is discharged at low velocity. The proportion discharged as a function of time in run No.3 is plotted in Fig.3.

A comparison between runs No.1 and 3 (same temperature but different hole diameter) shows that discharge through the larger hole takes place in about 39 min and through the smaller hole in about 45 min. In both runs, the proportion discharged is 53% (1 200 kg of UF₆ is discharged).

Since the expansion of the cylinder contents was considered to be adiabatic, the proportion discharged is clearly defined by the initial temperature. The influence of reasonable variations in the initial temperature is illustrated by the table below, which applies to a hole diameter of 1 in (runs 2 - 4).

<table>
<thead>
<tr>
<th>Initial temp. ($^\circ C$)</th>
<th>Initial press. (atm)</th>
<th>UF₆ discharged (kg)</th>
<th>Proportion discharged (%)</th>
<th>Discharge time (min)</th>
</tr>
</thead>
<tbody>
<tr>
<td>90</td>
<td>3.13</td>
<td>1117</td>
<td>49.1</td>
<td>45.1</td>
</tr>
<tr>
<td>100</td>
<td>4.00</td>
<td>1195</td>
<td>52.5</td>
<td>44.8</td>
</tr>
<tr>
<td>110</td>
<td>5.03</td>
<td>1271</td>
<td>55.8</td>
<td>44.1</td>
</tr>
</tbody>
</table>

The mean discharge rate $\bar{G}$ and the maximum discharge rate $G_{\text{max}}$ will be:

<table>
<thead>
<tr>
<th>Initial temp. ($^\circ C$)</th>
<th>$\bar{G}$</th>
<th>$G_{\text{max}}$</th>
<th>$G_{\text{max}}/\bar{G}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>90</td>
<td>0.41</td>
<td>0.80</td>
<td>1.95</td>
</tr>
<tr>
<td>100</td>
<td>0.44</td>
<td>1.01</td>
<td>2.30</td>
</tr>
<tr>
<td>110</td>
<td>0.48</td>
<td>1.25</td>
<td>2.60</td>
</tr>
</tbody>
</table>

The interesting feature from the risk aspect is the ratio of the maximum to the mean discharge rate, i.e. $G_{\text{max}}/\bar{G}$. This
ratio provides a rough indication of the concentration in the front part of the plume and the mean concentration. The time for the discharge rate to fall from its maximum value to its mean value is about 9 min at an initial temperature of 90°C, 10.5 min at 100°C and 11.5 min at 110°C. In all runs, the uranium hexafluoride started to crystallise within 15 minutes (the flat part in Fig.2 and 4). The discharge rate during the crystallisation period is constant at 0.37 kg/s at a diameter of 1 in and 0.68 kg/s if the hole diameter is assumed to be 1 11/32 in. The discharge rate in the case of the 1 in hole is shown in graph 4. The fraction of the cylinder content which is discharged before crystallisation starts varies between 16% at an initial temperature of 90°C and 27% at 110°C.

The discharge calculation is somewhat conservative, since the process in the cylinder has been assumed to be adiabatic. The reduction in the proportion discharged caused by heat dissipation to the surroundings through the cylinder walls is of the order of 4% (slightly below 100 kg less UF₆ discharged) under the following conditions:

- The heat transmission resistance in the cylinder wall and inside the cylinder is ignored.
- Cooling takes place by free convection in air on the outside (a = 10 W/m²°C).
- The ambient temperature is 20°C.

3. **Sequence of events after discharge.**

Uranium hexafluoride reacts very quickly with water as follows:

\[
\text{UF}_6(g) + 2 \text{H}_2\text{O}(g) \rightarrow \text{UO}_2\text{F}_2(s) + 4 \text{HF}(g)
\]

Owing to the high reaction rate, the limiting factor in the reaction of UF₆ with the humidity in the air is considered to be the availability of water in the form of airborne humidity. A mathematical treatment of the transformation of UF₆ in the gas plume formed inevitably leads to extensive calculation work which is outside the scope of this study. A mathematical treatment of this nature must include the diffusion of water vapour in air, the diffusion of uranium hexafluoride in air and the influence of turbulence and temperature gradients. We shall instead discuss the water quantities necessary for complete transformation of UF₆ to UO₂F₂.

In the initial stage, the discharge rate is relatively high. Assuming that the hole is 1 in in diameter, the initial flow is approx. 1 kg/s. This corresponds stoichiometrically to a water demand of 0.1 kg/s for complete transformation. The mean water consumption during the discharge time is approx. 0.05 kg/s for complete transformation.
The water content of the air is dependent on the temperature and the weather conditions (relatively humidity) and can easily be calculated by means of the vapour pressure table (see table below). On the basis of the water content in the air, an assessment can then be made of the air volume necessary for complete reaction of the UF₆ to take place.

Water content in air (kg/m³).

<table>
<thead>
<tr>
<th>RH %</th>
<th>Temp. °C</th>
<th>0</th>
<th>10</th>
<th>20</th>
<th>30</th>
</tr>
</thead>
<tbody>
<tr>
<td>20</td>
<td>9.7·10⁻⁴</td>
<td>1.9·10⁻³</td>
<td>3.5·10⁻³</td>
<td>6.1·10⁻³</td>
<td></td>
</tr>
<tr>
<td>60</td>
<td>2.9·10⁻³</td>
<td>5.6·10⁻³</td>
<td>1.0·10⁻²</td>
<td>1.8·10⁻²</td>
<td></td>
</tr>
<tr>
<td>100</td>
<td>4.8·10⁻³</td>
<td>9.2·10⁻³</td>
<td>1.7·10⁻²</td>
<td>3.0·10⁻²</td>
<td></td>
</tr>
</tbody>
</table>

Air volume (m³/s) necessary for complete reaction at the mean discharge rate.

<table>
<thead>
<tr>
<th>RH %</th>
<th>Temp. °C</th>
<th>0</th>
<th>10</th>
<th>20</th>
<th>30</th>
</tr>
</thead>
<tbody>
<tr>
<td>20</td>
<td>52</td>
<td>26</td>
<td>14</td>
<td>8.2</td>
<td></td>
</tr>
<tr>
<td>60</td>
<td>17</td>
<td>8.9</td>
<td>5.0</td>
<td>2.8</td>
<td></td>
</tr>
<tr>
<td>100</td>
<td>10</td>
<td>5.4</td>
<td>2.9</td>
<td>1.7</td>
<td></td>
</tr>
</tbody>
</table>

Total air volume (m³) necessary for complete reaction.

<table>
<thead>
<tr>
<th>RH %</th>
<th>Temp. °C</th>
<th>0</th>
<th>10</th>
<th>20</th>
<th>30</th>
</tr>
</thead>
<tbody>
<tr>
<td>20</td>
<td>140 000</td>
<td>70 000</td>
<td>38 000</td>
<td>22 000</td>
<td></td>
</tr>
<tr>
<td>60</td>
<td>46 000</td>
<td>24 000</td>
<td>14 000</td>
<td>6 700</td>
<td></td>
</tr>
<tr>
<td>100</td>
<td>27 000</td>
<td>15 000</td>
<td>7 800</td>
<td>4 600</td>
<td></td>
</tr>
</tbody>
</table>

Under extremely humid summer conditions, the air volume necessary corresponds to a sphere with a radius of 10 m (or a cone with an apex angle of 10° and a length of 83 m) to convert all of the uranium hexafluoride to uranyl fluoride. If dry winter weather is instead assumed, the corresponding sphere of air would have a radius of 31 m and the cone would have a length of 260 m.
In humid weather at the height of summer, it would therefore appear probable that most of the uranium hexafluoride would be converted to $\text{UO}_2\text{F}_2$ in the immediate vicinity of the discharge point (within $100^2\text{m}$), whereas during the winter or in dry weather, part of the $\text{UF}_6$ will probably be spread outside the plant boundary.

Two additional questions, which have not yet been discussed, also have an effect on the spreading of uranium in the surroundings. One of these concerns the cooling of $\text{UF}_6$, and the consequent formation of aerosols and crystallisation. The other question is the particle size of the $\text{UO}_2\text{F}_2$ crystals, i.e. how quickly these can be expected to settle on the ground. The fact that the reaction between $\text{UF}_6$ and the moisture in the air is very fast suggests that the particle size could be very small and that the crystals could then be transported across long distances by the wind. Since the basis for determining the spreading of uranium is uncertain, it is assumed in the consequence calculations that most of it is entrained by the wind, primarily in the form of $\text{UO}_2\text{F}_2$ particles.

4. Reaction in air.

Uranium hexafluoride reacts with the water in the air and forms uranyl fluoride and hydrogen fluoride as follows:

$$\text{UF}_6 + 2 \text{H}_2\text{O} \rightarrow \text{UO}_2\text{F}_2 + 4 \text{HF}$$

The reaction takes place instantaneously. The limiting factor is the amount of water vapour in the air. As mentioned earlier, this study does not include complete treatment of the actual reaction of $\text{UF}_6$, and the calculation is based on the assumption that complete transformation of $\text{UF}_6$ takes place.

As mentioned earlier, the reaction leads to the formation of hydrofluoric acid (HF) and uranyl fluoride ($\text{UO}_2\text{F}_4$). The radiological toxicity consists only of uranium which occurs here in soluble form as uranyl ions ($\text{UO}_2^{2+}$). The chemical toxicity is due to the hydrogen fluoride, the fluoride ions and the uranyl ions.

Radiological toxicity.

Under certain conditions, uranium accumulates in the skeleton, where it has a biological half-life of 300 days. The radiological toxicity increases with increased enrichment of the uranium. The highest permissible concentration in air for persons engaged on non-radiological work is $3 \times 10^{-12} \mu \text{Ci/cm}^3$. The maximum permissible single intake of uranium by breathing is 2.5 mg/day and by swallowing, 150 mg/day. At low enrichment, the critical organs are the kidneys.
Chemical toxicity.

Hydrofluoric acid at room temperature is a colourless liquid with a pungent smell. The acid is entirely soluble in water. In gaseous as well as liquid form, the acid attacks the eyes, skin and mucous membranes. The acid is classified as a digestive poison, a respiratory poison and a nervous poison.

The hygienic limit for hydrogen fluoride in air is 3 ppm. The inhalation of vapour or gas corresponding to 50 ppm for 30 - 60 minutes is lethal.

Hydrofluoric acid reacts very readily with oxidising as well as reducing agents, and hydrogen is liberated in the reaction. The acid also reacts violently with metals and hydrogen is liberated. The hydrogen is very flammable and may form explosive mixtures with air.

Since it is soluble, uranyl fluoride owes its toxicity to that of the uranyl ion and that of the fluoride ion.

When breathed in, soluble uranium (UF₆ and UO₂F₂) is quickly transported to the body by the blood. A large proportion is decomposed in the urine. But significant quantities will be retained by the kidneys and skeleton.

If soluble uranium is swallowed, only a small proportion will remain in the digestive tracts. Of this quantity, about 1% will be transported further in the blood.

The primary risk in the inhalation of compounds containing natural and low-enriched uranium is the chemical injury caused to the kidneys rather than the radiological toxicity. A poisoning symptom is the presence of albumen in the urine. On chronic inhalation, the chemical toxicity of uranium is comparable with that of lead. The lethal dose for animals varies between 0.1 mg U/kg of body weight and 20 mg U/kg of body weight. In the case of human beings, this value can be extrapolated to around 1 mg U/kg of body weight.

The toxicity of fluoride is due to it occupying the spaces of hydroxide ions in the apatite lattice of the skeleton. This leads to bone embrittlement.
5. Concentration and dispersion

The concentration at ground level along the centre line of the plume can be calculated from:

$$\kappa_o = \frac{M}{2\pi \sigma_y \sigma_z u t} \exp \left( - \frac{h^2}{2 \sigma_z^2} \right)$$

(kg/m$^3$)

where $M = $ quantity discharged (kg)
$h = $ height of discharge (m)
$t = $ duration of discharge (s)
$\sigma_y$ and $\sigma_z$ = horizontal and vertical dispersion parameters respectively (m)
$u = $ wind velocity (m/s)

Since no weather statistics are available for Västerås, the weather situation is compared with that applicable to Ägesta. These statistics are considered to be the best available in the country (6) for this type of calculation.

The value of $\kappa_o$ for Ägesta is specified in (1) as a function of the weather stability parameter $\lambda$ and the distance $X$ from the point of discharge. Converted to a discharge of about 1 200 kg of UF$_6$, the values specified in Table I will be obtained. These values specify the integral concentration given in kg/m$^3$ for various distances from the point of discharge.

The conversion to concentration in kg/m$^3$ is shown in Table II. This has been calculated on the basis of the mean discharge flow of 0.44 kg UF$_6$/s from the cylinder, which has been obtained from the discharge calculations in the preceding chapter.

The weather stability parameter $\lambda$ corresponds to various weather situations and wind velocities in accordance with the table below:

<table>
<thead>
<tr>
<th>Weather type</th>
<th>$\lambda$</th>
<th>$u$ (m/s)</th>
<th>frequency, %</th>
</tr>
</thead>
<tbody>
<tr>
<td>Neutral</td>
<td>-</td>
<td>5</td>
<td>44</td>
</tr>
<tr>
<td>Almost neutral</td>
<td>0.5</td>
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<td>19</td>
</tr>
<tr>
<td>Fairly stable</td>
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<td>4</td>
<td>17</td>
</tr>
<tr>
<td>Relatively stable</td>
<td>1.7</td>
<td>3</td>
<td>12</td>
</tr>
<tr>
<td>Stable</td>
<td>2.3</td>
<td>2</td>
<td>5</td>
</tr>
<tr>
<td>Very stable</td>
<td>2.9</td>
<td>1</td>
<td>3</td>
</tr>
</tbody>
</table>
Fig. 5 - 8 have been produced from these values and the tables in Appendices 2 and 3. Fig. 5 - 6 indicate the integral concentration as a function of the cumulative frequency, and Fig. 7 - 8 show the discharge concentration as a function of the cumulative frequency. All graphs include the distance from the discharge point as a parameter.

The integral concentration and the discharge concentration under average weather conditions (50% cumulative frequency) and unfavourable weather conditions (95% cumulative frequency) have then been read from Fig. 5 - 8 for the various distances from the discharge point. These concentrations are tabulated in Appendices 4 and 5. However, it should be pointed out that the values in the immediate vicinity are very uncertain.

As regards dilution of the plume in the direction of discharge, Tables III - IV show that the concentrations fall off by a factor of 38 between 0.25 and 10 km from the point of discharge under average weather conditions (50% cumulative frequency), whereas they fall off by a factor of no more than 8 under unfavourable weather conditions (95% cumulative frequency).

In the immediately adjacent zone, the difference between the concentrations under average and unfavourable weather conditions is negligible. However, the differences increase with the distance from the discharge point, and at a distance of 10 km, the concentration is a factor of 10 higher under unfavourable weather conditions than in average weather.

Relatively large amounts of data must be processed for accurate calculation of the dilution of the plume perpendicularly to the direction of discharge, i.e. the horizontal width of the plume, at various distances from the point of discharge, but this is considered to lie outside the scope of this study.

However, the width of the plume has been approximately calculated using the graphs in (1). The curves for 50% and 95% cumulative frequencies have been plotted in graph 25 in the above reference. The 50% case was found to correspond well to the curve for neutral weather ($\lambda = \infty$) and the 95% case corresponds up to 0.5 km from the discharge point to fairly stable weather ($\lambda = 1.1$) and beyond 0.5 km, to relatively stable weather ($\lambda = 1.7$). The plume widths at the various cumulative frequencies and distances were then read from graph 17 and graph 34 in reference (1). These values are tabulated in Table IV. The plume widths indicate twice the distance from the centre line to the point at which the concentration has fallen off to 1% of the concentration at the centre line of the plume.

5.2 Consequences to the general public.

People who are within the plume will be exposed to uranium as well as fluorine in soluble form which they will absorb primarily by breathing.

The duration of the discharge in this particular case is 45 minutes. The respiratory rate has been assumed to be $1.25 \text{ m}^3/\text{h}$. 
The calculations have been concerned with people who are in the dispersion zone of the plume throughout the discharge time.

Uranium quantities inhaled.

Most of the uranium is inhaled in the form of UO$_2$F$_2$. After reaction with water, 1 kg of UF$_6$ corresponds to approx. 0.9 kg of UO$_2$F$_2$ which, in turn, corresponds to approx. 0.7 kg of U.

The uranium is in soluble form throughout. The critical organs are then initially the kidneys and only when the doses are large will the uranium reach the skeleton.

A fraction of 0.25 of the material inhaled is deposited in the lungs and is absorbed by the blood. The amounts of uranium which are absorbed by the human body at various distances from the discharge point are shown in Table VI. This table illustrates that the amount of uranium inhaled is 1 - 2 powers of ten below the lethal dose (approx. 1 mg/kg of body weight) at 50% as well as 95% cumulative frequency. In the immediately adjacent zone, the values are presumably somewhat too low. It is very difficult to calculate the concentration accurately within this zone, since the actual discharge height due to the plume lift is difficult to determine. However, there is no doubt whatever that it is obviously unhealthy to be within this adjacent zone. On the other hand, further away from the discharge point, the amount inhaled is below the permissible limit of 2.5 mg for individual inhalation of soluble uranium.

It would be desirable to carry out a determination of the relationships prevailing in the immediately adjacent zone, to calculate the actual plume lift, etc., in order to obtain more reliable values of the uranium and fluorine contents in this zone.

Quantities of fluorine inhaled.

All fluorine is treated as hydrofluoric acid, since two-thirds are inhaled in this form and since the remaining one-third will form hydrofluoric acid in the body.

When it has reacted with water, 1 ml of UF$_6$ corresponds stoichiometrically to 6 mol of fluorine. Thus, 1 kg of UF$_6$ corresponds to 0.33 kg of hydrofluoric acid. The contents of hydrofluoric acid at various distances from the point of discharge have been calculated on the basis of the hypothetical UF$_6$ concentration (as if no UF$_6$ had reacted) in Table V. These values are given in Table VII.

The hygienic limit value for hydrofluoric acid is at 2.5 mg/m$^3$. As shown by the table, the contents on discharge are well below this value outside the immediately adjacent zone. The calculations are also considered to be reliable in this case. As mentioned earlier, the basis for the calculations for the immediately adjacent zone is fairly weak, and the contents may be appreciably higher here than is indicated by the table. Great care should therefore be taken in the immediate vicinity of the
discharge point and a gas mask should be used in the decontamination work.

6. **Proposals for the decontamination work.**

As soon as discharge is detected, people should be evacuated from the immediately adjacent zone to a safe area.

The decontamination personnel should work in full-body protective suits and gas masks.

It is presumably impossible to collect the gas in a free gas cloud. Hydrogen fluoride as well as uranyl fluoride are readily soluble in water, and spreading of the cloud can possibly be reduced by spraying it with scattered jets of water. The substances will then be deposited on the ground surface. Water jets can also be used for affecting the direction of travel of the cloud, if this is considered desirable.

The hydrogen fluoride and uranyl fluoride deposited on the ground surface can be neutralised by means of slaked lime. People who are suspected of having inhaled hydrogen fluoride or uranyl fluoride should quickly be taken to a hospital. In the case of hydrogen fluoride poisoning, the patient should be provided with fresh air or, if possible, oxygen, should rest and should be kept warm. The symptoms of hydrofluoric acid poisoning are heavy coughing, shortage of breath and a pronounced general effect.
References


6. SMHI, Norrköping. Personal contact.
### Table I

Time-integral of concentration as a function of the weather stability parameter at different distances from the source.

<table>
<thead>
<tr>
<th>( \lambda ) (km)</th>
<th>(-\infty)</th>
<th>0.5</th>
<th>1.1</th>
<th>1.7</th>
<th>2.3</th>
<th>2.9</th>
</tr>
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<tr>
<td>0.1</td>
<td>(1.8 \times 10^{-2})</td>
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<td>0.25</td>
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<td>-</td>
</tr>
<tr>
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</tr>
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### Table II

Concentration as a function of the weather stability parameter at different distances \(X\) from the source.

<table>
<thead>
<tr>
<th>( \lambda ) (km)</th>
<th>(-\infty)</th>
<th>0.5</th>
<th>1.1</th>
<th>1.7</th>
<th>2.3</th>
<th>2.9</th>
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<td>-</td>
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<td>-</td>
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<td>(1.2 \times 10^{-5})</td>
<td>-</td>
</tr>
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</table>
### Table III

**Integral concentration (kgs/m²) at 50 and 95 % cumulative frequency.**

<table>
<thead>
<tr>
<th>Distance (km)</th>
<th>50 % cumulative frequency</th>
<th>95 % cumulative frequency</th>
</tr>
</thead>
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<td>0.1</td>
<td>$1.5 \cdot 10^{-2}$</td>
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<tr>
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### Table IV

**Plume-width at 50 and 95 % cumulative frequency at different distances from the source.**

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<tr>
<th>Distance (km)</th>
<th>Plume-width (m) 50 % cumulative frequency</th>
<th>Plume-width (m) 95 % cumulative frequency</th>
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<td>600</td>
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</tr>
<tr>
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</table>
Table V
Concentration (kg/m³) at 50 and 95 % cumulative frequency.

<table>
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<th>Distance (km)</th>
<th>50 % cumulative frequency</th>
<th>95 % cumulative frequency</th>
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</thead>
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</table>

Table VI
Amount of uranium deposited in the lungs of a man at different distances from the source.

<table>
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<th>Distance (km)</th>
<th>50 % cumulative frequency</th>
<th>95 % cumulative frequency</th>
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<tr>
<td></td>
<td>grams U deposited</td>
<td>grams U deposited</td>
</tr>
<tr>
<td>0.1</td>
<td>1.3·10⁻³</td>
<td>1.6·10⁻³</td>
</tr>
<tr>
<td>0.25</td>
<td>3.1·10⁻³</td>
<td>3.8·10⁻³</td>
</tr>
<tr>
<td>0.5</td>
<td>1.8·10⁻³</td>
<td>3.6·10⁻³</td>
</tr>
<tr>
<td>1.0</td>
<td>9.5·10⁻⁴</td>
<td>2.4·10⁻³</td>
</tr>
<tr>
<td>2.0</td>
<td>4.3·10⁻⁴</td>
<td>1.6·10⁻³</td>
</tr>
<tr>
<td>3.0</td>
<td>2.9·10⁻⁴</td>
<td>1.1·10⁻³</td>
</tr>
<tr>
<td>4.0</td>
<td>2.1·10⁻⁴</td>
<td>8.1·10⁻⁴</td>
</tr>
<tr>
<td>5.0</td>
<td>1.6·10⁻⁴</td>
<td>6.4·10⁻⁴</td>
</tr>
<tr>
<td>6.0</td>
<td>1.4·10⁻⁴</td>
<td>5.3·10⁻⁴</td>
</tr>
<tr>
<td>7.0</td>
<td>1.2·10⁻⁴</td>
<td>7.6·10⁻⁴</td>
</tr>
<tr>
<td>8.0</td>
<td>1.1·10⁻⁴</td>
<td>6.9·10⁻⁴</td>
</tr>
<tr>
<td>9.0</td>
<td>8.6·10⁻⁵</td>
<td>5.2·10⁻⁴</td>
</tr>
<tr>
<td>10.0</td>
<td>8.1·10⁻⁵</td>
<td>4.7·10⁻⁴</td>
</tr>
</tbody>
</table>
Table VII

Concentration of hydrofluoric acid in air at different distances from the source.

<table>
<thead>
<tr>
<th>Distance (km)</th>
<th>mg/m³ hydrofluoric acid 50 % cumulative frequency</th>
<th>mg/m³ hydrofluoric acid 95 % cumulative frequency</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.1</td>
<td>1.8</td>
<td>2.1</td>
</tr>
<tr>
<td>0.25</td>
<td>4.3</td>
<td>5.3</td>
</tr>
<tr>
<td>0.5</td>
<td>2.5</td>
<td>4.9</td>
</tr>
<tr>
<td>1.0</td>
<td>1.4</td>
<td>3.1</td>
</tr>
<tr>
<td>2.0</td>
<td>0.59</td>
<td>2.2</td>
</tr>
<tr>
<td>3.0</td>
<td>0.40</td>
<td>1.5</td>
</tr>
<tr>
<td>4.0</td>
<td>0.30</td>
<td>1.1</td>
</tr>
<tr>
<td>5.0</td>
<td>0.23</td>
<td>0.96</td>
</tr>
<tr>
<td>6.0</td>
<td>0.19</td>
<td>0.86</td>
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<td>7.0</td>
<td>0.17</td>
<td>0.79</td>
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<td>8.0</td>
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<td>0.76</td>
</tr>
<tr>
<td>9.0</td>
<td>0.13</td>
<td>0.73</td>
</tr>
<tr>
<td>10.0</td>
<td>0.11</td>
<td>0.66</td>
</tr>
</tbody>
</table>
Temperature as a function of time.
Starting point 100°C.
Hole diameter 1".

Figure 2
Percent of cylinder content discharged as a function of time.
Starting point 100°C.
Hole diameter 1".
Release rate as a function of time.

Starting point 100°C.
Holediameter 1".
Integral concentration (kgs/m$^3$) = f (cumulative frequency)

1. 0.1 km from the source.
2. 0.25 km from the source.
3. 0.5 km from the source.
4. 1.0 km from the source.
5. 2.0 km from the source.
6. 3.0 km from the source.
7. 4.0 km from the source.
Integral concentration (kgs/m$^3$) = $f$ (cumulative frequency)

1. 5.0 km from the source.
2. 6.0 “ “ “ “.
3. 7.0 “ “ “ “.
4. 8.0 “ “ “ “.
5. 9.0 “ “ “ “.
6. 10.0 “ “ “ “.

Figure 6
Concentration (kg/m³) = f (cumulative frequency)

1. 0.1 km from the source.
2. 0.25 " " " "
3. 0.5 " " " "
4. 1.0 " " " "
5. 2.0 " " " "
6. 3.0 " " " "
7. 4.0 " " " "
Concentration (kg/m³) = f (cumulative frequency)

1. 5.0 km from the source.
2. 6.0 " " " " .
3. 7.0 " " " " .
4. 8.0 " " " " .
5. 9.0 " " " " .
6. 10.0 " " " " .
Safety analysis of UF$_6$ transportation

by

F. Israel,  P. Pages
ABSTRACT

A mathematical model has been performed in order to assess the risk associated to transportation system and to help in comparing alternative Safety measures. Based on accident data files kept by several involved organisations it uses two delicate notions:

- the severity indicator
- the failure probability associated to a given severity

The model applied to UF6 transportation with a kit of severity indicators has furnished interesting results.

A particular study of the fire severity indicator is needed in order to take into account hot spot risks.
INTRODUCTION

In order to assess the risk intranspor ting radioactive materials, we have developed a methodology suitable to any transport node. Such a study has to provide us with realistic estimates of all possible environment accident severities has to be able to take into account any change in future transportation system and finally has to help us in comparing alternative safety measures on the basis of their cost-efficiency rate.

The methodology applied to UF6 transportation points out the main difficulty of such a work: finding good severity indicators and a suitable formula relating each severity value to a probability of failure.

1. RISK ANALYSIS MODEL

In the range of a given transport mode, the risk of breaking a container (expressed as a number of failed containers per year) may be reached by using the equation:

\[ R = T \times P \times \sum_{j} P_j \times C_j \]

where

- \( R \) is the overall risk of breaking a container (number of failure/year).
- \( T \) is the assumed traffic (in vehicle x km/year).
- \( P \) is the accident rate in number of accident/(vehicle x km).

and finally \( \sum_{j} P_j \times C_j \) is the probability of breaking a container as an accident \( j \) has occurred.

This value is calculated by summing for all possible accident sequences (scenario) the product \( P_j \times C_j \),

where \( P_j \) is the probability of occurrence of the scenario \( j \) considered.

and \( C_j \) measures its consequences in terms for example of number of broken Containers per accident.
2. Methodology

The calculation of the R. value may be divided in three steps.

2.1. Analysis of accident data files.

Analysis of such data files, kept by several involved organizations such as (Ministry of transportation, Railway Society, etc.) provide:

- all possible accident sequences (scenarios) and for each, the probability of occurrence: \( P_j \).

- in terms of consequences of a given scenario; the probability of a given consequence severity. This means that during each scenario the container is submitted to various efforts (fire, collision, puncture, etc.). A severity indicator \( g \) may be associated to each kind of effort and the file analysis provides the probability \( f(g) \) for the container to be submitted to an effort of a given severity.

2.2. The second step is to point out a statistical relationship between accident severity indicators and container behaviour.

For each kind of effort, \( f_o(g) \) shall be the probability of failure of a container submitted to an effort of severity \( g \).

And at the end of this second step, we are able to calculate for a given kind of effort: \( C_j \) by summing for all values of the severity indicator \( g \) in the range \( G \):

\[
C_j = \int_{g \in G} f(g) \times f_o(g) \, dg
\]

This value is calculated for all kinds of efforts experienced by the container during the scenario \( j \) and the value \( C_j \) taken into account is the biggest one.

2.3. The third step is to calculate the overall risk associated to a transport mode assuming values of \( T \) and \( P \) and using the previously calculated value

\[
\sum_{j} P_j \times C_j
\]

3. Severity Indicators

For both rail and road transport modes we will consider first, accidents not involving fire.

As regards to road accident severity, the statistical analysis of accident data involving trucks carrying hazardous materials
led us to consider two indicators. The first one concerns impact. It expresses the energy theoretically assigned to an inelastic collision between two bodies:

\[ g_1 = 0.5 \times M \times V^2 \]

where \( M \) and \( V \) are respectively the reduced mass (a particular mass being assumed in the case of a collision with a fixed object) and the relative velocity between the two colliding objects. The second indicator concerns the puncture effort:

\[ g_2 = V/R \]

where \( V \) is the above mentioned relative velocity and \( R \) is the radius of the puncturing probe.

In the case of train accidents, we keep a single severity indicator which is not directly connected to a physical expression of accident forces, but obtained from a statistical analysis (discriminant factorial analysis) of data on observed damage to railcars involved in train accidents (particularly the number of damaged railcars). Figures 1, 2 and 3 show the adjustment of the observed indicator distributions to exponential type distributions such as Weibull or simple exponential laws. Such laws express in each case the conditional probability distribution of the severity indicator, knowing that an accident has occurred without a fire ensuing. For example, figure 1 shows the probability density function of impact severity, i.e. the probability (per unit interval of the indicator) for a truck of experiencing an impact type accident of the severity indicated.

\[ f(x) = \left( \frac{1}{2\alpha} \right) \exp\left( -\frac{x}{\alpha} \right) \exp (0.5) \]
\[ \alpha = 3.68 \]

Figure n°1: Truck accident. Probability density function of impact severity given a collision or overturn.
1.0

\[ y = \left( \frac{x}{a} \right)^2 \exp \left( -\frac{x}{a} \right) \]

\[ a = 2.4 \]

Figure n°2: Truck accident. Probability density function of puncture severity given a collision or overturn.

\[ y = \left( \frac{1}{2a} \right)^2 \exp \left( -\frac{x}{a} \right) \times 0.5 \]

\[ a = 9 \]

Figure n°3: Train accident. Probability density function of accident severity given a derailment or collision accident.
The severity of fires in both considered transport modes is estimated by a single indicator: the total fire duration, i.e. starting from the first call for help up to the complete extinction of the fire. Figures 4 and 5 show the adjustment of obtained distributions. As above, adjusted distributions are conditional, knowing that an accident has occurred with a fire ensuing.

**Figure n°4**: Train accident. Probability density function of fire duration given a train fire.

**Figure n°5**: Truck accident. Probability density function of fire duration given a truck fire.
4. CONSEQUENCES OF ACCIDENTS

Whatever the type of accident is - collision or overturn or fire - we consider a simple function \( f_0(g) \) expressing the conditional probability of a package failure, given a certain level of severity, i.e. a particular value \( g \) of the severity indicator.

\[
f_0(g) = 1 - e^{-\lambda g} \quad \text{(collision type)} \quad \text{or} \quad 1 - e^{-kt^2} \quad \text{(fire with duration t)}.
\]

\( \lambda \) and \( k \) are parameters expressing the package resistance to the forces it has to sustain. Those parameters are related to the mean failure rate observed on accidents involving packages of the type under study. When such data are lacking, they can be estimated, for example, by referring to the failure rates observed on industrial packages of a similar type and by dimensional analysis methods.

In the case of a fire, a failure threshold is obtained below which the package is considered to survive. Thus it is necessary to estimate first the time threshold, then at least a point on the curve relating fire duration and the associated failure probability.

5. RESULTS OF THE MODEL APPLIED TO UF6 SHIPMENTS

5.1. In the absence of further informations, we have supplied values about shipment traffic accidents based upon traffic and accident data about truck traffic on highways in 1975 and total freight train traffic in 1973-1975.

\[
\text{for truck: } P = 9 \times 10^{-7} \text{ accident per veh x km}
\]

\[
\text{for train: } P = 1.44 \times 10^{-7} \text{ accident per rail car x km.}
\]

5.2. Different traffic were assumed for two years: 1975 and 1980 (Eurodif in full operation).

Natural UF6 shipments were assumed to an amount of 460 12 ton cylinders for 1975 (90 by truck the rest by train) 1300 cylinders for 1980 (280 by truck) shipments of enriched UF6 are made exclusively by truck. Such shipments are transported on an average distance of 1000 km.
5.3. Risk of shipping UF$_6$ and risk sensitivity evaluations

Results concerning UF$_6$ shipments in France are shown in Tables 2 and 3; it can be seen that the risk level (estimated annual frequency of container failures) lies between two values differing by an order of magnitude: $5 \times 10^{-4}$ and $5 \times 10^{-3}$ (for 1975) according to the transport mode. The effects of a number of safety measures concerning natural UF$_6$ shipments are assessed: truck shipments with escort, direct trains, additional outer packaging. The risk reduction associated with the adoption of a protective outer packaging (*) - within either transport mode - is by far the most important: 2 to 4 fold decrease. This measure appears therefore as the most effective for transport safety. A comparison is made between train and truck shipments for both petroleum products and natural UF$_6$. The relative safety levels (all on a per vehicle x km basis) of these different shipments are seen on the graph of Figure 6. Within either mode, shipping UF$_6$ is safer than shipping petroleum products by a factor of 6. Moreover, for both these hazardous materials, shipping by train is 12 times as safe as shipping by truck.

Table no 1: Shipments of UF$_6$

<table>
<thead>
<tr>
<th>Traffic (10$^3$veh x km)</th>
<th>Natural UF$_6$</th>
<th>Depleted UF$_6$</th>
<th>Enriched UF$_6$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Truck</td>
<td>100</td>
<td>280</td>
<td>0</td>
</tr>
<tr>
<td>Train</td>
<td>180</td>
<td>510</td>
<td>128</td>
</tr>
</tbody>
</table>

Table no 2: UF$_6$ shipments. Risk of container failure in 1975.

<table>
<thead>
<tr>
<th>Container failure (x 10$^{-4}$)</th>
<th>Natural UF$_6$</th>
<th>Depleted UF$_6$</th>
<th>Enriched UF$_6$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Standard shield</td>
<td>Protective shield</td>
<td>Direct train</td>
</tr>
<tr>
<td>Train</td>
<td>4.7</td>
<td>1.3</td>
<td>3.8</td>
</tr>
<tr>
<td>Truck</td>
<td>37.</td>
<td>9.</td>
<td>-</td>
</tr>
</tbody>
</table>
Table n°3: UF₆ shipments. Risk of container failure in 1980.

<table>
<thead>
<tr>
<th>Container failure (x 10⁻⁴)</th>
<th>Natural UF₆</th>
<th>Depleted UF₆</th>
<th>Enriched UF₆</th>
</tr>
</thead>
<tbody>
<tr>
<td>Train</td>
<td>13.4</td>
<td>10.</td>
<td>-</td>
</tr>
<tr>
<td>Truck</td>
<td>103.</td>
<td>57.</td>
<td>13.4</td>
</tr>
</tbody>
</table>

Figure n°6: Relative risk of shipping UF₆ (48 Y container) and refined petroleum (R.P.) Products by train and truck.

6. FUTURE IMPROVEMENTS

I would like now to discuss about the improvements to bring to the model and especially, I would like to point out the work we are doing to improve the fire severity indicators in connection with packaging behaviour under fire conditions.

At the moment, the only fire severity indicator is fire duration. Moreover, the behaviour of container is an S-type law \( f_{\text{S}}(t) = 1 - e^{-kt^2} \) assuming that for fire durations greater than 90 minutes the probability of packaging failure is .9. Fire extent and temperature are not taken into account and particularly the problem of hot spots is not considered.

(*) This protective shield is assumed independent of the containment vessel and increasing the package resistance to accident forces (either thermal or mechanical).
This major problem is now studied and specific tests are done to see:

1) whether it is a real problem for 48Y containers and if it is,

2) what are the characteristics of hot spots (extent, and temperature) to get a packaging failure after a given time and with a given probability.

7. CONCLUSION

The results furnished by the model has to be considered carefully; keeping in mind that
1) the chosen severity indicators could have been different especially for fire,

2) the relationship effort severity $\Rightarrow$ failure probability is not easy to define since specific tests all along the severity range are necessary to reach the value of failure probability associated to a given severity.

Any how, the model is completed and even if the results are not precise enough they allow to conclude:

- shipping by train is largely safer than shipping by truck,
- the use of an outer protective packaging for natural UF$_6$ in 48Y cylinder, is a safety measure more efficient than the use of direct trains or escort.

This last point shall be clarified using the results of tests actually performed in Cadarache.
SUMMARY
by the Session Chairman
C. Jouannaud
SESSION II A

In three papers release scenarios were discussed, the fourth paper in this session concerned the probability of transport accidents.

1. The safety problems associated with the handling and storage of UF₆. By Dr. W. Thomas

This paper deals with the safety problems associated with the handling of UF₆ in a large fuel fabrication plant.

After giving data on the toxicity of HF and uranium components, and scenario of an accidental release of UF₆ inside the building, it evaluates the consequences for the environment. It concludes that there are no serious consequences for the environment. For the personnel inside the premises this accident could be more serious, possibly lethal, if there is no possibility of prompt evacuation.

The second scenario is a plane crash on the UF₆ storage area. In the more pessimistic conditions, there is no serious problem at a distance of more than 1 kilometer. The criticality accident has been evaluated (moreover the probability being very low) and it was concluded that the consequences for the environment would be acceptable.


The authors explain the transfer model used in France to evaluate the dispersion of pollutants in the environment. This model is applied to UF₆, assuming that HF and UO₂F₂ are indifferent to gravity for two types of diffusion: normal and weak.

Answering a question of Mr. F. Irons (B.N.F.L.), Mr. Gérard explained that these two types of diffusion cover the six types used in Great Britain.
3. **Atmospheric dispersion and consequences of a UF₆ release caused by valve rupture on a hot 30B cylinder.** By Miss A.M. Ericsson (Sweden)

The scenario is the valve rupture on a 30B cylinder heated to 100°C in the "12 o'clock" position. (Gaseous UF₆ escaping). It is assumed that about 50% of UF₆ is discharged in 45 minutes (1 inch hole). In outdoor conditions, it is assumed that the zone of real danger would be restricted to very close of the source only (less than 100 m).

**Conclusion and discussion**

1) these three papers insist on the problem of the lack of knowledge on the behaviour of UF₆ with air moisture.

   - composition of the solid phase physical form of HF (droplets?).
   - Thermal effect of the chemical reaction on the plume.
   
   So, the evaluation of the risk for the environment is assumed to be pessimistic.

2) different types of scenario are made for the accident, and the scenario, and the probability of accident (not always evaluated) depend on the design of the plant.

   Generally, the more severe consequences occur from an outdoor release in the liquid form in the case of a plane crash on a UF₆ storage area followed by a fire of long duration.

   In the case of an accident inside the building, the consequences for the environment would not be dangerous outside the boundaries of the site.

3) the safety evaluations of damages are assumed as maximum credible accidents.

   It would be useful to have a probabilistic evaluation of such accidents.

4. **Safety analysis of UF₆ transportation**
   By Mr. F. Israël and Mr. P. Pages (CEA, France).

   A mathematical model is applied to UF₆ transportation. It concludes that shipping by train is 12 times safer than shipping by truck, and the adoption of a protective shell would improve safety by a factor of 2 to 3.
Studies are in progress to improve the severity indicators in connection with experiments on the behaviour of the containers under fire conditions. Also comparisons will be made with the experience on accidents with as dangerous materials as chlorine and ammonia.
SESSION II B

UF₆ BEHAVIOUR

EXPERIMENTAL VERIFICATIONS
Fundamentals of UF₆ accidental release
by
H. Bouzigues, M. Mezin
FUNDAMENTALS OF UF₆ ACCIDENTAL RELEASE

by H. BOUZIGUES *

M. MEZIN **

E. MESTRE *

presented at the

CSNI specialists meeting on the Safety Problems Associated with the Handling and Storage of UF₆

on June 28, 1978

at Boekelo (N.L.)

* COGEMA - 26100 PIERRELATTE

** CEA - DCh/DPI - BP 510 - 75752 PARIS - CEDEX 15
I. - INTRODUCTION.

The study of the physico-chemical properties of UF$_6$ date back from the research and developments made on the separation of uranium isotopes in the early forties.

Today, uranium is the base material of nuclear energy whether military or civilian and increasing amounts of this compound are processed world-wide. Under these conditions, and for the sake of safety, it was found mandatory to standardize handling, transport and storage of UF$_6$ in order to enter dutifully the era in which very large quantities will be handled.

But, the standardisation of methods must rest on the thorough knowledge and the taking into account of some properties of UF$_6$ which have not been underlined sufficiently in the past.

This is reflected in the results of critical studies of accidents which have occurred in the past in France as well as in the U.S.. It appeared that a large scale release of liquid UF$_6$ may lead to the death of individuals close to the source and result in the pollution of large areas around plant site.

This paper summarizes the main properties of UF$_6$ useful in the assessment of safety of specific procedures or in the explanation of the behavior of this fluid when tons are released. Let us consider first the thermodynamic properties:
II. - THERMODYNAMIC PROPERTIES OF UF₆

The most useful properties are summarized in Table I. They refer to high purity (UF₆ > 99.5 %) UF₆.

At room temperature, uranium hexafluoride is a colorless, transparent solid which sublimes without melting. UF₆ vapor behaves essentially as a perfect gas.

The phase diagram (Fig.1), in the conventional Clapeyron system of coordinates, shows a triple point at a temperature of 64.052 °C, and a pressure of 1541 mbars, i.e. somewhat above atmospheric pressure. The critical temperature is between 217 and 249°C and the corresponding pressure between 44 and 66 bars. At atmospheric pressure, the equilibrium sublimation temperature is of 56.4 ± 0.2 °C. This means that, contrarily to other fluids like chlorine and ammonia handled in large quantities, liquid UF₆ cannot exist in a stable form at atmospheric pressure.

When liquid UF₆ is released for instance from a failed vessel, part of it will vaporise spontaneously in the air. The energy required for vaporisation results first in the cooling of the liquid, then in the crystallisation of the remaining UF₆. The UF₆ cristallised will sublimate slowly at a rate governed by heat transfer.

The phase diagram displays also a fusion curve with negative slope. This is a result of Clapeyron's equation:

\[
\frac{dP}{dT} = \frac{\Delta H_F}{T (V_{liq.} - V_{sol.})} = -23 \text{ bars/K}
\]

Thus, the melting of UF₆ crystals results in a 35 % increase in volume.

When a UF₆ container is heated, melting may occur locally. Due to the large expansion at the phase change, this may induce in the metal stresses in excess of its elastic limit and result in the failure of the envelope.
This phenomenon is likely to occur under fire conditions in account of the large temperature gradients encountered.

Also, during the transfer of UF₆ between two cylinders or vessels, crystallisation of UF₆ may occur if gaseous UF₆ comes in contact with cold walls, valves or piping. This can hamper the transfer operations by plugging the lines or lead to subsequent trouble if the filling limits of the vessel are exceeded. For each type of vessel or cylinder, the volume is set with a minimum uncertainty of 5% based on UF₆ of at least 99.5% purity and a maximum temperature of 121 °C. If the filling is not done with checking of the mass transferred, there is a possibility of replacing liquid UF₆ by solid UF₆ of higher density with a subsequent risk of a hydrostatic rupture of the closed vessel or cylinder upon heating. In France, this type of incident has been experienced on several instances on small amounts of UF₆.

III. - MOLECULAR DATA AND TRANSPORT PHENOMENA.

Some physico-chemical properties of UF₆ enable to understand its behavior in its various states during heat or mass transfer operations concerning safety.

The main data are recalled in table 2. It shows that regardless of its physical state, UF₆ behaves like a heavy molecule.

When crystallised, UF₆ particles are dense and abrasive, their hardness resulting both from the covalent nature of the bonds and from the smallness of the crystalline lattice. It should be also noted that the linear expansion coefficient of the solid is twenty times larger than that of steel.

Above the triple point, the liquid is quite dense and very fluid, its kinematic viscosity being half that of water. The latter enables the liquid to be easily transferred by gravity. Moreover, liquid UF₆, in account of its lack of permanent bipolar momentum, has a...
low surface tension. Thus, liquid UF$_6$ can easily be sprayed and a large entrainment of particles occurs together with tumultuous boiling.

Such a behavior results in the weakness of the cohesion forces between the molecules of the liquid.

It is also apparent in the volatility, an important property in mass transfer. [4]

III.1 - Heat transfer. [5]

Gaseous or liquid UF$_6$ is a poor heat conductor. At 50°C, the thermal conductivity of air is three times that of UF$_6$ gas. Compared to water, liquid UF$_6$ has a heat transfer coefficient that is only .27 times as large. Finally, steel transfers heat a hundred fold faster than solid UF$_6$. Thus, in a container, a 1 cm layer of UF$_6$ crystals on the walls reduces drastically heat transfer.

UF$_6$ is neither easy to heat when crystallised nor easy to cool when liquid. About twelve hours are required for 120°C steam to fully melt a 12 ton cylinder. It takes at least 3 days at room temperature to be sure that all the UF$_6$ has become solid after filling the same cylinder.

III.2 - Volatility. [4]

The relative volatility of chemical species can be assessed by comparing the vaporisation enthalpies as they express the cohesion energy of the liquid state. That of UF$_6$ is rather high which enable purification to be made either by distilling the liquid or sublimating the solid.

Refering to the phase diagram, let us consider the liquid in equilibrium with its vapor. If, at a given temperature, the pressure above the liquid is suddenly decreased, the liquid will be in a metastable state. A fraction of the liquid will vaporise...
and, under adiabatic conditions, the vaporisation heat will be taken from the energy content: the liquid phase will cool down until the equilibrium temperature at the new pressure is reached. If the pressure drops steadily, vaporisation will proceed, the temperature of the liquid decreasing down to the triple point.

At this point, the system will remain invariant till the liquid has completely disappeared by being either vaporised or crystallised. The ratio of these two quantities is simply given by the ratio of the vaporisation $\Delta H_{LV}$ and fusion $\Delta H_{LS}$ enthalpies. It is equal to:

$$\frac{\Delta H_{LV}}{\Delta H_{LS}} = 1.5$$

As soon as all the liquid has disappeared, sublimation proceeds with subsequent cooling down of the solid until it comes in thermal equilibrium with the surroundings.

In some cases, this phenomenon can be very rapid as its rate rests on three independent parameters, namely:
- the area of the liquid
- the diffusivity $\text{UF}_6$-air
- the $\text{UF}_6$ concentration gradient.

However, the latter, closely dependent on volatility remains the governing factor for the duration of the metastable state of the solid or liquid phase at atmospheric pressure.

It can be shown that, between 121°C and the triple point, volatility which can be estimated from the variation in the thermodynamic potential, varies significantly. Indeed:

$$\Delta G = \Delta H_v - T \Delta S$$

with

$$\Delta S = \frac{\Delta H_v}{T} + R \ln \frac{P_{\text{sat.}}}{P_{\text{atm.}}}$$

.../...
the term $\frac{R \ln \frac{p_{\text{sat}}}{p_{\text{atm}}}}{p_{\text{atm}}}$ showing the disequilibrium in the liquid phase.

$\Delta G$ values are respectively:
- at 121 °C : $-1474$ cal./mole
- at 64 °C : $-269$ cal./mole

Thus, at 64 °C, volatility is much less than at 121 °C as the potential variation is reduced by a factor of 5.

This result shows the large temperature dependance of the amount vaporised instantaneously.

The fraction of the initial mass of UF$_6$ vaporised adiabatically until equilibrium is reached, can be calculated as follows:

i: From an initial amount of $M$ molecules, a quantity $m$ is vaporised. The unvaporised liquid cools from 121 °C down to 64 °C, the triple point; the heat balance is:

$$m \Delta H_{\text{LV}} = (M - m) C_{\text{PL}} \Delta T$$

ii: The system is invariant and will remain at 64 °C until all liquid UF$_6$ is transformed into solid and vapor. $m'$ molecules are vaporised:

$$m' \Delta H_{\text{LV}} = \left[M - (m + m')\right] \Delta H_{\text{LS}}$$

iii: Then solid UF$_6$ sublimes, cooling the solid down to 56 °C where its vapor pressure reaches atmospheric pressure. $m''$ molecules are vaporised in this process:

$$m'' \Delta H_{\text{SV}} = \left[M - (m + m' + m'')\right] C_{\text{PS}} \Delta T$$

iii: The hot solid is then in equilibrium with atmospheric pressure. Its temperature will decrease down to room temperature at a rate controlled by the diffusion of UF$_6$ gas and heat transfer.

When considering other fluids handled in large quantities such as chlorine or ammonia, basically the same phenomena occur but the rates differ.
Indeed, the triple point of these fluids lies well below atmospheric pressure and they remain liquid down to low temperatures. Thus, after an initial flashing due to superheating, vaporisation will cool the liquid down to temperature where its vapor pressure will be well below atmospheric. So, if surface and diffusion, the other two parameters governing vaporisation remain constant, the rate of vaporisation will be slower and the amount released in the initial flash smaller. In the case of UF₆, the coordinates of the triple point as well as the 1.5 ratio between vaporisation and crystallisation enthalpies inhibit temporarily the decrease in the temperature, hence in the pressure (Fig. 2).

III.3 - Adiabatic expansion of UF₆ gas.

In UF₆, the value \( \gamma \) of the specific ratio \( C_p/C_v \) unusually low, in account of the large number of atoms making up the molecule. The use of Laplace's equation (\( PV\gamma = \text{constant} \)) and of the ideal gas law enable the cooling down of hot UF₆ gas in an adiabatic expansion to be computed with a \( \gamma \) value of 1.06. The expansion of UF₆ gas at 100° C from 3000 down to 760 mm Hg induce a cooling of 29° C only. Under these conditions, the expansion of the vapor has very little effect indeed on the reduction of the quantities released in the air.

This cooling is small in comparison to that experienced with diatomic gas where, for the same initial and final pressures, a cooling of about 120° C would be attained. But UF₆ being generally transferred into an evacuated vessel, operators experience the plugging of the valve or of the interconnecting piping by UF₆ crystals if their heating is insufficient.

III.4 - Diffusion in air.

The data given in the literature refer to molecular diffusion of UF₆ in air but they are of little use in safety assessment.
Measurements on the diffusion of UF\(_6\) in dry nitrogen at given pressures and temperatures have recently been made in a laboratory in Pierrelatte. Apparent diffusion coefficients have been derived by the "Division de Chimie" at C.E.A, using Fick's law. These coefficients (table 3) show that UF\(_6\) behaves like a dense gas hardly mixing with air. The use of the turbulent diffusion law gives a large horizontal component as can be foreseen from the values obtained with nitrogen. Thus, in case of a massive UF\(_6\) release, a rapid spreading close to the ground of a dense cloud around the source must be expected.

This would not necessarily be the case in a turbulent environment and in presence of humidity due to the thermal effects from UF\(_6\) hydrolysis reactions.

As a consequence, working areas would be best protected using a ventilation "per descensum", as is generally the case whenever heavy vapors are involved.

IV. CHEMICAL PROPERTIES.

IV.1 - Fluorination

Uranium hexafluoride is an oxidising and fluorinating agent, but, at a lesser degree than elemental fluorine. It reacts with most metals leading quite often to the formation of a protective layer of fluorides which slow down, or even stop the attack. This is the case with nickel, copper, aluminum and iron.

The reactivity of UF\(_6\) towards fluorine acceptors is that of a rather strong fluorinating agent. When reacting, UF\(_6\) generally loses two atoms of fluorine. But the enthalpy and kinetics of reaction in gaseous state do not lead to detonations as would be the case with fluorine and chlorine trifluoride. However, when mixed in a condensed state with organic compounds, it may,
due to its high reactivity, constitute an explosive mixture.

Two extreme cases are worth mentioning:

- A mixture of Freon 11 (C Cl₃F) with liquid UF₆ explodes, giving Freons 13 and 14 (C ClF₃ and CF₄ respectively), these two compounds having low critical temperature.

- A mixture of liquid UF₆ with Talpa 30 vacuum pump oil (essentially aliphatic and naphtenic hydrocarbons) reacts with little pressure increase due to the fluorination of double bonds and the breaking off of the (CH₂)ₙ chains producing practically no gaseous compounds.

The risks associated with these types of reactions are prevented in enrichment plants by the specification of a maximum of 100 vpm molar of chlorofluorocarbons in the feed.

IV.2 - Reactions with water.

Water reacts vigorously with UF₆ to produce uranyl fluoride and hydrofluoric acid.

Where safety is concerned, two reactions must be mentioned:

- In a condensed state
  \[
  \text{UF}_6 (s) + 2 \text{H}_2 \text{O} (l) \rightarrow \text{UO}_2^{++} + 2 \text{F}^- + 4 \text{FH} (g)
  \]
  with a measured \( \Delta H = -50.5 \text{ Kcal/mole} \).

- In gaseous phase
  \[
  \text{UF}_6 (g) + 2 \text{H}_2 \text{O} (g) \rightarrow \text{UO}_2 \text{F}_2 (s) + 4 \text{FH} (g)
  \]
  with a \( \Delta H = -37.5 \text{ Kcal/mole} \).

The first reaction is important under accident conditions when one attempts to control a UF₆ release by dousing with water to prevent the spreading of uranium-fluorine compounds into the environment. But this method cannot be used undiscriminately and, as will be shown below, it can be justified in very few instances only.
The second reaction occurs every time UF₆ is released into the environment. Dense white clouds contain UO₂F₂ aerosols and HF. The size of the particles in the cloud is around .5 microns and their settling is very slow; this type of aerosol is dangerous for the lungs. From the stoichiometry of the reaction, and the HF smell threshold, UF₆ can be detected at .3 to .8 ppm (4 to 12 mg/m³).

After a release of gaseous UF₆, the ratio of fluorine to uranium detected on the ground increased widely with the distance from the source. This means that UO₂F₂ particles must settle out and HF be carried further away.

It has been shown that even for a concentration as low as 10⁻¹⁰ g/m³, visibility does not exceed 90 cm. In this kind of aerosol, the weight ratio of UO₂F₂ to HF is around 4; thus, only 23% of the weight of the aerosol is HF. Unreacted UF₆ may be present at concentrations below 20 mg/m³. As, only a few hundred grams of UF₆ released in a closed facility will create a toxic and blinding cloud for the operators. There is a risk that they would not readily find the emergency exits. And the shortness of the stay in a UF₆ atmosphere is an important factor for survival. Toxicity values for UF₆ and HF are given in table IV.

The maximum permissible concentration of HF in air is 1.9 ppm, i.e. 2.4 mg/m³ or 3 vpm.

It is the HF concentration which is the controlling factor in the survival: in the span of time required for inhaling a lethal dose of HF, only one tenth of the chemical dose for uranium will be attained and much less of its radioactive dose.
V - RADIOCHEMICAL PROPERTIES.

The toxicity of uranium is of chemical nature whenever enrichment is less than 5% U\textsubscript{235}.

The three isotopes of uranium found in nature are α emitters with long periods. But most of the β activity associated with uranium come from 234\textsuperscript{Th} and 234\textsuperscript{Pa}, daughter products of U\textsubscript{238}; the remaining β activity coming from Thorium 231 [117]

\[
\begin{array}{cccccc}
238\textsuperscript{U} & \xrightarrow{\alpha} & 234\textsuperscript{Th} & \xrightarrow{\beta} & 234\textsuperscript{Pa} & \xrightarrow{\beta} & 234\textsuperscript{U} \\
92 & 4.1 \times 10^9 y & 90 & 24.1 d & 91 & 1.2 \text{ min} & 92 & 2.5 \times 10^5 y
\end{array}
\]

and the radiation from Protactinium is quite hard (2.3 MeV).

During some operations, the β activity associated with uranium may become separated, for instance by distillation and be deprived from the shielding effect of uranium. Lead shielding may then become necessary. On the other hand, the freshly distilled uranium does not exhibit the activity associated with secular equilibrium. It is practically reached in about 3 months as it is the period of Thorium 234 that controls its rise.

As U\textsubscript{238} is the naturally most abundant isotope and the specific activity of 234\textsubscript{Th} is quite high, thorium is very dangerous as far as air contamination is concerned. The risk factor depends upon two parameters:

- The biologic risk depending upon the radiation spectrum and the time taken for body elimination.
- The risk of inhalation depending upon the specific activity [127].

About 1 curie of thorium and protactinium are in equilibrium with ca 3 tons of natural uranium. But the corresponding mass of Th + Pa is only 43 micrograms. Thus, the risk associated to 234\textsubscript{Th} and 234\textsubscript{Pa} is similar to that of 239\textsuperscript{Pu}. However, radiothorium 234 is always fixed onto powdery uranium solids which reduce their probability of inhalation.
VI - MASSIVE UF₆ RELEASE.

A massive UF₆ release may result from the failure of a vessel or its associated piping containing liquid UF₆. A temperature of 12¹⁰ C, the maximum permissible on cylinders, is assumed. The thermodynamics involved are described below for a release as a vapor and as a liquid through the opening left by a failed valve.

VI.1 - Vapor release rate.

For a release as a gas, let us assume an opening of \( S \text{ cm}^2 \) in a thin wall and pressures respectively \( P_1 \) and \( P_2 \). The mass flow rate \( D \) is then expressed by:

\[
D = S \left( \frac{P_2}{P_1} \right)^{\frac{1}{\gamma}} \sqrt{\frac{2 \gamma}{\gamma - 1}} P_1 V_1 \left[ 1 - \left( \frac{P_2}{P_1} \right)^{\frac{\gamma - 1}{\gamma}} \right]
\]

Fig. 3 shows the variation of the flow rate to the atmosphere as a function of the temperature of the remaining liquid. Under adiabatic conditions, cooling down of the liquid is observed as vaporisation proceeds. Fig. 4 shows the gas flow rate from a 12 tons 45Y cylinder, initially at 12¹⁰ C after a valve failure, the opening having an area of 11.4 cm².

The phenomenon can be described along the three following time periods:

i : A short initial period of about 30 minutes during which 3.4 tons of vapor are released at a rate varying from 2.65 to 1.2 kg/sec.

ii : As the first UF₆ crystal appears, a second period about twice as long, where 3.34 tons of UF₆ vapor are released at the constant flow rate of 1.2 kg/sec until no liquid remains.

iii : A third period where 152 kg only of solid UF₆ sublime at a slow rate until the pressure within the cylinder reaches atmospheric. No more release then occurs.
In all, 6.9 tons of UF₆ have been released, i.e. 58% of the initial amount over a total time of about 80 minutes (Fig. 5). These results agree with data obtained in laboratory.

VI.2 - Liquid release rate.

Using the same basic assumptions as for a vapor release, the flow rate is given by:

\[ D = S \rho K \sqrt{\frac{2 g}{h + \frac{P_2 - P_1}{g}}} \]

where \( K \) is the contraction coefficient of the liquid (assumed at .65).

\( h \) the head of liquid above the opening

\( g \) the gravity.

Fig. 6 shows the amount of liquid released versus time for a 48 Y cylinder lying horizontally and having its valve at the lowest. It takes only four minutes for 80% of the liquid contained, that above valve level, to spill on the ground. This is a very short time to take counteractive measures. The only efficient one, would be to place the valve opening in the gas phase, the rate of release being then much slower.

As soon as it leaves the cylinder, the liquid will flash adiabatically, 2.7 kg of UF₆ being vaporised every second, spreading droplets and crystals around until the remaining solid (42% of the liquid that flowed out) is at atmospheric pressure, the three steps described earlier taking place in rapid sequence.

The diffusion of UF₆ in the air being slow and UF₆ being a dense gas spreading out close to the ground, the area for heat transfer with the surroundings may increase rapidly. Thus, when the initially dry vapor will cool down at temperatures below 56° C, UF₆ crystals will be formed and part of the UF₆ released in the initial flash will settle down in crystalline form around the source. The percentage thus extracted from the "cloud" depends...
essentially on the initial temperature of the flash, on the
temperature of the surroundings and on heat transfer, essentially
with the ground. This is an important phenomenon as far as safety
is concerned: the fraction of the vapor which will crystallise
will be proportionately more important when the liquid flashing
is close to the triple point. With liquid UF₆ at 80°C, close to
90% of the UF₆ should be found in the vicinity of the source.

The importance of the area contaminated depends upon the turbulence
of the atmosphere at the time of the release as well as upon the
temperature of the liquid.

With air containing humidity, the UF₆ crystals formed may be
coated with hydrates of uranyl fluoride \( \text{UO}_2\text{F}_2 \cdot x\text{HF} \cdot y\text{H}_2\text{O} \).

VII - STUDY OF AN ACTUAL RELEASE.

VII.1 - The incident.

The incident happened on July 1\text{st}, 1977 on a 48 Y cylinder
containing 8,327 kg of liquid UF₆ at around 95°C after a liquid
phase sampling.

The cylinder was resting on a 
\textit{berth}, close to the ground next to
the sampling building, its valve being at the lowest position.
During handling, a mishap occurred and the valve still connected
to the sampling building via a flexible tubing, broke flush
with the cylinder wall leaving an opening slightly under one inch
diameter: its area was 3.84 cm².

The release of toxicants took place over nearly one hour.
Using fire fighting equipment, 13.5 m³ of water were sprinkled
on and around the source over about 1/2 hour. Moreover, 600 kg
de liquid carbon dioxide were sprinkled 15 - 20 minutes after the
initial burst.

A total of 7,106 kg of UF₆ were released to the atmosphere according
to a weighing of the cylinder plugged at the end of the release

.../...
which showed a residual amount of 1,721 kg of UF₆.

VII.2 - Data collected.

About thirty locations could be used for sampling near ground level and at distances ranging up to 50 km from the source. Very few of them were located on the axis of the plume and were able to integrate the pollution. The most representative ones are indicated in table 5. Measurements of ground deposits have been made and data have been extrapolated using conventional atmospheric diffusion codes \(^{14}\).

The three following points are worth stressing:

With a wind velocity of 10 m/sec,

i : the amount of uranium detected in the atmosphere is only a small fraction of the UF₆ vaporised.

ii : Hydrogen fluoride from UF₆ hydrolysis can be traced as expected in an area extending 15 km downwind.

iii : The fluorine to uranium weight ratio in the ground contamination becomes significantly large beyond 1000 m.

At the locations where the highest concentrations have been detected, chemical dangers were small. As far as the radioactivity danger is concerned, it being associated to uranium deposited on the ground, 150 mrem could be attained on kidneys, at a distance of 600 m downwind from the source. But this point lies within the controlled area of plan site.

VII.3 - Discussion of data.

The flow of liquid UF₆ at 95° C lasted for 11 minutes in the abovementioned valve opening. The hottest vapor was produced...
during the initial flash. Representing 16% of the total amount released, due to its energy content, it was prone to be carried the furthest away from the source.

The amount that evaporated was less than 48% of the amount released, 33% of which were vaporised at the triple point.

Thus, the bulk of the amount vaporised was evaporated at a temperature only 8°C above the freezing point at atmospheric pressure (56°C). It seems proper that most of that vapor would have condensed in the vicinity of the source.

The uranium concentration found agree with such an assumption as diffusion code show that less than 6% of the uranium vaporised could be found in the atmosphere and it may be assumed that the remaining 94% settled near the source. On the other hand, the quantities of HF detected indicate a significant hydrolysis of the UF₆ released. In account of the rate of the release and of the humidity content of the air, the quantities found at long distances can only be explained by the deluge of water.

Otherwise, the hydrolysis of 7 tons of UF₆ would have required its perfect mixing with 10^5 m³ of air containing 1% humidity by weight.

As it appeared from the logbook, fire hoses were used early against the source. But their effect must have widely contributed to the hydrolysis of the UF₆ vapor in the air and crystals on the ground. The quantities of uranium analysed in the ground after the incident are in agreement with the above scheme.
CONCLUSIONS.

As we have shown, a massive release of $\text{UF}_6$, in account of the thermodynamic potential involved, is very difficult if not impossible to control.

Safety must then rely essentially on prevention. Firstly, only equipment of proven quality and in agreement with international regulations must be used. Secondly, operators should proceed only in compliance with well-established and proven procedures. Finally, the various operations should be performed only by well-trained operators under adequate supervision.

Prior to any heating, the equipment must be checked to be in good condition. This concerns particularly the valves. Moreover it will be checked that the filling limits of cylinders or vessels have not been exceeded.

Handling of cylinders insufficiently cooled should be reduced to a minimum and to short distances only. Valves on cylinders being handled or transported should be well-protected against mechanical shocks as they constitute the Achille's heel of heavy cylinders. These are delicate to handle, in account of their weight, the more so when they still contain a fraction of $\text{UF}_6$ still liquid. Keep in mind the long time for a $\text{UF}_6$ container to freeze completely.

A cylinder should be used with its valve at the lowest only to be emptied out, the uppermost position, being the safest, being adopted otherwise.

Incident or accident fighting is very uneasy when dealing with a massive release due to the density of the cloud and to the toxicity of the vapor. As can be inferred from the rates of the releases, the first measure to be taken is to evacuate speedily people without gas masks and suitable protective clothing, away from the cloud.

.../...
Then, the only suitable measure is to attempt to plug the opening by sprinkling liquid CO$_2$ to freeze UF$_6$. However this can be successful only if the leak rate is small enough. However the overall cooling effect given by CO$_2$ favors the settling down of the UF$_6$ vaporised.

The cooling effect obtained with water is much less as its temperature cannot be as low as that of dry ice and heat transfer is hindered by the presence of UF$_6$ crystals. Moreover, the reaction between UF$_6$ and water releases heat. Finally, criticality control cannot always be guaranteed when using this fluid.

Nevertheless, water can be successfully used downwind of the source to bring down the fumes by having the plume cross a wall of water.

Afterwards, UF$_6$ or its hydrolysis products remaining on the ground should be covered with hydrated lime or alumina prior to their removal.
TABLE 1.

At 64° C

<table>
<thead>
<tr>
<th>Property</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Enthalpy of vaporisation (liquid)</td>
<td>7.038 Kcal/mole</td>
</tr>
<tr>
<td>Enthalpy of fusion (solid)</td>
<td>4.588 Kcal/mole</td>
</tr>
<tr>
<td>Enthalpy of sublimation (solid)</td>
<td>11.63 Kcal/mole</td>
</tr>
<tr>
<td>Heat capacity (solid)</td>
<td>43.49 cal/mole°C</td>
</tr>
<tr>
<td>Heat capacity (liquid)</td>
<td>43.59 cal/mole°C</td>
</tr>
<tr>
<td>Heat capacity (vapor)</td>
<td>31.5 cal/mole°C</td>
</tr>
<tr>
<td>Specific heat ratio $C_p/C_v$</td>
<td>1.062</td>
</tr>
</tbody>
</table>

TABLE 2

<table>
<thead>
<tr>
<th>Property</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Molecular weight</td>
<td>352</td>
</tr>
<tr>
<td>Density : solid at 20.7° C</td>
<td>5.090 kg/m³</td>
</tr>
<tr>
<td></td>
<td>4.920 kg/m³</td>
</tr>
<tr>
<td></td>
<td>3.674 kg/m³</td>
</tr>
<tr>
<td></td>
<td>3.404 kg/m³</td>
</tr>
<tr>
<td>Liquid at 100 °C</td>
<td>12.1</td>
</tr>
<tr>
<td>Relative density vapor (air)</td>
<td>3.2 $10^{-4}$/° C</td>
</tr>
<tr>
<td>Cubic expansion coefficient of liquid</td>
<td>2. $10^{-3}$/° C</td>
</tr>
<tr>
<td>Kinematic viscosity liquid at 67.2° C</td>
<td>0.200 centistokes</td>
</tr>
<tr>
<td>Surface tension liquid at 100° C</td>
<td>13 dynes/cm</td>
</tr>
</tbody>
</table>
### TABLE 3

**UF$_6$ VAPOR DIFFUSION COEFFICIENT IN NITROGEN**

<table>
<thead>
<tr>
<th>Turbulent vertical (upwards)</th>
<th>$D_{1,2} = 0.2 \text{ cm}^2/\text{sec.}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Turbulent vertical (downwards)</td>
<td>$D_{1,2} = 90 \text{ cm}^2/\text{sec.}$</td>
</tr>
<tr>
<td>Turbulent horizontal</td>
<td>$D_{1,2} = 600 \text{ cm}^2/\text{sec.}$</td>
</tr>
</tbody>
</table>

### TABLE 4

**TOXICITY OF UF$_6$**

<table>
<thead>
<tr>
<th>Light incident</th>
<th>$0.3 \text{ g/m}^3$</th>
<th>Irritation of mucous membranes</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$0.5 \text{ g/m}^3$</td>
<td>Attack of the respiratory tract. Tissues destroyed may be regenerated.</td>
</tr>
</tbody>
</table>

| Severe incident | $1 \text{ g/m}^3$ | Recovery possible for exposure less than 1/2 hour but after effects on bones and teeth. Death for exposures between 1/2 and 1 hour. |

<p>| Lethal dose     | $3 \text{ g/m}^3$ | Death by acute edema of the lungs after an exposure of a few minutes. |</p>
<table>
<thead>
<tr>
<th>Temperature T °C</th>
<th>121</th>
<th>110</th>
<th>100</th>
<th>90</th>
<th>80</th>
<th>70</th>
</tr>
</thead>
<tbody>
<tr>
<td>ΔG (Kcal/mole)</td>
<td>-1.5</td>
<td>-1.25</td>
<td>-1.0</td>
<td>-0.8</td>
<td>-0.6</td>
<td>-0.4</td>
</tr>
<tr>
<td>% vaporised T → 64 °C</td>
<td>35</td>
<td>29</td>
<td>24</td>
<td>18</td>
<td>11</td>
<td>3.8</td>
</tr>
<tr>
<td>% vaporised at triple point</td>
<td>25</td>
<td>28</td>
<td>30</td>
<td>32</td>
<td>35</td>
<td>37.5</td>
</tr>
<tr>
<td>% sublimated 64° C → 56° C</td>
<td>1.1</td>
<td>1.2</td>
<td>1.3</td>
<td>1.4</td>
<td>1.6</td>
<td>1.7</td>
</tr>
<tr>
<td>Total vaporised %</td>
<td>61</td>
<td>58</td>
<td>55</td>
<td>51</td>
<td>48</td>
<td>43</td>
</tr>
</tbody>
</table>

Nota: The isotherm expansion of the vapor has been neglected
### TABLE 6

**COMPONDED CONCENTRATIONS AT GROUND LEVEL**

<table>
<thead>
<tr>
<th>DISTANCE (km)</th>
<th>.6</th>
<th>1</th>
<th>3</th>
<th>6</th>
<th>18</th>
<th>50</th>
</tr>
</thead>
<tbody>
<tr>
<td>CONCENTRATION</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>FLUORINE</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>(gram. second)</td>
<td>39</td>
<td>12</td>
<td>1.2</td>
<td>.27</td>
<td>.04</td>
<td>.002</td>
</tr>
<tr>
<td>(m³)</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>URANIUM</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>(gram. second)</td>
<td>9.8</td>
<td>3.6</td>
<td>.72</td>
<td>.24</td>
<td>.05</td>
<td>.02</td>
</tr>
<tr>
<td>(m³)</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>F/U by weight</td>
<td>.23</td>
<td>.61</td>
<td>1.6</td>
<td></td>
<td>5.5</td>
<td>12.5</td>
</tr>
<tr>
<td>on the ground</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

* values measured
Fig. 1 - PHASE DIAGRAM
INFLUENCE OF THE TEMPERATURE OF LIQUID UF₆ ON THE AMOUNT VAPORIZED AT ATMOSPHERIC PRESSURE

Fig: 2
Specific Flow rate
Kg/sec.m²

UF₆ VAPOR FLOW RATE THROUGH AN OPENING
VS. LIQUID TEMPERATURE

Fig. 3

\[
\frac{D}{3} = \left(\frac{P_1}{P_f}\right)\frac{\sqrt{2}}{n} \left(1 - \left(\frac{P_1}{P_f}\right)^\frac{2}{3}n\right)
\]

Pᵣ pressure atmosphérique
Vapor flow rate
(Kg UF₆/min.)
UF₆ VAPOR FLOW RATE FROM A 48 Y CYLINDER AT 121°C
THROUGH A 1.5" I.D. OPENING.

Time (minutes)

0  20  40  60  80  100
3000  4000  5000

First crystal appears
Last drop of liquid disappears
End of release

Fig. 4
Amount released
(Kg UF₆)

UF₆ VAPOR RELEASED FROM A 48 Y CYLINDER AT 121°C
THROUGH A 1.5" I.D. OPENING.

Fig. 5
LIQUID UF₆ RELEASED FROM A 48 Y CYLINDER AT 121°C THROUGH A FAILED VALVE.

Amount released (Kg UF₆)

Fig. 6
$C_t \text{ max (sec/m}^3) = f(d)$

**Fig. 5**

Normal diffusion - Moderate wind to low wind.
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Experimental approach of main parameters for safety evaluation of uranium hexafluoride industrial utilizations

by

A. Raggenbass, M. Berlin, J.C. Malet
EXPERIMENTAL APPROACH OF MAIN PARAMETERS FOR SAFETY EVALUATION OF URANIUM HEXAFLUORIDE INDUSTRIAL UTILIZATIONS

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The safety studies of uranium hexafluoride industrial installations can be made with accident scenarii. Knowledge of some physicochemical characteristics is absolutely necessary to realise such scenarii.

In some cases, only an experimental determination can provide these data:

- physical behaviour of gaseous UF₆ transformation conditions of liquid uranium hexafluoride into gas,
- uranium hexafluoride hydrolysis by water in various conditions,
- reaction between uranium hexafluoride and kerosene,

experimental conditions and results are given in this paper.
Three types of accidents were particularly studied during elaboration of the safety report of Eurodif installation and the report on uranium hexafluoride transportation conditions:

(1) Gaseous uranium hexafluoride release (10 tons) after an accidental rupture of vessel in an industrial building.
(2) Rupture of a transportation container filled with liquid uranium hexafluoride.
(3) Transportation accident followed by release of a fairly large amount of liquid uranium hexafluoride.

I. SCENARIO OF ACCIDENTS

From the initial conditions of the accident up to the consequences on workers or environment it is possible to build in any case a scenario with several steps inside. These scenario and steps studied below, set a certain number of questions and the answers to these questions were researched in an experimental program.

I.1. Vessel rupture in an industrial structure -

The first question is: how is uranium hexafluoride (very heavy gas), mixed with a neutral gas?

The second question is: what is the hydrolysis course of uranium hexafluoride
- with water vapour in atmosphere,
- with a water pulverisation (in case of water pipe rupture),
- with a sheet of water.

The third question is: what are the conditions of reaction of uranium hexafluoride with available kerosene in case of aircraft crashes?

The knowledge on the answers to the above questions allow to determine:

... / ...
- the rate and the characteristics of polluting emission,
- the nature of polluting compounds through the hole made by the missile.

After that, the study of atmospheric dispersion of these polluting products to man is possible to be carried out.

I.2. Accidental rupture of a transportation container filled with 10 tons of liquid uranium hexafluoride. [1]

This accident may occur when uranium hexafluoride as liquid or gas is drawn off to an installation. Stop-valve rupture was selected as the main cause of this type of accident. Recent accidents demonstrated that this point of view was realistic.

The thermodynamic data on uranium hexafluoride and particularly its triple point in the equation of state at 66°C temp. and 1.5 bar pressure point out that the liquid phase is unstable under atmospheric pressure; in consequence ejection outside of the vessel under several bars of pressure gives instantaneously a transformation of this liquid in gas and solid. This reaction is completely athermal near the triple point, consequently no limitation of speed is possible by the heat control of the phenomenon. The ejection speed of gas or liquid was calculated [2].

Nevertheless we found useful to make experiments on the above phenomena to confirm theoretical studies.

For the liquid release outside the installation the relative proportion of gas and solid which is 65% of gas with a starting temperature of 100°C, may be slightly different according to external conditions. After this step we find again the same problem as in the first type of accident, that is to say:

- hydrolysis by atmospheric vapour,
- or hydrolysis by water pulverisation,
- or hydrolysis by sheet of water.
I.3. **Transportation accident followed by release of a fairly large amount of liquid uranium hexafluoride.**

The most probable scenario is as follows:

To obtain a significative release of UF₆ with an accident during transportation of solid uranium hexafluoride, heat must be furnished. Consequently the transportation accident must be associated with an heterogeneous fire. In these conditions, it is possible to have a failure of the container due to the appearance of liquid uranium hexafluoride pressure (dilatation of more than 30% in solid → liquid transformation), then hexafluoride vaporization immediately after destruction of the container.

II. **EXPERIMENTAL PROGRAM**

During 1976-1977, an experimental program has been developed in close collaboration between PIERRELATTE Laboratories, Eurodif and Safety Laboratories of Protection and Nuclear Safety Institute in CADARACHE, on:

1. experimental diffusion of uranium hexafluoride in an inert gas;
2. experimental determinations of physical characteristics of liquid uranium hexafluoride release;
3. uranium hexafluoride hydrolysis
   - by atmospheric water vapour,
   - by water pulverization,
   - by a sheet of water;
4. reaction of uranium hexafluoride on kerosene;
5. destruction of an uranium hexafluoride container by hydrostatic pressure of liquid UF₆.

In the present paper, only points from 1 to 4 have been developed, the fifth point has been developed in another paper of this seminar.
II.1. Experimental diffusion of uranium hexafluoride in an inert gas.

Made with external detection of uranium using the $\gamma$-ray of $^{235}$U in a cylindrical vessel, this experimental series gave the following results:

- below the UF$_6$ inlet, mixing with nitrogen was made almost immediately,
- above the inlet, diffusion of UF$_6$ gas is less than 1 cm/min.

II.2. Experimental determination of physical characteristics of liquid uranium hexafluoride release.

Experimental conditions:

The experiment was held in an air tight metal test cell of 22 m$^3$ (Fig. I). About 2 kg of liquid uranium hexafluoride at 100$^\circ$C, were released in the test cell, through a valve of 0.8 cm$^2$ in section. Movies were made of gas release and of container weight evolution. Vaporization of 1250 g was achieved in 10 seconds, 88% in weight of UF$_6$ (1750 g) vaporized in 20 seconds.

It is clear that the gas jet carries droplets of liquid which disappear within a few centimeters. The theoretical behaviour of liquid UF$_6$ is completely confirmed by this experiment.

II.3. Studies on hydrolysis conditions of gaseous uranium hexafluoride.

All the experiments were held on the 22 m$^3$ test cell equipped with an air conditioning system. The instrumentation allowed:

- granulometric analysis of the aerosols
- infra-red analysis of the gaseous hydrogen fluoride and uranium hexafluoride
- measurement of the temperature

II.3.1. Hydrolysis by atmospheric water vapour

Experimental conditions (Fig. II)

The test cell contained air with a relative humidity of 85%. A plexiglass container of gaseous uranium hexafluoride was suddenly opened. The water vapour and the hexafluoride were therefore in contact.

... / ...
RESULTS
=======
The formation of white fumes made the atmosphere opaque.
The various measurements, Uranium concentration in the aerosols in the upper part of the test cell (Fig. III), and concentration of gaseous HF and UF₆ in the container (Fig. IV) prove that the hydrolysis reaction duration was about 50 minutes.

No rise in temperature was evidenced. In the upper part of the box, the existence of gaseous hydrogen fluoride was not established. More than 70% of the uranium remained in the initial vessel.

Consequences on the accident scenario:

(a) Consequences on the dispersion model
- Hydrofluoric acid concentrations in air will be smaller but the pollution duration will be longer,
- only few percents of initial uranium will be involved in the diffusion mechanism in the granulometric range where particles behave like a gas,
- we cannot hope that the temperature effect will result in raising the polluting cloud.

(b) Consequences on man
- near the pollution source, workers can be injured by UF₆ itself,
- in the experiment conditions, there is a pollution of the environment by a fog of liquid hydrofluoric acid but not by gaseous HF.

II.3.2. Uranium hexafluoride hydrolysis by water pulverisation.

Experimental conditions (Fig. V)

This experiment was carried out in a dry atmosphere.
A plexiglass container filled with a mixture of dry air and gaseous uranium hexafluoride was opened and simultaneously, water was projected into the container by using a jet situated at the bottom of it. The jet was adjusted so as to get a cloud of very fine droplets. All the water introduced into the container stayed inside it. The quantity of water was calculated so as to react stoechiometrically with the hexafluoride (115 ml of water and 1.1 kg of UF₆).
EXPERIMENTAL RESULTS
====================

In this case the hydrolysis reaction was very fast. We detected gaseous hydrofluoric acid in the test cell (half of the theoretical yield of the reaction)
- 60% of uranium remained in the container,
- the gas temperature in the plexiglass container increased by 40°C during experiment (Fig. VI).

II.3.3. Gaseous uranium hexafluoride hydrolysis by sheet of water.

Experimental conditions
Less than 1 kg of gaseous uranium hexafluoride have been released above a sheet of water of 20 liters (0.6 m²) during five hours in a wet atmosphere (Fig. VII).

RESULTS
=======

The main results of this experiment are:
- we did not obtain a significant temperature increase,
- no significant amount of gaseous fluorhydric acid was detected in the atmosphere of the box.

An important fog appeared almost immediately after the start of the reaction and we found 2/3 of initial uranium
1/3 initial fluor in the sheet of water.

II.3.4. General results from hydrolysis experiments.

These first series of experiments point out the importance of experimental approach for the safety studies on uranium hexafluoride.

(1) to prevent potential hazards associated with UF₆ utilization,
(2) to obtain efficiency in intervention.

Some data are now better known:
Uranium hexafluoride hydrolysis by atmospheric water vapour is not so fast we thought before. We do not find gaseous hydrofluoric acid in the products of reaction; 95% of the total uranium involved in the reaction remain near

... / ...
the reaction point. Other characteristics need further investigations, particularly the knowledge about uranium hexafluoride hydrolysis by water pulverization is of great importance to define possible intervention conditions.

II.4. Gaseous uranium hexafluoride action on kerosene

Contact between uranium hexafluoride and kerosene may happen during an airplane crash or a transportation accident. Therefore an experiment on this chemical reaction was useful.

Experimental conditions: 0,75 kg of gaseous uranium hexafluoride have been poured on a sheet of kerosene (10 liters 0,6 m²). The experimental set-up was the same as for the hydrolysis by a sheet of water.

RESULTS
=======
- no temperature increase was noticed,
- no gaseous hydrofluoric acid was detected in the containment; we noted the formation of a solid compound (containing UF₄) found at the bottom of the vessel.

CONCLUSIONS
=============
In our experimental conditions, we do not obtain:
- an explosive reaction,
- the production of gaseous and polluting compounds.

III - GENERAL CONCLUSIONS

During the safety studies needed by industrial utilization of uranium hexafluoride, many data were not available and experimental verifications and experimental determinations were required.

Our works in this field are not complete. But results already in our possession seem very useful for teams in charge of risk prevention associated with UF₆ handling and for teams in charge of intervention preparation.

... / ...
We saw that liquid uranium hexafluoride is very rapidly released and converted into gas,

- in case of important accidents with uranium hexafluoride, cooling (by a cooled inert gas for example) would transform gaseous UF$_6$ in solid UF$_6$ with important limitations for evaporation and hydrolysis rates.

- for a good definition of pollution source, we have a better understanding of the nature and formation rate of polluting gas provided by uranium hexafluoride hydrolysis by the atmospheric vapour; we have not the same knowledge when liquid water is poured accidentally or intentionally on uranium hexafluoride releases.

- From a toxicological point of view, the evaluate real risks to workers and the environment during an important release of uranium hexafluoride we know now that:

  - UF$_6$ as a compound may exist during almost one hour in a confined area; we have to know the toxicity of UF$_6$ to perform the accident evaluation,

  - as no gaseous FH was evidenced, but droplets containing FH were found to exist as a toxic fog, the problem is to know the real toxicity of such fogs,

  - on the contrary, at a fair distance of the pollution emission point, the fluor compound toxicity is the only hazard.

... / ...
VAPORISATION OF LIQUID UF₆, EXPERIMENTAL SET-UP

![Diagram](image)

HYDROLYSIS OF GASEOUS UF₆ BY WATER VAPOUR, EXPERIMENTAL SET-UP

![Diagram](image)
CONCENTRATION OF URANIUM AÉROSOLS IN THE TEST CELL

Fig: 3

U concentration (g/m³)

Sampling point 1
Sampling point 2

CONCENTRATION OF GASEOUS HF AND UF₆ IN THE TEST CELL. SAMPLING POINT 3

Fig: 4

Gaseous UF₆ (g/m³)

Gaseous HF (g/m³)

UF₆
HF
HYDROLYSIS BY A PULVERISATION OF WATER
EXPERIMENTAL SET-UP

Fig: 5

MEAN TEMPERATURE
IN THE 240 LITERS PLEXIGLASS CONTAINER

Fig: 6
HYDROLYSIS BY A SHEET OF WATER
EXPERIMENTAL SET-UP

Fig: 7

Test cell
H₂O vapour = 0.34 g/m³

Aerosols analysis
H₂F₆ analysis

Aerosols analysis
H₂F₆ analysis

Sheet of water: 20L, 0.6 m²

Water sampling
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DSN-76/281, mai 1976.


SUMMARY

by the Session Chairman

dr. M. Banaschik
In an era of expanding technology and safety consideration of the physical and chemical properties of UF₆ several studies have been made to better describe safety parameters for the handling and transport of uranium hexafluoride.

With the projected expansion of the nuclear power reactor program and increased UF₆ flows, these studies become increasingly important as bases for specifications and regulations.

As it was pointed out the uranium hexafluoride is a highly reactive substance. It reacts chemically with water, and most organic compounds forming soluble reaction products.

It does not react with oxygen, nitrogen or dry air.

Gaseous UF₆, when released to the atmosphere, reacts with the atmospheric moisture to form HF gas and particulate UO₂F₂ which tend to settle on the surface. The corrosive properties of UF₆ and HF are such that exposure to a severe release can result in skin burns and temporary lung impairment. The discharge rates are a very sensitive function of the phase conditions. The safety analysis of UF₆ handling indicates a need for an experimental verification of the behaviour of UF₆ during a release.

The presented papers about the experimental verification of UF₆ behaviour have shown that the theoretical approach is very conservative.

Therefore further investigations has to be initiated in the following safety problem fields in order to quantify the safety margins in the UF₆ handling:

- heating-up mechanism of UF₆
- behaviour of UF₆ during large release of fluid UF₆
- dispersion of chemical reaction substances as UF₆
- dispersion and deposition of UO₂F₂
- dispersion and critical concentration of HF.
SESSION III
INCIDENT EXPERIENCE
Experience on UF₆ - handling in uranium manufacturing processes

by

F. Schinzer, R. Hille
EXPERIENCE ON UF₆-HANDLING IN URANIUM MANUFACTURING PROCESSES

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Hanau 11

For more than 10 years the fuel element plant NUKEM GmbH in Hanau has been practising the handling and storage of uranium in the form of UF₆. It is a matter of UF₆ with depleted, low and highly enriched uranium. Furthermore, NUKEM has developed sampling procedures for UF₆ to determine the U 235 and the uranium content. The essential outcome of this operating experience has proven that with appropriate safety precautions the handling and storage of UF₆ can be carried out without danger.
Since more than ten years NUKEM has fabricated fuel elements especially for Material Test Reactors (MTR) and High-Temperature Reactors (HTR). The uranium used in these types of elements is enriched in U-235 up to 93%. It is usually supplied in the form of uraniumhexafluoride (UF₆). The container used is the type 5A with a diameter of 127 mm and a length of 762 mm, the contents can be up to 25 kg UF₆ or 16 kg Uranium. At some times 50 containers of this type had to be stored at NUKEM simultaneously.

At the beginning of the fuel fabrication the UF₆ must be converted into the desired chemical form e.g. metal for MTR-fuel and U/Th-oxide for HTR. The container must be heated to evaporize the UF₆ which flows through pipes into a reaction chamber containing perchlorethylene (C₂Cl₄). There UF₄, an insoluble salt stable in air is formed. This can be brought into metallic form by the reduction with calcium metal or to an oxide by a treatment with water vapor at higher temperatures.

For MTR-Fuel elements the uranium is mixed with aluminium metal and rolled to plates. For HTR-fuel the oxide is dissolved in nitric acid mixed with thorium nitrate casted into ammonia to form small spherical particles which are sintered. Besides the production of high enriched uranium fuels NUKEM also processes and stores low enriched, natural and depleted uranium. This uranium is mainly processed for research and development purposes and as far as depleted uranium is concerned for production of shielding devices. The storage of low enriched, natural and depleted uranium is done under Federal Government contract.

In the last years we have stored the following quantities of UF₆:

- High enriched up to 1,25 tons UF₆ in 50 containers.
- Since 1971: low enriched uranium, 340 tons in 154 containers (30 A and B). The amount increased in 1975 to 600 tons in 272 containers.
- Natural and depleted uranium, the latter in 12 containers of the type 48A.

The storage of high enriched UF₆ is performed in physically protected areas under conditions to ensure that no criticality can occur. The low enriched UF₆ is stored in a normal warehouse. The depleted UF₆ is deposited in the open air just surrounded by a fence. The containers are stored without the "overpack" used during transport as a fire protection. Neither a special protective coating nor other special security devices are used and needed during the storage.

The number of UF₆-containers handled in the last ten years is approximately 708.

Up to now, we have never observed any release of UF₆ from the containers including the valves. The corrosion of the outer side after several years of storage in the open air is meaningless, and no danger of leakage exists.

There have been several experiments to determine the corrosion of UF₆ to the inner wall. Independent of the type of steel the rate of corrosion was estimated to be at maximum 0.13 μm/year. So also from this side no dangerous effects are to be expected.
Particular measures are necessary during the evaporation of UF₆ from the containers for the purpose of sampling as well as for the first step of fabrication. During these operations three accidents occurred in the past. The consequence of these incidents is, that today the evaporation takes place in a special container which is a second barrier against UF₆ releases. Several safety devices are used to detect, control and reduce the possible UF₆ leakage. During sampling of liquid UF₆ difficulties arise if pipes and valves are not uniform in temperature, therefore a special isolated box with a fan heater has been developed to insure a safe sampling.

As already mentioned three UF₆ leakages occurred at NUKEM in former times. The first event happened in August 1966. During evaporation of a 12-inch cylinder with low enriched uranium, the heating of the connection pipe failed. The UF₆ condensed and blocked the line. The operators now tried to clean the line by external heating. During this operation the pipe bursted and UF₆ was released. The reaction products of UF₆ with the air moisture, UO₂F₂ and HF, could easily be identified as a white cloud. Parts of the cloud escaped from the working room through the open door. They remained visible to a distance of approximately 10 m.

The two workers involved immediately left the room. One of them put on a ventilated protective suit, entered the room, and closed the container valve half an hour after the beginning. The total amount released was about 50 kg UF₆. The workers were examined by a physician. Slight irritations of the bronchia were detected. Blood and urine samples were taken. There were no signs of any other injury.

The first urine samples showed an increased level of uranium (approx. 50 pCi/24 h) which was reduced to much lower values (< 2 pCi/24 h) after two days. This effect was seen in all further cases. We know now that the biological halftime of UO₂F₂ is in the order of half a day and the excretion in almost all cases is finished after 24 hours. The radiological impact therefore is very low. In the event described no limits were exceeded. After the accident the involved working room had to be decontaminated. In the surroundings the contamination was not measurable.

A second accident with big amounts of UF₆ involved occurred at a Saturday in September 1969 during a test being performed in the open air. The experiment was part of the qualification programme for a safety container developed for 30" cylinders. A 30 inch cylinder filled with 280 kg depleted UF₆ was placed in the safety container and heated by steam. The output for the condensed water was controlled by a pH-meter which had to close the steam input and output valves if acid was measured. After the 30 inch cylinder was heated its valve was opened and UF₆ flew into the safety container. The steam valves closed as planned and evaporation from the safety container to the
process vessel began as foreseen. After approximately 10 minutes the steam output valve opened, due to a defect of the ph-detector and UF₆ was released through this relatively big opening. One of the operators was already equipped with compressed air breathing and a protective suit. He tried to stop the release but, because of difficulties, especially with the sight conditions, he was not successful before 5 minutes. In this time about 200 kg UF₆ escaped. The visible cloud of UO₂F₂ and HF was approx. 25 m high and 200 m long, due to low wind velocities. Measurements of contamination showed higher values up to a distance of 50 m. A street and a building just opposite the test place had to be decontaminated afterwards. No measurable activity had been detected outside the plant site (fence distance approx. 300 m). No changes on vegetation were observed. No one of the test observers was injured by this accident. Immediate medical treatment included inhaling of neutralisation solution for HF. Urines were gathered and analysed for uranium, no extraordinary values were detected. A little event with a release of 20 g UF₆ had happened in March 1969. A flexible steel hose cracked during decanting natural UF₆ in the development laboratory. Only the amount of UF₆ in the tube was released. Already this small amount filled the laboratory with smoke. But analyses of the urine of the two people in the room did not show increased uranium values. The airborne activity was reduced by the fume hoods of the laboratory providing an air exchange rate of 5 per hour. Picture 1 shows the decreasing concentration. The decrease approximates an exponential function. The analyses were done by measurement of short time air samples in a flow counter. A low activity level was reached 2 h after the event. Before this time working without breathing protection was impossible. In picture 2 typical curves of results of urinary uranium concentration is given. The quick decrease after intake of UO₂F₂ is evident. A slight increase of excretion will normally happen in the morning of the following day. After one day the concentration will reach low and relative constant values. The biological halftime is below 24 h, according to our experience.

The experience at NUKEM is that safe handling and storage of UF₆ is attainable if besides technical measures just mentioned some further precautions are foreseen. A UF₆ release is easily detected because the white fog is already seen when the HF concentration is below 1,5 ppm. The maximum permissible HF concentration, according to german rules, is 3 ppm. After such release has been detected the area should entered only with protective suit, compressed air breathing, gasmasks as appropriate. Shower-baths should be at hand for the extensive douching of the persons involved to remove the HF contamination. Spare dressing should also be available to allow a complete exchange for the people involved. Provided the persons working in an area where UF₆ is stored or handled are well acquainted with the safety procedures, some of which are mentioned above, even an accidental UF₆ release will not cause dangerous injuries.
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Figure 1: Uranium concentration in air after an accidental UF₆-release
Figure 2: Uranium concentration in urine of 4 workers involved in an accidental UF₆-release
UF₆ release experience and prevention in United States gaseous diffusion plants by G.T. Hull
UF₆ RELEASE EXPERIENCE AND PREVENTION IN UNITED STATES GASEOUS DIFFUSION PLANTS

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ABSTRACT

The three United States gaseous diffusion plants have been in operation for a combined total of over 80 years, and in that period have produced or processed many thousands of tonnes of UF₆. Releases of UF₆ have occurred; however, release frequency has decreased sharply since the early period of plant operation. In assessing the impact and consequences of these releases, it is most significant that there has never been a serious injury caused by UF₆ exposure from a gaseous diffusion plant and no release has caused a serious disruption of plant operations.

INTRODUCTION

The three United States uranium enrichment gaseous diffusion plants are owned by the Government and have been operated by private contractors since completion of construction. Union Carbide Corporation has operated the plant at Oak Ridge, Tennessee, since 1944 and the plant at Paducah, Kentucky, since 1952. Goodyear Atomic Corporation has operated the plant at Portsmouth, Ohio, since 1954.

The low pressure operation of the diffusion barriers, compressors, and associated piping in the diffusion cascade requires more concern with inleakage of moist air than of release of UF₆ from this very reliable continuous operation. This cascade equipment is unique to the diffusion plant so it does not provide illustrations of prevention of UF₆ releases useful to other UF₆ handlers. For these reasons, this paper discusses only the type of releases of UF₆ that could occur in any type plant handling UF₆. These common operations are vaporization of UF₆ from cylinders as feed, withdrawal of UF₆ into cylinders as product or tails, sampling of UF₆ contained in cylinders, and transfer of UF₆ between cylinders. In addition to the diffusion plant cylinder operations, feed plants for UF₆ manufacture have been operated at all three plants for varying periods of time, with the major production occurring in the Paducah feed plant. Cylinder handling releases at these feed facilities have been included in the release data since feed manufacture is an integral part of nuclear fuel production.

The paper includes a description of actual release incident examples, and the conclusions drawn by the author.

RELEASE EXPERIENCE

An analysis of the release experience at the three plants immediately reveals a definite trend, with releases occurring more frequently in the first few years of operation. The number of releases then decreased sharply and has remained at low levels for a number of years. This trend is undoubtedly due to the improvement in design, operation, maintenance of the UF₆ cylinder handling facilities, and continued training of the work force after the startup of each plant. As experience and know-how were gained at each plant, the technology of UF₆ handling steadily improved. Continuous efforts have been expended to make improvements to prevent release of UF₆. This effort continues today, with the three plants sharing their knowledge to further decrease the potential for releases.

This paper discusses UF₆ conditions commonly encountered in nuclear fuel production - UF₆ up to 5% U-235 in Model 30 and 4.5% U-235 in Model 48 cylinders. Releases of higher U-235 isotopic assay material represent different problems in regard to the value of material and criticality and are not within the scope of this paper.
In order to place the number of releases into proper perspective, a conservative estimate was made of the number of cylinder hookup and disconnect operations during the years of operation at the three plants. The total number of cylinder hookup and disconnect operations is estimated to be over 650,000 and the number of valve operations to be over 1,950,000. The total number of significant releases was about 60, making the frequency of releases occurring per cylinder operation less than 1 in 10,000. If the experience of the early years of operation were removed and the numbers compared for only the last 10 years, the frequency would be even lower.

Causes of releases are many and varied, and each release has its own characteristics. However, they may be categorized into the three general areas of cylinders, valves, and piping manifolds including the tubing connections between cylinders and manifolds, commonly called pigtails. Experience at all three diffusion plants is quite similar with slight variations in release frequency and cause due to differences in operating conditions and equipment. Experience at the Paducah Plant has been selected for analysis.

Figure 1 is a graph detailing the number of UF\textsubscript{6} cylinder handling releases at Paducah since the beginning of plant operations. It illustrates clearly the improvement trend and the small number of releases in recent years.

Table I is a breakdown of the Paducah releases by cause. Valves and pigtails are vulnerable parts of the system, causing the majority of the releases. It is also of interest that liquid UF\textsubscript{6} was involved in practically all of the releases. No significant releases have occurred with cylinders containing completely solidified contents.

**DESCRIPTION OF RELEASES**

Following are brief descriptions of events and releases which have occurred at the gaseous diffusion plants. They have been selected as representative of the primary causes of releases, as shown in Table I.

1. **Valves**

   **Incident** - A release occurred in 1952 in a feed vaporizer during the sampling operation. A Model 30 cylinder was placed in a feed bath with the valve in the 6 o'clock position. The UF\textsubscript{6} in the cylinder had been liquefied and the cylinder was connected to the sampling manifold. When the operator attempted to open the valve to withdraw the sample, he noted UF\textsubscript{6} escaping through the valve. He quickly closed the valve, but gas continued to escape. He then attempted to close the valve more tightly, and the two-piece valve came completely apart. The UF\textsubscript{6} pressure blew the stem and seat assembly out of the valve body causing the liquid UF\textsubscript{6} release. The building in which the feed vaporizer was located was evacuated after several unsuccessful
attempts were made by emergency personnel equipped with respiratory protection to freeze down the cylinder with CO\textsubscript{2} extinguishers. The cylinder was removed from the bath and taken outside the building where the valve hole was plugged and the cylinder frozen down with dry ice.

Discussion - This release was caused by an equipment failure, fracture of the union nut holding the two-piece valve together. The valve design was later changed to the present one-piece valve, and all two-piece valves were removed from cylinders and replaced with the improved valve—thus eliminating the problem. This release demonstrates, however, that a liquid release inside a building presents a difficult type to control, primarily because of the visibility problems that are encountered.

Incident - A release occurred in 1953 in a vertical hot water bath vaporizer. A Model 30 cylinder was in the vertical position in the bath. While the operator was moving the overhead crane in the vicinity, the sling and hook struck the valve of the cylinder, cracking the nipple. The damaged valve immediately began to leak UF\textsubscript{6}; and when attempts were made to stop the leak, the valve was completely broken off. The cylinder was cooled down and the release stopped by driving a tapered wooden plug into the valve opening.

Discussion - This incident demonstrates the vulnerability of cylinder valves to damage from external and sometimes unexpected sources. Striking a cylinder valve with another cylinder or through improper crane operations is a very probable type of event. Cylinder valves have been struck and damaged by slings, bath lids, and mobile cylinder handling equipment. Many actions have been taken to alleviate this hazard. Cylinder skirts and valve shields are now in use which protect the valves. A removable cylinder valve protector is also used. Crane and mobile equipment operators are licensed and trained in the necessity for the careful operation of their equipment. Protection of cylinders, particularly while the UF\textsubscript{6} is in the liquid state, is also a significant factor in the storage pad design and layout. Figures 2 and 3 show typical valve damage.

2. Pigtails

Incident - A release occurred in 1955 in a UF\textsubscript{6} withdrawal room. Liquid UF\textsubscript{6} was draining through a copper pigtail into a Model 43 cylinder on a scale cart when the cart axle broke. The cylinder rolled out of its saddle as the cart frame dropped to the floor. This fall sheared the pigtail and the cylinder rolled so that the valve was now in a position below the liquid level of the cylinder. A remotely operated emergency block valve in the liquid drain line was promptly closed, thus stopping the release from the drain manifold. The release from the cylinder, however, presented a more difficult problem since visibility in the room was obscured. Emergency personnel in impermeable suits were employed to attach a hitch to the end of the cylinder and were successful in dragging it outside the room with the use of a
crane. Once outside, the problem was diagnosed and the cylinder valve was closed -- immediately stopping the release.

Discussion - This release was caused by an equipment failure. Examination of the cart axle at the break point revealed a weak spot from a weld repair performed at the factory where the carts were manufactured (Figure 4). As a result of this accident, the following actions were taken:

a. Larger axles were installed on all scale carts, and pipe supports extending down to about one centimeter from the floor were installed on each corner, thus preventing a cart from dropping more than this distance if an axle break should occur.

b. All UF₆ cylinder handling equipment must be designed with ample safety margins.

c. All equipment manufactured internally or purchased must be carefully inspected for any flaws and defective workmanship.

d. Equipment must be designed with safety devices included where possible to prevent component failure from causing an accident.

3. Cylinders

Incident - In 1960 a Model 48 cylinder located in a temporary vaporization facility in a large building ruptured due to internal hydraulic pressure. The release occurred just two weeks before the facility was scheduled to be taken out of service. Operation of a newly constructed autoclave-type vaporization facility was soon to be initiated. Standard practice at that time was to vent noncondensibles from full feed cylinders into a low pressure system before heating the cylinders with steam for feed purposes. This properly filled cylinder was erroneously valved into a high pressure feed line and left for three hours in this condition. During this time the cold cylinder acted as a condenser and was backfilled with approximately 907 additional kg of UF₆, overfilling the cylinder by 766 kg.

Unaware that the cylinder had been overfilled with liquid and solid, leaving insufficient vapor space for liquid expansion, the operator valved off the cylinder from the manifold and applied steam heat. After two hours the cylinder wall ruptured from the hydraulic pressure resulting from the overfilled condition. The vaporizer cover utilized a water leg seal and permitted UF₆ to escape into the building. The cover was removed and emergency personnel brought the release under control by use of cooling water and filling the vaporizer bath with dry ice. The vaporizer bath helped to contain the release, and
the cylinder was refrigerated in the bath for 48 hours prior to the removal. The cylinder had bulged within the stiffening rings and had a 0.95 cm by 10.2 cm crack near the midpoint of the cylinder. A patch was welded over the crack similar to Figure 5.

Discussion - The inadequate design and inherent hazards of the facility had been recognized, and a new facility was nearing completion. Existing instrumentation currently used in autoclave-type vaporizers prevents backfilling from occurring. An important rule, however, to avoid cylinder rupture is to never apply heat to a cylinder having a closed cylinder valve. The cylinder should be connected to a manifold with a relief or surge system and pressure-monitoring instrumentation.

Conclusions

1. Four general categories may be used to classify the potential for large UF₆ releases.
   a. Minimum Potential
      UF₆ in solidified state in cylinders in storage.
   b. Moderate Potential
      A release from a valve or pigtail positioned above the cylinder liquid level so that only gaseous UF₆ is escaping. This is the most common type of release and is also relatively easy to control.
   c. Increased Potential
      A release from a valve or pigtail positioned below the cylinder liquid level so that liquid UF₆ is escaping. This type release generally occurs in the sampling or transfer operation and is more difficult to control.
   d. Maximum Potential
      A release from a cylinder puncture or rupture below the liquid level.

2. Release frequency has sharply decreased as a result of experience gained from actual events and utilization of better equipment and improved designs.

3. With properly trained emergency personnel and modern equipment, most releases can be controlled in a reasonable time and almost all can be mitigated to some extent. The technology used today and the experience learned from initial operations, as well as future
improvements planned in release prevention, containment, and control, are described in a paper by A. J. Legeay entitled "Handling of UF₆ in U. S. Gaseous Diffusion Plants." [1]

4. The gaseous diffusion plant release experience indicates that safety, health, and environmental consequences can be prevented. Consequences to date have been economic due to the value of the material lost and the costs of decontamination and uranium recovery.

5. With properly designed facilities of the latest technology, the probability of a UF₆ release can be minimized; and with well-trained and equipped emergency personnel, actual occurrences can be brought under control promptly and safely.

6. Advance preparation, practice drills, and simulated rehearsals are effective means of achieving a well-trained work force which can recognize potential problem areas in handling of UF₆ and take effective actions if a UF₆ release occurs.

7. The most serious problems which merit additional study are the potential for foreign material (such as hydrocarbons) in cylinders, and the problems involved in moving cylinders of liquid from withdrawal or sampling points to safe storage locations, including the protection of the cylinders until the UF₆ is solidified.

REFERENCE

1. Legeay, A. J., Handling of UF₆ in U. S. Gaseous Diffusion Plants (U), Union Carbide Corporation, Nuclear Division, Oak Ridge Gaseous Diffusion Plant, Oak Ridge, Tennessee, June 1978 (K/P-6452).
<table>
<thead>
<tr>
<th>CAUSES OF RELEASES INVOLVING UF₆ CYLINDERS</th>
<th>NO. OF RELEASES</th>
</tr>
</thead>
<tbody>
<tr>
<td>1. VALVES</td>
<td>9</td>
</tr>
<tr>
<td>2. PIGTAILS AND MANIFOLDS</td>
<td>10</td>
</tr>
<tr>
<td>3. CYLINDERS</td>
<td>7</td>
</tr>
<tr>
<td><strong>TOTAL</strong></td>
<td><strong>26</strong></td>
</tr>
</tbody>
</table>
CYLINDER VALVE DAMAGE

FIGURE 2
CYLINDER VALVE DAMAGE

FIGURE 3
BROKEN AXLE ON SCALE CART

FIGURE 4
RUPTURED CYLINDER REPAIRED WITH PATCH

FIGURE 5
<table>
<thead>
<tr>
<th>No.</th>
<th>Causes of Releases Involving UF$_6$ Cylinders</th>
<th>No. of Releases</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.</td>
<td>Valves</td>
<td>9</td>
</tr>
<tr>
<td>2.</td>
<td>Pigtails and Manifolds</td>
<td>10</td>
</tr>
<tr>
<td>3.</td>
<td>Cylinders</td>
<td>7</td>
</tr>
<tr>
<td></td>
<td>Total</td>
<td>26</td>
</tr>
</tbody>
</table>
An experience of accidental release of UF₆
by
A. Ducouret
AN EXPERIENCE
OF ACCIDENTAL RELEASE OF UF₆

A.J. DUCOURET, docteur ès sciences
Production manager at Comurhex plant
Pierrelatte (France).
On 1st July 1977, as a result of a handling error, the content of a 48 Y cylinder, amounting to some 7.1 tonnes of liquid uranium hexafluoride, spilled onto the ground. Nobody was injured.

What I have to say is not concerned with determining the cause of, or the means of preventing, an accident of this sort. My purpose is to state the facts, consider the action taken and means employed to neutralize the products of the spillage, make an effort to understand the reactions of those present on the spot and, finally, set out the effects of this accidental release of uranium hexafluoride on the environment.

At the same time, I shall attempt to arrive at one or two guiding principles, the application of which might contribute to improve the effectiveness of action to be taken in the case of a massive escape of uranium hexafluoride.
1 - THE FACTS

The time: 14.30 on a warm and sunny afternoon, outdoor temperature 26°C, with a north-south wind blowing at a speed of approximately 9 meters per second.

A sample of uranium hexafluoride had just been drawn off in the liquid phase from a 48 Y container holding 8827 kg of hexafluoride. For doing so, use had been made of a mobile steam-heated oven located outside a building.

The sampling operation had been completed and the situation was as follows:

- heating of the oven had been discontinued,
- the temperature of the container was 90-95°C,
- the uranium hexafluoride was at a pressure of 3.7 bars,
- the cylinder valve was in the six o'clock position,
- the valve was closed but still connected up to the sample line.

At that moment, a handling operation involving the use of a fork elevator to remove the top cover of the oven caused the valve to rupture at the threads. The uranium hexafluoride in the container immediately started to spill out onto the ground. A very heavy fog formed and, within the space of a few seconds, it was no longer possible to take any remedial action. The duration of the flow of liquid can be estimated at 10 to 15 minutes. Once the level of the liquid hexafluoride had fallen to that of the valve opening the hexafluoride continued to escape as a gas.

A wooden peg was introduced 30 minutes after the start of the spillage, at which time 1727 kg of hexafluoride remained in the container.
The number of persons in the immediate vicinity of the accident was small. Operating personnel were taken unawares and hesitant as to the action needed.

14.30 : alarm raised.

The COGEMA safety organization, with which COMURHEX has an agreement for intervening at its adjoining plant, was alerted after six minutes had passed. Production staff were placed on alert in the plant installations and day personnel were evacuated to the assembly point.

The plant safety personnel donned their self-contained protective equipment but were unable to take any action since visibility in the area of the container was totally obstructed by a very substantial and heavy fog.

14.41 : help arrives.

The emergency squad, fitted out with self-contained protective equipment, brought two big fire water hoses into action in an attempt to lay the fog and cool the container. The contact between the water and liquid uranium hexafluoride had the opposite of the desired effect, the fog thickening and expanding.

14.49 : carbon dioxide used.

A carbon dioxide gun was brought into action. Almost twenty minutes had then elapsed since the start of the spillage and the flow of liquid had probably stopped.

14.56 : escape of uranium hexafluoride arrested.

Using the carbon dioxide had enabled the leak to be plugged with a solid mass of uranyl fluoride and carbon dioxide.

The size of the foggy cloud gradually reduced as the carbon dioxide spread over the ground and prevented contact and exchange with the air.
Lime was used to block the flow of water into the rainwater run-off drains.

A wooden peg was positioned in the hole a few minutes later by the safety personnel.

15.22 : No further emission.
Escape checked.
Neutralization with lime continues.

The quantities of materials employed in bringing the escape under control were:

- 13.5 m³ of water, which is approximately twenty times the quantity required for the hydrolysis of the uranium hexafluoride spilled,
- 600 kg of carbon dioxide,
- 8 tonnes of lime.

The area surrounding the 48 Y container, on which virtually all the solid uranium compounds settled, did not exceed 1000 m².

The next few days were spent in decontaminating the area on which solids had deposited. The topsoil was removed to a depth of ten centimeters and trees in the contaminated area were felled. All the materials removed (soil, wood, rubbish, etc.) were stored in a large number of drums.

2-2 Comments

After this description of the sequence of events, I should now like to make one or two comments on the action taken.

In the first instance, I feel the personnel present were taken unawares and this contributed to the delay in raising the alarm.

The density of the fog generated handicapped ability to act effectively and some time was taken to locate the precise source of the leak.

.../...
With the benefit of retrospection, and after due consideration, the use of the two big water hoses to cool the cylinder would seem to have been ill-advised. This in fact contributed to the hydrolysis of the uranium hexafluoride and formation of the hydrogen fluoride fog. There was clear evidence of an updraught caused by the heat released by hydrolysis. The buildings located north of the source of emission were invaded by the fog as a result of the updraught and of the eddies caused by the wind. Again on reflection, it seems that it would have been better to set up the fire hoses downwind and form a curtain of water to lay the hydrogen fluoride fog.

The use of carbon dioxide was particularly effective because of its much greater cooling effect and the fact that it blanketed the liquid uranium hexafluoride with a layer of gas, thus limiting the rate of hydrolysis.

The use of carbon dioxide thus assists solidification as crystalline uranium hexafluoride or uranyl fluoride.

The use of lime was quite certainly highly beneficial under the circumstances since it formed solid, water-insoluble compounds of fluorine and uranium. This explains why there was no pollution of the water-underground.

However, I should have reservations about spreading lime directly on liquid uranium hexafluoride since this would give rise to a violent and highly exothermic chemical reaction, thus favouring the evaporation and hydrolysis of the hexafluoride.

The question is, therefore: how can a mass of liquid uranium hexafluoride covering a large area be dealt with effectively?

I can offer no hard and fast reply, but only a personal opinion.
With this reservation, I should be tempted to advocate the use of substantial quantities of carbon dioxide in order to bring about crystallization of the hexafluoride and limit hydrolysis. The need is then to fix the hexafluoride as a solid, which can only be done by spreading a solid material of high specific surface capable of absorbing the uranium hexafluoride without undue evolution of heat. Alumina might be a possibility here. This still leaves the problem of neutralizing the resulting compound.

This experience shows that under no circumstances is it advisable to hose water directly onto liquid uranium hexafluoride. Water should however be used to lay the fog of hydrogen fluoride formed.

3 - REACTIONS OF PERSONNEL

My intention is not to attempt a detailed analysis of the reactions of those present, but more to point to those of significance within the context of an accident of this nature.

When the alarm was given, the installations were shut down without undue haste and in an entirely safe manner. All plant personnel gathered at the point of assembly north of the area affected and certain of them left the plant.

This accident, the possibility of which had in fact at one time been envisaged, but was no longer taken very seriously, came as very much of a surprise. Cylinders of uranium hexafluoride had been handled for fifteen years without incident. The factor of surprise was not conducive to the best organization of necessary remedial action. If a recommendation is invited here, it is that the existence of this kind of hazard must be kept permanently in mind, even if experience has shown the probability of its materializing to be very slight.

One thing which may be noted, however, is that those present, aware that no one was directly endangered, had the good sense not to attempt any action without the necessary protective equipment. This is why no injuries were recorded.
The fire water hoses initially employed had exactly the opposite of the desired effect and this came as an unpleasant surprise in view of the fact that the literature generally recommends spraying with water. A number of initiatives were subsequently taken:

- water was replaced by carbon dioxide,
- lime was used in the first instance to block access to the drains.

It was then found to be highly effective in arresting not only the water but also the fluorine and uranium materials, whereupon it was decided to cover the whole of the affected area.

I should also like to mention the reaction of two men working in a control room ten meters north-west of the source of the escape. When the alarm was given, they found themselves being enveloped by a thick fog. Their first impression was that this was an escape of steam but then they noted that there was no condensation on the windows. They then supposed that something had gone wrong in the main plant to the north. They shut down the workshop in question, donned their masks and decided to leave the control room and head south. Visibility being nil, they followed the fence southwards, which meant that they plunged into the thickest part of the fog. After a dash of forty meters, the fog started to lift and lighten, they saw a patch of light to their left and were able to make their way out of the fog.

Only one of the two men was contaminated since his mask was not perfectly tight fit on his rather thin features.
Two potential physiological hazards were to be feared, arising from exposure to:

i) inhalation of uranium,
ii) hydrogen fluoride.

To take the case of uranium first, the position can, very roughly, be summed up as follows: where a subject inhales a soluble compound of uranium

- 25% of the uranium inhaled is exhaled in the next breath,
- 50% is initially retained in the upper respiratory tract, then passes into the digestive tract, and is finally excreted in the stool,
- 25% is absorbed in the lung air cells and passes into the blood, to be subsequently and rapidly eliminated in the urine. It is estimated that 80% of this quantity, i.e. 20% of the quantity of uranium absorbed is excreted in the urine in the course of the first twenty-four hours following intake.

It is generally accepted that the maximum permissible intake of a soluble compound of uranium is 2.5 mg uranium/day, which corresponds to 500 μg uranium evacuated in the urine within twenty-four hours or, again, since the volume of urine passed is ca. 1.5 litres per twenty-four hours, an average concentration of uranium in urine of 300-350 μg uranium/litre.

Again, chronic or acute uranium poisoning can lead to albuminuria should the intake be such that the concentration of uranium in urine amounts to 5-7 mg uranium per litre of urine.

In the case considered, all the personnel of the COMURHEX and neighbouring plants were asked immediately after the incident to supply samples of urine. The occasion was also taken to supply each person concerned with bottles for collection of urine over the immediately subsequent 24-hour period.

275 people supplied immediate samples of urine and urine collected over the next 24 hours (Category A),
84 supplied only immediate samples of urine (Category B),
90 supplied only the urine collected over the following 24 hours (Category C),
449 people were examined and 724 samples analysed.

Résultats are tabulated below:

<table>
<thead>
<tr>
<th>Uranium content of urine supplied (µg)</th>
<th>no of subjects</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>category A</td>
</tr>
<tr>
<td>0 to 49</td>
<td>130</td>
</tr>
<tr>
<td>50 to 99</td>
<td>86</td>
</tr>
<tr>
<td>100 to 199</td>
<td>48</td>
</tr>
<tr>
<td>200 to 299</td>
<td>7</td>
</tr>
<tr>
<td>300 to 499</td>
<td>2</td>
</tr>
<tr>
<td>500 to 699</td>
<td>1</td>
</tr>
<tr>
<td>900 to 999</td>
<td>1</td>
</tr>
<tr>
<td>TOTAL</td>
<td>275</td>
</tr>
</tbody>
</table>

Only those results relating to subjects in category A are significant.

Observed contamination of urine was found to be higher than the permissible maximum in two cases. The toxic concentration was not however reached and the two employees concerned did not therefore show any subsequent physiological disorders.

Turning now to the hazard represented by hydrogen fluoride, no symptoms affecting the skin or respiration were observed.

In concluding this report of the medical examination undertaken, I would mention the difficulties encountered.

On 1st July, all the samples involved were taken at the French Atomic Energy’s specialist laboratory at Pierrelatte.
Quite apart from the need for immediate availability of several hundred sample bottles, it is not hard to appreciate the problems posed by having to deal at the same time and at the same place with 450 people, in addition to which a considerable number of those concerned were immigrant workers who had to be instructed in how to collect their urine over a 24-hour period.

It was also observed that a number of samples had been contaminated at the time of sampling.

5 - EFFECTS ON THE ENVIRONMENT

The aggregate results recorded by the fixed monitoring stations throughout the passage of the toxic cloud, lasting less than 20 minutes, provide a fairly accurate idea of the quantities of hydrogen fluoride and uranium released into the atmosphere. These were 330 kg of uranium and 1600 kg of hydrogen fluoride.

It also proved possible to map uranium and fluoride pollution by means of OWEN gauges and triethanolamine-impregnated filters sited at various parts of the plant, plus analyses of samples of plant life.

In the case of hydrogen fluoride, the "workforce" limit (2.4 mg/m³) was exceeded up to a distance of 1200 meters from the starting point during the emission.

In the case of uranium, contamination amounting to 10 mg/m² was observed up to 600 meters from the source of emission.

Beyond this distance, deposition of uranium was slight and can have no adverse consequences in terms of possible human consumption of insufficiently washed vegetable matter.

Again, samples of water taken south of the source of emission showed that permissible fluoride/uranium limits had not been exceeded in the case of water likely to be supplied to drinking water facilities.
This has been an account of the effects of an escape of uranium hexafluoride which may be considered as one of the biggest it is possible to contemplate.

Additionally, the circumstances of the escape were particularly unfavorable in that it occurred in the open air, thereby causing maximum pollution of the environment - even though the wind was a determining factor in diluting and dispersing the toxic cloud.

It has to be emphasized that no bodily injuries occurred, nor were there any appreciable affects on the environment at distances of more than 600 meters.

I would make two final remarks:

First, regarding the liquid-phase sampling of the content of 48 Y cylinders of uranium hexafluoride. Quite clearly, the first precaution is not to carry out this operation with the cylinder valve at the 6 O'clock position. There is also however the question of whether it is really essential to take samples of uranium hexafluoride in the liquid phase rather than limiting sampling to the gaseous phase in view of the hazards surrounding the liquid-phase sampling technique. What in fact is the advantage of liquid-phase sampling, given that the most stringent quality specifications relate to those compounds which exhibit highest volatility, i.e. which will tend to concentrate in the gas phase?

Secondly, there is the question of the physical location of the incident. As noted, this occurred in the open air and, as also noted, nobody was injured.

It is not difficult to imagine what the consequences would have been if the escape of uranium hexafluoride had occurred in a confined space with personnel present. It is no doubt true that the practice of not handling liquid uranium hexafluoride in the open air ensures a greater degree of protection of the environment, but is this not to some extent at the expense of the safety of the workforce? What can be done by someone present in a confined space in the event of a substantial escape of uranium hexafluoride? Must personnel be required to wear self-contained protective apparatus at all time?

These are the questions which now need to be answered.
SUMMARY
by the Session Chairman
G.T. Hull
SESSION III

Session III dealt with UF₆ release experience. It contained three papers as follows:

F. Schinzer - NUKEM - Experience on UF₆ handling in uranium manufacturing processes.

G.T. Hull - Union Carbide - UF₆ release experience and prevention in United States gaseous difussion plants.

A. Ducouret - Comurhux - An experience of accidental release of UF₆.

The three papers presented discussed in some detail seven actual UF₆ release incidents which have occurred in UF₆ handling processes.

Conclusions:

1. Experience of these releases demonstrated that very slight personnel or environmental consequences resulted from any of the releases.
2. Releases can happen unexpectedly and properly trained personnel can take the correct actions not only to protect themselves but to stop or control the release to the extent possible.
3. Well trained and equipped emergency forces should be maintained and kept ready to act immediately on notification of a release.
4. Many thousands of depleted UF₆ cylinders have been stored outside for many years with no problems and negligible internal or external corrosion.
5. After the releases described, local decontamination was necessary but there were little effects on off site vegetation or water supplies.
6. No personnel injuries or any consequence resulted from the release.
7. A liquid release inside a confined space presents a difficult problem to combat because of the visibility problem resulting from the dense UO₂F₂ fog formed.
8. Release frequency is very low considering the number of cylinder operations performed.

Areas for future work

1. Develop methods for detection of contaminants such as hydrocarbon oil in cylinder before filling.
2. Study methods of protection during moving and storage of liquid filled cylinder.

3. Study methods for dealing with released liquid UF6 covering a relatively large area.

4. Develop method of sampling cylinder without the necessity for having the valve in the most dangerous 6 o'clock position.

5. Determine if gaseous sampling would not be satisfactory.

6. Study the question that while handling liquid UF6 in the open presents a greater environmental risk, it presents a lower risk to the safety of the work force.
PANEL DISCUSSIONS
SUMMARY, CONCLUSIONS AND RECOMMENDATIONS
BY THE PANEL CHAIRMAN: G.B. HART
Initially the five session chairmen presented summaries and conclusions from their sessions. The Panel Chairman then proposed three questions for discussion which had been compounded from the session conclusions and from questions directed to the panel from the floor. The three questions were:

1. Are we satisfied that present technology and operating procedures are adequate to present UF₆ releases or is it desirable to work towards, for example, reduced liquid UF₆ handling and improved protection against inadvertent liquefaction as in a fire? In this connection, would it be feasible for feed and withdrawal streams to be in the vapour/solid phases? Can gaseous sampling be substituted for liquid sampling? If these are not considered feasible are there any ways of reducing the vulnerability of these activities (e.g. increased cooling time).

2. Do we know enough about the behaviour of UF₆ and its reaction products to be able to predict with confidence the consequences of any conceivable accident? If not in what area is more information needed?

   a) Atmospheric dispersion
   b) Biological effects.

   We need to note the varying results of theoretical treatments, leading to possibly different conclusions and the varying limits referred to for permissible levels of HF and UO₂F₂ in the atmosphere.

- 3. -
3. Is further work necessary on methods of

a) protecting the work force against hex escape? e.g. location of control rooms, emergency equipment and/or facilities which can be recommended for dealing with a release, evacuation routes and training,

b) minimising the effects of an accident

(i) by reducing the quantity and duration of release
(ii) by improved methods of clearing up the mess.

Following a brief introduction by a panel member, each of these topics was discussed generally. It was agreed that, although present standards were good, further improvements in all areas were desirable. Particular emphasis was placed on the hazards arising from liquid UF₆ handling: it was felt that the most hazardous operation in this area was liquid sampling. Suggestions were made for reducing the hazard and the necessity for liquid sampling was questioned. It was felt that procedures had not been optimised between commercial and safety considerations and it was concluded that CSNI could perform a valuable role in initiating such optimisation studies among operators, contractors and regulatory authorities. It was also agreed that further experimental work on hex releases, carried out on small scale models, would be valuable. Finally, the meeting concluded that CSNI could provide a valuable service in disseminating information about hex releases and it was recommended that a system should be introduced to this effect.

Because of lack of time, it was not possible to pose and answer specific questions on the papers presented at the previous sessions and the Chairman requested questioners to contact authors directly.
CLOSING REMARKS

by

C.J. van Daatselaar

(Meeting Chairman)
Ladies and Gentlemen,

With these final discussions in the panel this specialist meeting on the safe handling of UF₆ has come to an end. When the programme group started to work out the initiative of the CSNI to have an expert meeting on the safety aspects of handling, transport and storage of UF₆, and the first results of the call for papers came in, I personally was a bit disappointed. Certainly not because of the quality of the papers sent in before the closing date, but because of the limited number of authors that offered a contribution. Also the number of experts that applied for participation to the meeting did not show an explosive character.

Now looking at the final results of the meeting I must say that I am very satisfied. The number of participants exceeded by 50% the number we had in mind as a maximum. Also the number of papers, besides their excellent quality, were quantitatively of the right number to fill this two and a half days meeting, leaving reasonable time for questions and answers. Furthermore because we spent (or had to spend) all our time in this comfortable meeting place, there was additional time for shoptalking.

I do not have the intention to summarize at this moment the results of our meeting. I think in the panel session the most important aspects have been put in proper order.

I sincerely hope that everyone of us who participated in this meeting has the feeling that although he is already an expert, learned something in addition. Anyhow, among other things I learned that a much shorter expression exists for uraniumhexafluoride.

The meeting made it very clear that the most hazardous situation arises when the hex is in the liquid state. With the defence in depth principles and the training of personnel as discussed during the meeting, the probability of an accident with serious consequences is however very remote.

- The -
The theoretical assessments of the consequences of large releases of UF₆ showed that in fact there is a large safety margin. So I do think that this meeting gave evidence to the statement I made in my opening remarks that UF₆ can be handled without undue risk to employees or the public.

Ladies and gentlemen, I think I may speak also on your behalf when I express my gratitude to the Nuclear Energy Agency of the O.E.C.D. that agreed with the proposal of the Committee on the Safety of Nuclear Installations to have this meeting.

Now also on behalf of the O.E.C.D. I like to thank our host Urenco Netherlands Operations, presented here by the Director Mr. Stocksclader, who made it possible to have this meeting in our cloudy country, and to Mr. Kell and his assistants for all the technical arrangements that made our meeting and our stay here in hotel Boekelo a pleasant one.

Then of course I like to thank the members of the Programme Group, the speakers and the chairmen of the different sessions and last but not least all the participants, for making this meeting so successful.

With the wish that you have a good trip home, I declare the meeting closed.
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Mikkola, I.  
TVO Power Company  
(Finland)

Norsworthy, D.F.  
UKAEA  
(United Kingdom)

Oppenheim, C.  
Kernkraftwerk Kaiseraugst  
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De Winter, H.M.J.  
Comprimo B.V. (Netherlands)

Ziza, C.  
Comurhex (France)