AN EXPERIENCE ON DRY NUCLEAR FUEL REPROCESSING IN THE CZECH REPUBLIC

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Abstract

Experience of Czech research and industrial organisations in the development and realisation of pilot-plant technological line FREGAT-2 for dry reprocessing of fast reactor spent fuel is presented. The FREGAT-2 line is based on the fluoride volatility method. The possibility of using this experience of Czech organisations for developing technologies for selected areas of the transmutation process and the first results of technological research in this field in the Czech Republic are discussed.

Introduction

The development of dry reprocessing methods for spent fuel was motivated in the past by the assumed commercial utilisation of fast breeder reactors. Their application in the power industry can be economically efficient only in the case of a closed fuel cycle. However, reprocessing of fast reactor spent fuel brings about a number of specific difficulties in comparison to the reprocessing of spent fuel from thermal reactors. They are caused, for example, by higher burn-up, shorter cooling time resulting in a higher amount of energy released by the fuel, higher concentration and amount of plutonium, different cladding material, presence of metallic sodium, different fission products composition etc. Therefore, countries which were planning the introduction of fast reactors also were attempting to develop suitable methods for reprocessing, because the hydrometallurgical PUREX process was not suited to the fast reactor spent fuel.

One of the alternative processes, also being developed in the former Czechoslovakia in the 1970s and 1980s was the fluoride volatility process based on the separation of uranium and plutonium in the form of volatile hexafluorides from fission products, the majority of which form non-volatile fluorides. The process was developed in Czechoslovakia in co-operation with the former Soviet Union.

Fluoride reprocessing technology line FREGAT-2

The fluoride reprocessing consists of the following main operations:

1) Fuel elements decladding.
2) Transformation of the fuel to a powder.
3) Fluorination of the fuel.
4) Separation of uranium and plutonium fluorides.
5) Purification of the products obtained.

According to the agreement between the former Czechoslovak Atomic Energy Commission (now State Office for Nuclear Safety) and State Commission for Atomic Energy of the USSR it was agreed that the Soviet organisations will develop the technology and apparatus for items 1 and 2 and the Czechoslovak organisations for items 3 and 4. Item 5 will be worked out together, the purification of uranium on the Czechoslovak side and the purification of plutonium on the Soviet side.

The Nuclear Research Institute Rez was the contractor for the Czechoslovak side.

The Nuclear Research Institute Rez had a good qualification for the development of the fluoride reprocessing. It was at that time the only institution in the former Eastern bloc, except for the Soviet Union, where the fluorine gas production by anhydrous hydrogen fluoride electrolysis from potassium hydrogenfluoride melt had been managed.

Basic research in the field of volatile fluorides – for example of niobium, ruthenium, molybdenum, plutonium and wolfram, was the main subject studied in the Fluorine Chemistry Department of the Nuclear Research Institute Rez in the 1960s and early 1970s.

In the second half of the 1970s, the work on development and realisation of the pilot-plant technological line called FREGAT-2 was started. This line was intended for the fluoride technology verification by the way of experimental reprocessing of spent fuel from the Russian research fast reactor BOR 60.

The development and realisation of FREGAT 2 line was carried out in co-operation of the Nuclear Research Institute Rez with a number of Czechoslovak research and industrial organisations.

The most significant of them were:

- Chemoprojekt, Praha – design and engineering organisation which worked out the project of the entire technology including the measurement and control of the process.
- Institute of Nuclear Fuels, Zbraslav – design of special measuring and control instrumentation (manometers, pressure-vacuum gauges, flowmeters, closing and control valves). These instruments were designed for the fluorine, fluorides and hydrogen fluoride containing media.
- Research Institute of Chemical Equipment, Brno - design of all technological apparatuses and auxiliary equipment and manufacture of some of the apparatuses. Most of the apparatuses of the FREGAT-2 line which were in contact with fluorine, were made from nickel or Monel metal.
- Kovohute, Rokycany – production of nickel and Monel metal materials for the production of apparatuses.

Most of the technological apparatuses were manufactured in the engineering shop of the Nuclear Research Institute Rez. The work with nickel and its alloys as well as the necessary flaw detection methods were to be mastered in order to make products of required quality.

Spent fuel reprocessing in the FREGAT-2 line was based on the following principles.

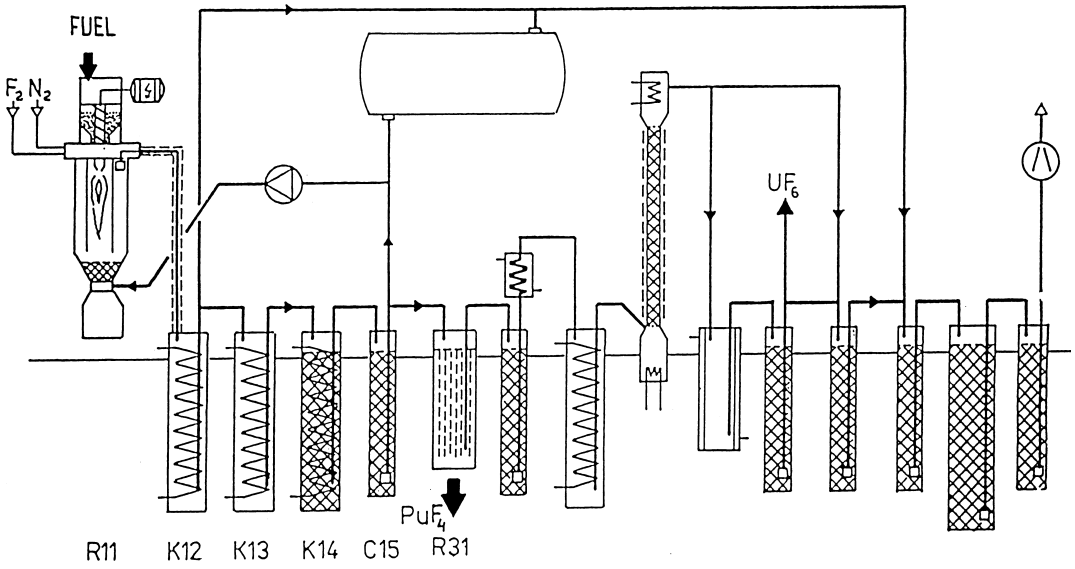
Powdered ceramic spent fuel is dosed in the nitrogen atmosphere by a worm doser into the preheated reactor R11 – fluorinator and at the same time fluorine gas also is fed. A reaction takes place in the flame, during which the uranium oxides react to form gaseous UF_6, mostly plutonium to gaseous PuF_6, a great majority of fission products form solid fluorides dropping to the fluorinator bottom and only a small part of fission products (Nb, Ru, Tc, Mo, I) reacts with fluorine under gaseous fluorides formation and will accompany uranium and plutonium together with rare gases.

Three condensers are situated after the fluorinator. The first one, K12, is intended for uptaking niobium and ruthenium. Uranium and plutonium fluorides should condensate in condensers K13 and K14 at -60 and -80 °C. It was assumed that a complete fluorination of Pu will not take place. Therefore, secondary fluorination of fuel in the reactor R11 was introduced by the way of fluorine circulation in a part of the apparatus by means of a circulating pump.

After the K13 and K14 condensers are filled with uranium and plutonium hexafluorides, the condensers are heated and the evaporated fluorides are introduced into the reactor R31, where thermal decomposition of PuF_6 to solid PuF_4 is taking place, whereas volatile UF_6 passes through the reactor. In this way, uranium is separated from plutonium.

UF_6 is then purified by column distillation to remove molybdenum, iodine and technetium fluorides.

Figure 1. **Technological diagram of FREGAT-2 line**



Czech industrial enterprises and research institutions produced in the 1980s the following main technological apparatuses and instrumentation:

- Worm and vibrating dosers for powdered spent fuel with a capacity of 1-3 kg fuel/hour.
- Flame fluorination reactors.
- Condensers for condensation and evaporation of fluorides.
- Sorption columns.
- Distillation columns for the distillation of a UF_6 - MoF_6 - IF_5 mixture.
- Closing and regulation valves for fluorine and fluoride media.
- Flowmeters for fluorine and fluorides for the flow rates of 0-600 l/hr and 0-4 000 l/hr.
- Pressure-vacuum gauge for the range of 10-200 kPa.
- Differential manometers for the range of ± 1 kPa.
- Auxiliary equipment for the hot cell.

This equipment, individual apparatuses and then the complete assemblies were subjected to functional and technological tests in the Nuclear Research Institute Rez. Depleted U_3O_8 was used as a fuel in these tests. A few tens of kg of U_3O_8 were used in these experiments. After completion of the tests the entire technology was transported to the RIAR Dimitrovgrad in the Soviet Union to make experiments in hot cells.

Because the ideas about an early application of fast reactors in the nuclear power industry were re-evaluated in the second half of the 1980s and also due to the Chernobyl accident, the experimental work on the FREGAT-2 line was discontinued in 1998 and all the apparatuses and equipment, which was the property of the Czechoslovak side were returned after decontamination to the Nuclear Research Institute Rez.

However, we succeeded in the meantime to test the function of most of the apparatuses by using experiments with depleted uranium and of most of the measuring and regulating circuits, too [1,2].

Although the experimental programme on the FREGAT-2 line was discontinued in 1998, the functionality of the majority of apparatuses and equipment and of the remote control was demonstrated and the soundness of most of the technological operations was confirmed. Czech research and industrial organisations acquired during the execution of the project FREGAT-2 good experience in the field of a complex development of a special radiochemical technology.

Fluoride technology for the transmutation programme

At present, Czech research and industrial organisations intend to use the above experience in the planned national programme of transmutation technology development. Study of various aspects of the ADTT and a research and development programme in this field is being carried out in the Czech Republic for 3 years already, most of all within the scope of the national programme of deep geological repository development. This programme is financially subsidised by the State and development of transmutation technologies within this programme represents to some extent the support of an alternative technology.

Some of the Czech research and industrial organisations take part in the preparation of the fully independent national programme of transmutation technologies development. First of all, it is the Nuclear Research Institute Rez plc, SKODA – Nuclear Machinery, Czech Power Company, Institute of Nuclear Physics of the Czech Academy of Sciences and other organisations that were participating in the development of the FREGAT-2 fluoride reprocessing technology.

Establishment of an experimental laboratory for research and development in the field of transmutation technologies is assumed within the Nuclear Research Institute Rez plc. Experimental studies of a limited extent on the determination of physico-chemical properties of melts of selected fluoride salts mixtures were carried out at this place. At present, assembly of the first experimental loop filled with a fluoride salt melt is carried out together with the SKODA - Nuclear Machinery company. Three of such loops are planned to be realised. They will be used, first of all, for the material and design studies of the transmutation technology. The laboratory experiments in the field of suitable fluoride melt preparation and its behaviour has been started and technological experiments of the first main separation steps of the spent fuel before transmutation are planned and prepared. Selected parts of FREGAT-2 equipment will be used for this investigation.

Conclusion

The Czech Republic as a country operating and constructing nuclear power plants and treating problems of spent nuclear fuel is interested in the international co-operation in the field of transmutation technologies development. A co–operation already exists at the level of individual institutions and is directed to the information exchange between the Czech organisations mentioned and research institutes in the U. S. A., France, Sweden, Italy, Russia, Korea and others.

Development and realisation of transmutation technology can be considered practicable at present within the scope of a wide international co-operation. It cannot be figured that the Czech Republic, as a relatively small country, could participate financially to a deciding extent on this research. However, within the international co-operation at the level of government and research institutions the tasks could be allocated in a way that each of the participants will take part on the

investigation of some selected problems. Czech research and industrial organisations are prepared to participate in such a co-operation. Experience from the fluoride reprocessing technology development is a good basis for this co-operation.

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