OECD Nuclear Energy Agency
Nuclear Science Committee

The First Information Exchange Meeting on Nuclear Production of Hydrogen

Paris, France
2-3 October, 2000

at OECD Headquarters,
2 rue André-Pascal, Paris

Abstracts

Meeting Organization

Organizing Committee

D. C. Wade	ANL, USA (Chair)
E. Arthur	LANL, USA
G. Marucci	ENEA, Italy
M. Ogawa	JAERI, Japan
M. Salvatores	CEA, France

International Scientific Members

A. Guigon	CEA, France
M. Hori	Nuclear Systems Association, Japan
J. M. Kendall	IAEA
M. Lecomte	Framatome, France
G. Marcus	DOE, USA
C. Nordborg	OECD/NEA
S. Shiozawa	JAERI, Japan

Meeting Secretariat

K.Suyama	OECD/NEA

Meeting Programme and Schedule

Monday 2 October

08:30 - 09:00 Registration

Session 1: 21st Century Energy Perspectives(Chaired by D.Scott)

09:00 - 09:15 **Welcome**
by *Ph. Savelli*

09:15 - 09:45 **Building Sustainable Energy Systems: The Role of Nuclear-Derived Hydrogen**
by *H. Rogner and D. Scott*

09:45 - 10:15 **Role of Nuclear Energy in the Long-Term Global Energy Perspective**
by *M. Hori*

10:15 - 10:30 Coffee break

10:30 - 11:00 **On Decarborisation: Historically and Perspectively**
by *C. Marchetti*

11:00 - 11:30 **Potential of Multi-Purpose Liquid Metallic Fueled Fast Reactor (MPFR) as a Hydrogen Production System**
by *H. Endo, H. Ninokata, A. Netchaev and T. Sawada*

11:30 - 12:00 **Will the Nuclear Production of Hydrogen be Socially Acceptable ?**
by *A. Waltar*

12:00 - 14:00 Lunch break

Session 2: R&D Programmes Ongoing and in Development(Chaired by S. Johnson)

14:00 - 14:30 **Present status of JAERI's R&D on Hydrogen Production Systems in HTGR**
by *M. Ogawa*

14:30 - 15:00 **Overview of CEA Studies on Hydrogen Production and Related Prospects for Nuclear Power**
by *J. Agator, A. Guigon and P. Serrecombe*

15:00 - 15:30 **Hydrogen Programme in Italy**
by *R. Vellone*

15:30 - 16:00 Coffee break

16:00 - 16:30 **IAEA Activities on the Nuclear Production of Hydrogen**
by *J. Kendall*

16:30 - 17:00 **Research Activities on High Temperature Gas-Cooled Reactors (HTRS) in the 5th EURATOM RTD Framework Programme**
by *J. Martin-Bermejo and M. Hugon*

Tuesday 3 October

Session 3: Physics and Chemistry of Hydrogen Production Methods(Chaired by A.Guigon)

09:00 - 09:30 **Open Questions and Research Topics on Nuclear Hydrogen**
by W. Stoll

09:30 - 10:00 **Hydrogen Production by Thermal Decomposition of Methane: An Assessment and an Experimental Program**
by A. Aiello, G. Benamati, G. Gherardi and G. Marucci

10:00 - 10:30 Coffee break

10:30 - 11:00 **A Study on the Thermochemical I-S Process**
by *K. Onuki*

11:00 - 11:30 **The Pyrolysis of Methane over Liquid Metal to Form Hydrogen and Carbon**
by C. Marshall, M. Lewis, L. Leibowitz , D.Lewis

11:30 - 12:00 **Hydrogen Permeation Barrier Development**
by A. Aiello, C. Fazio and G. Benamati

12:00 - 14:00 Lunch break

Session 4: Applications of Nuclear Technology for Hydrogen Production(Chaired by M.Lecomte)

14:00 - 14:30 **Hydrogen for Transportation: Available Canadian Technology**
by A. Miller

14:30 - 15:00 **A Proposed Modular-Sized, Integrated Nuclear and Hydrogen-Based Energy Supply/Carrier System**
by D. Wade, R. Doctor, B. Spencer, K. Peddicord, C. Boardman and G. Marucci

15:00 - 15:30 **Contributions to a Safety Analysis for a Hydrogen Production System with HTGR**
by K. Verfondern, R. Moormann and W. Breitung

15:30 - 16:00 Coffee break

16:00 - 16:30 **Hydrogen Production Associated to the Treatment of Nuclear Waste**
by A. Rahier, A. Fonteyne, M. Klein, M. Ponnet, J-P. Pirard and A. Germain

16:30 - 17:00 **Thermochemical Hydrogen Production with a High Temperature Gas Cooled Reactor**
by G. Besenbruch, L. Brown, J. Funk and S. Showalter

Session 5 : Discussions and Preparation of Recommendations for the Path Forward(Chaired by D.Wade)

Monday 2 October

Session 1: 21st Century Energy Perspectives

Chairman D. Scott

Building Sustainable Energy Systems: The Role of Nuclear-Derived Hydrogen

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D. Scott
University of Victoria, Canada

The prospect of planet-wide climatic instabilities, caused by anthropogenic changes to atmospheric gases, will be the most critical *environmental* threat to global prosperity and economic security during the 21st Century. In our view, any rational appraisal of the scientific data and its analysis leads to this conclusion—although there remains a large, well-funded and diverse group of special interests that have mounted a major effort to discredit concern about climate destabilization. It must be expected that, as evidence continues to accumulate in support of anthropogenic climate destabilization, the efforts of these critics will increase proportionally. In this paper we review features of the 21st century energy system—how that system evolved and where it seems to be taking us, *unless* there are clear and aggressive multi-national initiatives to mitigate and then reverse the contribution by today's energy system emissions to this unprecedented threat to civilization's continued, healthy development.

The extensive deployment of the two non-carbon based energy currencies electricity and hydrogen—which we will call hydricity—will lie at the heart of any realistic reduction of anthropogenic carbon dioxide (CO_2) emissions. Of these two, hydrogen will be the newcomer to energy systems. Popular thinking has identified renewables as the category of energy sources that can provide electricity and hydrogen in sufficient quantities—although many lay-persons do not realize the need for a chemical currency to allow renewables to power the most important market, transportation. However, by applying the most elementary numeracy, renewables (while able to make important contributions) cannot realistically provide the magnitude of energy that will be required. The massive deployment of nuclear fission is the only realistic option. For the benefit of civilization we must hope that popular thinking comes to recognize that nuclear may be the "greenest" of energy sources—in time for it to be deployed to prevent climate catastrophe.

This paper highlights many of the issues surrounding these questions and issues.

Role of Nuclear Energy in the Long Term Global Energy Perspective

M. Hori
Nuclear Systems Association,Japan

The paper will discuss the long term global energy perspectives and role of nuclear energy as the major energy source including supply of non-electric energy.

Looking over the coming century, such global problems as population growth in developing countries, increase of energy consumption, and the resulting increases of carbon dioxide content in the atmosphere as well as other environmental problems make it important to secure energy sources with low environmental impact, high economic competitiveness, and long sustainable supply to satisfy worldwide energy demand.

Nuclear fission is the only demonstrated energy source to meet the above requirements. It remains the most certain option of the energy sources available of maintaining the future global environment and economy. The continuous and sustainable supply of nuclear energy can be attained by introducing the fast breeder reactors and its fuel recycle in due timing.

Electric power is the major application of nuclear energy, though electricity comprises about 30% of the primary energy consumption at present. To make nuclear energy useful as the major energy source in the future, it is necessary to extend the nuclear energy application to non-electric purposes.

The advanced nations should restructure their nuclear R&D program based on the vision that nuclear energy would become the major energy source in the middle of the 2lst century. There are many tasks -- technological, institutional, societal and international -- for making nuclear energy as the major energy source. Synergistic approaches with cooperative interaction among such possible measures as technological development, institutional measures, multinational projects and commercial venture would be important.

On Decarborisation: Historically and Perspectively

C.Marchetti
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CO_2 emissions are considered a threat to earth's climate stability. Various recipes I did propose during the last 30 years to cure them are reported.

The long term solution for a compact and sustainable primary energy source in the multiterawatt range is identified in terms of thermochemical hydrogen from nuclear heat.

The basic drive is that nuclear reactors and chemical plants have important economies of scale that make one Tw energy islands difficult to compete with.

I will also show that the stop in nuclear construction during the last decade is a natural effect of Kondratiev cycles and that nuclear energy should be expected to resume speed in the next few years.

Potential of Multi-Purpose Luquid Metallic Fueled Fast Reactor (MPFR) as a Hydrogen Production System

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H. Ninokata, A. Netchaev and T. Sawada
Tokyo Institute of Technology, Japan

1. Requirement on the Nuclear Energy

Nuclear energy is an only effective candidate to be replaced to the fossil fuel in the next century. In order to establish the alternative energy source by utilizing the nuclear energy, the future nuclear power plant should meet the following requirements:

i. Multiple energy conversion capability at higher temperature condition not only to an electricity generation with high efficiency but also to the hydrogen production for the fuel of the means of the transportation,
ii. Extended siting capability near by the energy consumption area assured by the elimination of the on-site refueling (no-refueling concept),
iii. Fuel breeding capability and incineration capability of the long lived fission products(LLFP), and
iv. Passive safety capability to eliminate the energetics due to re-criticality events at the time of severe accidents.

2. Concept of the MPFR

An aim of this paper is to describe the characteristics of the basic concept of the Multi-Purpose liquid metallic fueled Fast Reactor system (MPFR). The MPFR introducing the U-Pu-X(X: Mn, Fe, Co) liquid metallic alloy satisfies all of conditions above based on the following characteristics of the liquid metallic fuel:

i. Expanding the coolant temperature range during the reactor operation between 550ºC to 1100ºC promising the hydrogen production based on the thermal H_2O dissociation and the electricity production with high efficiency,
ii. Elongation of the core life time due to the FP separation characteristics in the fuel alloy (neutronic effect) and by the elimination of radiation damage of fuel material such as swelling(physical effect), and
iii. Elimination of the re-criticality phenomena at the core disruptive accident due to the liquid fuel discharge from the active core without compaction motion.

The MPFR will be developed step by step. First step of the development is a sodium cooled MPFR with the core outlet temperature of 650 ºC to confirm the principle of the MPFR applied to the hydrogen production by using the thermo-chemical reaction such as Mg-I cycle/1/. The second step of the development is the lead cooled system with the temperature range over 1000 ºC, which is the target plant of the MPFR applicable to the hydrogen and electricity production using AMTEC and to the hydrogen production based on the UT cycle with high efficiency/2/.

The MPFR is categorized in the small reactor with the thermal power of 150MW for the sodium cooled system and 300MW for the lead cooled system. The MPFR consists of separated "energy unit" and "core unit" and piping system with isolation valves between two sections. The concept of the separated energy and core units, even in the case of the small reactor, aims at realizing the transportable core unit system including the fresh fuel assemblies and core internals from the factory to the site in the completely fabricated form. It should be emphasized that the used core unit can also be transported after the completion of the core lifetime with the concrete reactor vault used as the radiation shield during the transportation. The total mass of the core unit and reactor vault is smaller than about 200 ton for the sodium cooled MPFR.

The energy unit can be used as IHX, SG for the electricity generation and as the thermo-chemical reactor for the hydrogen production, which is optionally applicable to the required energy conversion dependent on the siting condition. Figure 1 shows the example of the hydrogen production reactor by using the Mg-I cycle.

3. Core Characteristics of the MPFR

In the case of the sodium cooled MPFR, the liquid metallic fueled core with U-Pu-X alloy can be operated at the inlet/outlet coolant temperature condition of 500/650 ºC. The candidate component metal X will be the transition metals such as Mn, Co, and Fe. In the case of the U-Pu-Mn alloy, the melting point is ranging from 750 to 800 ºC dependent on the Pu enrichment between 10-20-%. The fuel pins are bundled in the large-scale hexagonal fuel assembly with the effective diameter of about 30cm.

For the lead cooled MPFR, the liquid metallic fueled core with U-Pu alloy is operated at the inlet/outlet coolant temperature condition of 950/1150 ºC. In the case of the U-Pu alloy, the melting point range is between 800 ^{o}C and 900 ^{o}C depending on the Pu concentration. The core characteristics are summarized in Table 1.

The core lifetime is over 10 years for the MPFR, and is accomplished by the reduced power density of about 180w/cc (linear heat rate of 100W/cm). The core lifetime will be more elongated by employing the selective venting fuel pin to escape the rare fission gas and by expecting the effect of the density separation between fuel alloy and FP elements produced in the fuel matrix

4. Safety Feature of the MPFR

The safety feature of the MPFR is characterized by the passive shutdown capability against the scrum failure accident such as ATWS. The possibility of the core disruption events can be strongly limited due to the negative feedback induced by the axial thermal expansion of the liquid fuel in the pin structure.

Even in the fuel pin failure phase of the ATWS, the core wide fuel relocation and re-criticality events can be eliminated by the early fuel discharge from the degraded fuel pins to the out-of core region, and the permanent neutronic shutdown will be established during the stage of a local core damage.

Reference

/1/ H.Endo, H.Ninokata, A.Netchaev, and T. Sawada, "A CONCEPT OF THE MULTI-PURPOSE LIQUID METALLIC FUELED FAST REACTOR SYSTEM (MPFR)," Int. Symposium on the Global Environment and Nuclear Energy System (GENES-III),Tokyo, Japan, Dec.(1999).

/2/ A. Netchaev, T. Sawada, H. Ninokata and H. Endo, "LONG LIFE MULTIPURPOSE SMALL SIZE FAST REACTOR WITH LIQUID METALLIC-FUELED CORE," Int. Symposium on the Global Environment and Nuclear Energy System (GENES-III),Tokyo, Japan, Dec.(1999).

Table 1. Reactor Core Design Specifications

Item	*Specification*
Thermal power	300 MW
Average core power density	186 W/cc
Fuel material	Pu-U alloy
Coolant material	Lead
Structural material	Coated Tantalum
Control mechanism	7 B_4C control rods
Active core height	700mm
FP separation region height	700mm
Core diameter ID/OD	1350mm/1390mm
Radial reflector thickness	400mm
Coolant channel OD/ID	10.0mm/7.6mm
Pin pitch-to-diameter ratio	1.4
Number of coolant channels	7816
Coolant temperature Out/In	1150^{o}C/950^{o}C
Coolant mass flow rate	10183 kg/sec
Core material volume fractions	F60%/C25%/S15%
Coolant velocity in the channel	2.6 m/sec
Core operational lifetime	over 10 years

Will the Nuclear Production of Hydrogen be Socially Acceptable?

A. Waltar
Texas A&M University, USA

Nuclear power appears well poised as a source of primary energy to produce the prodigious amounts of hydrogen that will very likely be needed within the new century to service our transportation sector. But if nuclear power is to grow to the proportions needed for such a task, it is important to remove the primary barrier that has impeded the full implementation of commercial nuclear power in the last century; namely, wide-scale public acceptance. In this paper we focus on the four primary impediments (safety, waste disposal, proliferation, and radiation health effects) and suggest ways that the linkage of nuclear energy to the production of hydrogen may either exasperate or mitigate these obstacles to public acceptance. We conclude that whereas such barriers will likely erode in time, the primary gains to achieving public acceptance may arise from clearly articulating the incredible ***benefits*** associated with nuclear technology as a whole. By employing modern communication techniques such as Decision Analyses in articulating these benefits, and doing so early on, we believe nuclear-generated hydrogen could become a popularly supported technology to ensure the mobility that modern civilization has come to enjoy and demand.

Monday 2 October

Session 2: R&D Programmes Ongoing and in Development

Chairman *S. Johnson*

Present Status of JAERI'S R&D on Hydrogen Production System in HTGR

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The Japan Atomic Energy Research Institute (JAERI) has conducted research and development (R&D) of nuclear heat utilization systems of a High Temperature Gas cooled Reactor (HTGR), which are capable to meet a large amount of energy demand without significant CO_2 emission to relax the global warming issue. The High Temperature Engineering Test Reactor (HTTR) with a thermal output of 30 MW and a reactor outlet coolant temperature of 950 C, which is the first HTGR in Japan, has been just constructed by the JAERI, and its first criticality was achieved in November 1998. The power rise test is being performed in the HTTR and the 30MW full power will be attained on January 2001.

A demonstration program on hydrogen production was started on January in 1997 as a study consigned by Science and Technology Agency. A hydrogen production system connected to the HTTR is being designed to be able to produce hydrogen of about 4000 Nm^3/h by steam reforming of natural gas, using a nuclear heat of 10 MW supplied by the HTTR. The HTTR hydrogen production system is first connected to a nuclear reactor in the world, hence an out-of-pile test was planned to be carried out prior to the demonstration test of the HTTR hydrogen production system. In order to confirm controllability, safety and performance of key components in the HTTR hydrogen production system, the facility for the out-of-pile test is under construction on the scale of approximately 1/30 of the HTTR hydrogen production system. It is equipped with an electric heater as a heat source instead of the nuclear heat supplied by the HTTR. The out-of-pile test will be performed for four years from 2001. Also, a hydrogen permeation test and a corrosion test of a catalyst tube of the steam reformer are being carried out to obtain basic data for the design of the system. Check and review of the demonstration program in the HTTR will be made in around 2000 from the view point of economy and technology, then the HTTR hydrogen production system will be constructed, and tested from around 2008.

In parallel to this steam reforming hydrogen production test, hydrogen production by thermochemical water splitting, so-called IS process has been studied at JAERI as one of the most promising heat utilization systems in the future. Continuous and stoichiometric production of hydrogen and oxygen for 48 hours was successfully achieved with a laboratory-scale apparatus mainly made of glass. Following this achievement, a larger scale apparatus with hydrogen production rate of 0.05 m^3/h is being manufactured from 1999 to obtain data of chemical characteristics in a close-cycle test. The test in each component, steady state and transitional state will be started from October 2000 to 2004. The R&D on the materials and increase of thermal efficiency is inevitably necessary for the success of the IS process. The coupling to the HTTR for the demonstration test is to be planned after the steam reforming system.

The R&D on hydrogen production is consigned to JAERI by Science and Technology Agency of Japanese government.

Overview of CEA Studies on Hydrogen Production and Related Prospects for Nuclear Power

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A. Guigon
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1. Place of hydrogen as energy carrier among 21rst century energy sources

Hydrogen will probably become, with electricity, one of the two major energy carriers of the 21rst century, for its potential to play a key role in the evolution of the society towards a sustainable energy system minimising the use of fossil fuel to restrict the emission of greenhouse gases.

Hydrogen may provide energy storage solutions for off-peak electricity generation, in particular non-fossil electricity such as nuclear or hydro electricity, and for intermittent renewable energy sources such as wind energy or solar energy. It may also provide fuel cells with clean feedstock for different applications such as the traction of vehicles and the decentralized generation of electricity and/or heat.

This paper will first provide some indications about the possible place of hydrogen in the energy system over the next decades and the potential impact on the share of fossil, nuclear and renewable energy sources in the future energy mix ; the analysis will consider the future prospects for the development of the fuel cells markets, for transportation and stationary applications, for the development of hydrogen-based technologies, and for the technical and economical progress for hydrogen storage and production technologies, including gasoline partial oxidation and water electrolysis.

Key issues will also be analysed, such as the conditions of possible transition scenarios from a fossil to a non-fossil-based economy, taking into account the existing and growing non-energy hydrogen market.

In this perspective, the situation of France, with a share of 75% of nuclear electricity, is of particular interest.

2. Nuclear power and its potential for hydrogen generation :

Hydrogen today is a byproduct of the carbon industry (oil or chemical industry) and therefore, its production is also associated to the generation of CO_2 ; the use of this process for massive hydrogen generation for transport or stationary applications would require to trap the CO_2 to limit the environmental impact. It is therefore foreseeable that the first generation of hydrogen extraction techniques using gas, oil and coal, will trigger at the same time researches on CO_2 limitation means.

The demand for hydrogen is likely to increase with the development of fuel cells, in particular low temperature fuel cells, at the beginning of the second half of the 21st century. At this time, the reduction in the hydrocarbon and natural gas reserves - which are at the same time used as a basic material for the production of hydrogen and for the energy of hydrogen extraction - will lead to rather extract hydrogen from water or biomass with non fossil energies.

Candidate energies for this second generation processes may be nuclear and renewable energies, with more flexibility for nuclear facilities to meet massive hydrogen needs and to adjust the thermal output (power, temperature) to specific requirements of thermal processes for hydrogen production.
Nuclear energy, with 4^{h} generation reactors capable of saving natural fuel resources for the long term, thus appears as a promising and sustainable option for the production of both electricity and hydrogen over centuries, when natural hydrocarbon and natural gas reserves will become scarce.

Given the extensive use of nuclear energy in France, the CEA, with other research laboratories have developed a special interest in exploring, among other solutions, the potential of nuclear power to meet increasing needs of hydrogen with limited environmental impact.

3. Research on hydrogen generation and storage:

After pointing out the context in which the CEA undertook studies on hydrogen 30 years ago, the paper will review the current research on hydrogen generation, storage and risk mitigation, and will tentatively address questions on nuclear technologies and safety principles relevant to hydrogen co-generation plants.

Research at the CEA is being currently conducted in 4 directions:

- modelling and performance assessment of promising processes which are at the laboratory stage : thermochemical cycles, high temperature electrolysis derived from the technology of high temperature fuel cells (SOFC), hydrocarbon plasma splitting, and plasma electrolysis,
- technical and economic comparison of these processes with the steam reforming of natural gas, which is used at an industrial scale,
- control of the technology of processes through the modeling of the process engineering,
- proposal of a safety approach applicable to the production on the nuclear site,
- predimensioning of production units based on various processes and comparative evaluation of their performances and cost.

Research and development work on hydrogen storage technologies, including high pressure tank and carbone nanotubes structures, is being carried out besides.

4. Review of key questions for nuclear hydrogen generation :

The processes exclusively based on electricity, make it possible to uncouple the production of hydrogen from the nuclear site and candidate power plants are not subject to additional constraints, besides the objectives assigned to the next generation reactors to progress in economic competitiveness, safety, minimisation of long-lived radioactive waste, proliferation resistance and compliance with a wide diversity of fuels to save the natural fissile and fertile fuels.

The processes making use of heat from the reactor lead to a closer interaction between the reactor and the production plant, and the following questions have been identified to investigate the feasibility, the performances and the safety requirements of nuclear hydrogen generation plants :

- which hydrogen production processes are best suited to nuclear power ?
- may some of these processes be implemented on operating nuclear plants ?
- how do various reactor technologies (LWRs, HTRs…) rank for dedicated hydrogen production or co-generation with electricity and/or process heat ?
- how can the potentialities of high temperature reactors best be used ?
- what are the additional and specific requirements put by on-site hydrogen production on standard nuclear safety criteria ?

These questions call for research work in the following areas which may be the subject of valuable international exchanges :

- development of physical and economic modelling tools for a consistent evaluation of performances and economics of nuclear hydrogen production plants based on different processes,
- development of international standards for safety, design, construction, operation, and maintenance of nuclear hydrogen production plants,
- development of key technologies such as specific materials (standards of testing method, dimensioning, corrosion and long-term behaviour),
- evaluation of the needs for a specific safety approach and associated instrumentation *(detection of hydrogen leakage…)* and safety systems *(mitigation systems of hydrogen risk…),*
- investigation of adequate technologies for large storage facilities and massive hydrogen transport.

Hydrogen Programme in Italy

R. Vellone
ENEA, Italy

Hydrogen can become an important option in the new millennium. It provides the potential for a sustainable energy system as it can be used to meet most energy needs, without harming our environment**.** Hydrogen has in fact the potential for contributing to the reduction of climate changing emissions and other air pollutants as it exhibits clean combustion with no carbon or sulfur oxide emissions and very low nitrogen oxide emissions. Furthermore it is capable of conversion directly to electricity in systems such as the fuel cells with no pollution.

Today a widespread use of this fuel is not still feasible because of economic and technological barriers.

In Italy a national program to facilite the introduction of the hydrogen as energy carrier is on going. This program has the aim to promote, in an organic frame, a series of actions regarding the whole hydrogen cycle. It foresees the development of technologies in the areas of production, storage, transport and utilization. Researches are also addressed to the development of technologies for separation and sequestration of CO_2.

The program is shared both by public organizations (as the research institutions and the universities) and national industry (oil companies, electric and gas utilities, research institutions).

Hydrogen can be used as a fuel, with significant advantages, both for electric energy generation/cogeneration (thermodynamic cycles, fuel cells) and for transportation (internal combustion engine, fuel cells). A focus of research will be on the development of fuel cell technologies. The fuel cells presents all characteristics to result a key technology in a future economy based on hydrogen.

During the initial phase of the project, hydrogen will be derived from fossil sources (natural gas), in the second phase it will be generated from renewable electricity or nuclear energy.

The presentation will provide an review of the hydrogen program and highlight future goals.

IAEA Activities on the Nuclear Production of Hydrogen

J. Kendall
International Atomic Energy Agency, Austria

High temperature process heat has long been viewed as a potential application of High Temperature Gas Cooled Reactors, with hydrogen production being one of the applications of primary interest. Thus most of the IAEA activities related to nuclear production of hydrogen have taken place in the Gas Cooled Reactor Project of the IAEA Nuclear Power Technology Development Section. This paper provides an overview of the Gas Cooled Reactor Project and the activities of the project of primary relevance to hydrogen production. Specific activities addressed in more detail include a co-ordinated research project on design and evaluation of heat utilisation systems for the high temperature engineering test reactor in Japan, and the production of a technical report providing a review of hydrogen production by nuclear power.

Research Activities on High Temperature Gas-Cooled Reactors (HTRS) in the 5th EURATOM RTD Framework Programme

J. Martin-Bermejo and M. Hugon
European Commission DG RTD,Belgium

There are strong indications in the scientific community that our society is moving inexorably towards hydrogen as the ultimate clean power source of the future. One of the main reasons is the rapid development of the fuel cell technologies. Fuel cells hold great promise in delivering very efficient and clean energy for a wide range of applications (e.g. electricity production, transport, telecommunications), and most of them use hydrogen as fuel. The potential of fuel cells has been recognised since the inception of the European Union (EU) RTD multi-annual Framework Programmes as it is reflected in the increasing budgets allocated to that end in the specific programmes for non-nuclear and renewable energies (e.g. JOULE, THERMIE). Most of these RTD activities address the technical challenges and the complex issues related to the design of the fuel cells themselves. However, one of the main questions (i.e. where will the hydrogen that powers them come from?) remains yet to be answered. A possible response could be nuclear energy, and, in particular, High Temperature Gas-cooled Reactors (HTRs).

The strategic goal of the 5^{th} multi-annual (1998-2002) EURATOM RTD Framework Programme (FP5) is to help exploit the full potential of nuclear energy in a sustainable manner, by making current technologies even safer and more economical and by exploring promising new concepts. This programme is being implemented through "indirect actions", i.e. research co-sponsored and co-ordinated by DG RESEARCH of the European Commission (EC) but carried out by external public and private organisations as multi-partner projects, and "direct actions", i.e. research sponsored and undertaken directly by the EC Joint Research Centre (JRC). One of the areas of research of the "*nuclear fission*" key action of FP5 is "Safety and efficiency of future systems", and it has as an objective to investigate and evaluate new or revisited concepts for nuclear energy that offer potential longer term benefits in terms of cost, safety, waste management, use of fissile material, less risk of diversion and sustainability.

After the first call for proposals of FP5 (deadline October 1999), several projects related to HTRs were retained by the EC services. They address important issues such as HTR fuel technology, HTR fuel cycle and HTR materials In the next call for proposals (deadline January 2001 the EC expects important HTR-related items not covered by the first call (e.g. power conversion systems and system analysis) to be addressed. The EC also expects proposals for strategy studies and/or thematic networks for the assessment of applications of nuclear energy other than electricity production such as hydrogen production. These actions could provide a preliminary indication of the potential of these applications and help the EC in the definition of future RTD needs and priorities.

Tuesday 3 October

Session 3: Physics and Chemistry of Hydrogen Production Methods

Chairman ***A.Guigon***

Open Questions and Research Topics on Nuclear Hydrogen

W. Stoll
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Nuclear Power presently meets some apprehension in the Western World. Its use is practically limited to electricity production only. In the process of harvesting fission energy the primary energy produced in the MeV-range is thermodynamically downgraded to the 400°C-level. This process, as used in current LWR, has severe material stability limitations and is coupled to conventional steam turbines, mainly because of their widespread technical availability and the general utility experience background with the steam-to-electric-power-system.

The routine approach to Hydrogen would be :Nuclear power – electricity - electrolysis. This has been demonstrated, but the overall efficiency is low (standard LWR: 28 to 34% thermal to electrical efficiency, electric power to hydrogen : 50% efficiency at best). Direct thermo-chemical processes, which have an overall efficiency surpassing those 14 to 17 %, should be easy to find, but they have the drawback, that the dissociation equilibrium of water into hydrogen and oxygen needs some 2500°C, to be measurably shifted towards the separated gases. Present nuclear power systems cannot supply this temperature level. If the standard thermo-chemical approach was to be used, it is necessary, to split the energetic requirement for water dissociation into several steps, where each of which promotes the reaction stepwise on a lower temperature level only.

Among others C. Marchetti has published several of those cycles, none of which is simple and most of them, because of utilizing poisonous and costly (Mercury) and/or halogenous compounds, are risky and have a high corrosion potential. They also all need for some steps the high temperature level of a HTGR.Maximum perceived temperature limits for this reactor type range up to some 900°C. There are limits in the heat exchanger material (only Siliconcarbide seems feasible) and the secondary cooling medium presently could only be high temperature steam, at best. As there is no real economic incentive, the system has nowhere ever matured to an industrial facility yet, while there is still scientific interest in Japan, China and Russia.

On the nuclear side there are practically no activities in the direction of direct (thermal) hydrogen production anymore, since the methane-cracking activities by the German JÜLICH Research Center have been stalled.

From all that there seems to be a general impasse in the onset.
Besides the technical problems it has to be noted, that the general direction of the research, as focussed in the hydrogen society activities (in the US and elsewhere) is supported by circles in a juxtaposition to nuclear energy, to stay clear from the perceived risks of that technology. Therefore the preferred energy basis comes from the “soft” spectrum, like Solar, Wind and Hydro-energy. It is also more oriented

towards the solving the problems of Hydrogen engines, batteries, the storage and the transport of hydrogen (Hydrides, pressure vessels). This is reflected in the "Strategic Planning for the Hydrogen Economy" , issued by the national Hydrogen Association (=NHA), seated in New York, in Jan.1998.

The primary energy sources between the present chemical Hydrogen fabrication (from coal and/or Hydrocarbons) and the soft energies, like solar energy have still a wide cost- and efficiency gap, which keeps hydrogen from "soft" sources economically inattractive.

Nuclear hydrogen would fit well in between and would be "Greenhouse-beneficial" as well.
Research topics, outlined in more detail in the paper, center around a literature search and a practical evaluation of new low-energy thermodynamic processes, the potential use of fission spikes and radiation energy for radiochemical Hydrogen production, the material science around helium-tight heat exchanger materials at high temperatures and the nuclear energy substitution in the thermal Hydrogen production from fossil fuel, e.g. in the form of nuclear process steam.

While this temperature level cannot be reached with standard nuclear power stations, it is nevertheless occurring to a high fraction in the primary fission fragment energy spectrum.

There were early experimental "chemical" reactor experiments (Harteck, Bonhoeffer, Bethe, at the Rennselaer Polytechnical University (NY), in the early 60es), to utilize this energy by passing gas mixtures over fiber-glass strands, impregnated with highly enriched Uranium, in a reactor environment. Also the gamma-radiation was used as an energy source for a chemical reaction (Dow-Chemical produced ethyl-bromide from ethane and Bromine for some years in a special high gamma energy reactor). Also the high G-value (0,44) for alpha´s, found in water dissociation, points to that possibility.

Some new ones, probably a bit more elegant, are (documented here by some of the more recent publications, which are not the only ones, the :H_2-H_2O-HBr -system with silica membranes (S.Morooka, S.S.Kim, J.21(3), 1996, pp 183-188), The Iodine – Sulfur closed cycle (H.Nakajima, K.Ikenoja, J.24(7),1999, pp.603-612, The catalytic splitting of HCl and of H_2S (E.I.Onstott, J.22(4), 1997, pp. 405-408, and the bicarbonate-formiate reaction cycle (O.I.Onsager, M.S.A.Brownrigg, J. 21(10), 1996, pp. 883-885

The other additional approaches fall in to three categories :Undertunneling the thermodynamic equilibrium by the particular sorption capability and an "uphill"-diffusion of the group of membranes in the system Pd-Ag-Ni, by special foils or layers, (F.A.Lewis, J. 20(1),1995, pp. 35-41, and R.V.Bucur, J.22(2-3), 1997, pp.141-144, and also catalytic activities, lowering the reaction temperature on Zeolithes(M.Momirlan, J.22(12), 1997,pp. 1133-1136) and :on Ni^{II}-ferrites, (Y.Tamura, N.Hasegawa, et al., J. 21(9),1996, pp. 781-787) and mixed systems, where the starting material is carbonaceous e.g. Methan, et al., (B.Gaudernack, J.23(12), 1998, pp. 1087-1093) and the thermal decomposition energy directly or around some intermediate reactions (e.g. with ZnO) is introduced via heat, (A.Frei, J.20(10), 1995, pp. 793 – 780) also nuclear heat, saving some C to be burnt to CO_2 (more generally in: C.Mitsugi, A,Harumi, F.Kenzo, J. 23(3), 1998, pp. 159-165)

Most of the other reaction schemes are dependent on high temperature, photo-ionisation or microbial action (Halobacterium Halobium and others) , all more or less off-springs of solar light.
Among the people, being active in this field, could be the authors quoted above, and some scientists of them and in addition ,which are quoted in several topics, as :

Prof. Cesare Marchetti, now at IIASA,
F.Kenzo, Jap. H_2-program,
J.Rothstein, Nuclear energy and hydrogen economy,
E.Bilgen, UT-3 thermochemical Hydrogen,
M. Lennartz, Hydrogen Program Jülich/Germany,
V.A.Goltzov, together with the present chairman of Hydrogen society,
T. N.Veziroglu (a good summary of the present state of the art in :
"Hydrogen Energy progress XII, by J.C.Bolcich, T.N. Veziroglu, Proceedings of the 12th World Hydrogen Energy Conference, ISBN 987-97975-3-2)
K.J.Kim, HBr-system,
M.S.A.Brownrigg, formiate-way to H_2,
P.F.Clarke, H_2S-thermal decomposition,
R.V.Bucur, Pd-diffusion.

But since this time practically no new publications covering this field have turned up.
Prof. Marchetti has followed the field in a much broader, philosophical manner from his present position at IIASA in Laxemburg/Vienna:

1) C.Marchetti, On Society and Nuclear Energy, Report EUR 12675 EN, Commission of the European Communities, Luxembourg, Dec.1988
2) C.Marchetti, Nuclear Energy and its Future, in: Perspectives in Energy, 1992, Vol. 2, pp. 19-34
3) C.Marchetti, On Decarbonisation: Historically and Perspectively, IIASA Laxemburg, prepared for HYFORUM 2000.

Hydrogen Production by Thermal Decomposition of Methane: an Assessment and an Experimental Program

A. Aiello, G. Benamati, G. Gherardi and G. Marucci
ENEA, Italy

Today, steam catalytic conversion of methane seems to be the most attractive hydrogen production technology:

$$CH_4 + H_2O \rightarrow CO + 3H_2$$
$$CO + H_2O \rightarrow CO_2 + H_2$$

All the carbon is converted in CO_2, and the gas has to be sequestered under the ocean or underground (depleted oil or gas wells), to mitigate the CO_2 greenhouse gas climate change, increasing significantly the hydrogen cost.

Moreover, in a natural gas (methane) pyrolysis only black carbon and hydrogen are produced according to the reaction CH_4=C+2H_2. The higher temperatures required and the low efficiency are the main disadvantages of discontinuous thermal pyrolysis process, but new continuos production processes, based on liquid metals, are under investigation.

Thermal decomposition of methane requires temperatures above 700°C, and higher pressures favour higher rates of reactions. The endothermic energy to perform the reaction is only 75.3 kJ per mol of methane to produce two moles of hydrogen at 25°C and the efficiency of hydrogen production by continuos thermal decomposition process seems to exceed 50%. The value of pyrocarbon cannot be ignored, and it can modify the economical balance of the process. Pyrocarbon can be sold to the market and the cost vary significantly from 300 to 4000 $ per ton.

We are examining a thermal methane decomposition using a molten lead bath. Theorically, bubbling methane in liquid metal heated to temperatures between 700°C and 900°C could decompose over 90% of methane. The carbon could be separated on the top of liquid metal bath.

A testing facility to verify the process feasibility is under construction.

A Study on Thermochemical I-S Process

K. Onuki
Japan Atomic Energy Research Institute, Japan

Thermochemical hydrogen production from water using high temperature nuclear heat has a potential to become an important technology to realize the so-called "Hydrogen Energy System" where hydrogen will play a key roll as a secondary energy in complementary with electricity, the dominant energy carrier of the present energy system. A thermochemical process utilizes several endothermic- and exothermic-chemical reactions, where all the reactants except water are cyclically used in the process. The process absorbs high temperature heat in the endothermic reactions, produces hydrogen and oxygen, and disposes low temperature waste heat in the exothermic reactions. In other word, it works like a chemical engine to produce hydrogen from water using nuclear heat.

Japan Atomic Energy Research Institute (JAERI) has conducted a research and development (R&D) of one version of thermochemical processes named I(Iodine)-S(Sulfur) process. The process utilizes the following three chemical reactions; endothermic sulfuric acid decomposition for oxygen production, hydrogen iodide decomposition for hydrogen production, and exothermic Bunsen reaction for the acids production. The process was first proposed by General Atomics, and has been studied in several research institutions. At JAERI, a "closed-cycle" continuous hydrogen production by IS process was successfully demonstrated in laboratory with the production rate of 1 L-H_2/h for 48 hrs. Next target is to carry out the closed-cycle hydrogen production under more efficient conditions, namely, to carry out the Bunsen reaction step at elevated temperatures, where the concentration and also the separation efficiency of the acids could be increased. At present, a laboratory-scale experimental apparatus for the study is under construction. In parallel, a study to improve the process using membrane technologies is in progress, which aims at establishing an efficient hydrogen iodide decomposition step having a simpler and milder flowsheet compared to the previous ones. Thirdly, selection and development efforts on the materials of construction are under way, which should bare the corrosive process environments.

In the meeting, the present status and the perspective of these studies will be presented.

The Pyrolysis of Methane over Liquid Metal to Form Hydrogen and Carbon

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A calculational and experimental study was undertaken to determine whether a liquid metal could be used as a heat source for the pyrolysis of methane to carbon and hydrogen. The thermodynamics and energetics for production of hydrogen by methane pyrolysis, followed by generation of electricity in a fuel cell, were determined to be favorable. The financial penalty from the sequestration of carbon is only about $100 per ton of carbon produced. Substitution of this process for combustion of natural gas by a turbine would make a major contribution to reducing greenhouse gas emissions.

Hydrogen Permeation Barrier Development

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The control of hydrogen losses in an hydrogen production industrial plant is of crucial importance especially for its safety implications. High temperatures and pressures required in several processes, and corrosive process fluids can enhance drastically the intrinsic permeation characteristics of metals and alloys.

To reduce the hydrogen permeation and hydrogen mechanical properties dependence a low permeation coating can be realised on structural materials. Better performances can be achieved using alumina rich coatings.

Several deposition techniques are under investigation but Chemical Vapour Deposition and Hot Dipping seems to offer good performances. In this work is examined the effectiveness of different hydrogen permeation barriers using time dependant method in gas phase and in liquid metal phase.
Testing devices PERI and Vivaldi are presented.

Tuesday 3 October

Session 4: Applications of Nuclear Technology for Hydrogen Production

Chairman ***M. Lecomte***

Hydrogen for Transportation: Available Canadian Technology

A. Miller
Atomic Energy of Canada Ltd, Canada

Just as the 20th Century may be viewed by historians as the CO_2 Century, converging trends strongly suggest that the 21st Century will either be the H_2 Century, or the century in which the World's climate experiences severe disruption (or worse) and the World economy collapses.

Economically, the period of abundant cheap energy (oil) is ending while the rising price of natural gas indicates that its current vogue will be ephemeral. In parallel to cost pressures on the supply side of hydrocarbons, concern is switching to alarm over effects on global climate of the by-product emission of greenhouse gases (GHGs), principally CO_2, from burning of carbonaceous fuels. Given the economic needs and aspirations of the huge populations of developing countries, even sharply higher efficiency in the use of carbonaceous fossil fuels offers only a short-term palliative.

Various non-carbon, sustainable sources of energy are being developed but only nuclear power and electricity from mature hydroelectric dams have a track-record of significant reduction in GHG emissions. In 1995, Canada's CANDU® reactors reduced GHG emissions by around 100 million tonnes, an avoidance of about 14% from all Canadian man-made sources. World-wide nuclear today avoids CO_2 emissions of over one billion tonnes each year. If applied just to base-load electricity generation, expansion of nuclear quite quickly saturates the potential for power-generation emissions avoidance and world CO_2 emissions grow as automobile, truck and industrial fuel use expands.

To reduce emissions further and allow sustainable development, hydrogen is the obvious emissions-free means both to provide electricity for traditional applications and to extend fuel substitution into the transportation sector. Note that, in Canada, CO_2 emissions from the transportation sector are almost double those from electricity production, which is also true or becoming so in many countries as they switch to the free-enterprise model. Applying hydrogen from nuclear (and any other near-zero GHG-emitting energy sources) to transportation doubles or triples nuclear's leverage to help reduce the GHG emissions problem.

Obviously, hydrogen produced by steam reforming of natural gas offers little advantage in total cycle GHG emissions over normal automobile engine technology. Hydrogen produced via methanol is even less attractive and would be inferior even to battery-IC hybrid technology.

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The Canadian position is that GHG reduction is urgently required and Canada has agreed to target significant CO_2 reductions, albeit with much continuing and vociferous discussion. We and the World cannot wait for development of radical new technologies.

Canada is a leader in modern energy technologies, and in balancing energy production and exploitation with environmental concerns. Canada is well placed to apply proven or near-proven technologies: as well as the CANDU reactor, Canadian companies are world leaders in the development of the component technologies for application of hydrogen to transportation. Ballard's fuel cells are the reference design for most of the world's car makers. Stuart Energy Systems has developed a low-cost electrolysis unit for consumer use, also in collaboration with major car makers—avoiding the need to develop a new distribution infrastructure. Dynetek is a world leader in low-weight, high-pressure cylinders for hydrogen storage.

Phased and sensible application of fuel cells to Canadian transportation could avoid more than another ~100 Mt CO_2 each year.

Canada is a country of low population density and large distances, which makes the electrification of railways largely uneconomic. However, fuel cells powered by liquid hydrogen would be an attractive way to displace diesel-powered locomotives, preferably leveraged by transfer of freight away from roads and onto rail.

Another opportunity that has been identified in Canada is large-scale electrolytic production of hydrogen and oxygen combined with production of heavy water. Assuming that the transition to a post-hydrocarbon economy is evolutionary rather than abrupt, further development of the oilsands in northern Alberta is likely. Reserves there exceed that of the largest Gulf nations, but require extensive energy for extraction and hydrogen for upgrading to refined crude. Extraction and processing of bitumen consume large quantities of steam (at pressures within the reach of a CANDU reactor), electricity, hydrogen and oxygen. CANDU reactors can provide this combination, with the advantage of further elimination of CO_2 emissions, perhaps by 20 Mt CO_2 per year or more. Economic analysis of the co-generation of steam, hydrogen, electricity, heavy water and oxygen streams show this to be technically viable and environmentally attractive.

Overall Canada is well placed to both achieve significant emissions reductions, by introducing new technology and innovative adaptations of synergistic nuclear , hydrogen and carbon-based energy use.

A Proposed Modular-Sized, Integrated Nuclear and Hydrogen-Based Energy Supply/Carrier System

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K. Peddicord
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General Electric Company, USA

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ENEA, Italy

A system is proposed for the utilization of nuclear energy to produce hydrogen, electricity, potable water and other marketable resources in a self-contained, modular plant featuring zero carbon emissions. The reference process for hydrogen production (and oxygen byproduct) is the Sulphur-Iodine thermochemical water cracking cycle. This cycle requires process heat at up to 900? C temperature which is provided by a fast neutron spectrum, heavy liquid metal cooled converter reactor designed for near autonomous operation during a 15-year refueling interval. The reactor is radically simplified and of modular construction for economic competitiveness, and is additionally designed for proliferation resistance and passive safety. It features maximum fission conversion of fuel and actinides for long core operating lifetime, minimum waste, and sustainability. The reactor heats a gaseous heat transport medium for operation of the water cracking cycle, A waste heat is used for desalinization and process heating. The potable water produced is used for local consumption and to feed the water cracking process. A fraction of the hydrogen and oxygen products of this process are used to power a combustion turbine cycle for high efficiency base or load-following electricity generation. The main portion of the hydrogen is available to power fuel cells envisioned to be used in the transportation sector.

Contributions to a Safety Analysis for a Hydrogen Production System with HTGR

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A comprehensive safety and risk assessment for a nuclear hydrogen production system necessitates the definition of a proper methodology to analyze all steps of conceivable event sequences. The accidental potential of such a combined chemical and nuclear complex is given by its radionuclide inventory in the nuclear system and the presence of feed and product gases, if inadvertently released, to form combustible gas mixtures in the secondary or tertiary circuit of the nuclear reactor. Its risk is thus considered higher than that of a conventional nuclear reactor. The quantitative safety analysis comprises the decent investigation of the whole plant with respect to accidental conditions or other abnormal system states.

In this presentation, research activities will be described which cover some parts of a probabilistic safety and risk analysis (PRA) for a hydrogen production plant combined with a High-Temperature Gas-Cooled Reactor (HTGR). In the 1980s, the Research Center Jülich has conducted a safety analysis for two conceptual designs of an HTGR under the modified conditions of process heat applications, the 50 MW AVR-II and the 170 MW variant of the HTR-Module. The focus was laid upon the analysis with respect to its nuclear hazardous potential. The specific features of a process heat HTGR are a higher temperature level in the core because of the higher gas outlet temperature and a lower system pressure to adjust to the pressure conditions in the secondary and tertiary circuit. The tools of a probabilistic risk assessment have also been successfully applied to non-nuclear, flammable gas carrying systems. The systems investigated were the autonomous hydrogen production, storage, and consumption demonstration facility PHOEBUS and the ELBA test vehicle equipped with a 5 kW fuel cell. For both FZJ projects, the hazardous potential and accident-initiating events were identified and the frequencies of safety-relevant event scenarios were estimated.

In a nuclear hydrogen production system, a link is given between the chemical and nuclear system via the secondary helium circuit, thus enabling feed/product gases in case of respective pipeline ruptures to accumulate inside the nuclear containment jeopardizing the nuclear primary circuit in case of an explosion. For the Japanese HTTR, steam reforming of methane is the top candidate method for demonstrating nuclear H2 production. The product gas will be a mixture of H2, CO, CO2, residual H2O, and still unreformed methane. At the Research Center Karlsruhe, an experimental program has been started in the 12 m Hydrogen Detonation test Tube Facility to investigate the combustion behavior of H2-CO-air mixtures under different conditions. The first tests conducted with a fuel (H2 + CO) fraction of 11

% and 18 %, respectively, in a fully confined, partially obstructed geometry have demonstrated a significant retarding effect of the CO. The observation of slower flame acceleration rates and longer acceleration paths indicate longer induction and reaction time scales in comparison to combustion tests with pure H2 fuel under the same conditions.

An impact from outside to the HTTR hydrogen production system is given by the possible accidental formation and explosion of a flammable gas cloud in the open atmosphere resulting from leakages in the chemical complex including the main components steam reformer and storage tanks for LNG or hydrogen. The preceding step of the evolution of a methane-air gas mixture is, e.g., the spillage of the cryogenic liquid LNG after a pressure vessel failure of the storage tank or pipeline rupture onto the ground. A state-of-the-art calculation model has been developed at FZJ, which allows the description of the transient behavior of the cryogenic pool and its vaporization. The code was validated against LNG spill tests from the literature and against own experiments with the release of liquid hydrogen. As an example, the results of a predictive study will be given, in which the behavior of different cryogenic liquids, LH2, LN2, LOX, LNG, was compared based on a released amount of 40 m^3 representing the typical quantity of a tank truck.

Future work: A prerequisite for the analysis of the consequences of vapor cloud formation is the precise knowledge of the plant design and its safety management including the safety of storage and transportation as well as the safety of processing. In particular the mitigation measures need to be well defined because of their significant influence on the scenario of an accident. Combustion modes to be investigated should include boiling liquid expanding vapor cloud explosion (BLEVE), unconfined vapor cloud explosion (UVCE), jet fire, transition deflagration to detonation (DDT).

Hydrogen Production Associated to the Treatment of Nuclear Waste

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A constantly increasing attention is being paid to nuclear waste treatment activities. Several treatment processes can be set up, which are susceptible to produce hydrogen. For safety reasons, one generally tries to avoid hydrogen production whenever possible. However, the opposite approach coping with the possible use of hydrogen as energy vector may also be considered. Associating hydrogen production to waste treatment processes should also be recommended because the economics of the said processes would be favoured though the valorisation of hydrogen, thereby increasing the competitiveness of the waste treatment. The present paper stresses this possibility through two examples, both already involving electrochemical techniques. The first example addresses the case of the decontamination of metallic pieces arising from either the exploitation or the dismantling of nuclear installations. The second example focuses on the destruction of organic waste by electrochemical mediation.

For this purpose, one very effective process consists in dissolving around 8 to 20 µm of the thickness of the metal, using a solution containing around 0.025 mol.L-1 Ce4+ and 0.025 mol.L-1 Ce3+ in 1M sulphuric acid. This method allows oxidising stainless steels under controlled conditions. During the decontamination, Ce4+ is being reduced to Ce3+. To minimise the waste effluent, the solution is being recycled through the anodic compartment of a filter-press electrolyser where Ce3+ is re-oxidised to Ce4+. The reduction of protons to elemental hydrogen occurs in the cathodic compartments, which are fed with a 1 M solution of sulphuric acid. A Nafion® 324 membrane is used as separator. A typical installation devoted to the decontamination of metals produced during the dismantling of a small 10.5 MWe PW reactor would produce 13 Nm3 of hydrogen per hour, under nominal operating conditions.

The SCK-CEN has successfully tested this approach at laboratory scale. The hydrogen evolution proceeds smoothly in the cathodic compartments, while a full model describing the anodic selectivity has been developed. As defined, the process may be easily adapted to recover the hydrogen. A collector line equipped with a demister and a filter may be connected to e.g. a gettering unit for safe storage under the form of hydrides. Compressing the hydrogen in view of storing it in pressure tanks is also convenient.

In this process, strong oxidising Ag++ species are generated electrochemically in nitric acid, using an adequate electrolyser. Next, the oxidising fluid is brought into contact with the organic waste. If the latter is miscible with the anolyte, then the electrogeneration of the Ag++ species and the subsequent oxidation reactions involving the organic waste can occur into the electrolyser. It is known that many organic molecules are completely oxidised to CO2 and H+ by the Ag++ species, which are reduced to

Ag+. Maintaining a current flow through the electrolyser ensures the regeneration of the argentic species and the further reaction with organic molecules until the latter are completely oxidised. The whole process runs below 100 °C and may be used even for oxidising dangerous materials (e.g. explosives or PCBs).

Here also, a second reaction occurs in the cathodic compartments of the filter-press electrolyser. Again, a Nafion® 324 membrane is used as separator. Generally, one chooses nitric acid for both. This means that the cathodic reaction leads to the formation of huge amounts of NOx. An interesting idea is to replace the nitric acid by sulphuric acid in the cathodic compartments. Thereby, the cathodic reaction becomes the reduction of protons and hydrogen is formed again. Experimental investigation of this configuration shows that additional R&D work is required to achieve the goals while limiting the energy requirements. Indeed, we measured a relatively high voltage drop across the membrane (up to 0.9 V) using current densities below the recommended values for the silver II process. This voltage drop is responsible for only part of the cell voltage increase observed when passing from the usual configuration ((+) Pt ; (HNO3 + AgNO3)/(HNO3 + AgNO3) ; SS304 (-)) to the new situation ((+) Pt ; (HNO3 + AgNO3)/(H2SO4) ; SS304 (-)). Next, the selectivity of the membrane appears to be insufficient. Small but detectable amounts of Ag+ migrate to the cathodic compartments where they are reduced to elemental silver. Interesting hysteresis behaviour of the cell voltage is also observed. All these experimental results will be detailed and discussed in the final paper.

It is possible to combine hydrogen production with electrochemical waste treatment processes. In the case of the decontamination of metallic pieces, only minor changes to the existing processes would be required to reach the goals. On the other hand, the mediated oxidation of organic waste requires additional R&D efforts in order to valorise the cathodic current through hydrogen production.

Thermochemical Hydrogen Production with a High Temperature Gas Cooled Reactor

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Currently there is no large scale, cost effective and environmentally attractive hydrogen production process available for commercialization, nor has such a process been identified. The U.S. Government through the Nuclear Energy Research Initiative (NERI) is funding a three-year development program to determine the potential for efficient, cost-effective, large-scale production of hydrogen using high temperature heat from an advanced nuclear power station.

This report will address the first phase of the project. In this phase we evaluated thermochemical processes which offer the potential for efficient, large-scale production of hydrogen from water, in which the primary energy input is high temperature heat from an advanced nuclear reactor. The result of the first phase was the selection of two processes for further detailed consideration.

The work in Phase 1 consisted of:

A detailed literature search and development of a database of all published thermochemical cycles.
A first round screening process to reduce the initial list to 20-30 cycles.
A second round of screening to reduce the number of cycles to 3 or less.
A report describing the results of Phase 1.

Ten databases were searched and over 800 literature references were located, representing over 100 thermochemical water-splitting cycles. After two rounds of screening two cycles were selected for final consideration: the adiabatic UT-3 cycle and the sulfur-iodine cycle. The adiabatic UT-3 cycle was invented at the University of Tokyo and has been extensively studied in Japan by a number of organizations. Its predicted efficiency is 40%. In a combined hydrogen/electric plant, overall efficiencies as high as 50% have been projected. The sulfur-iodine cycle has been under development since the early 1970s and improvements proposed by various researchers should further increase the already high predicted efficiency (52%) of this cycle.

Considering that a significant effort is already under way on the UT-3 cycle in Japan, we selected the sulfur-iodine cycle for further development in the USA.

We are working on establishing a cooperative program with our Japanese colleagues to foster joint development of both the UT-3 and the sulfur-iodine process. This may allow future selection of one thermochemical water-splitting cycle for scale-up and demonstration.

Our effort in Phases 2 and 3 will be aimed at incorporating the proposed improvements into the sulfur-iodine cycle and at adopting this process to a high temperature advanced nuclear reactor.

Tuesday 3 October

Session 5 : Discussions and Preparation of Recommendations for the Path Forward

Chairman *D.Wade*