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THE UNITED STATES FOREIGN RESEARCH REACTOR SPENT NUCLEAR FUEL ACCEPTANCE PROGRAM: PROPOSAL TO MODIFY THE PROGRAM

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ABSTRACT

The United States Department of Energy (DOE), in consultation with the Department of State (DOS), adopted the *Nuclear Weapons Nonproliferation Policy Concerning Foreign Research Reactor Spent Nuclear Fuel* in May 1996. The policy was slated to expire in May 2009. However, in October 2003, a petition requesting a program extension was delivered to the United States Secretary of Energy from a group of research reactor operators from foreign countries. In April 2004, the Secretary directed DOE undertake an analysis, as required by the National Environmental Policy Act (NEPA), to consider potential extension of the Program. On December 1, 2004, a Federal Register Notice was issued approving the program extension. This paper discusses the findings from the NEPA analysis and the potential changes in the program that may result from implementation of the proposed changes.

1. Introduction

DOE, in consultation with the Department of State (DOS), adopted the Nuclear Weapons Nonproliferation Policy Concerning Foreign Research Reactor Spent Nuclear Fuel in May 1996. The program was scheduled to expire in May 2009. The Foreign Research Reactor (FRR) Spent Nuclear Fuel (SNF) Acceptance Program is a critical component of U.S. nonproliferation efforts. The primary objective of the program is to reduce and eventually eliminate highly-enriched uranium (HEU) of U.S.-origin from civil nuclear applications worldwide. Since 1996, the Acceptance Program has conducted 31 shipments involving 27 countries in the safe return of 6,445 spent nuclear fuel elements to the United States for management at Department of Energy (DOE) sites in South Carolina and Idaho, pending final disposition in a geologic repository.

2. Global Threat Reduction Initiative

In May 2004, the FRR SNF Acceptance Program was incorporated as a key element in DOE's newly established Global Threat Reduction Initiative (GTRI). GTRI's mission is to identify, secure, remove or facilitate the disposition of vulnerable, high-risk nuclear and other radiological materials around the world ---as quickly and expeditiously as possible---that pose a threat to the United States and the international community. GTRI objectives include to:

- accelerate and complete the repatriation of U.S.-origin research reactor spent HEU fuel from locations around the world;
- work in partnership with Russia to repatriate Russian-origin HEU fuel from Russian-supplied research reactors;
- work to convert the cores of civilian research reactors that use HEU to use low-enriched uranium (LEU) fuel; and
- work to identify other nuclear and radiological materials and related equipment that are not yet covered by existing threat reduction efforts and rapidly address the most vulnerable facilities.

The National Nuclear Security Administration (NNSA) has made remarkable progress since this new initiative was launched last year. Representatives from approximately 100 countries attended the successful Global Threat Reduction Initiative International Partners' Conference last September. In addition, under GTRI and the Russian Research Reactor Fuel Return program, NNSA has worked closely with the IAEA, the Russian Federation, and other countries to repatriate fresh HEU from Uzbekistan and the Czech Republic to Russia over the past 6 months. More than 10 other joint projects under GTRI are in the planning stages.

Although the Acceptance Program has demonstrated significant progress, participation and shipping rates have not met initial expectations. As of the beginning of FY2005, DOE had received only about 35 percent of the material eligible for return as estimated in the Final Environmental Impact Statement on a Proposed Nuclear Weapons Nonproliferation Policy Concerning Foreign Research Reactor Spent Nuclear Fuel [1] (FRR SNF EIS).

Several factors have contributed to lower than expected receipts. Several countries have burned fuel less rapidly than was expected in 1996. In addition, two countries (Belgium and France) decided to process their SNF at the COGEMA facility in France, and consequently have not participated in the program. These two countries were expected to contribute a significant percentage of the fuel elements projected for acceptance by 2009. However, it is deemed appropriate and acceptable to process SNF to LEU material. Finally, as discussed below, delays have been experienced in the development of alternative fuels. Under the GTRI, NNSA is working to overcome these obstacles by accelerating the development of higher-density LEU fuels, and accelerating the return of eligible U.S.-origin HEU spent nuclear fuel from research reactors.

3. Low-Enriched Uranium (LEU) fuel development

The international community has experienced technical delays in the development of an alternate "processable" LEU fuel. The development and qualification of LEU fuel that can be processed is important to ensure FRRs are able to continue reactor operation in the foreseeable future and ensure proper disposition of SNF. Extending the Acceptance Program ensures a backend solution is maintained while progress continues in eliminating HEU in research reactor fuels.

The most predominant LEU fuel being used today is uranium-silicide LEU fuel developed in the 1980s by DOE. Although processing studies concluded that the uranium-silicide fuel could be successfully processed at the Savannah River Site (SRS), disposition through a repository was chosen in lieu of processing at the SRS unless the physical condition of the fuel warranted a more immediate disposition. To date, no SNF received from the FRR SNF Acceptance Program has been dispositioned by processing in the SRS H-Canyon Separation Facility. The United Kingdom Atomic Energy Agency (UKAEA) demonstrated in laboratory scale trials [2] that uranium-silicide LEU fuel could be processed at its facility in Dounreay, Scotland. However, the capital investment in new equipment required for large-scale processing operations was determined to be uneconomical, and the facility was shutdown in 1998. COGEMA in La Hague France is the only remaining major potential processing facility. COGEMA has not processed uranium-silicide fuel on a commercial scale, and the facility would also require additional equipment for efficient processing operations. However, COGEMA is now accepting a limited quantity of uranium-silicide LEU fuel for processing, and is investigating the possibility of increasing the quantity that could be accepted for processing [3].

4. Proposal to modify the acceptance program

During the DOE-sponsored Reduced Enrichment for Research and Test Reactors (RERTR) Conference in Chicago in October 2003, a group of reactor operators and supporters (40 petitioners from 17 countries) prepared and delivered a petition to the U.S. Secretary of Energy. The petition requested that DOE extend the FRR SNF Acceptance Program until new LEU fuels become available, allowing reactor operators to convert to a LEU fuel type that has an identified disposition path. On

February 11, 2004, in a speech to the National Defense University, the President of the United States stated, “We will help nations end the use of weapons-grade uranium in research reactors.” On April 14, 2004, the Secretary of Energy directed the DOE’s National Nuclear Security Administration (NNSA) to consolidate the U.S. FRR SNF Acceptance Program within its nonproliferation mission, further directing the appropriate offices within DOE to initiate actions necessary to extend the FRR SNF Acceptance Program’s fuel acceptance deadline.

As a result of these actions, DOE conducted an evaluation on the effects of extending the FRR SNF Acceptance Program and adding the Open Pool Australian Lightwater (OPAL) Reactor (formerly referred to as the Australian Replacement Research Reactor), documented in the Supplement Analysis for the Foreign Research Reactor Spent Nuclear Fuel Acceptance Program [4] in accordance with the National Environmental Policy Act (NEPA) regulations. DOE and the U.S. Department of State proposed to modify the FRR SNF Acceptance Program by:

- Extending the expiration date for irradiation of eligible spent fuel either 5 or 10 years, from May 12, 2006, to May 12, 2011, or May 12, 2016;
- Extending the acceptance date for eligible spent fuel either 5 or 10 years, from May 12, 2009, to May 12, 2014, or May 12, 2019; and
- Extending eligibility to Australia’s Replacement Research Reactor (RRR) for participation in the Acceptance Program.

The amount of eligible SNF would remain the same as identified in the original EIS, about 22,700 elements or about 20 metric tonnes of heavy metal (MTHM). The proposed program extension would not change other requirements contained in the EIS assumptions and EIS Record of Decision. However, target material (fuel for isotope production such as Technicium-99) and damaged spent fuel currently can only be treated in H-Canyon at SRS. Current DOE plans call for the SRS H-Canyon facilities to be maintained in operable condition through 2010 pending a review of the facility and DOE’s initiatives. Therefore, target material and damaged spent fuel would not be eligible for acceptance beyond 2009 under the proposed extension.

5. Analysis of the proposed action

The Supplement Analysis discusses the impact of the proposed action on the shipment schedule projected and analyzed in the FRR SNF EIS, and compares the likely effects that the proposed action would have on environmental resources to those analyzed in the EIS. The comparison focuses primarily on the radiological effects on human health and safety. Human health and safety is considered to be the most important resource, and radiological considerations are the most likely variables, to be affected by the proposed action. The Supplement Analysis also discusses the most significant assumptions used in the FRR SNF EIS to analyze the radiological effects on human health and safety, the conservatism of these assumptions based on experience gained from actual shipments to date, and assumptions that need to be changed because of the proposed action or because of new information available since the publication of the FRR SNF EIS in 1996. Key considerations evaluated in the Supplement Analysis include shipping schedules; normal and accident radiological consequences for marine, port, and ground transportation situations; and impacts to the management sites. A summary of these considerations is provided below:

Shipment schedule: The Supplement Analysis determined that shipment schedule projected over the extended period is expected to remain substantially lower than the constant shipping rate assumed in the FRR SNF EIS.

Normal and Accident Radiological Consequences: The Supplement Analysis discusses the conservatism built into the original EIS radiological impact analysis and analyzed changed assumptions and actual conditions since the EIS. The evaluation of the normal and accident radiological consequences included bounding SNF fission product activity, transport packages used in the program, projected change in population density over the extended period, actual

transport package external dose rate from experience derived since the program's inception, and the effect of receipt of the OPAL SNF. These factors were applied to analysis of marine transport, port activity, and ground transportation impacts. Accident and incident-free impacts of marine transport, port activity, and ground transportation activities were considered. The study found no significant increase in risk to marine shipping crews, port workers, or ground transportation workers as a result of the proposed program extension. The study found the proposed extension could increase the radiological risk to the general public near the port of entry or along ground transportation routes primarily due to population increases since 1996, but the risk remained within or slightly above the low risk calculated in the FRR SNF EIS.

Impacts at SRS and the Idaho National Engineering and Environmental Laboratory: The Supplement Analysis determined that the proposed extension would not impact the total quantity of candidate SNF and that the SNF mass and receipt schedules would remain within DOE site-specific limits. The study also determined that the risk to workers and the general public from the addition of the Australian's OPAL Reactor fuel assemblies would be too small to impact existing environmental impact calculations.

6. Conclusion

The Supplement Analysis determined that an extension of the FRR SNF Acceptance Program for 5 or 10 years and inclusion of the Australian's OPAL Reactor would not constitute a substantial change in action relevant to environmental concerns. Therefore, additional NEPA documentation to the FRR SNF EIS would not be required. The program extension was approved within DOE on November 23, 2004 and a formal issuance of an extension of the program occurred by the issuance of a Federal Register Notice [5] on December 1, 2004.

The OPAL Reactor was added to the Supplement Analysis and the subsequent Record of Decision changed to accommodate a unique situation encountered by the Australian Nuclear Science and Technology Organization (ANSTO) as the HEU-fueled High Flux Australian Reactor (HIFAR) is replaced with the LEU fueled OPAL. Conversion from HEU fuel to LEU fuel is effectively occurring as the HEU fueled HIFAR is shutdown and the LEU fueled OPAL is brought on-line. ANSTO has participated heavily in the development of high-density uranium-molybdenum LEU fuel, serving to benefit continued development of this fuel for other FRRs. Because the uranium-molybdenum LEU fuel will not be available in the timeframe required for commissioning of OPAL it is expected to operate for a limited period of time using uranium-silicide LEU fuel. Without confirmed processing options for all of this fuel, the Australian government has requested that the fuel be accepted under DOE's FRR SNF Acceptance Program until processable uranium-molybdenum fuel becomes available.

Subsequent to DOE's determination that additional NEPA documentation to the FRR SNF EIS would not be required, DOE issued a change to the FRR SNF EIS Record of Decision. The change notice to the Record of Decision allows the program to be extended for an additional 10 years and includes the participation of the Australian's OPAL Reactor. The issuance of a change notice to the Record of Decision also allows DOE to proceed in development of an implementation plan and guidance for the Acceptance Program extension. This information will be shared with FRR participants and other interested parties.

Many currently participating FRR operators will not need to change their current shipping strategies and participation and shipping schedule will continue as currently planned. However, some FRR operators may need to change their participation schedule based on specific operational needs and requirements. It is not the intent of the FRR SNF Acceptance Program or the GTRI mission to prevent or disrupt the needed operation of a research reactor. The Program intends to assist reactor operators in conversion to LEU fuels while providing an acceptable disposition path for SNF involving uranium that was enriched in the United States. In accordance with the objectives of GTRI and the allowances provided in the FRR SNF Acceptance Program extension, it is expected that FRRs that are currently

using HEU fuel will convert to use LEU fuel as soon as possible with the understanding that SNF with uranium enriched in the United States can be dispositioned in the United States until May 13, 2019 provided the fuel is not irradiated after May 13, 2016. Operational strategies will be evaluated on a case-by-case basis based on these new considerations.

As a result of these new GTRI activities, DOE is evaluating the FRR SNF Acceptance Program Fee Policy to determine how to accelerate and solidify shipment schedules.

The United States remains committed to supporting worldwide nonproliferation goals such as those for which this program was designed. Accepting eligible fuel now rather than later remains a primary focus of that commitment. The United States hopes to work with all remaining eligible research reactors to plan for shipments of their eligible spent fuel, and DOE continues to support research reactor operators' needs and seeks to meet any interested parties to further discuss the program

7. References

[1] Department of Energy, Final Environmental Impact Statement on a Proposed Nuclear Weapons Nonproliferation Policy Concerning Foreign Research Reactor Spent Nuclear Fuel, DOE/EIS-0218F, February 1996

[2] Cartwright, P., 1996, "Reprocessing of LEU Silicide Fuel at Dounreay." Proceedings of the 19th International Meeting on Reduced Enrichment for Research and Test Reactors, Seoul, Korea, October 7-10, p. 418, United Kingdom Atomic Energy Agency, Dounreay, Scotland.

[3] Auziere, P., 2004, "UMo Spent Fuel Acceptance Treatment at La Hague Plant." Transactions of the 8th Topical Meeting on Research Reactor Fuel Management, Munich, Germany, March 21-24, p. 131, COGEMA, La Hague, France.

[4] Department of Energy, Supplemental Analysis for the Foreign Research Reactor Spent Nuclear Fuel Acceptance Program, DOE/EIS-0218-SA-3, November 2004

[5] Federal Register Notice, Volume 69, No 230, Wednesday, December 1, 2004, Pages 69901 – 69903, Revision of the Record of Decision for a Nuclear Weapons Nonproliferation Policy Concerning Foreign Research Reactor Spent Nuclear Fuel.

R&D FOR THE FINAL DISPOSAL OF IRRADIATED RESEARCH REACTOR FUEL ELEMENTS

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ABSTRACT

R&D is going on in the Research Centre Jülich to investigate the long-term behaviour of U/Al and U₃Si₂ based fuels in salt, clay and granite repositories. Results from leaching experiments with irradiated U/Al fuel elements in salt brines show, that the fuel corroded within some months and that the mobilised radionuclides were trapped by the formed secondary phases. Treatment of these secondary phases with fresh brine resulted in a quantitative remobilisation of Cs and Sr, while Pu was not affected at all. A phase component of these corrosion products was identified as Mg-Al-Cl-hydrotalcite. The ability of this double layer hydroxide to trap ⁹⁹Tc and ¹²⁹I was investigated.

Recently leaching experiments with non-irradiated U/Al and U₃Si₂ based fuel elements in clay pore water (Mont-Terri type) have started and the first results show that both types corroded. Work is in progress.

1. Introduction

Quite a great number of different types of spent fuel from research reactors exist in Germany. The general policy is to send fuel from MTRs and TRIGAs of US origin back to the USA as long as possible. An alternative option is reprocessing in Great Britain or France. For those fuels which cannot be returned to the USA or which will not be reprocessed with respect to economical reasons, a back-end option has been developed in Germany. It consists in dry storage in special casks and, later on, direct disposal in deep geological formations [1]. As deep geological formations salt, granite and clay are considered. In granite and clay formations pore water is always present but in a salt repository a water ingress and the subsequent formation of salt brines is considered as accident scenario. In the presence of these aquatic phases the corrosion of the fuel container and then the direct contact to the fuel itself is possible. The arising question “If the fuel element will corrode, what will happen to the radionuclides” must be answered. This question in mind, first leaching experiments with a metallic U/Al-fuel element in salt brines and in granite water (Grimsel-West type) were performed [2]. From leaching experiments with granite water it was shown, that the U/Al-fuel element corroded so slowly that no corrosion products were obtained. Calculated by the radionuclide content found in solution a time period of 10³ years is assumed for a complete dissolution of the fuel element.

In contrast high corrosion rates of the U/Al-fuel element were obtained in salt brines, especially in magnesium chloride rich brines. The addition of cast iron (basic material of the fuel element container) had an acceleration effect. A complete dissolution of the fuel was observed within a couple of months. Under aerobic as well as under anaerobic conditions the bulk of released radionuclides were trapped by the formed corrosion products (secondary phases). As a crystalline phase component of the secondary phases a Mg-Al-Cl-hydrotalcite was identified [3].

As a results of these data new work packages were established in November 2003 and they can be summarised as follows:

- Topic 1: Identification of parameters which determine the remobilization rates of the trapped radionuclides.
- Topic 2: Syntheses of a Mg-Al-Cl-hydrotalcite and subsequent sorption experiments with repository relevant radionuclides

Topic 3: Leaching experiments with non-irradiated and irradiated U/Al- and U₃Si₂-fuel elements in repository relevant aquatic phases

First results from the work packages specified are presented subsequently.

2. Experimental set-up

The experiments performed to determine the remobilization rates of trapped radionuclides were performed in hot cells. Corrosion products used for these studies were obtained by complete corrosion of an irradiated U/Al-fuel element plate (40 x 20 x 1.36 mm) in MgCl₂-rich brine in the presence of iron under anaerobic conditions at 90 °C. To study dilution effects aliquots were treated with MgCl₂-rich brine. The influence of the ionic strength was studied by the use of different concentrated MgCl₂-brines. The fraction of inventory was determined by complete dissolution of some aliquots in 8 M HNO₃ solution. All samples obtained were analysed radiometrically.

The experimental set-up for the syntheses of the Mg-Al-Cl-hydroxalite is described in [4]. The radioactive iodine and technetate solutions used were prepared from standard stock solutions. Sorption experiments were performed according to the batch technique. All samples were stored in glass tubes with occasional shaking under argon atmosphere for two days. Then all samples were filtered (450 nm) and the pH was measured. Aliquots of the solutions were analysed radiometrically. The solids were washed, dried and analysed by XRD and FT-IR. Blank experiments were performed too, indicating that the adsorption of I⁻ and TcO₄⁻ on the glass walls was negligible.

Leaching experiments with the non-irradiated U/Al- and U₃Si₂-fuel elements were performed in glass autoclaves at 90°C in 400 ml of MgCl₂ brine solution or in clay pore water (Mont Terri type) in the presence of 10 g FeCl₂ under anaerobic conditions. The dimension of the U/Al plate was 28 x 20 x 1.5 mm with an amount of 1.20 g U total (U-235-enrichment: 89%). The dimension of the U₃Si₂ plate was 40 x 20 x 1.36 mm with an amount of 1.44 g U total (U-235-enrichment: 0.21%).

3. Results and discussion

Topic 1:

As it was pointed out the U/Al-fuel element corroded in MgCl₂-rich brine. The radionuclides were mobilised first and then trapped by the formed corrosion products. In view of the long disposal

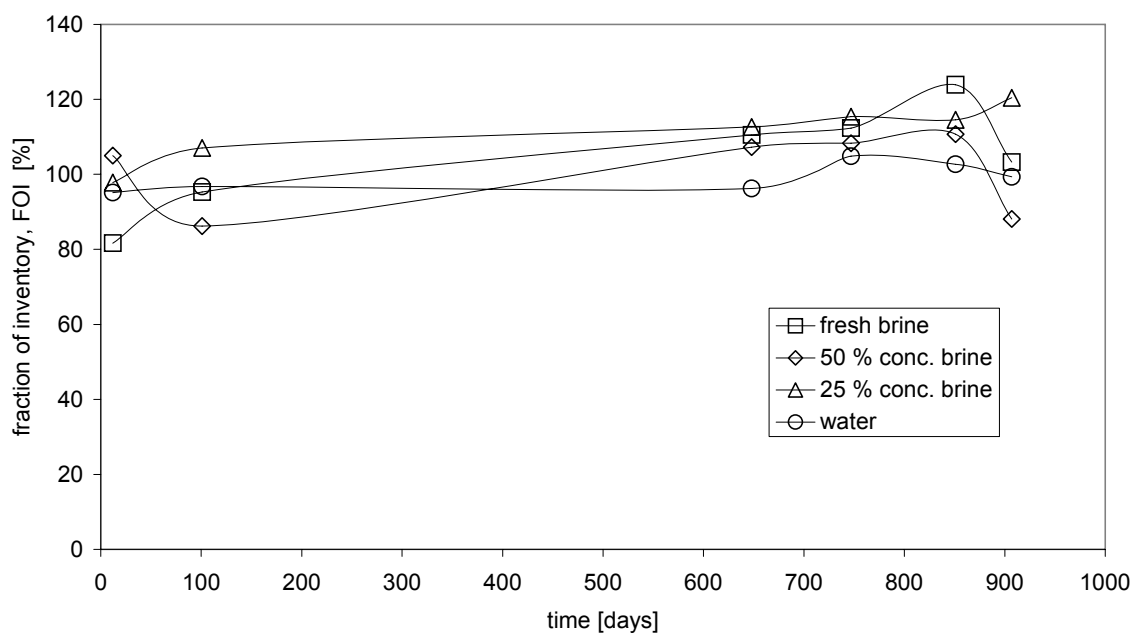


Fig.1 Remobilization rates of Cs in different concentrated salt brines and in water at 90°C

time the behaviour of the radionuclides must be investigated under various geochemical conditions. Therefore the obtained secondary phases were aliquoted and then treated with “fresh magnesium chloride brine” to study the effect of dilution. Other aliquots were treated with different concentrated magnesium chloride brines to study the effect of ionic strength. As it is shown in Fig. 1 the radionuclide Cs is released quantitatively and the release does not depend on the ionic strength. The same results were obtained for strontium. On the other hand Pu, a long-lived actinide, which contributes strongly to the long-term dose rate of the spent fuel waste, is not affected at all. This different behaviour can be explained by the different chemistry of Cs and Pu. Cs can only act as a monovalent cation and has a poor complex chemistry. Pu can be present in different valent states and it has a wide complex chemistry.

Topic 2:

The secondary phases may have a significant influence on radionuclide retardation. Therefore it was of interest to identify these secondary phases. Besides amorphous parts crystalline phases like bischofite, goethite and Mg-Al-Cl-hydrotalcites were identified. Especially the Mg-Al-Cl-hydrotalcite is of great interest. This substance also referred as anionic clay material is able to fix anionic species as well as cationic ones. To study the sorption effects with repository relevant long-lived radionuclides we first synthesised this Mg-Al-Cl-hydrotalcite and determined all analytical data. From these results the formula of the Mg-Al-Cl-hydrotalcite could be derived as: $\text{Mg}_3\text{Al}(\text{OH})_8\text{Cl}_{0.88}(\text{CO}_3^{2-})_{0.063} \cdot 2.4 \cdot \text{H}_2\text{O}$. Starting sorption experiments we first tested the ability to retard anionic species like iodine and technetate. For the Mg-Al-Cl-LDH (molecular weight: 314 g/mole) we calculated an anion exchange capacity of 2.8 E-03 mole/g (we did not consider the amount of carbonate ions present in the interlayer) for monovalent anions, which corresponds to 2.8 meq/g. That means, that for the used Mg-Al-Cl-LDH at most 2.8 E-03 mole/g of chloride in the interlayer can be exchanged by other monovalent anions.

In kinetic studies iodine adsorption onto the Mg-Al-Cl-hydrotalcite reached equilibrium within 2 days and the adsorption rate reached 50%. The same experiments performed in 0.01 M MgCl_2 solution led to a decrease in adsorption down to 30 %. Moreover in a 0.1 M MgCl_2 -solution no adsorption took place. These results can be explained by the effect of competition anion. In our case chloride is the competition anion. If a high chloride concentration is given, no exchange of iodine takes place because, compared to the iodine ion, the chloride ion has the higher affinity towards the Mg-Al-Cl-hydrotalcite. The sorption of technetate as function of time was studied too. Technetate behaved like iodine and again it was proven that chloride has the highest affinity towards the Mg-Al-Cl-hydrotalcite.

The sorption of technetate and iodine was investigated as function of pH as well, and it was found that there is no dependency on pH in the used pH-range between 3.5 and 8. Then the adsorption isotherms were obtained by plotting the amount of metal ions adsorbed on Mg-Al-Cl-LDH, Cads against metal concentration in solution at equilibrium, Ce.

The adsorption data were fitted to the Dubinin-Radushkevich (D-R) equation [5].

The mean energy of sorption is the free energy change when one mole of ion is transferred to the surface of the solid from infinity in the solution. The isotherms did fit the D-R-equation well as presented for iodine in Fig. 2. The value of the mean energy of the sorption in 0.01 M MgCl_2 solution was found to be about 8 kJ/mole for iodine and for technetate and this is in the range for ion exchange reactions, i.e., 8-16 kJ/mole. The sorption capacity found by the D-R isotherm in 0.01 M MgCl_2 solution was determined to be 1.97E-04 mole/g for iodine and 5.53E-06 mole/g for technetate. Compared to the calculated maximum anion exchange capacity of 2.8 E-03 mole/g these results predict that it is possible to exchange 7 % of the interlayer chloride by iodine in 0.01 M MgCl_2 solution. In the case of technetate only 0.2% of the interlayer chloride can be exchanged.

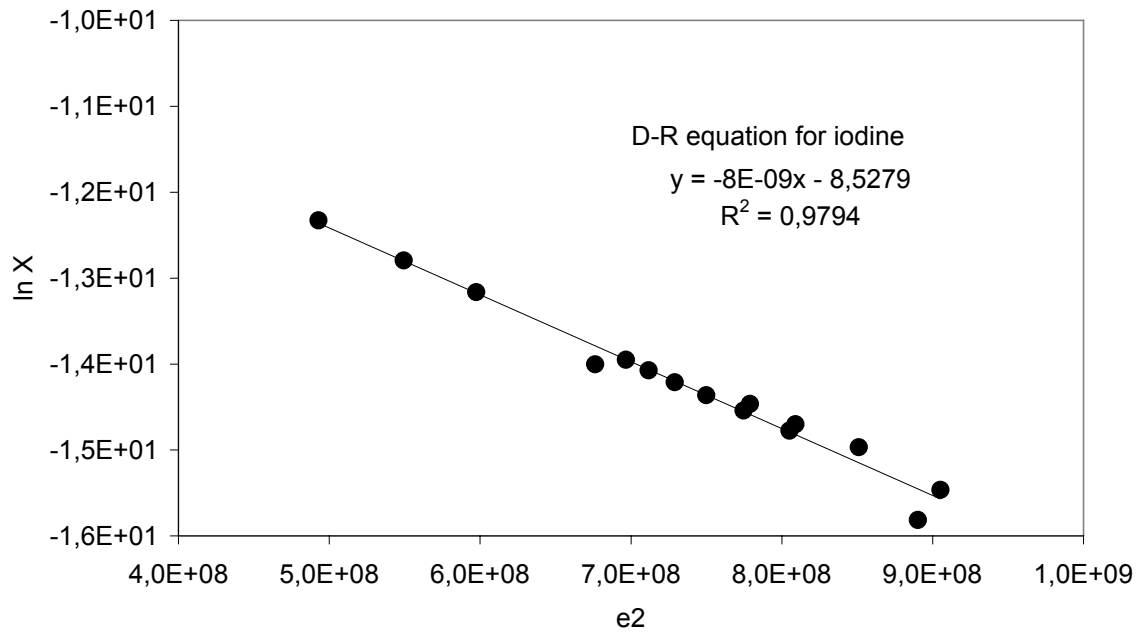


Fig.2 D-R Equation for iodine in 0.01 M MgCl₂ solution

Topic 3:

In the third work package the corrosion behaviour of U/Al- and U₃Si₂-fuel elements in repository relevant solutions should be investigated. First experiments started with non-irradiated U₃Si₂-fuel elements and with non-irradiated metallic U/Al-fuel elements in MgCl₂-rich brine and in clay pore water (Mont Terri-type). The H₂ formation can be regarded as indicator for the progress of the corrosion (Fig. 3). After 100 days, the H₂ production of the corroded U₃Si₂-fuel plate reached the same value in salt brine as in clay pore water. Nevertheless in salt brine a higher corrosion rate is observed in the first 40 days. Then equilibrium is reached whereas in clay pore water no equilibrium is reached yet.

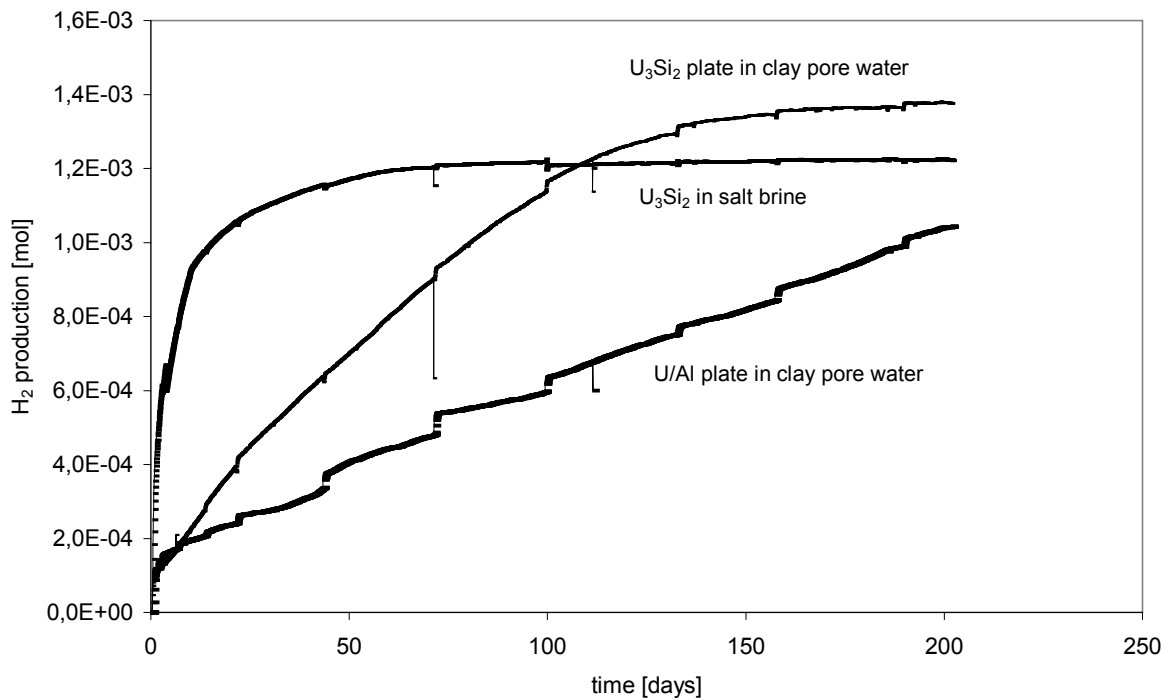


Fig. 3 H₂-pressure development in clay pore water (Mont-Terri type) and in MgCl₂-brine

The corrosion behaviour of the U/Al-fuel element in clay pore water is similar to the corrosion behaviour of the U_3Si_2 -fuel element but with half of the intensity.

4. Conclusions and outlook

From the first results of the present work the following main points can be summarised:

- Secondary phases, formed by corrosion of an irradiated U/Al-fuel element in salt brine, were separated and treated with different concentrated salt brines and water. It was found that Cs and Sr were quantitatively released by dilution. It should be pointed out that in view of long-term safety aspects these mobile radionuclides will not be retarded by the secondary phases when the geochemical milieu will change. The actinide Pu is not affected yet.
- One crystalline phase component of the corrosion products, a Mg-Al-Cl-hydrotalcite was synthesised and it was shown that iodine and technetate were retained by means of ion exchange reactions. Nevertheless it should be kept in mind that in the presence of high chloride concentrations no exchange reaction would take place, because chloride as competition anion has the higher affinity towards the Mg-Al-Cl-hydrotalcite.
- A fast corrosion of the non-irradiated U_3Si_2 -fuel element in salt brine and in clay pore water (Mont Terri-type) was observed.
- Compared to the U_3Si_2 -fuel element the corrosion of the non-irradiated U/Al-fuel element in clay pore water (Mont Terri-type) occurred with half of the intensity.

In our future work the remobilization rates of other long-lived radionuclides will be investigated. The sorption experiments with the Mg-Al-Cl-hydrotalcite will be enlarged to other repository relevant radionuclides. The experiments performed with the non-irradiated fuel elements will mainly be used to characterise the formed secondary phases. Then leaching experiments with irradiated fuel elements are planned in hot cell facilities. Here the distribution between the radionuclides in solution and in the formed corrosion products is of interest.

5. Acknowledgment

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REPROCESSING U-Mo SPENT FUELS: DISSOLUTION EXPERIMENTS ON NON-IRRADIATED AND IRRADIATED MATERIALS

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ABSTRACT

U-Mo fuels, containing up to 10 mass% of molybdenum, are one of the most studied low enriched uranium fuels (less than 20 mass% of ^{235}U). Although their reprocessability is ascertained, more precise reprocessing conditions are now under studies. The process would consist at first in a specific dissolution in nitric acid media. The obtained solution could then be diluted into standard UOx type fuels dissolution solution. Uranium and plutonium could be selectively recovered from this feed solution using adapted PUREX process (liquid/liquid extraction using tributyl phosphate).

The first step of this process have been extensively studied :

- dissolution tests in nitric acid media have first been conducted on non-irradiated U-Mo powder to define the optimal experimental conditions ;
- then dissolution kinetics have been studied on non-irradiated pieces of U-Mo fuels according to the previously defined conditions. Solubility limits of the different elements (especially molybdenum) have also been explored ;
- at last, dissolution tests on pieces of U-Mo spent fuels (irradiated inside Osiris French reactor) have been carried out to check the previously defined dissolution conditions and compare the behaviour of the irradiated and non-irradiated materials.

The following steps of the process : U and Pu purification from the U-Mo dissolution solution diluted into standard UOx type fuel dissolution solution, fission products solution concentration and waste management are still to be implemented.

1. Introduction

The definition of the U-Mo fuels reprocessing process is a part of the French qualification program [1]. The proposed scheme consists, first of all, in a dissolution in a nitric acid medium. The obtained solution is then diluted into standard UOx fuel dissolution solutions before treatment by the PUREX process. Despite the experience acquired by the reprocessing of U-Mo alloys containing from 0.5 to 1 weight % of molybdenum, it was necessary to obtain additional data in relation to the RTR fuels (composed of 7 % of Mo) and the chosen process. Therefore a complete R&D program, focussed on the dissolution step, was defined and conducted first on non-irradiated materials. The optimal dissolution conditions that were thus defined were then qualified with UMo plates irradiated inside the French Osiris reactor [2].

2. Studies on materials

The influence of Al, U and H⁺ concentrations on the solubility of Mo as well as the dissolution rate versus the temperature and the nitric acid concentration were first studied on UMo powder. Only the aluminium concentration was found to have a significant effect on molybdenum solubility [3]. In order to obtain a solution free of precipitate, the aluminium concentration should be less than or equal to 15 g/L while the UMo concentration is also 15 g/L. These results have led to the definition of the maximum amount of fuel by dissolution batch.

Afterwards, experiments carried out on pieces of fuel plates showed a complete dissolution and a meat dissolution rate ranging from 200 to 250 mg.cm⁻².h⁻¹ (at boiling temperature : about 107 °C), the acidity effect being insignificant [3].

3. Studies on irradiated materials

3.1 Description of the UMo spent fuel used

Among the three UMo fuel plates irradiated inside the French Osiris reactor (IRIS 1 experiment [2]), only the one containing 7 w% of molybdenum (Fig.1) was used for dissolution tests. Its physical characteristics before irradiation are described in Table 1 whereas the corresponding post-irradiation characteristics are detailed in Table 2.

	Plate characteristics
	Mo 7 w%
Length, mm	641.5
Width, mm	around 73.3
Thickness, mm	around 1.28
U mass, g	150.38
Mo mass, g	12.5
Al total mass, g	130.7

Tab 1. Initial physical characteristics of UMo fuel plate before irradiation.

	Post-irradiation characteristics
	Mo 7 w%
Reactor output date	01/22/201
Cooling time, year	2
Irradiation duration, day	268
M _{235U} , g	14.18
M _{□Pu} , g	0.929
MF, g	15.109
α activity, GBq	7.71
β γ activity, TBq	20.75
Residual power, W	2
Neutron flux, n/s/4π	534

Tab 2. Post irradiation characteristics of UMo fuel plate.

3.2. Dissolution tests

3.2.1 Description of the experiments

The following operating conditions, defined after studies on non-irradiated materials were applied :

- dissolution in nitric acid medium, at boiling temperature,
- initial $[H^+] = 5 \text{ M}$,
- initial $[Al] = 3 \text{ g/L}$ to take into account the aluminium contribution of the structural device supporting the plates in reactor,
- recombining NOx gases,
- targeted concentrations : $[Al] = 15 \text{ g/L}$, $[UMo] = 15 \text{ g/L}$,

First tests were conducted on small amounts of material (less than 5 cm long) in small vessels (Figure 1) whereas the last one, dedicated to validation of the operating conditions, was realized on one third of UMo plate (20 cm long) inside a five litre vessel (Figure 2).

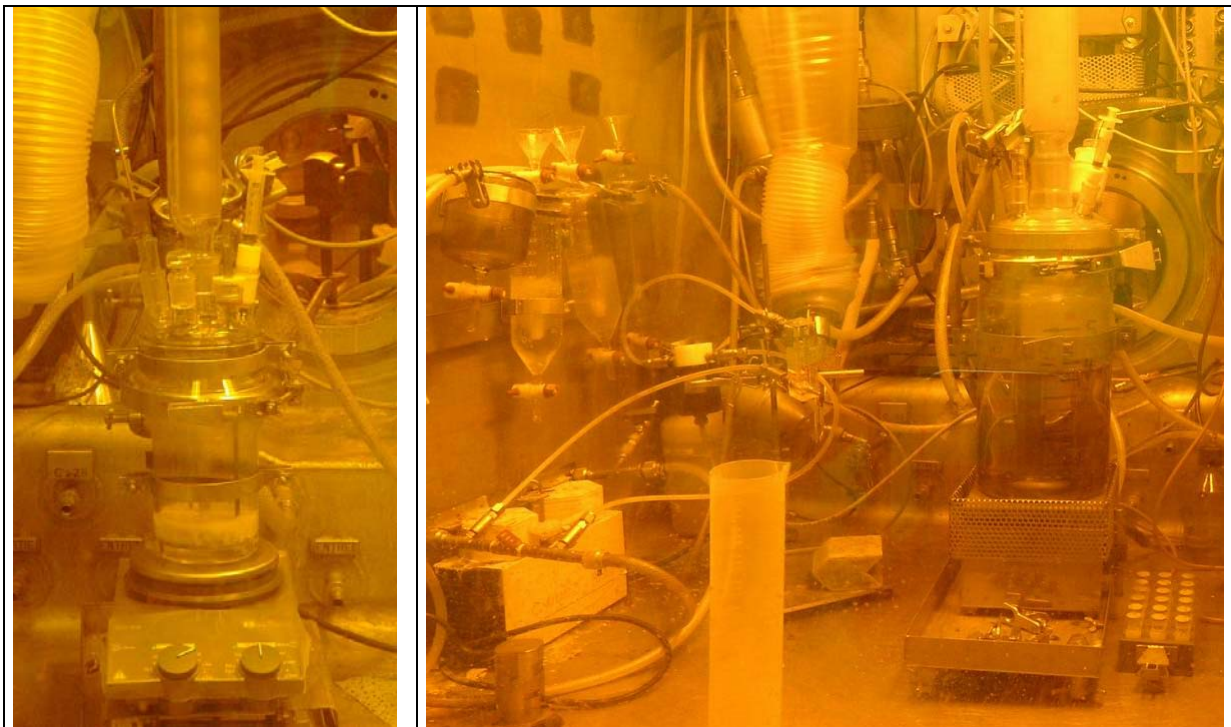


Fig 1. Small scale dissolution vessel Fig 2. Large scale dissolution apparatus

More over one small scale test was conducted on a cladding piece, sampled outside the core zone. Therefore, as there is no analytical interference between the aluminium coming from the cladding and the one from the core, the cladding dissolution kinetics determination was optimized.

3.2.2 Kinetics results

The dissolution kinetics obtained on irradiated materials (dissolution curves on Figures 3 and 4) can be compared to the one on fuel (Figure 5) : the obtained dissolution rates, both on core and cladding, are rather similar (Table 3).

Irradiation seems to have no increase on aluminium dissolution rate, contrary to previously observed phenomenon on thermally treated samples [3].

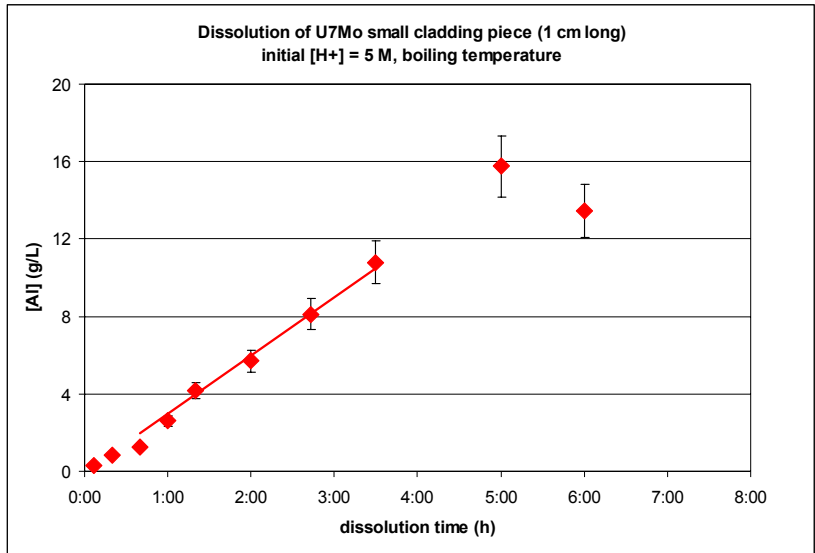


Fig 3. Dissolution kinetic of UMo spent fuel : only aluminium alloy cladding piece

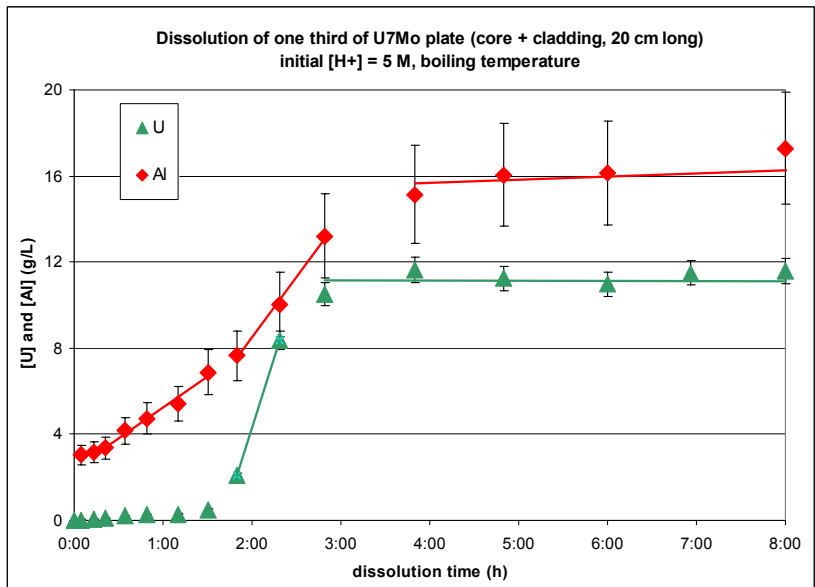


Fig 4. Dissolution kinetic of UMo spent fuel : core + cladding piece

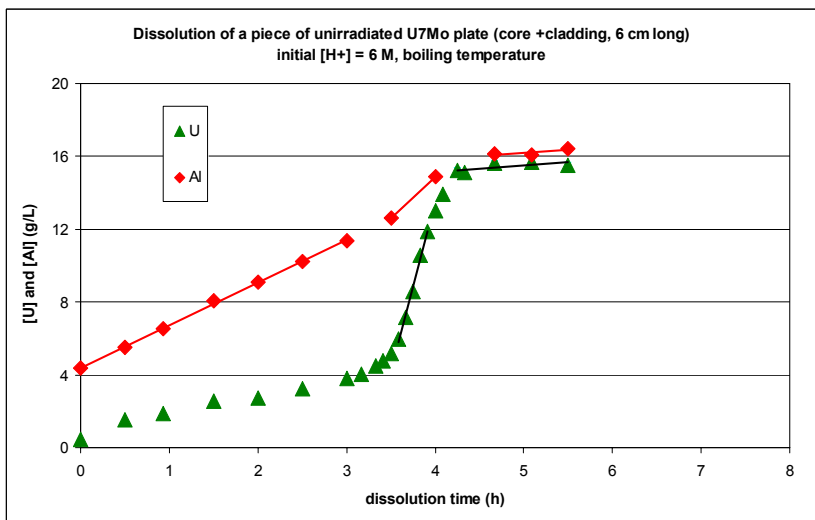


Fig 5. Dissolution kinetic of non-irradiated UMo plate : core + cladding piece

	Tests on non-irradiated materials			Tests on irradiated material	
	Core + cladding			Only cladding	Core + cladding
Temperature (°C)	Boiling temperature				
Initial [H ⁺] (N)	6.1	7.3	4.8	5	5
Final [H ⁺] (N)	4.1	5.2	2.8	-	2.8
Cladding aluminium dissolution rate (mg.cm ⁻² .h ⁻¹)	~30			32	34
Uranium dissolution rate (mg.cm ⁻² .h ⁻¹)	195	180	185	-	192
Meat dissolution rate (mg.cm ⁻² .h ⁻¹)	235	220	225	-	240

Tab 3. Comparison on dissolution kinetic on irradiated and non-irradiated materials.

Insoluble species and solutions aging

Insoluble species were especially studied during the last “validation” dissolution test conducted on one third of UMo plate. While dissolving at boiling temperature, the dissolution solution appeared to be rather clear. Then after one night cooling it became slightly cloudy. Filtration upon a 0.3 µm filter led to an amount of insoluble species representing less than 0.6 w% of the initial spent fuel introduced into the dissolution vessel. These insoluble species were found to be composed of more than 90 w% of molybdenum and aluminium.

The filtrated dissolution solution was then kept inside glass flask for months. While aging it showed no evolution and remained totally clear. New filtration after one month aging (upon 0.3 µm filter) showed no significant amount of insoluble species.

4. Conclusions

Although reprocessability of UMo spent fuel was ascertained, more precise reprocessing conditions were studied, mainly concerning the dissolution step. Experiments were carried out first on non-irradiated materials and then on UMo plates irradiated inside the French Osiris reactor. Provided the dissolution solutions are diluted enough to take into account the rather low solubility limit of molybdenum, clear and stable dissolution solutions can be obtained, with quite a little amount of insoluble species. Dissolution kinetics of non-irradiated and irradiated materials were found to be very similar : about 30 mg.cm⁻².h⁻¹ for cladding and 220-240 mg.cm⁻².h⁻¹ for the core.

The following steps of the process: U and Pu purification from the U-Mo dissolution solution diluted into standard UOx type fuel dissolution solution, fission products solution concentration and waste management are still to be implemented.

5. References

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HABOG: ONE BUILDING FOR ALL HIGH LEVEL WASTE AND SPENT FUEL IN THE NETHERLANDS. THE FIRST YEAR OF EXPERIENCE

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ABSTRACT

COVRA N.V. operates a site for the treatment and storage of all types of radioactive waste in the Netherlands. In 2003 the facility for storage of high level reprocessing waste and spent fuel from research reactors came into operation. The building is a piece of art. In a technical sense, as it was designed to withstand events such as earthquakes, flooding and aircraft crashes; but also in a literal sense, its provocative design was meant to evoke a discussion about the public aspects of radioactive waste management.

Construction took place from 1998 until 2002 and active operation at the end of 2003. The technical experience is positive. Nine MTR2 containers with spent fuel were treated and stored and 28 canisters with vitrified reprocessing waste were stored. The experience with the communicative function is positive also. Since the opening of the facility, the amount of visitors to the COVRA site more than doubled.

1. Introduction

COVRA N.V., the Central Organisation for Radioactive Waste, manages all radioactive waste produced in the Netherlands. Its statutory task is to execute the Dutch policy on the management of radioactive waste. This policy includes the long-term storage of all types of waste above ground for a period of at least a hundred years. COVRA treats and stores all categories of radioactive waste, including TENORM, spent fuel from research reactors and vitrified waste from reprocessing, on a specially designed site of 25 ha at the industrial area of Vlissingen-Oost, [1,2].

Direct disposal is not yet feasible in the Netherlands. Deep geologic disposal has not found general public acceptance and the small quantities involved prohibit an environmentally sound and economically feasible construction of the repository. However, the small volumes of waste do not require an immediate final solution. The small quantities of waste can easily be isolated from the environment by storing them for a long time in buildings. The natural decay during this period reduces the radioactivity, notably for the low and medium level waste. The long-term storage also buys time for the development of future international or regional disposal solutions, or radically new techniques to remove the hazardous constituents. Moreover, a capital growth fund has been established to use the available time to build up sufficient financial resources to soundly construct, operate and finally close a repository.

The choice for long-term storage above ground is not a “wait and see” option. This follows directly from the integral parts of the policy: the establishment of the capital growth fund and the transfer of the ownership of the waste to COVRA. This policy does not leave the burden of waste generated today to future generations. Only the execution of the disposal is left as a task for the future.

2. Safety of the design

For the long-term storage of high-level radioactive waste above ground, the treatment and storage building, the so-called HABOG (the Dutch acronym), was developed. Safety was a matter of great

concern in the design of the HABOG. The criteria for a safe design can be divided into two groups, relating to the source of possible disturbance of the safe situation: a) external influences, and b) internal influences.

External influences

All accidents with a frequency of occurrence larger than once per million years were taken into account:

- Earthquakes with a strength 6,5 on the Mercalli scale;
- Plane crash of a F16-A Falcon Fighter;
- Flooding resulting in a water level reaching over more than 10 metres above normal sea level for the Netherlands;
- Gas cloud explosions, caused by an accident of a passing ship loaded with liquid gas at a distance of a few miles;
- Severe windstorms with wind velocities of 125 m/s.

Obviously, the design criterion was that these accidents should not cause any radiological damage to the environment.

Internal influences

The design of the building should be such as the impact on the environment and the employees are ALARA. In the design high levels of radiation and heat production must be considered:

- High radiation levels:
- Protection by heavy construction: the wall thickness of the building is 1,7 metres of concrete;
- Remote control, and a hot cell for carrying out several operations such as overpacking of canisters, opening of canisters and decontamination;
- Back-up devices and continuous detection.
- Heat production:
- Cooling by natural ventilation.

Apart from significant spare store capacity, the long-term storage period also requires special measures relating to safety:

- Storage wells of austenitic stainless steel with molybdenum to reduce susceptibility to pitting corrosion, considering the elevated salt levels in the air in proximity of the sea;
- A (empty) third storage module to inspect the storage wells in the other two and if needed to remove and replace them. After the canisters have been relocated in the spare module, the well can be removed vertically and replaced by a new one.

3. Beauty of the design

The design of the building is based on safety and practical and economic use. Originally, the colour setting of the building was chosen to match the other buildings on the site: white and blue and some red accents. Figure 1 shows that the colour setting changed radically during the construction of the HABOG.

This radical change started with the idea to create a special wall painting in the storage hall for vitrified waste and spent fuel, which is part of the guided tours of the COVRA facilities. The special painting should evoke questions and discussion while the people are standing right above the high-level waste. In close collaboration with a local artist, this idea developed, and the painting grew into a complete building that is a piece of art in itself. The outside of the building is painted orange. The formulas of Einstein and Planck are painted in green, representing the process inside.



Fig 1. The HABOG

The building is to be repainted every 20 years in a slightly lighter shade so that after 100 years the colour of the outside walls is white. The fading of the outside colour reflects the decay of the heat production of the hottest waste inside.

Inside, photomontages of a beautiful, local landscape are more than virtual windows to the outside. The montages are located at four different places in the HABOG: the first in full colour, the subsequent ones each missing a base colour (see Fig 2). The beauty of the landscape shows the reason to build such facilities as the HABOG. The double horizon reflects that radioactive waste management requires looking further than a conventional time-horizon. The decay of the colour obviously reflects the decay of the radioactivity. The last black-and-white photograph is located in the storage hall for vitrified waste and spent fuel. It is portrayed on a layer of gold leaf to symbolise the value to society of the process in which the HABOG plays an integral part [3,4].

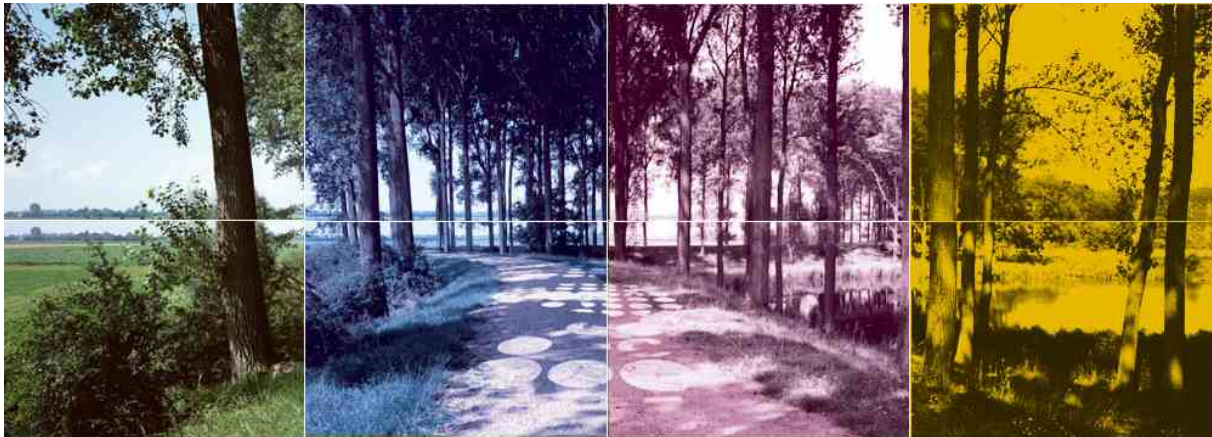


Fig 2. Metamorphosis: an artist's impression of radioactive waste management

4. Types of waste and capacity

In the design of HABOG, a distinction was made between heat and non-heat generating waste. The former requires cooling; for the latter a well-shielded area is sufficient. The canisters with heat generating waste are stored in vertical storage wells, which are cooled by natural (passive) ventilation. There is no contact between the circulating air and the canisters: temperature differences and draft drive the air through annular shaped stainless steel jackets placed around the wells (see Fig 3).

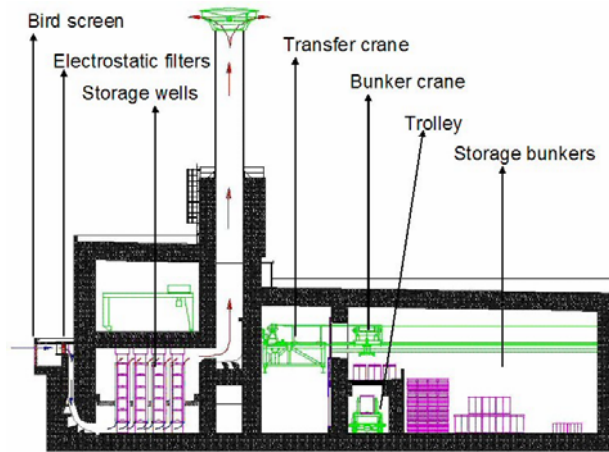


Fig 3. Cross-section of the HABOG building

Heat generating waste

Heat generating waste is produced by the two nuclear powers plants in the Netherlands: the Dodewaard plant (shut down in 1997) and the Borssele plant. Their spent fuel is reprocessed by respectively BNFL (Sellafield, Great Britain) and COGEMA (La Hague, France) and send to the HABOG as vitrified residues.

In addition, two research reactors in the Netherlands also produce high-level radioactive waste: one at the Delft University of Technology and a second in Petten. The latter is owned by the European Community (JRC), operated by NRG and mainly used for the production of molybdenum. The HABOG accepts the spent fuel from the research reactors without further treatment in a basket, which contains 33 fuel assemblies, welded tight in a canister.

Non-heat generating high level waste

A broad range of non-heat generating wastes can be stored in the HABOG:

- Drums with cemented hulls & ends from reprocessing spent fuel from COGEMA. This waste is to be returned now as compacted waste in CSD-C canisters;
- Drums with cemented technological waste from COGEMA. At this moment, the specification for compaction of this type of waste is not yet approved, but a return as compacted waste, mixed up with the hulls & ends, is foreseen;
- Bituminous waste from the reprocessing of spent fuel at COGEMA;
- Drums with cemented hulls & ends from the reprocessing of spent fuel at BNFL;
- Drums with high-level waste from research institutes, and from decommissioning of nuclear reactors.

Capacity

The dimensions of the HABOG are 90 by 45 by 20 metres (80.000 m³). The building consists of three vaults for storing heat generating waste and 3 bunkers for the storage of non-heat generating waste. According to the license COVRA has to be able to unload a complete vault or bunker for inspection of the vault or bunker (see also section 2). This means that only 2 of the 3 vaults and 2 of the 3 bunkers may be completely loaded.

The designed capacity of the HABOG is:

- 270 canisters with vitrified waste from reprocessing;
- 70 canisters, each containing 33 spent fuel elements;
- 600 drums with high level waste non-heat generating.

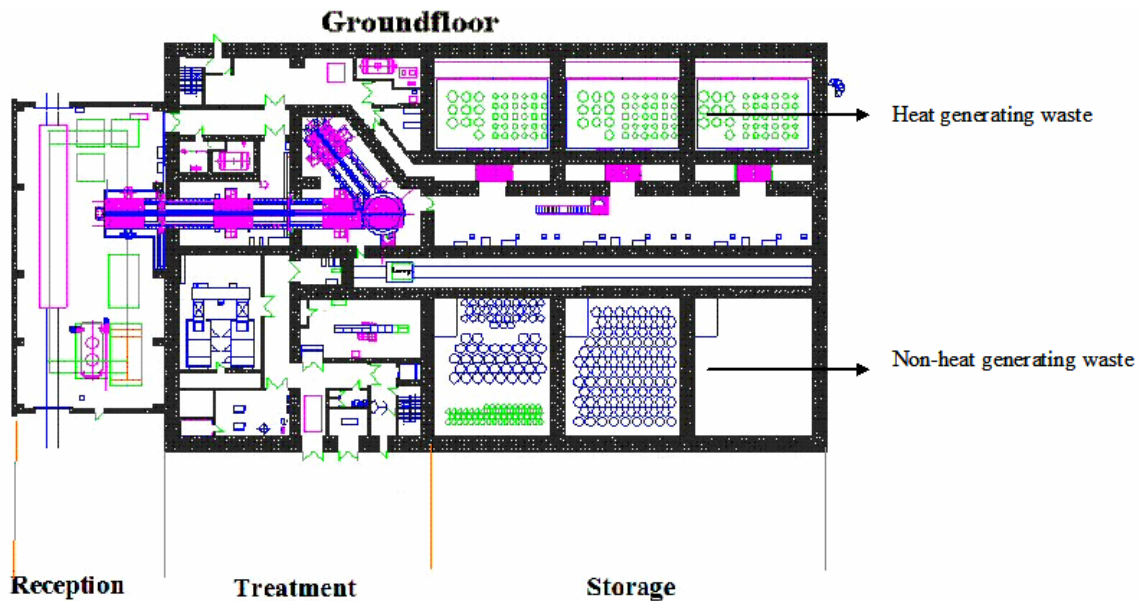


Fig 4. Ground floor of the HABOG building

This means a total storage capacity of 750 m³ of waste (corresponding to less than 1% of the total volume of the building) which will be shipped to the facility in 150 transports.

5. Construction

The licensing and construction of the HABOG took a long time, reflecting both the social and technical complexity of safe above ground storage of high-level radioactive wastes. The design of the building was carried out between 1987 and 1993. The licensing procedure took place between 1994 and 1999. After the engineering work initiated in 1999, the actual commissioning started in 2002 and ended with the official opening by HM Queen Beatrix in 2003.

The construction began with the preparation of the underground. In the south-western region of the Netherlands the soil in many places exists of soft layers and all buildings have to be piled. Due to the extreme design basis events (earthquake and aircraft crashes), piles are very complex and expensive. Piles are designed for vertical loads and not for horizontal loads generated by an aircraft crash or earthquake.

To save time and money, an easier foundation method was devised. The soil was excavated to 6 metres below ground level and replaced with high-grade sand for soil improvement. A pre-loading mountain 8 metres high was erected on top (equalling the exact weight of HABOG) to ensure compaction of deeper layers, and removed after six months. Settlement before, during and after was closely monitored.

After finishing the infrastructure, the first concrete was poured in 1999. The concrete was highly reinforced: on a total of 32.000 m³ concrete 5500 tons of reinforcement has been used. In addition, limestone-based concrete was used instead of the normal gravel-based concrete. Limestone improves the properties of the concrete: it contains no salt and leads to better shrinkage properties.

6. The first year of experience

Since the start of the active operation, in November 2003 one MTR2 container with spent fuel of the research reactors was transported, unloaded, packed and stored in the HABOG. In 2004, eight more MTR2 transports with spent research reactor fuel and one TN28 flask, containing 28 canisters with

vitrified residues from the reprocessing of spent fuel from the nuclear power plant Borssele were handled in HABOG.

The complete cycle for the treatment of a MTR2 transport container, loaded with 33 spent fuel elements from research reactors, takes about 5 days. For preparation, testing of the equipment and cleaning the installation another 5 days are needed, which makes in total 10 working days per campaign. Up to now, no contamination has been detected in the hotcell. Even the gripper, which takes the basket out of the flask, has not been contaminated.

A cycle for the treatment of a TN28 transport flask, loaded with 28 canisters with vitrified waste from reprocessing, takes about 15 working days including reception and return of the transport flask. The delivery and product documentation of the waste is checked by COVRA at the La Hague plant. After the control of documentation, the canisters are physically checked before loading into the flask. This check consists of a visual control, contamination and a dose-rate control. After reception of the canisters, the same controls are carried out during the unloading of the canisters at COVRA.

After the unloading of the first TN28 flask with vitrified waste, a dose rate of 5 microSievert/h was measured, due to scattering at the air inlet. In the design it was foreseen, based on calculations, that the expected dose rate would be 17 microSievert/h. The value could be penalising for the dose rate limit at the boundary of the site.

During office hours, four operators operate the facility. In addition to their operational tasks, they are also responsible for a significant part of the first-line maintenance, the equipment testing and the cleaning of the facility.

In the first year of operation only minor problems occurred:

- Some small problems in the software of control system, which have been fixed;
- Power supply rails of the trolley had to be replaced;
- The electrostatic filters that treat the incoming cooling air cause false alarms and are not operating to our satisfaction, due to moisture during fog and rain;
- The guiding of one of the heavy shielding doors is not correct. The guiding mechanism based on chains is replaced by one based on rollers, but the rollers cause some problems as well.

Experience with the communicative function of the building is also very positive. People are really intrigued by the facility. This results in the wish to come and visit the facilities. COVRA has always welcomed visitors, but now a complete new group of society shows interest. The art function is a good start for communication. Since the opening in 2003 the amount of visitors more than doubled as compared with earlier years.

7. The future

The storage of the high-level radioactive waste is foreseen for at least 100 years. The period is divided in an active and a passive operation phase. During the active operation phase, which in principle will end in 2015, the building is loaded. In the period between 2015 and 2100, the passive period, no activities occur in building. The central control room at the COVRA site does surveillance. After this passive period, it has to be decided to unload the storage facility and to bring the waste to a final repository or use new techniques to process and/or store the waste. The storage capacity and duration of the active period was based on the operational life spans of the nuclear power plants (Borssele and Dodewaard) and the research reactors in the Netherlands. Dodewaard was shut down in 1997 and the closure of Borssele was originally planned at the end of 2003. However, after construction of the HABOG, politics decided to keep Borssele in operation as long as safety and economics permit. This means that the storage capacity for reprocessing waste is likely not to be sufficient and need to be increased in the future. The modified HABOG should be ready in about 10 years from now.

8. Conclusions

In the Netherlands a multipurpose facility for high-level radioactive waste is in operation. Vitrified reprocessing waste and spent fuel from research reactors can safely be handled and stored for a period of at least 100 years.

The first year of experience shows that, the facility operates very well. Radiation levels inside and outside the building as well as the temperatures in the storage vault are as expected.

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SPENT FUEL FROM THE FINNISH TRIGA RESEARCH REACTOR IN THE SURROUNDINGS OF BWR SPENT FUEL FINAL DISPOSAL REPOSITORY. SAFETY ASSESSMENT AND COMPARISON TO THE RISKS OF THE BWR FUEL

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ABSTRACT

The Finnish Triga reactor, a 250 kW research reactor, has been in operation since 1962. According to the current operating license of our reactor we have to achieve a binding agreement between our Research Centre and the domestic Nuclear Power Companies about the possibility to use the Olkiluoto final disposal facility for our spent fuel. Naturally there is also the possibility to make an agreement with USDOE about the return of our spent fuel back to USA. In case of the domestic final disposal solution the main safety aspects, which have to be analyzed and compared to the spent fuel coming from the nuclear power plants, are the criticality safety, the solubility of the fuel ($UZrH_x$) to water and the existence of some moving and long-lived radioactive isotopes. The criticality safety calculations show that it is possible to load safely all the TRIGA fuel elements in one heavy final disposal canister. A simple safety analysis for the Triga fuel has been carried out in order to evaluate the long term risks of the final disposal. For the analysis a few scenarios from the TILA-99 safety assessment have been chosen. These scenarios will give a good picture of the potential risk of disposed Triga fuel compared to BWR fuel. TILA-99 safety assessment includes about 100 calculated different scenarios for the spent fuel so it's not reasonable to calculate them all for the Triga fuel. The main result is that the risks from the final disposal of Triga fuel are minor compared to BWR mainly due to smaller activity inventories.

1. Introduction

The FiR 1 reactor, a 250 kW Triga reactor, has been in operation since 1962. The main purpose to run the reactor has been lately the Boron Neutron Capture Therapy (BNCT). The BNCT work dominates the current utilization of the reactor: three or four days per week for BNCT purposes and the rest for other purposes such as the neutron activation analysis and isotope production.

According to our current operating license we have to achieve this year (2005) a binding agreement between VTT and the domestic Nuclear Power Plant Companies about the possibility to use the Olkiluoto final disposal facility for our spent fuel. In this case we can continue the operation of the reactor as long as there is reasonable work to do and the funding is in order [1]. Naturally we can also make an agreement with the USDOE. Especially after the announcement of the coming extension of ten years of the return policy the US alternative is nearly equal compared to the domestic one. For later negotiations aiming to the binding agreement about the domestic final disposal we are making safety studies about the long term behaviour of the spent TRIGA fuel in the final disposal surroundings. The main safety aspects, which have to be analyzed and compared to the spent fuel coming from nuclear power plants, are the criticality safety, the solubility of the fuel ($UZrH_x$) in water and the existence of some moving and long-lived radioactive isotopes.

For the final repository the spent fuel will be encapsulated in airtight copper canisters and situated in the bedrock at a depth of 500 m. The safety of this deep underground repository is based on multiple

natural and engineered barriers. Each canister contains 12 normal BWR fuel assemblies. The present concept for 127 Triga fuel elements is that the elements will be loaded in special containers, which have the same outer dimensions as the BWR fuel assemblies. Figure 1 describes the heavy final disposal canister.

The Triga fuel is much more reactive compared to the spent BWR fuel and therefore the Triga fuel can not be situated so tightly in the final disposal canister. The Triga fuel containers will be situated in the outer zone of the canister and the inner zone will be left empty. In practice the empty positions will be loaded with dummy assemblies made of cast iron. The criticality safety calculations show, however, that it is possible to load safely all the TRIGA fuel elements in one final disposal canister [2].

In this study the potential risks associated with final disposal of Triga fuel is in this study analysed on the basis of comprehensive safety assessment TILA-99 [3].



Posiva Oy



Posiva Oy

Fig 1. Final disposal canister

2. Fuel to be disposed off

The canister containing Triga fuel differs from Olkiluoto BWR fuel in several ways:

- The amount of uranium is only 1.3 % of the uranium contents of the BWR fuel from Olkiluoto.
- The amount of zirconium is 260 kg in the Triga canister compared to 1000 kg in a BWR canister.
- The Triga fuel in the canister is an alloy of uranium (27.5 kg), zirconium (260 kg) and hydrogen (~ 3 kg).
- The Triga canister contains also 45 kg of graphite reflectors at both ends of the rods.

Graphite is considered to be hazardous for copper in some circumstances. It doesn't cause any difference, however, compared to BWR fuel, when forming the scenarios for the safety analyses, as the copper canister is assumed to be damaged as a result of outside corrosion processes.

The inventories of radionuclides and stable isotopes are the most important differences between Triga and BWR fuels. The inventory of Triga fuel has been calculated with a 3-dimensional MCNP-model [4] and the inventory of Olkiluoto with ORIGEN-model [5]. The activity inventory of a canister containing 127 rods of Triga fuel (steel cladding assumed) has been compared to that of a canister containing 12 elements of Olkiluoto BWR fuel in Table 1.

Nuclide	Triga (Bq)	Olkiluoto (Bq)	Nuclide	Triga (Bq)	Olkiluoto (Bq)
H-3	$1.0 \cdot 10^{11}$	-	Np-237	$1.1 \cdot 10^8$	$1.2 \cdot 10^{11}$
C-14	$3.3 \cdot 10^8$	$7.3 \cdot 10^{10}$			
Cl-36	$4.4 \cdot 10^7$	$3.1 \cdot 10^9$	Pu-239	$2.8 \cdot 10^{11}$	$2.9 \cdot 10^{13}$
Se-79	$6.0 \cdot 10^8$	$3.5 \cdot 10^{10}$	Pu-240	$8.8 \cdot 10^{10}$	$4.1 \cdot 10^{13}$
Sr-90	$6.5 \cdot 10^{13}$	$2.9 \cdot 10^{15}$	Pu-242	$1.0 \cdot 10^7$	$1.7 \cdot 10^{11}$
Zr-93	$3.6 \cdot 10^9$	$2.1 \cdot 10^{11}$			
Tc-99	$2.0 \cdot 10^{10}$	$1.1 \cdot 10^{12}$	U-238	$3.5 \cdot 10^8$	$2.5 \cdot 10^{10}$
Pd-107	$2.9 \cdot 10^7$	$1.1 \cdot 10^{10}$	U-234	$1.1 \cdot 10^{10}$ *	$2.1 \cdot 10^{11}$
Sn-126	$5.5 \cdot 10^8$	$6.9 \cdot 10^{10}$			
I-129	$3.4 \cdot 10^7$	$2.8 \cdot 10^9$	U-235	$4.5 \cdot 10^8$	$2.4 \cdot 10^9$
Cs-135	$1.6 \cdot 10^9$	$5.0 \cdot 10^{10}$			
Cs-137	$6.9 \cdot 10^{10}$	$4.4 \cdot 10^{16}$			

Tab 1. Activity inventories of a canister containing Triga fuel (127 rods with steel cladding, burnup 35 MWd/kgU) and of a canister containing Olkiluoto BWR fuel (12 elements 2.14 tU, burnup 36 MWd/kgU). Inventories at the closure of the repository (30 yr cooling time); contribution of short lived are added to some daughters.
* U-234 inventory of Triga fuel is assumed to be 30 times that of U-238 which roughly coincides with the effect of isotopic enrichment.

It can be seen directly from Table 1 that the inventory of the Triga fuel is small compared to the Olkiluoto BWR inventory. Thus it's quite evident that the canister containing Triga fuel can't cause greater risk than the BWR fuel. There is one difference, the relatively higher inventory of U-235 (enrichment 20 %) and U-234 compared to U-238 in the Triga fuel. When applying the solubility limit of uranium (as it is always the case) the concentration of U-234 and U-235 in water inside the canister will be higher than in the case of BWR fuel. The amount of stable isotopes in the Triga fuel has been estimated from MCNP-model results separately. This data is important when applying the solubility limit inside the canister.

3. Scenarios to be analysed

The four scenarios to be analysed are from the TILA-99 safety assessment. The first two scenarios are the so called base cases:

- SH-sal50: Initially a small hole in the canister (5 mm^2), saline groundwater (present at Olkiluoto) and median flow rate of groundwater. Fuel matrix is assumed to degrade within about one million years.
- DC-ns50: Canister suddenly severely damaged after 10 000 years, non-saline ground water (due to land uplift) and median flow rate of groundwater. Fuel matrix is assumed to degrade within about one million years.

In order to find out the effect of some extreme conditions, the following two sensitivity analyses from TILA-99 have been chosen:

- DC-vhfsal: Canister suddenly severely damaged after 10 000 years, saline ground water and very high flow rate of groundwater. Fuel matrix is assumed to degrade within about one million years.
- DC-vhfsalficm: Canister suddenly severely damaged after 10 000 years, saline ground water and very high flow rate of groundwater. Fuel matrix is assumed to degrade evenly between 10 000...20 000 years. (ficm an acronym from "finish instant coffee model").

The comparison of the two very high flow and saline chemistry scenarios will reveal the effect of the differences of Triga and BWR fuel material. The other data (diffusion coefficients etc.) for calculations including instant release fraction from fuel (e.g. 6 % for iodine) are from TILA-99.

4. Results

The total dose rates and doses from three most important nuclides from Triga fuel and BWR fuel are presented in Table 2. The relations between doses from Triga fuel and BWR fuel are presented in Table 3.

	tmax (yr)	max (Sv/yr)	1st nuclide	(Sv/yr)	2nd nuclide	(Sv/yr)	3 rd nuclide	(Sv/yr)
Median flow scenarios								
SH-sal50 - Ol. BWR	$8.4 \cdot 10^5$	$1.7 \cdot 10^{-9}$	I-129	$1.3 \cdot 10^{-9}$	Cs-135	$3.6 \cdot 10^{-10}$	C-14	$1.3 \cdot 10^{-10}$
- Triga	$8.6 \cdot 10^5$	$3.7 \cdot 10^{-11}$	I-129	$1.6 \cdot 10^{-11}$	Sn-126	$1.3 \cdot 10^{-11}$	Cs-135	$1.1 \cdot 10^{-11}$
DC-ns50 - Ol. BWR	$1.1 \cdot 10^4$	$3.2 \cdot 10^{-8}$	I-129	$2.9 \cdot 10^{-8}$	Sn-126	$5.7 \cdot 10^{-9}$	C-14	$4.4 \cdot 10^{-9}$
- Triga	$1.1 \cdot 10^4$	$3.8 \cdot 10^{-10}$	I-129	$3.6 \cdot 10^{-10}$	Se-79	$7.1 \cdot 10^{-11}$	Pa-231	$4.8 \cdot 10^{-11}$
High flow saline chemistry scenarios (vhfsalficm; fuel degraded in 10 000 years)								
DC-vhfsal - Ol. BWR	$6.1 \cdot 10^5$	$2.7 \cdot 10^{-6}$	Ra-226	$2.7 \cdot 10^{-6}$	I-129	$2.1 \cdot 10^{-7}$	Pa-231	$4.2 \cdot 10^{-8}$
- Triga	$4.9 \cdot 10^5$	$1.3 \cdot 10^{-7}$	Ra-226	$1.2 \cdot 10^{-7}$	Pa-231	$7.6 \cdot 10^{-9}$	I-129	$2.5 \cdot 10^{-9}$
DC- vhfsalficm - Ol. BWR	$2.9 \cdot 10^5$	$7.1 \cdot 10^{-6}$	Ra-226	$7.0 \cdot 10^{-6}$	I-129	$2.3 \cdot 10^{-7}$	Pa-231	$4.2 \cdot 10^{-8}$
- Triga	$2.7 \cdot 10^5$	$3.6 \cdot 10^{-7}$	Ra-226	$3.5 \cdot 10^{-7}$	Pa-231	$7.7 \cdot 10^{-9}$	I-129	$2.8 \cdot 10^{-9}$

Tab 2. Maximum indicative dose rates and the most important nuclides

<i>Scenario</i>	Relation of doses BWR fuel/Triga fuel	Dose rate from Triga fuel (Sv/yr)
SH-sal50	46	$3.7 \cdot 10^{-11}$
DC-ns50	84	$3.8 \cdot 10^{-10}$
DC-vhfsal	21	$1.3 \cdot 10^{-7}$
DC-vhfsalficm	20	$3.6 \cdot 10^{-7}$

Tab 3. Relation between doses from Triga fuel and BWR fuel in the calculated four scenarios

From the Table 2 it can be seen, that in the base cases (SH-sal50 and DC-ns50), where I-129 is dominating the dose rate is proportional to the inventories in Table 1. Nuclide Pa-231 is more important in Triga fuel as the relative amount of parent nuclide U-235 is higher (enrichment 20 % in Triga fuel).

The most important and interesting result is that in the high flow scenario (DC-vhfsalficm), where the fuel is assumed to degrade within 10 000 years, the dose rate from the Triga fuel is $3.6 \cdot 10^{-7}$ Sv/yr. This rate is lower than the dose rate ($2.7 \cdot 10^{-6}$ Sv/yr) from Olkiluoto BWR fuel in DC-vhflowsal scenario, where the degradation of fuel is assumed to last about one million years. Thus the potential faster degradation of Triga fuel (UZrH_x alloy) is of minor importance.

5. Conclusions

According to the results presented in this work, the doses resulting from the final disposal canister of Triga fuel are lower than from the canister of the Olkiluoto BWR fuel and no safety problems have appeared during the study. The comparison has been made between one BWR fuel canister and the only Triga canister. In reality there will be more than 1000 BWR and PWR canisters in the final disposal repository and only one Triga fuel canister. The results are important for the negotiations between our Research Centre and the company Posiva Oy, which is taking care of the whole final disposal arrangements of the spent fuel from the Finnish nuclear power plants. Also the authorities are interested in the results, because they follow our research work connected to the decommissioning plans of our reactor.

6. References

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UPDATE ON NAC'S TRANSPORTATION PROJECTS

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ABSTRACT

The Global Threat Reduction Initiative (GTRI) was announced in early 2004 by then U.S. Secretary of Energy Spencer Abraham and was reinforced by an international meeting in Vienna in September 2004 of countries supporting non-proliferation efforts. Under GTRI, the Foreign Research Reactor Program (FRR) will continue and irradiated fuel shipments under the Russian Research Fuel Return Program (RRRFR) will begin. These programs will result in a projected increase in research reactor shipments in the coming years.

NAC owns and operates a fleet of eight NAC-LWT casks, which can transport up to 42 MTR fuel elements each. For the past 30 years, NAC has been supporting the Department of Energy with non-proliferation initiatives. Our teams have loaded casks in more than 25 foreign countries. Recent incidents of terrorism and ongoing threats have confirmed the necessity to ensure the security of high-enriched uranium and remove it from sensitive locations. Tremendous government efforts were initiated in 2004 in order to execute intra-governmental agreements for reactor conversion and material removal. As a result of these agreements, NAC will continue to provide its cask lease and transportation services to the Department of Energy. We expect to see very challenging shipments in the coming years. We are committed to these non-proliferation efforts as well as meeting our commercial customers' needs for transportation of irradiated fuel. The recent acquisition of NAC by USEC Inc. strengthens NAC's resources and joins NAC with a corporation with a strong non-proliferation orientation as the U.S. Government's Executive Agent under the highly successful U.S. - Russian Megatons to Megawatts program. This presentation summarizes our planned activities for the coming years with respect to GTRI initiatives.

1. Introduction

Last May the U.S. Secretary of Energy launched a comprehensive Global Threat Reduction Initiative (GTRI) to secure and remove high-risk nuclear and radiological materials that continue to pose a threat to the United States and the international community. In order to achieve this, the Secretary has directed the National Nuclear Security Administration (NNSA) to consolidate and accelerate the department's nuclear material removal efforts. NNSA's Office of Global Threat Reduction is responsible for major non-proliferation programs, including the Foreign Research Reactor Program (FRR) and the Russian Research Reactor Fuel Return Program (RRRFR). These programs involve a significant number of spent nuclear fuel shipments from research reactors around the world back to the originating countries – the United States and Russia.

A related effort applying to U.S. research reactors has been initiated to effect conversion of remaining HEU cores to LEU material, and to more expeditiously return spent fuel from on-site storage at research reactors to DOE storage sites.

NAC has been supporting the U.S. Department of Energy for the last 30 years in the removal of sensitive materials from various countries. In addition, NAC services U.S. commercial reactors by providing transport of small numbers of PWR and BWR pins for post irradiation examination as well as shipment of irradiated materials in connection with facility decommissioning. This paper will evaluate the potential demand for transportation services including support to these key programs:

2. FRR Program

Program Dynamics

In November 2004, the U.S. Secretary of Energy announced the extension of the FRR program until May 2019 for fuel irradiated prior to May 2016 expanding candidate fuel returns. Historically, the FRR program has executed three to four consolidated shipments per year averaging five to six casks per shipment. However, several reactors that have provided large fuel throughputs have or will shut down in the near term. In our judgment, beyond the two or three coming years, the combined effects of the extension of the program duration (allowing more flexibility in return date), the shutdown of major reactors, and the increased emphasis from GTRI will mostly cancel themselves but in total will result in a somewhat reduced demand yielding a reduced shipment rate or a similar rate with reduced casks per shipment.

Other Than High Income (OTHI) Country Perspective

The Secretary's GTRI initiative has placed added emphasis on achieving the objectives of the FRR program. As a result, discussions have resumed with many OTHI country reactor operators utilizing HEU cores regarding alternatives that might not previously have been available to them (i.e. funding for replacement of HEU cores with LEU fuel). If these efforts are successful and all the OTHI countries operating HEU reactors participate in the return program, a total of around 600 MTR fuel elements representing approximately 17 cask loads and 400 TRIGA fuel elements representing two to four casks loads would be shipped to the United States.

In addition, a few shipments from OTHI countries operating LEU fuel elements should be expected. However, the number of shipments is expected to be lower than historical levels as a large effort has already been completed during the first seven years of the program implementation, and a number of the participating reactors have ceased operation.

High Income (HI) Country Perspective

During the next three years a significant number of fuel elements from HI countries are scheduled for delivery at Savannah River Site: The European Commission has decided to return part of the U.S.-origin fuel to the United States from the Nuclear Research Group (NRG) in Petten, The Netherlands. One to two shipments should be completed in 2005. Several months ago, Studsvik's management in Sweden decided to shut down their reactor. Approximately 500 fuel elements remaining on the site will be returned to the United States in the next three years. Also, as the replacement OPAL reactor comes on line in Australia, ANSTO will ship the remaining irradiated fuel from HIFAR reactor (about 500 fuel elements) back to the United States. After this very active period, the yearly average number of shipments is expected to be lower than historical levels unless other reactors are shut down or revise their spent fuel management policy.

3. RRRFR Program

The Russian Research Reactor Fuel Return (RRRFR) is a cooperative effort of the United States, the Russian Federation and the International Atomic Energy (IAEA) to repatriate Russian fresh and spent nuclear fuel from research reactors operating with Russian origin fuel. Under this agreement close to 20 countries are eligible to receive financial and technical assistance from the United States and IAEA in order to ship their fresh and spent research reactor fuel to Russia for safe and secure management. While fresh HEU fuel shipments have been successfully performed, spent nuclear fuel shipments present a more complex technical challenge due to the nature of the fuel, casks and certifications needed, extended period since shipments were conducted and relative inexperience of participating reactor operators. After several years of planning, they are expected to start shortly.

The RRRFR is a very challenging program as about 15,000 fuel elements representing many different fuel designs are targeted to be returned to Russia by 2010. More than 80% of the fuel is enriched to greater than 20% Uranium-235. The candidate reactors are spread among three continents: Europe, Asia and Africa. The existing fleet of Russian SNF transportation casks (the TUK-19) for research

reactors has a very limited capacity of four fuel elements per cask. In order to achieve the goal to repatriate the 15,000 fuel elements by 2010, additional cask types will need to be committed to the program. This has resulted in a program to certify the NAC-LWT cask fleet for the various Russian fuel forms. The NAC-LWT was selected because of its capacity and very flexible loading system, combined with the fact that NAC-LWT Cask has been previously analyzed and licensed for Russian fuel forms during the shipment of Iraqi and Georgian spent fuel. Additionally, in connection with the Iraq campaign, a total of six NAC-LWT casks have been successfully unloaded at the Mayak facility. We believe that upon completion of the licensing effort, the capacity and adaptability of the NAC-LWT cask fleet can easily support two yearly shipment campaigns for the Russian Program while continuing supporting the FRR shipments.

Excluding the Vinca fuel in Serbia Montenegro, which represents approximately 8,000 fuel elements and which utilizes an atypical smaller size fuel element, the RRRFR Spent Fuel Program represents around 280 cask loads to be shipped to Mayak (assuming an average of 25 fuel elements per cask). Considering the extended sea and/or land shipment duration from the reactor sites to Mayak, program management will be extremely challenging. Selection of the most efficient routes and optimum casks for each facility depending of the type and number of fuel elements, facility infrastructure, shipment combination with casks originated from different countries and selection of transport mode will be key for the success of the Program.

2. NAC's Fleet/Equipment/Capacities/Capabilities relative to RRRFR utilization

NAC owns and operates a fleet of eight NAC-LWT casks capable of transporting up to 336 MTR type fuel elements within a single shipment. The NAC-LWT is licensed by the U.S. Nuclear Regulatory Commission (NRC) and also by the U.S. Department of Transportation (DOT) for international shipments. It has received validation by more than 20 foreign countries. The NAC-LWT cask is a very flexible cask licensed for various contents such as MTR and TRIGA fuel elements, PWR and BWR fuel assemblies or rods and other types of research fuel rods. It also has been analyzed for Russian IRT-2, EK-36, EK-10, and EK-NU, and Tammuz fuel assemblies.

Last January, a revised Certificate of Compliance was issued by the NRC to extend the NAC-LWT certificate for five additional years (until February 2010) and to update the certificate in compliance with TSR-1 (IAEA 96). While the adoption of the TSR-1 regulations by the U.S. NRC (implemented October 2004) allowed a three-year time period for compliance, NAC immediately upgraded the certification and will perform the first shipment under the -96 certificate this spring.

NAC also has a fleet of five NLI-1/2 casks than can be utilized for fuel shipments as specified in its existing certificate, or if authorized by special arrangement. The NLI-1/2 is the predecessor cask to the NAC-LWT and duplicates much of its loading flexibility, procedures, and capacity.

The unique loading flexibility of the NAC-LWT (an extensive complement of equipment has been designed and manufactured to facilitate fuel transfer in facilities with limited pool and handling infrastructure) permits expedient adaptation of the cask to facilities such as the RRRFR participants that may not have performed a spent fuel shipment in their lifetime. This loading flexibility is also a key element in meeting the requirements of the RRRFR program as equipment design to resolve cask-loading interfaces can result in a long and costly process.

NAC's 20 years of experience in international shipments of spent nuclear fuel by road, rail, sea and air, offer substantial benefit to the program's efficiency and safe execution.

4. Near Term Perspective (coming year)

NAC will continue substantial involvement in the return shipments of SNF under the FRR Program. The European Commission has awarded a contract to NAC to ship back to Savannah River Site up to

700 HEU fuel elements. A first shipment of five NAC-LWT casks from the Nuclear Research Group's (NRG) Petten reactor will take place this year.

In 2004, NAC received a contract from Reaktorinstitut Graz, Austria to ship the HEU and LEU Argonaut plates back to Savannah River Site using one NAC-LWT Cask. We are expecting to perform the shipment in 2005 in combination with another shipment from the Mediterranean Sea.

For the OTHI countries, NAC is the designated DOE transportation contractor. NAC provided technical support to the Department of Energy earlier this year to assess research reactor spent nuclear fuel transportation located in Europe and South America as these reactors are planning to utilize the opportunity to return their HEU fuel elements to the United States in the near future.

Another key element for spent nuclear fuel element transportation is the licensing activity. The casks are licensed for detailed content and any material deviation is subject to a license amendment. Obtaining a license amendment can be a very long process especially for international shipments as more than one Competent Authority is involved in the process. In 2005, NAC will support significant licensing efforts for the FRR and RRRFR Programs. NAC will prepare a license amendment to accommodate some Australian fuel elements from HIFAR with a design for a modified basket. Also, the NAC-LWT has previously been licensed to transport Russian Origin fuel types when NAC performed a shipment of Russian fuel from Iraq back to Mayak in 1993 and from Georgia in 1998. As mentioned above, NAC has been contracted to perform fuel analysis in order to submit a license amendment covering the Russian Origin fuel types and be ready to ship in 2006. Based on using existing basket designs, it is our current assessment that the NAC-LWT will be able to ship a minimum of 28 elements per cask loading and as many as 140 per cask.

Also, NAC has been and will continue supporting the IAEA to prepare for the shipment of irradiated fuel from the reactor of VINCA in Belgrade, Serbia Montenegro. Due to the limited infrastructure of the facility and the degradation of the fuel, the NAC-LWT is a good candidate for the return-shipment as the use of the NAC-LWT would limit re-canning and handling operations at the site. The NAC-LWT is licensed to transport highly degraded fuel due to its very low leak rate design.

5. Domestic U.S. Prospective

The DOE emphasis on shipment of U.S. university research reactor fuel resulted in five NAC-LWT cask shipments during 2004. A similar number is expected during the coming years as storage basins are emptied, older casks are retired, and additional domestic reactors are shut down. These shipments are performed by motor carrier and are of short duration relative to FRR and RRRFR durations. With proper planning, the shipments can be interspersed with the foreign shipment campaigns on a non-interference basis.

As mentioned above the NAC-LWT casks can accommodate PWR and BWR fuel assemblies and rods. In connection with fuel performance programs, NAC also expects to support a few shipments in 2005 from U.S. nuclear power plants to US and other foreign hot cells.

6. Looking ahead

Looking beyond the short-term, NAC will continue its involvement in the return of US origin fuel back to the US under the FRR Program. As described above, we expect that the two to three coming years will be very active with shipments from Europe, Australia and OTHI countries.

In addition, the RRRFR Program will start its implementation. The NAC-LWT should be used in support of the existing Russian casks and other potential casks as needed. At this stage of the Program, it is premature to provide schedule information. However, based on the time period foreseen for completion of the Program, NAC expects to be in a position to provide casks for at least two shipment campaigns per year.

7. Conclusion

With the continuation of the FRR Program and beginning of the RRRFR, we expect that the number of shipments from research reactors will generally increase. However, we see a good transition between the FRR Program and RRRFR Program in the two coming years as the number of shipments performed under the first one should decrease and shipments under the second one should grow.

NAC's existing fleet can perform four multi-cask international campaigns a year while still supporting a number of individual domestic US shipments. With effective integrated program management including careful planning, licensing anticipation, diligent allocation of trained man-power, regular communication with all the entities involved in the Programs, NAC's existing cask capacity will be sufficient to meet the demand.

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