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RAPHAEL PROJECT - HTR SPECIFIC WASTE CHARACTERIZATION PROGRAMME

M.H. Mouliney / **AREVA NC**

P. Bros / **CEA 30207 Bagnols/Cèze, France**

78141 Vélizy, France A. Sneyers / SCK CEN J. Fachinger / FZJ

D. Roudil / **CEA 30207 Bagnols/Cèze, France**

2400 MOL, Belgium

F. Cellier / **AREVA NP 69456 Lyon, France**

D-52425 Juelich, Germany

D. Millington / AMEC NNC WA168QZ Knutsford, UK

K. Vervondern / FZJ D-52425 Juelich, Germany

T.J. Abram / **NEXIA WA36AS Warrington, UK**

ABSTRACT

In the frame of the RAPHAEL European Project, the back end of the fuel cycle is addressed concerning mainly open cycles : direct disposal of spent fuel or of separated constituents (fuel matrix, particles, compacts or pebbles). One of the objectives is to obtain more precise experimental data allowing to predict the fuel behaviour for direct geological disposal conditions.

The global effort is shared between both bibliographic and experimental work including :

the establishment of an international comparison of contamination characterization levels and disposal specifications to propose a list of conditioning and/or decontamination requirements,

a compilation of existing data about HTR spent fuel characterization,

the acquisition of physical, chemical, radiological states and inventory data of irradiated and fresh fuels with LEU kernels.

The characterization programme will be carried out in the Atalante Facility (CEA) in France, using samples provided by Juelich center in Germany (FZJ). Both destructive and non destructive analysis will be performed with pebbles and coated particles. This paper describes in detail both characterization techniques and objectives and gives an overview about the equipments that will be used.

INTRODUCTION

The RAPHAEL project addresses the viability and performance issues of the modular Very High Temperature Reactor (VHTR). This innovative system is not only meant at competitive and safe electricity generation, but also at providing industrial process heat, in particular for hydrogen

production. It offers significant advantages in terms of safety (inherent safety features), environmental impact (robust fuel with no significant radioactive release), resource utilization (high efficiency and use of any fissile/fertile material), and cost effectiveness (due to the simplification arising from inherent safety). The technical part is structured in sub projects. One is dedicated to the back end of the fuel cycle and covers the present work.

Different basic options for VHTR spent fuel management are shown on figure 1 (annex A). Path "A" addresses an open cycle with direct disposal of spent fuel taking benefit from the ceramic encapsulation of the fission products (FP) and minor actinides (MA). However, this option leads to rather large volumes to be disposed, in a final repository. Due to the 'dilution' of the spent fuel in the fuel element, this type of waste can be assumed as medium-level waste with limited heat generation.

Significant volume reduction could be achieved by separating the fuel graphite matrix from the fuel compacts or from the coated particles (path B). The two fractions can then be treated differently because the graphite may be treated as low-level waste. The spent fuel needs special conditioning or back-fill material before being disposed as high-level waste. Despite the disintegration of the fuel element, this path still represents an open cycle. Both options (A&B) need precise knowledge on the leaching behaviour of the matrix – even if disposed separately – and of the multi-barrier coated particle fuel. Thus, the general objective is to obtain more precise experimental data allowing to predict the fuel behaviour for direct geological disposal conditions.

Finally, a third option (path C) will allow to close the fuel cycle by reprocessing spent HTR fuel. The work focuses only on open cycles.

DISPOSAL SPECIFICATIONS AND REQUIREMENTS

The waste management routes depend strongly on the national context and classification of the discharged spent fuel. Comparisons has been made with the situation in France, UK, Germany, Belgium and US to identify converging denominators for future specifications, requirements and handling of HTR spent fuel.

Identification of common denominators :

A comparison of the radioactive waste classification schemes and disposal routes reveals many converging criteria, but also some significant areas of difference between the national schemes. The schemes are summarized and compared in Table 1 (Annex C).

Among the EU states, the Belgian and French schemes are very similar and are closely related to the EU classification scheme, which is in itself based upon the generic IAEA recommendations. These schemes formally recognize the lifetimes of the predominant radionuclides within waste packages, and segregate low and intermediate level waste into short-lived and long-lived categories, on the basis of whether the half-lives of these nuclides are less-than or greater-than 30 years respectively. These correspond to the EU LILW-SL and LILW-LL categories, and are linked to the planned disposal routes. EU LILW-SL, Belgian Category A waste, and French short-lived LLW and ILW are all suited to surface disposal, whereas EU LILW-LL, Belgian Category B waste, and French HLW and long-lived ILW will require geological disposal.

The Belgian scheme formally quantifies the division between short-lived (Category A) and long-lived (Category B) waste on the basis of a concentration criterion which identifies maximum volumetric activity concentrations for a set of key long-lived nuclides. A second criterion which assesses waste activity against the total radiological capacity of the disposal site is also applied.

The French scheme distinguishes between low-level and intermediate level long-lived waste, and places graphite waste into the former of these categories, for which a dedicated disposal facility is planned. The lower limit for High Level Waste is defined on the basis of volumetric decay power generation in the Belgian scheme, which requires waste with decay power densities in excess of 20 $W/m³$ to be classified as Category C waste. This is roughly equivalent to the activity-based limit in the French scheme of 10^8 Bq/g.

The French and Belgian schemes, and the EU scheme upon which they are based therefore classify waste according to the planned disposal route for package waste items, and make a direct link between the properties of the waste package and the acceptance criteria of the disposal route.

The UK classification scheme is not directly related to the disposal route, and does not address the half-lives of the constituent radionuclides in the same manner as the other EU schemes reviewed. An activity criteria is used to categorize wastes as low-level waste and intermediate level waste whereas the EU scheme uses a lifetime criteria to divide these wastes into short-lived (LILW-SL) and (LILW-LL). The EU scheme, and those of Belgium and France are based upon packaged waste items, whereas the UK scheme classifies waste on the basis of its raw characteristics prior to packaging and without reference to the disposal route. On this basis, additional site-based conditions for acceptance are enforced at the UK surface disposal facility at Drigg, and low/intermediate level graphite waste from conditioned HTR fuel would have to be assessed against these limits. There is no disposal route for ILW in the UK, although there are activities in progress to produce a short-list of options and recommendations for further study and investigation, so it is not possible to anticipate the acceptance criteria for any eventual facility.

The US commercial radioactive waste classification system has four categories of waste recognized by the US DOE, and six levels adopted for the classification of commercial waste, which can to some extent be mapped onto the IAEA system. In a similar manner to the UK scheme, the US scheme categorizes waste on the basis of its origin and characteristics.

The German classification for disposal is not in accordance to the other EU states because Germany has decided to dispose all kind of radioactive waste in deep geological repositories. Therefore no distinction is required between low-level, medium-level and high-level waste as well as between long-lived and short-lived radionuclides (however such a classification is still used by the waste producers due to practical reasons for handling). A basic classification is made into heat generating and negligible heat generating waste. Thereby negligible heat generation stands for wastes, which have a negligible thermal effect upon the surrounding host rock. This is defined as a temperature increase below 3K in case of the Konrad mine, which is proposed as repository for negligible heat generating waste. Finally acceptance criteria inclusive maximum disposable nuclide inventories will be derived from disposal site-specific safety analysis, which lead to requirements with respect to the waste packages and waste forms. Especially the maximum disposable nuclide inventory for ¹⁴C causes problems for the ceramic waste arising from dismantling of the two German high temperature reactors AVR and THTR, because the 14 C inventory of these ceramics would utilize nearly the whole amount of ¹⁴C licensed for the Konrad repository.

Furthermore spent fuel is excluded from the group of negligible heat generating wastes, even if the spent fuel would meet this criterion after an extended cooling period. Therefore all spent fuel obtained during operation of AVR and THTR are designated for a repository considered for heat generating waste.

Having made a comparative review of several national classification systems, it is apparent that the EU scheme, which is itself an implementation of IAEA recommendations, forms a useful basis for discussing HTR spent fuel disposal within the context of the RAPHAEL-BF Sub-project, as it directly relates packaged waste characteristics to those of the disposal route. Whilst the UK scheme does not preempt the disposal route, and relies on additional site-based conditions for acceptance, it is less useful for providing a framework for discussing the generic disposal routes for waste arising from HTRs.

Specifications for disposal of future HTR waste :

As previously stated, EU States will have to report national waste inventories using the EU classification scheme, so it is proposed to discuss and define HTR waste management principles in the context of this scheme. This can then be mapped onto the individual national schemes as required.

The review performed in this document has confirmed that disposal routes for waste containing radionuclides with half-lives in excess of 30 years, and for waste with significant decay heat generation, corresponding to EU categories LILW-LL and HLW respectively, are not well established in some EU states eg UK, Germany but likely to converge on the same common denominators for geological disposal. The US scheme differs from the EU scheme, but can be mapped onto the IAEA scheme, and is partly based around the identification of Yucca Mountain as a final disposal site.

Untreated spent HTR fuel from a commercial power reactor will contain high initial concentrations of long-lived radionuclides and high levels of decay heat output and will therefore be classified as HLW in the EU scheme, and direct disposal will be the only disposition route available. A detailed study of the engineered barrier systems, beginning with the TRISO coating containing the fission products, and extending to the graphite matrix and the packaging container will be required, alongside that of the repository design, in order to determine acceptance criteria for spent HTR fuel.

If HTR fuel is reprocessed, it will be separated into constituent components, which will comprise block graphite and compacts, depending on the back-end treatment path followed. The disposal strategy that can be adopted for these constituents is imposed by the radiological characteristics and thermal output of the conditioned waste, and so these characteristics must be determined in more detail by the RAPHAEL-BF programme. The waste packages arising can be disposed in a surface repository only on the condition that concerned waste packages complies with the criteria equivalent to those for EU LILW-SL. For conditioned reprocessed HTR waste which belongs to EU LILW-LL or HLW, geological disposal is the only available strategy.

BIBLIOGRAPHIC SURVEY ON EUROPEAN DATA ON HTR SPENT FUEL CHARACTERISATION

Data have been collected at an European level. The information essentially came from the results already obtained in Germany, France, UK.

Data in Germany :

In Germany, two high temperature reactors were operated, the AVR test reactor in Juelich and the prototype reactor THTR – 300 in Hamm-Uentrop. Both were shut down but considerable experience has been gained during their operating period.

The AVR reactor which was operated over 21 yrs until 1988 served particularly the purpose to test and qualify HTGR pebble fuel. In the course of operation, more than 290,000 spherical fuel elements of 15 different types (carbide/oxide, BISO/TRISO, HEU/LEU) with more than 6 billion coated fuel particles were inserted into the core. Also the fuel element design changed soon from machined graphite shells to pressed matrix material.

The continuous measurements of coolant activities over the years have shown the steady improvement of fuel quality. The fraction of heavy metal contamination as the major contributor decreased from formerly 10^{-3} for the BISO fuel to the level of 10^{-5} for the modern TRISO fuel. On the other hand, the activity measurements also allowed the identification of failing coated particles and, in combination with comprehensive PIE of regularly and randomly discharged fuel element specimens, the identification of poorquality fuel charges. Mechanical damages observed included the so-called peeling effect, which could be traced back to an inadvertent air ingress event in 1971 which led to oxidation reactions. The number of broken fuel remained with 220 balls very low, most of them were of the machined shell type of the first core and the first reload showing a strong decrease in strength with increasing fast neutron fluence.

Spherical fuel elements and coated particles, respectively, were also - and still are - investigated with respect to the conditions of disposal in salt mines. An excellent long-term chemical resistance was found for intact coated particles, whereas activity release from leaching in a brine is only possible from particles with defective coatings. Works also included the investigation of temperatures and gamma-activities of storage casks filled with high-burn up AVR fuel.

In contrast, the THTR-300 was operated over 423 efpd based on one type of fuel with high-enriched uranium and BISO coated particles. The fuel was basically qualified in the AVR completed by irradiation tests in the Studsvik (R2) and Dragon reactor. Unlike AVR, the THTR operation was not designed for discharging fuel elements for the purpose of dedicated PIE.

All fuel from both AVR and THTR has been stored in CASTOR casks (approx. 2000 balls each) and given to interim storage sites.

Data in France:

In France, R&D programmes on HTR has been conducted in collaboration with foreign partners (General Atomics, Dragon project, FZJ – Juelich). Irradiation experiments were carried out in many test reactors : OSIRIS (Saclay), SILOE and SILOETTE (Grenoble), PEGASE, PEGGY, MARIUS, CESAR, RAPSODIE (Cadarache). The main aims were to study :

- the particle, compact or fuel block behaviour under irradiation,
- the fission products migration and deposition in case of particle failure,
- specific properties (graphite creep;…).

In major cases, FP migration in coated particles has been studied not for waste management but to get data about potential contamination of cooling gas in both normal operating and accident conditions.

It has been observed, about gaseous FP behaviour that :

- after emission in the kernel, gases go easily through the buffer,
- IpyC and SiC coatings retain gas to a temperature close to 1500°C,
- In the case of particles failure, gas migration into compact is fast.

About non gaseous FP :

- For metallic FP, IpyC only delays the migration and the first effective barrier is the SiC coating,
- In the case of SiC coating failure, FP migration in the graphite occurs by diffusion and absorption phenomena, depending on FP nature and graphite characteristics (impurities).

Data in UK :

Experience has been collected through the DRAGON reactor that operated between 1964 and 1975.

The UK has experience of the defuelling and decommissioning of existing HTR waste due to operation of the DRAGON Reactor Experiment (DRE). DRAGON was the pioneering experimental reactor of the OECD High Temperature Reactor Project and was situated at Winfrith in the UK. The reactor was designed and built as a fuel and materials test facility and was the world's first high temperature reactor. Criticality was achieved in August of 1964 and full design power of 20 MW was reached in April 1966. DRAGON finally shut down in September 1975.

The core of DRAGON consisted of hexagonal fuel elements containing a ring of six driver fuel rods, made up from annular graphite fuel compacts contained within graphite sleeves, and a central rod containing an experimental section. The six driver rods in each fuel element generally contained highly enriched $UO₂$, in the form of coated particles bonded into graphite compacts. A coated particle consisted of 0.8 mm $UO₂$ kernels encased within a TRISO coating, consisting of a silicon carbide layer sandwiched between two layers of pyrolytic carbon.

Following defuelling of DRAGON, 75,000 fuel compacts were stored untreated and were subsequently repackaged in stainless steel cans in 1998. These waste packages contain fuel consisting of uranium and uranium/thorium oxide and carbide kernels, graphite and some ZrC, covered with carbon and SiC layers to give 0.1- 0.25 mm particles. The fuel particles are mixed with graphite and compressed into compacts and some of the fuel has been disintegrated. Chemically, the composition of the packages can be summarized as follows :

- \sim 95% graphite/pyrocarbon
- \sim 5% heavy metal oxides and carbides (U/Th/Zr)
- 14 C - graphite and pyrolytic carbon
- Th Thorium oxide $\&$ thorium carbide (ThC $\&$ $ThC₂$)
- U Uranium oxide and uranium carbide (UC $\&$ $UC₂$)
- Pu Plutonium oxide and plutonium carbide (PuC)

There is no detailed publicly available experimental characterization or inventory data for the DRAGON waste packages, and published inventories are based on calculations using inventory codes. The DRAGON waste packages are the property of the UK Nuclear Decommissioning Authority (NDA) and, untypically for spent fuel, are classified in the UK as ILW as their heat generation rates are low enough not to require engineered cooling.

WASTE CHARACTERISATION

Spent fuel provisioning :

Considering first the low enriched uranium oxide kernels, fresh and irradiated samples have been identified in Juelich (pebbles, compacts and particles). The samples choice has been done. The main characteristics are indicated in table 2 (annex D).

Characterization :

Physical-chemical and radiochemical characteristics have to be known for interpreting the results of the studies on the long term behaviour of the spent fuel and the performances of the treatments for the separation of graphite from TRISO particles. It is necessary to get information about the localization and the distribution of the contamination.

Figure 2 shows the characterization programme. It is proposed to carry out both non destructive and destructive analysis.

For the acquisition of data in complementary of the existing ones, it is suggested to measure for each material :

- the morphological aspect (cracks, roughness, \dots) by optical microscope
- the structural properties by X rays diffraction
- the opened and closed porosities
- the localization of the contamination by EBM and microprobe
- the local chemical composition of the spent fuel (SEM and EPMA)
- the homogeneity of the $\beta\gamma$ contamination by γ collimation spectrometry (measure of the longitudinal distribution of the FP activities)
- the contents of U, Th, Pu and curium 244 by active and passive neutronic counting
- the inventory of the chemical and radiochemical composition by elementary analysis and radiochemistry after total dissolution of the solids.

Analysis will be performed, in the Atalante facility, vith equipment implemented in gloves boxes and hot cells.

Samples treatment and analytical techniques description :

Both elementary and microstructural particles analysis will be performed in hot cells. They need the development of a specific device for coating and polishing steps. A calibrated grid will allow to place at the bottom of a coating mould about 10 to 20 particles which will be fixed with an epoxy resin. After complete filling of the mould, a thickness indicator will allow to get information about the polishing step and to optimize it.

Complementary characterization will allow to compare perfomances and phenomena for various types of kernels, temperature and irradiation conditions :

- Fission products release quantification in the inner coating layers by electronic microprobe,
- Kernels X ray diffraction analysis to get information about lattice parameters after irradiation to study swelling under irradiation.

Thermal and physical parameters have to be determined in hot cells too. Specific heat (Cp) analysis of each particle constituent (SiC, PyC, kernels) allows to access both, for a temperature range between 300K and 1000K, to the annealed temperature of the different defects in the various networks and to the energy stored under irradiation because of fissions

damage. Using both density measurements and material diffusivity for various temperatures, thermal conductivity evolution will be assessed in this range of temperature.

Bibliographic data show an important evolution of both kernels and PyC coating densities. First, kernels swelling is observed with important porosity formation depending on the burn up and the temperature. PyC coating evolution depends of the fast neutron flux. Analysis carried out with both optical and electronic microscopes will allow to compare and complete these data.

CONCLUSION

The generation of waste from the operation of nuclear power plants, and specifically the VHTR one is an issue of major importance with regard to sustainable development, and receives much more attention than in the past when the main emphasis was on the technological development of the reactor systems and high conversion fuel cycles. In this field, an ambitious characterization programme is scheduled in Atalante facility with HTR samples provided by FZJ.

The objective of the RAPHAEL sub project on the back end of the fuel cycle is to study the characteristics and performance of HTR fuel with regard to behaviour in direct geological disposal conditions and is restricted to the oncethrough fuel cycle. Prior to these studies of disposal performance, an overview of the specifications and requirements for disposal of HTR waste has been obtained from the analysis of waste disposal legislation and classification schemes in operation within several EU states and US. Based on identified common denominators it then derives generic recommendations for disposal requirements.

Existing characterization data from past investigations in France, Germany and UK will be completed when FZJ samples will be transported to Atalante facility.

NOMENCLATURE

AVR : Arbeitsgemeinshaft Versuchs Reaktor DOE : Department Of Energy HLW : High Level Waste ILW : Intermediate Level Waste LEU : Low Enriched Uranium LILW : Low and Intermediate Level Waste LL : Long Lived category LLW : Low Level Waste RAPHAEL : ReActor for Process heat, Hydrogen And Electricity generation SL : Short Lived category THTR : Thorium High Temperature Reactor

ANNEX A

HTR FUEL BACK END OPTIONS

FIGURE 1 : FUEL CYCLE BACK END OPTIONS

ANNEX B

CHARACTERISATION PROGRAMME

FIGURE 2 : CHARACTERIZATION PROGRAMME.

Non destructive characterisationDestructive Characterisation

ANNEX C

SUMMARY AND COMPARISON

OF RADIOACTIVE WASTE CLASSIFICATION SCHEMES

Generic Disposal Routes

Landfill / Free Disposal Geological Disposal

ANNEX D

Samples characteristics

