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Preliminary synthesis and performance assessment

Executive summary

Important results were obtained in work package BF2 to assess waste conditioning techniques, waste package concepts and its disposal behavior: The direct disposal of fuel pebbles and compacts requires large container volumes (iron as hydrogen source). The separation of fuel kernels and embedding in more compact matrices like SiC, ZrO2 or glass as been shown to be technically feasible. Concerning long term performance it has been shown that these matrices are all very stable. In case of direct disposal without fuel kernel separation, radiolysis of porewater in graphite may create large quantities of CO2 and organic matter which will influence porewater chemistry. It may as well increase the gaseous part of C14 release.

Revisions

Rev.	Date	Short description	Author	WP Leader	SP Leader	Coordinator
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1 Introduction

Very high temperature gas cooled reactors (VHTR) will use sub-mm sized coated fuel particles embedded in the graphite matrix of fuel pebbles or block type fuel. The small TRISO fuel particles (kernels) surrounded by buffer carbon (BC), inner pyrolytic carbon (IPyC), silicon carbide (SiC) and outer pyrolytic carbon (OPyC) are designed to better confine gaseous and metallic fission products formed during the high temperature operation of the reactor. Direct disposal is one of the key waste management options. It is the focus of work package BF2 to identify optimal conditioning techniques for the direct waste disposal and to assess the performance under geological disposal conditions. The performance of both the conditioned waste product as well as the non-conditioned irradiated compacts or pebbles under disposal conditions will be evaluated.

2 Waste conditioning and waste package concept

2.1 Waste volume reduction by conditioning of kernels

One of the problems is the large disposal volume necessary for direct disposal of irradiated fuel pebbles or block type fuel. The disposal volume can be reduced significantly if the fuel kernels are separated from their graphite matrix and become embedded into a new more compact matrix. Three conditioning techniques for fuel kernels have been tested: embedding in SiC, in ZrO_2 and two vitrification methods: melting at 1200-1300°C under oxidizing conditions and sintering at much lower temperatures of 700°C. Electron microscope images have shown that all techniques except oxidative melting in glass have produced very compact embeddings but large particle densities have not yet been tested. The oxidative glass melting has led to oxidation of the coating layers and gas production.

But certain inconveniences have also been encountered for some of the other techniques: SiC and ZrO2 embedding involve very high temperatures. Conventional methods for SiC

production, such as liquid sintering or hot pressing are not applicable when processing spent coated particle fuel. These methods may damage the particles and lead to a release fission products, either due to the high processing temperatures (1800°C-2000°C) or the high pressure applied. In order to maintain integrity of the coated particles, a mild processing method for the production of the composite material is required.

Concerning encapsulation in glass, large-scale radioactive production experience and favorable disposal properties make glass a primary choice as confinement matrix. The lower operation temperature makes the use of sintered glass an interesting option to conserve as much as possible the strong confinement properties of the fuel particles. The product is probably capable to withstand groundwater attack for many hundreds of thousands of years. Alteration of by water will lead to the transformation of the glass into a dense gel like alteration phase, which most likely will maintain strong confinement properties of fuel particles.

To confirm or contradict these expectations, the aqueous stability of the conditioned waste products has also been investigated experimentally. The results show that the dissolution rate of the glass waste product is similar to that of current nuclear waste glass for reprocessed waste. ZrO_2 has a dissolution rate, which is about a factor of 1000 lower, but it is not yet sure whether the porosity of the waste product is sufficient low. Embedding in SiC would as well lead to products, capable to withstand a groundwater attack for a long time.

It shall finally be mentioned that the experiments performed do only give a first insight how SiC/ coated particle-compacts or vitrified particles can be produced, further examinations and possibly adaptations of the chosen parameters are necessary.

2.2 Container concept

Any disposal option requires also a reasonable container concept. The container may provide absolute isolation of the waste from groundwater form thousands to ten-thousands of years. The RAPHAEL project does not allow for any development work of container concepts, so existing container concepts will be used for a conceptional study. A starting point is the

CASTOR THTR container, which is proposed by GNS to be used both for interim storage and final disposal of HTR waste. The castor cask contains about 2100 graphite pebbles. However, with its 37 cm container wall thickness (cast iron) this is over-engineered as disposal container for clay formations. This is way too much iron (28 to) per spent fuel mass (about 20 kg if we consider 10g HM/ fuel pebble) . If we consider as reference (only for the sake of the argument here) a single AREVA HTR reactor of 600 MWt we have about 20000 compact fuel elements discharged after 60 yr of utilisation (Greneche, Szymczak, HTR meeting 2004, china) and 4.5 kg HM/ fuel element, we obtain a discharged fuel mass of 90 to HM in 60 yr. This corresponds to about 9 Mio fuel pebbles or 4300 Castor containers =1.2E5 to of cast iron or 1.4 Mio m3 hydrogen generation. (by slow long term corrosion). This mass of iron is about 1/3 of the total iron used in the actual french repository concept for the whole french nuclear power generation also for about 60 years.

A reduction of wall thickness would be useful. The Castor container contains an inner fuel can, which does not provide shielding, and may not be disposed directly due to radiation effects on the rock, fast corrosion etc.. but it seems reasonable to put this fuel can in an overpack container of about 11 cm of iron wall thickness. Such a thickness should allow for about 10000 yrs of tightness in a repository in clay formations. This thickness is also proposed by ANDRA in the french direct disposal concept for of spent LWR fuel. The total mass of iron in such a concept would be about 4 times less than the one used with the castor.

A disposal concept considering filling a container with fuel element pebbles (or balls) leads even with this wall thickness to an enormous amount of containers. Consequently, it is not realistic out of economic reasons and to avoid production of large quantities of hydrogen by container corrosion in groundwater, to consider also 11 cm wall thickness for this option Direct emplacement of the fuel can from the castor container seems possible but this would correspond to an instant loss of a very important barrier . A disposal concept considering a long-life time container filled with confined TRISO particles seems more realistic.

It was agreed to consider a slimmed version of a CASTOR type container (Laugh et al., 1997); the inside dimensions of the CASTOR container are kept, i.e. L = 1964 mm, diameter 640 mm, but its wall diameter is assumed to be 110 mm. The CASTOR container can accommodate 1

THTR canister. The inside dimension of the THTR canister are about L = 1600 mm and diameter 590 mm; the volume is 437 l.

2.3 The stability of the SiC coating under disposal conditions

The ongoing study pursues the aim to investigate whether or not and to what extend coatings of the fuel particles are able to keep their containment function under the long-term storage conditions in a spent fuel repository assuming that, at some point, a safe enclosure of the spent fuel is no longer given, and that the salt environment will have obtained immediate access to the fuel. In case of failed coatings, corrosion of the fuel and radionuclide release will start then. In the HTR/N and HTR/N1 project it has been shown that complete corrosion of the SiC layer by groundwater in absence of irradiation will probably take more than 10000 years. However, much earlier coating failure may occur caused by external mechanical stress and internal He-pressure buildup (about 0.35 Mpa after 40 yr). The PANAMA code is currently going to be adapted to predict coating failure coupling corrosion, pressure buildup and mechanical failure.
An additional problem is the formation of secondary phases by the interaction of SiC with water. Geochemical calculations have reproduced and explained the observation form literature studies: SiC is unstable in water under all conditions and SiO2 is expected to be the principal solid reaction product. The known high stability of SiC in water is in consequence probably the consequence of the formation of a protective SiO2 layer. Literature data have shown that SiC forms a very tight layer on the surface of SiC. This indicates the formation of a dense protective layer, but more work on reaction cinetics on SiC dissolution is necessary to confirm this hypothesis.

3 Performance analyses

The principal ways, how to assess the performance of the conditioned and non-conditionned wastes are today clear: (1) assess the barrier function of each of the different release barriers:

Graphite matrix or embedding matrix, SiC (or ZrC), PyC, matrix UO2 (or UCO) grain boundaries. (2) assess the relative importance of each of the barriers and the couplings between them for the boundary conditions of geological disposal such as thermal loads. (3) Calculate the overall impact. .

3.1 Coupling of reactions and processes leading to radionuclide release

3.1.1 Non conditioned irradiated fuel

A corrosion model has been developed in the project HTR-N, N1 to describe fuel kernel dissolution within the fuel pebble considering the following scenario: diffusion of water into the pebble in few years, failure of the SiC coating in about 10000 years, dissolution of Kernels in about 106 years under reducing and 10^{-4} years under oxidizing conditions. Solubility limits in the fuel pebble pore space are considered as well. The results show that the for conventional LWR fuel very important instant release fraction is much less important for HTR fuel. This is due to the stability of the SiC coating.

Chemical and mass transfer modeling of the reaction network governing radionuclide release from spent HTR/VHTR fuel under geological disposal conditions is currently ongoing. First results show for non-conditioned waste that radiolysis of pore water in the graphite matrix is an important process which may control both the dissolution rates of the graphite and as well pore water chemistry. Graphite corrosion rates even at low dose rates of 0.1 Gy/h are predicted to be after 3000 yr as high as 1.3E-7/yr. High carbonate and oxalate concentrations in pore water produced by gamma radiolysis may lead to high radionuclide mobility in graphite pore water. Radiolysis is main source for graphite oxidation/dissolution. Nevertheless, due to radioactive decay, the effect of gamma radiolysis is more or less limited to the first 500 years. The effect of alpha radiolysis is expected to be dominant in the long term but it is expected to be less important since, due to the small escape depth of alpha particles, alpha radiolysis is confined

to the interior of the fuel particle. Nevertheless, one might expect that the interaction of alpha particles with the PyC buffer layer may result in a significant production of organic matter for very long time periods. These observations significantly change the preliminary model developed during the HTR-N project, in which case no radiolysis was included in the calculations. The following effects might be envisioned:

(1) due to radiolysis carbonate concentrations in the graphite pore water will be higher than carbonate concentrations in the groundwater. This will increase the solubility of actinides in the pore water. It is likely that solubility constraints for radionuclides in the pore water must be omitted.

(2) Radiolysis produces as well large quantities of organic matter which may further increase mass transfer of radionuclides

(3) The formation of organic carbon by radiolysis may also increase the quantity of gaseous C14 released from the graphite. This may strongly increase the C14 migration rates towards the environment

3.1.2 Conditionned waste

Significant elements for a prediction of the RN release properties have been gathered: Basic matrix corrosion rates are available for glass, SiC and ZrO2. This will povided a lower bound for the rates by which embedded fuel particles may become in contact with diffusion groundwater. It is expected that the corrosion rates of the embedding matrix will be in the order of 10^{-5}/yr (case for glass) or lower (case of the other matrices), hence, the rate by which water comes on average in contact with the fuel particles will be slower that the rate by which the SiC coating may disintegrate (10000 yr). Hence, the SiC coating will probably provide no additional barrier function when comparing the relative importance with that of the matrix. Important reduction of release rates is also expected from the fuel particles. Data from BF3 have shown that hydrogen form container corrosion strongly reduce corrosion rates of irradiated fuel. Moreover, data from light water reactors show that there is a threshold for specific alpha activity in the fuel, below which increase of fuel dissolution rates by radiolysis

do not need to be considered. Considering that the embedding matrix will strongly delay groundwater contact with the fuel particles, radiolysis is most likely not a facto which needs to be considered in fuel dissolution rates (it may still be an effect in organic matter generation and in increasing the solubility of radionuclides by organic matter complexation).

Important questions on conditioned fuel still needs to be resolved before the end of the project:

(1) are sintered matrices (glass, ZrO2) sufficient dense (sufficient low porosity) to prevent any water diffusion towards the fuel particles without dissolution of the matrix

(2) once the embedding matrice has been transformed into secondary phases (after hundredth of thousands of years): will this matrix still protect the fuel particles.

(3) Calculate the diffusive mass transfer across a corroded embedding matrix.

Low matrix dissolution rates for glass are only valid if disposal temperatures are low once groundwater comes in contact with the glass matrix. This points to the importance of a reliable container concept assuring absence of groundwater/glass contact during the thermal period (first 1000 yr). The safety function of the container is more important for conditioned fuel particles than for the direct disposal for graphite pebbles since disposal temperatures are initially much higher.

3.2 Thermal calculations

One THTR canister can contain 2100 fuel elements (Laugh et al., 1997). This corresponds to 21 kg U. This is a very low amount of heavy metal for such a large container; e.g., within the Belgian radioactive waste management programme we consider containers with a comparable diameter and a length of about 5000 mm that contain 1840 kg U.

The thermal output of this container after 10 years cooling is only 50.3 W; if the containers are placed one after the other along the axis of a disposal gallery this corresponds to a thermal loading of the gallery of 25 W/m. For the disposal of vitrified HLW or spent fuel we normally

apply a maximum thermal loading of 300 W/m; this requires cooling times for the vitrified HLW of 50 or 60 years.

If we assume that the present Belgian nuclear power park (total amount of spent fuel with burn-up of 50 GWd/tHM is 4300 tHM) should be replaced by a HTR power park generating the same amount of electricity (burn-up 76 GWd/tHM), this would result in 1867 tHM of HTR spent fuel or 187 million fuel elements or 88 910 POLLUX containers. For comparison, the total HLW for the present PWR park is estimated to be 4277 COGEMA canisters in case of reprocessing, or about 2337 containers containing each 4 PWR fuel assemblies.

4 Conclusion and perspective

Significant progress in the buildup of a knowledgebase for the evaluation of the performance of conditioned and non-conditioned HTR fuel has been achieved. For unconditioned waste a significant modification of the model has been developed. For conditioned waste, basic stability data have been gathered by performance calculation still needs to be developed.