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Design study of an irradiation experiment with inert matrix and mixed-oxide fuel at the Halden boiling water reactor

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Abstract

An effective way to reduce the large quantities of plutonium currently accumulated worldwide would be to use uranium-free fuel in light water reactors (LWRs) so that no new plutonium is produced. To test such a new fuel under reactor conditions and in comparison with standard mixed-oxide (MOX) fuel, an irradiation experiment is planned at the Halden boiling water reactor. The behaviour of three fuel rods consisting of uranium-free fuel will be investigated together with three rods made out of uranium-plutonium mixed-oxide fuel in the same assembly. The fuel compositions were adjusted so that all rods produce a similar power. Because of the moderation with D_2O in the Halden reactor, two different surroundings of the considered assembly were examined to analyze the influence of the flux spectrum on the experiment. This showed that the influence of the spectrum on the material behaviour is negligible. The relation between assembly power and average neutron detector signal as well as the burnup or depletion function was calculated. The assumed power history was adapted to a usual LWR schedule. It is possible to reach a burnup of ~ 540 MW d kg⁻¹_{HM} with the uranium-free fuel and ~ 54 MW d kg⁻¹_{HM} with the MOX fuel after five years of irradiation, which is similar to the average burnup reached in commercial LWRs after four years of operation. © 1999 Elsevier Science B.V. All rights reserved.

1. Introduction

Due to economical reasons and lack of public acceptance, the nuclear fuel cycle is not effectively closed today. Consequently, UO_2 -fueled light water reactors (LWRs) continue to dominate the nuclear energy scene. Thus, plutonium stocks are increasing far beyond projected needs and becoming of considerable concern from a waste and non-proliferation viewpoint.

A spent fuel management strategy has direct influence on this issue and on resource consumption. To maximize energy extraction from a given amount of ore and to minimize waste, recycling of fissile materials from spent fuel is necessary. One way to utilize the energy content of spent fuel through reprocessing is the use of uranium-plutonium MOX [1–3]. This method allows one to increase from $\sim 0.6\%$ to $\sim 1.2\%$ the energy extracted from the initial fuel. However, it does not seem possible to effectively reduce the large quantities of plutonium that are accumulating worldwide by using MOX fuel alone because of the build-up of new plutonium from the uranium present. For this reason a oncethrough cycle with a uranium-free inert matrix fuel (IMF) has been proposed to reduce stocks of plutonium by using its energy content in current LWRs [4–10].

MOX recycling technology is being developed at the Korean Atomic Energy Research Institute (KAERI) on the basis of feasibility studies, particularly for higher burnup, trying to make MOX pellets for pressurized water reactors (PWRs) with an improved microstructure by using more efficient fabrication equipment and process technology. One part of the project 'Advanced Fuel Cycle' at the Paul Scherrer Institute (PSI) in Switzerland is the development of IMF with research groups in Japan and Italy [11,12]. The MOX fabrication method developed by KAERI can be applied also to

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zirconia-based IMF. Zirconium oxide stabilized in its cubic phase with yttrium is one possible material for the inert matrix. Moreover, it shows a well known, favourable neutronic behaviour.

An irradiation experiment with IMF and MOX fuel proposed by KAERI and PSI is planned in the Halden boiling water reactor in Norway as part of the OECD Halden Reactor Project. The Institutt for Energiteknikk is responsible for its organisation. The aim is to measure the thermal conductivity, the fission gas release and the behaviour of these fuels under irradiation conditions similar to those in current LWRs. For comparison, the proposed fuel rods will be irradiated together with one rod of 'short-binderless route' MOX fuel [13] (called standard MOX herafter) fabricated and supplied by British Nuclear Fuels. The three fuel types will be loaded in an instrumented fuel assembly (IFA) in the Halden reactor.

This paper deals with the design of this experiment based on neutronic calculations and comparisons with other fuel assemblies in the Halden reactor. One aim is to find an enrichment which makes the new fuels comparable with the standard MOX fuel. A power schedule is proposed and different reactor environments of the IFA are investigated. In all cases, the values of the average linear heat rates (ALHRs) of the studied fuel rods and the surrounding normal fuel assemblies used as drivers are calculated. Nuclear detector (ND) signals were derived and compared with measured values. Functions like the KG-factor, i.e. the relation between the assembly power and the ND signal, and the depletion functions which are used to convert the signals of the NDs into linear heat rates and to calculate the burnup, are also determined in the preparation phase of the experiment.

2. Geometry and materials

The Halden reactor is a boiling, heavy water cooled and moderated reactor with a thermal power of 18–20 MW. The D₂O moderator temperature is about 510 K with a pressure of ~3.3 MPa. The reactor is fueled with a combination of about 35 instrumented fuel assemblies belonging to the participants in the OECD Halden reactor project, containing the different experiments, and about 70 driver assemblies, so-called spikes, which mainly provide reactivity for operating the reactor. Each spike contains eight UO₂ fuel rods with an enrichment of 6 wt% ²³⁵U. All assemblies are surrounded by a 1 mm thick shroud with an inner diameter of 71 mm; the lattice pitch of the assemblies is 130 mm.

The burnup of the spikes is usually less than 34 MW d kg_{HM}^{-1} .² The fresh spikes are normally loaded in the

Fig. 1. Comparison of the average linear heat rates (ALHR) in a PWR and in the experiment in the Halden reactor (HBWR).

core centre (the inner three rings), while the spikes with higher burnups are placed in the periphery of the core. The maximum power of spikes in the core centre is ~ 300 kW corresponding to an axial linear heat rate of 450-500 W cm⁻¹. For the design calculations, spikes with a constant 3 burnup of 15 MW d kg⁻¹_{HM} were used. The isotopic number densities in the spike fuel were calculated with an average volume ratio of the core. These spikes were used to analyze two different geometries. In the first case, the considered fuel assembly was surrounded by one open ring of spikes. This geometry represents a typical situation of instrumented fuel assemblies in the Halden reactor where half of the nearest neighbour positions on the hexagonal lattice are occupied by spikes; the flux spectrum corresponds to the average spectrum in the reactor. This spectrum is more thermal than that of a UO₂ fueled LWR. The IFA surrounded by a closed ring ⁴ was considered to examine the influence of a more epithermal spectrum. To adapt this case to a more realistic situation in the Halden reactor, a second open ring was taken into account.

The neutron flux level is measured with vanadium NDs. There are usually three NDs in one horizontal plane (to measure the radial flux distribution) and in three or four axial positions. Thus, together with neutronic physics calculations, it is possible to define the power of the fuel rods during irradiation. Turbine flowmeters, inlet and outlet coolant thermocouples are used to calibrate the instrumented fuel assemblies and define a relation between power and the signal of the NDs, the so-called KG-factor.

The power schedule used for the burnup calculation corresponds to the power history of fuel assemblies in a current PWR. Fig. 1 compares the maximum average



 $^{^{2}}$ kg_{HM} means the weight in kg of heavy metal, i.e. uranium and plutonium for the present study, in the fuel.

³ The isotopic number densities in the spikes are kept constant during burnup calculations.

⁴ All neighbour positions are occupied by spikes.

linear heat rate in a PWR with the linear heat rate which was used during the calculation to define the power of the spikes, the required ND signals and the accessible burnup for the planned irradiation time.

3. Methods and data

For all calculations the two-dimensional transport code HELIOS was used [14,15]. This code allows almost every kind of two-dimensional geometry to be calculated and has been used at the Halden Project since January 1998. The cross-section library designed for LWR calculations has either 34, 89 or 190 neutron as well as 18 or 48 gamma groups and is based on the master library ENDF/B-VI. For the burnup calculations, 28 heavy nuclides are used in the depletion chains. Because 114 fission products are always treated explicitly, it is not necessary to lump fission products together.

3.1. Validation of the calculations for experiments in the Halden reactor

The power of an IFA is determined by measuring the water flow through the rig and the difference between inlet and outlet temperatures. The measurement of the power and the simultaneous measurement of the neutron flux with the NDs gives the relation between power and the detector signal in the observed rig. The ND signal itself is the current produced by the decay of ⁵²V which is a capture product of ⁵¹V. Taking this into account, there are two ways to calculate the average ND signal $I_{\rm ND}$ using HELIOS. One possibility is to define the signal with a measured calibration constant S and the thermal flux $\phi_{\rm th}$, where the signal measured in the Halden reactor is proportional to the thermal flux at the detector position: $I_{\rm ND} = S \cdot \phi_{\rm th}$ where $\phi_{\rm th}$ is the flux below the energy of 0.625 eV as calculated by HELIOS, and the value of the constant S has been determined to be 3.6×10^{-12} nA cm² s.

The relation between $I_{\rm ND}$, the vanadium absorption and the neutron flux in the NDs, can also be obtained by assessment of power calibration data. Therefore, three different instrumented fuel assemblies were evaluated in their environments. The ratio $R_{\rm abs}$ between the calculated vanadium absorption rate $V_{\rm abs}$ and the measured ND signal $I_{\rm ND}$ was determined as

$$R_{\rm abs} = \frac{V_{\rm abs}}{I_{\rm ND}} = 2.0 \times 10^{10} \text{ cm}^{-3} \text{ s}^{-1} \text{ nA}^{-1}.$$

Both methods give similar results, with a slightly higher ND signal for calculations using the integral R_{abs} value. In a case with ~4 wt% ²³⁵U enrichment, the difference in the ND signals calculated by both methods is ~0.8%; while for an IFA containing rods with MOX and rods with highly enriched uranium it is 2.6%, because of the

slightly harder flux spectrum. This shows that it is now possible to determine the KG-factor of an instrumented fuel assembly, i.e. the ratio between power and average ND signal, from nuclear physics calculations.

In addition, the depletion function can be calculated with the HELIOS code. This function gives the change of rod power during burnup in an IFA, for a constant ND signal, or flux level normalized to the beginning of life (BOL) value. The only influence on the depletion function comes from the flux spectrum. Therefore, when performing the calculations, it is necessary to take into account the surroundings of the IFA.

Using these results, four different fuel assemblies called IFA515-10 and IFA562-2/3/4 were investigated. IFA515-10 contains six highly enriched uranium rods, three without and three with gadolinium (8 wt% gadolinium highly enriched by 98.6% of ¹⁶⁰Gd). At BOL as well as at a burnup of about 60 MW d kg⁻¹_{HM} power calibrations were carried out for this IFA. The depletion function was calculated with HELIOS as well as the KG-factors for both points of calibration. The ratio of calculated and measured values of the KG-factors for IFA515-10 were 0.982 in the first case and 1.0 in the second.

These results show that it seems possible to accurately define the relationship between power and ND signal. However, the in-pile measured burnup still depends on the calculated depletion function in the case of IFA515-10. Therefore, IFA562-2 to IFA562-4 were examined because burnup determinations are available for one rod from loading 2 and one from loading 4. The comparison of the burnup as calculated using a KG-factor and depletion functions from HELIOS calculations, and the measured burnup, shows that the uncertainty of the calculated burnup was less than 4% for both rods.

Therefore, it is possible to calculate the depletion function as well as the KG-factor with a satisfactory accuracy, if the environment is adequately modelled. Consequently, it seems possible to prepare a realistic layout of the IMF/MOX experiment from the neutronic point of view and to predict some important measurable values.

3.2. Fuel compositions

In the planned IMF-experiment, a standard MOX fuel rod will be used as a reference for currently installed MOX fuels in LWRs. It was suggested that all rods should produce the same power during irradiation. Consequently, the fuel compositions given in Table 1 were determined after several HELIOS-simulations with an open lattice.

The plutonium contents were adapted to the power distribution in the fuel assembly at BOL. In these conditions, the linear heat rate of the IMF rods is $\sim 2.0\%$

compositions of the various rates in the river more experiment as they were used for the design calculations									
Fuel type	MOX	MOX	MOX	IMF	IMF	IMF			
U-cont. (g cm $^{-3}$)	8.449	8.450	8.486	_	_	_			
Pu-cont. (g cm $^{-3}$)	0.772	0.784	0.745	0.90	0.90	0.93			
²³⁵ U (at.% of U)	0.314	0.314	0.314	_	_	_			
Zr_{nat} (g cm ⁻³)	_	_	_	3.345	3.345	3.328			
Y (g cm $^{-3}$)	_	_	_	0.335	0.335	0.335			
$\operatorname{Er}(\operatorname{g}\operatorname{cm}^{-3})$	_	_	_	0.360	0.360	0.380			

Table 1 Compositions of the various fuels in the IMF/MOX experiment as they were used for the design calculations

higher than that of the MOX rods, while at end of life (EOL), i.e. after ~ 1000 days of irradiation, the power in the MOX rods is higher. In all calculations the plutonium had the following composition:

The part of 241 Pu was defined for July 1999 where the amount of 241 Am was calculated to be 4.3 wt% of the Puweight. The plutonium used for the rod with standard MOX fuel has a composition similar to the one given above. The amount of fissionable plutonium is 73.16 wt% of the total, slightly lower than in the other cases.

Since not all erbium isotopes are available in the HELIOS library, the following contributions were used for the calculations for IMF:

The density of the ZrO₂ was defined such that the final IMF-density is ~95% of the theoretical density. In addition ~10 wt% Y_2O_3 was taken into account as a stabilizer of IMF.

All fuel rods have an active length of 400 mm and a fuel diameter of 8.20 mm. The inner and outer diameters of the cladding are 8.36 and 9.50 mm, respectively, for all fuel types. Two MOX rods and one IMF rod contain an expansion thermometer in a central hole of 1.8 mm diameter. The other rods are equipped with pressure transducers and thermocouples.

4. Influence of the experimental environment

Usually, the surroundings of experiments in the Halden reactor are similar to the so-called open ring surrounding. This means that the experiment has higher moderation and so a more thermal flux spectrum than a normal LWR. Therefore, the considered IFA was investigated with a second, so-called closed ring environment. Fig. 2 compares the neutron flux spectrum in MOX fuel as a partial loading of one rod in a uranium-fueled PWR lattice, with spectra from both of the considered cases.



Fig. 2. Comparison of a calculated normalized flux spectrum of MOX in a uranium-fueled PWR with the spectra of an open and a closed lattice in the Halden reactor. Lethargy $= \log(E_0/E)$, with $E_0 = 10$ MeV.

Because of the better moderation in the case with the open ring, the thermal flux is $\sim 30\%$ higher than in the case of the closed ring, and more than 3 times higher than for the MOX fuel in the PWR environment. In the epithermal energy range, the case with the closed ring shows the highest flux, while the flux for the open ring lattice is similar to that in the MOX fuel of the PWR case. Because of the higher Dancoff factor [16] in the PWR lattice, the fast flux is larger here than in the Halden experiments. For these reasons, the results of the burnup calculations, like the isotopic compositions at EOL, are different in the three cases.

4.1. Burnup calculation

The two different surroundings of the fuel assemblies, i.e. the open and the closed ring environments, influence the behaviour of the IMF and MOX fuel during burnup. In order to compare this behaviour with the burnup behaviour of MOX-rods in a PWR environment, the depletion was calculated for all cases in the form of so-called 'CD-functions', which are defined as

$$CD = \left(\frac{P(t)}{A_{ND}(t)}\right) \left/ \left(\frac{P(t=0)}{A_{ND}(t=0)}\right)$$
(1)

where P(t) is the pin power and $A_{ND}(t)$ the vanadium absorption rate at time *t*.

For the calculation of the CD-function in the PWR case, the absorption rate of the UO_2 surrounding was used instead of the vanadium absorption rate. Fig. 3 shows the depletion functions for all three cases. In all cases, the CD-functions of the MOX rods were almost the same. This was also the case for those of the IMF rods, so that only one CD-function per surrounding and fuel type is shown.

In case of MOX fuel, due to the higher resonance capture rate in ²³⁸U because of the harder flux spectrum, the build-up of new ²³⁹Pu and also the higher fission rate of ²³⁸U, the CD-function decreases more slowly in the PWR case than under the conditions of the Halden reactor. The fastest decrease of the CD-function takes place for an IFA with an open ring, while the CD-function of the IFA with a closed ring lies in between.

For the IMF rods there is a similar situation. Here the resonance capture of ¹⁶⁷Er is higher in the cases with harder flux spectra. The ¹⁶⁷Er content of the open ring case is highest at EOL, with ~13% of the initial amount, and smallest in the PWR case, with ~7.5% of the initial amount. These differences may not be negligible from the neutronic point of view, but the main issue in this experiment is the material behaviour of IMF under irradiation. In that case, only the total amount of the elements are important and not the isotopic compositions. For both fuel components, erbium and plutonium, the changes with burnup are the same in all three scenarios and the distinctions at EOL are smaller than 1%. During burnup, the total amount of plutonium decreases by ~57% while the reduction of the erbium is only 1.5%.

These results show that the surroundings of the IMF/ MOX fuel assembly will have no effect on the material behaviour of IMF. But they also show that it is important to consider the environment of an IFA when calculating the depletion function.



Fig. 3. Depletion functions (CD) in various environments for IMF and MOX fuel.

The planned irradiation time of the IMF/MOX fuel assembly is 5 yr or about 1000 equivalent full power days (EFPD). The calculated burnup for the different fuels is about 490 kW d cm⁻³ in 1000 EFPDs using the power schedule of Fig. 1. Thus the planned burnup of the IMF is ~540 MW d kg⁻¹_{HM} and ~54 MW d kg⁻¹_{HM} for the MOX fuel.

5. Power peaking

For the Halden reactor, the planned experiment will be the first in which this kind of IMF will be irradiated and produce power. Therefore, it is necessary to observe the behaviour of this new fuel under low power and to rise to the maximum power only after one or two cycles of irradiation. The required ND signal (for the necessary flux level which will be higher than for fresh fuel) and the power of the spikes in the surroundings of this fuel assembly were calculated. All these values are summarized in Table 2 for both cases, the open and the closed ring environments.

It seems possible to reach the maximum average linear heat rate with accessible ND signals ⁵ for both surroundings. In the open ring case, this value is accessible with a maximum linear heat rate of the spikes of \sim 420 W cm⁻¹, while with the closed ring the maximum linear heat rate of the spikes is \sim 550 W cm⁻¹ which is the upper operating limit.

It has to be borne in mind that the fuel of the spikes has a burnup of 15 MW d kg⁻¹_{HM}. For spikes using fuel with a lower burnup, the average linear heat rate will increase and for spikes with a higher burnup it may not be possible to achieve the required flux level in the IMF/ MOX fuel assembly. From this point of view, the IMF/ MOX fuel assembly with an open ring environment is the more satisfactory solution because the maximum linear heat rate in the spikes is much lower than in the closed ring case, and it also seems possible to achieve the target linear heat rate for the IMF and the MOX fuel with lower burnups in the spikes.

6. Relation between power and ND signal

The relation between the power in an IFA, P_{IFA} , and the ND signal, I_{ND} , is called the KG-factor:

$$KG = P_{\rm IFA}/I_{\rm ND}.$$

Usually, this value will be measured at BOL for every IFA in the Halden reactor as described in Section 3.1. The KG-factor, together with the initial power distribution, the flux distribution measured by the NDs and

⁵ The maximum accessible ND signals are \sim 300 nA.

Table 2

Burnup, power and maximum average linear heat rates (ALHR) in the IMF/MOX fuel assembly and in the spikes, for two different environments of the fuel assembly

Parameter	Unit	Open ring	Closed ring
Burnup	kW d cm ⁻³	61	58
IFA-power	kW	112.4	112.4
Max. ALHR (IMF)	$W cm^{-1}$	475	475
Max. ALHR (MOX)	$W cm^{-1}$	465	465
Required ND-signal	nA	190	185
Max. ALHR (spikes)	$W \ cm^{-1}$	421	509/548 ^a

^a Values for the first/second ring.

the CD-function, allows one to calculate the average linear heat rate of the fuel rods. For the IMF/MOX fuel assembly, the KG-factor was determined to be 0.643 kW nA^{-1} for the open ring and 0.664 kW nA^{-1} for the closed ring surrounding. The relative initial power distribution was calculated to be:

Rod-1	Rod-2	Rod-3	Rod-4	Rod-5	Rod-6
MOX-1	IMF-1	MOX-2	IMF-2	MOX-3	IMF-3
0.99	1.01	0.99	1.01	0.99	1.01,

where the differences between the two different environments are negligible.

7. Conclusions

An IFA with six fuel rods was investigated using the 2-D transport code HELIOS, to determine the material composition of IMF and MOX fuel for an irradiation test in the Halden boiling water reactor. In a first step, the HELIOS code was validated for the calculation of instrumented fuel assemblies in the Halden reactor by re-assessing fuel assemblies which had already been irradiated in the reactor. It was shown that the relation between the power of an IFA and the measured signal of the NDs, as well as the depletion function of the fuel could be defined in a satisfactory way.

For the investigation of the burnup behaviour of this IMF/MOX fuel assembly, two different surroundings, consisting of an open and a closed ring of spikes, were considered. The results of these calculations were compared with calculations of a single IMF and single MOX rod in a current uranium-fueled PWR environment. These comparisons showed, on the one hand, that the surroundings have a large impact on the changes of isotopic compositions with burnup and consequently on the depletion function because of the different flux spectra. On the other hand, there is no influence on the variation of the material compositions during burnup: this is a very important conclusion from the point of view of the transferability of the results of this experiment. In all IMF rods, during the whole irradiation the

plutonium content decreases by 57% of the initial one, while the total amount of erbium is reduced by 1-2%.

The chosen power history of the IFA is adapted to that of a current PWR. After 1000 days of irradiation, the burnup reaches \sim 490 kW d cm⁻³ which is equivalent to \sim 540 MW d kg_{HM}⁻¹ for IMF and \sim 54 MW d kg_{HM}⁻¹ for MOX. Since this is the first irradiation of IMF in the Halden reactor, it is proposed that lower power be used in the first one or two cycles, in order to observe the behaviour of this fuel under operational reactor conditions. For this reason, the maximum power was determined at a burnup of about 60 kW d cm⁻³. For the maximum average linear heat rate of 475 W cm⁻¹ for IMF and 465 W cm⁻¹ for MOX, the required ND signal was calculated to be ~ 190 nA. The maximum average linear heat rate of the spikes reaches 421 W cm⁻¹ for the open ring lattice. For the spikes in the closed ring case it reaches \sim 550 W cm⁻¹; this represents the upper limit for the spikes. Because there is no influence from the flux spectrum on the material composition of the fuel in the fuel assembly, it is suggested that an open ring arrangement be used as the environment for the IMF/ MOX experiment.

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