

Inert matrix fuel behaviour in test irradiations

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Abstract

Among others, three large irradiation tests on inert matrix fuels have been performed during the last five years: the two irradiation tests IFA-651 and IFA-652 in the OECD Halden Material Test Reactor and the OTTO irradiation in the High Flux Reactor in Petten. While the OTTO irradiation is already completed, the other two irradiations are still ongoing. The objectives of the experiments differ: for OTTO, the focus was on the comparison of different concepts of IMF, i.e. homogeneous fuel versus different types of heterogeneous fuel. In IFA-651, single phase yttria stabilized zirconia (YSZ) doped with Pu is compared with MOX. In IFA-652, the potential of calcia stabilized zirconia (CSZ) as a matrix with and without thorium is evaluated. The design of the three experiments is explained and the current status is reviewed. The experiments show that the homogeneous, single phase YSZ-based or CSZ-based fuel show good and stable irradiation behaviour. It can be said that homogeneous stabilized zirconia based fuel is the most promising IMF concept for an LWR environment. Nevertheless, the fuel temperatures were relatively high due to the low thermal conductivity, potentially leading to high fission gas release, and must be taken into account in the fuel design.

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PACS: 28.41.Bm; 81.05.Je; 81.05.Mh

1. Introduction

In inert matrix fuel (IMF), plutonium is embedded in an U-free matrix. This allows to burn plutonium without breeding any new plutonium

by neutron capture in ²³⁸U. Thus, a more efficient consumption of plutonium is achieved compared to mixed oxide fuel (MOX). While reprocessing of spent UO₂ fuel is the key to an optimised utilisation of nuclear fuel, recycling of second and third generation plutonium as MOX in LWR seems economically unfavourable. Therefore, a once-through-cycle in an uranium-free IMF has been proposed for utilisation of this fissile plutonium. Suggestions of different types of IMF were based on neutronic

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evaluations, aspects of safety and non-proliferation as well as economic assumptions [1].

For reasons of acceptance and costs, the fuel assemblies should be designed to replace conventional MOX elements. To reduce power peaking, addition of a burnable poison proves to be necessary. Studies have identified erbium oxide as the most suitable candidate for this purpose [2,3]. Loading up to 12 IMF assemblies with optimised design in an LWR could be done following fuel management schemes that are quite compatible with current operational requirements (e.g. cycle length, soluble-boron concentration, etc.) [4]. Thus, the neutronics calculation tools employed were tested against a range of integral LWR experiments with PuO_2/UO_2 fuel, a series of burnable-poison reactivity worth measurements, as well as via comparisons of numerical results obtained in the framework of an international benchmark exercise for LWR IMFs [5,6]. The addition of thoria for neutronic reasons was also discussed as an interesting option [7–9].

The share of plutonium assemblies in a core can be reduced from one third to one eighth for the operation of an LWR in the so-called ‘self generated’ mode if an inert matrix material is used as carrier instead of depleted or natural uranium oxide [4]. Relative plutonium consumption rates for IMF assemblies were estimated to be 2.5 times higher than for MOX assemblies, underlining the considerable economic incentive for PWR cores with partial loadings of IMF assemblies [2]. Last but not least, less Pu-fuel assemblies with longer decay heat production must be stored in the spent fuels reactor pool. IMF has thus been confirmed to be an attractive option for a once-through LWR strategy [10].

Development programmes focusing on material technology with the emphasis on fabrication, characterisation, and irradiation of different IMF concepts have been started during the last decade in Europe, Japan, Russia, and the US [3,8–24]. Comparably early, an innovative type IMF was suggested that consists of plutonium dissolved in yttria stabilised zirconia (YSZ), a highly radiation resistant cubic phase with the potential of achieving high burnup [25]. Alternatively, the cubic structure of the zirconia matrix can be stabilized by calcia, CSZ [7]. The extreme insolubility of the matrix makes the fuel failure safe with regard to cladding defects during irradiation (no washout of fissile material in case of a clad failure) and with regard to direct waste disposal. The YSZ matrix is not redox sensitive as it has to be assumed for uranium

based fuel in the safety assessment of geological underground storage [26]. Additionally, the proposed YSZ based IMF is practically insoluble in aqueous and acidic solutions, both as the fuel and the produced spent fuel matrix. This is an effective barrier against misuse of the fissile material.

The major drawback of stabilized zirconia as a host matrix is the comparable low thermal conductivity [27,28]. Various countermeasures have been discussed: the application of annular pellets to reduce the fuel central temperature and the application of a heterogeneous concept, i.e. YSZ-based fuel particles embedded in a spinel matrix. Also, the addition of thoria or urania not only for neutronic reasons but also to increase the thermal conductivity was discussed. Although such fuel is then no longer a pure IMF, it will also be considered in this report. Different heterogeneous concepts were investigated in the course of the OTTO (Once-Through-Then-Out) project by JAERI, NRG, and PSI [29]. CSZ-based IMF with and without thoria additions is under irradiation in IFA-652, a joint Halden Programme experiment conducted in collaboration with ENEA (Italy). The homogeneous concept of YSZ-based IMF fabricated by two different routes was and is being tested in IFA-651, a joint Halden Programme experiment conducted in collaboration with PSI and the Korea Atomic Energy Research Institute (KAERI) [30].

Several reports have already been published on these irradiation tests (see references). However, an overview of these three tests and a comparison and general evaluation of the results have not been done so far. The aim of this report is to briefly describe the three mentioned IMF irradiation tests, to summarize the results available today, to compare them, and finally to draw conclusions for future IMF development.

Note: the burnup unit used in this report is kW d cm^{-3} ; conversion to the burnup unit of MW d kg^{-1} of heavy metal is obtained by dividing by the initial heavy metal density of standard UO_2 fuel, $\sim 9.17 \text{ g cm}^{-3}$.

2. Irradiation tests

2.1. The IFA-651 experiment in Halden

The objective of the Inert Matrix/MOX fuel experiment IFA-651 is to measure the general thermal behaviour of YSZ-based IMF under irradiation conditions similar to those in current LWRs. Of

particular interest are the thermal conductivity and its degradation with burnup, fission gas release (FGR), fuel densification, and swelling. The secondary aim is to compare the performance of IMF with that of MOX. IMF pellets have been fabricated in the Pu-laboratories at PSI. Rod fabrication and instrumentation as well as assembling the rig were done at the Institutt for Energiteknikk (IFE), Kjeller, Norway [31]. The IFA-651 rig originally contained a cluster of six rods: three rods with MOX and three rods with IMF. All fuel rods were equipped with fuel thermocouples and pressure transducers to measure the evolution of the pin inner pressure. Additionally, three rods were equipped with fuel stack elongation sensors. The irradiation test started successfully in June 2000. After the end of the first loading at end of May 2003, one IMF rod was discharged for PIE. The results of the PIE are to be published. The irradiation test is now continuing with two IMF rods and three MOX rods. An average assembly burnup of 300 kW d cm^{-3} was reached at end of 2004.

The test has contributed to the increasing knowledge base of IMF [30,32], together with out-of-pile experiments [33].

2.2. The IFA-652 experiment in Halden

The objective of the IFA-652 experiment is to investigate the in-pile behaviour of alternative IMF types, i.e. CSZ-based IMF with and without larger additions of thoria. The topics of primary interest are the same as for IFA-651. The secondary aim is to compare the performance of the two types of IMF with that of (U,Th) O_2 fuel. All fuel pellets were fabricated at IFE in Kjeller. For practical reasons, high enriched uranium (HEU) was used as the fissile material instead of Pu. The rig comprises six rods with three different fuel types: CSZ-based IMF with HEU (instead of Pu), CSZ-based IMF with HEU and 39.2 wt% ThO_2 , and (U,Th) O_2 fuel. All fuel rods were equipped with fuel thermocouples and pressure transducers to measure the evolution of the pin inner pressure. Additionally, three rods were equipped with fuel stack elongation sensors. The rig was loaded in June 2000 and has reached an average assembly burnup of more than 280 kW d cm^{-3} at the end of 2004.

2.3. The OTTO experiment in Petten

The objective of the OTTO experiment was to investigate the in-pile behaviour of YSZ-based

IMF both as solid solution and as particles of different sizes in a spinel matrix. For the OTTO experiment, six plutonium containing IMF segments were prepared for an irradiation experiment in the High Flux Reactor (HFR) in Petten together with a MOX segment. The IMF segments consist of micro- and macro-dispersed spinel targets as well as homogeneous YSZ-based IMF. Small amounts of either erbia or urania were added to each type of IMF to increase the Doppler effect. Homogeneous fuel pellets and IMF particles have been fabricated in the Pu-laboratories of PSI. The IMF particles were combined with spinel and fabricated to pellets at NRG in Petten, The Netherlands. Rod fabrication and instrumentation as well as assembling the rig were also done at NRG in Petten [29]. During irradiation, fuel and cladding temperatures were monitored by means of 24 thermocouples. Four central thermocouples measured the temperatures of three heterogeneous IMF targets and the MOX target. Neutron radiographs were taken from time to time to monitor the geometrical changes. The irradiation experiment was started in fall 2000 and was completed in December 2002 after 22 HFR cycles. PIE was then performed on all rods. The results of the PIE are to be published. All seven targets were irradiated up to a burnup of about 200 kW d cm^{-3} .

2.4. Comparison of experimental designs

A condensed overview of the different experimental designs is given in Table 1. The main differences in irradiation conditions are the target burnup, the coolant temperatures, and the different power histories. While for the OTTO irradiation the power was relatively low and steadily decreasing (except for the start-up and shut-down ramps), the power histories for the Halden irradiation were on a higher level and reflect the typical irregular Halden reactor operation history (see Fig. 1).

3. Experimental results

An overview of the main irradiation results is given in Table 2.

3.1. The IFA-651 experiment in Halden

For the IFA-651 experiment the following main observations can be made:

Table 1
The different experimental IMF-designs in a condensed overview

Experiment	IFA-651	IFA-652	OTTO
Objective	Comparison of YSZ-based IMF with MOX fuel	Comparison of CSZ-based IMF with (U, Th)O ₂ fuel	Comparison of homo/heterogeneous IMF concepts and MOX
Number of IMF targets	3	4	6
Composition ^a , and Pu ^{fis} - or U ^{fis} -density at BOI (g cm ⁻³)	(Er, Y, Pu, Zr)O ₂ ss; 0.75 (Er, Y, Pu, Zr)O ₂ ss; 0.75 (Er, Y, Pu, Zr)O ₂ ss; 0.60	(Ca, U, Zr)O ₂ ss; 0.98 (Ca, U, Zr)O ₂ ss; 0.98 (Ca, U, Th, Zr)O ₂ ss; 0.84 (Ca, U, Th, Zr)O ₂ ss;	(Er, Y, Pu, Zr)O ₂ ss; 0.37 (Y, Pu, U, Zr)O ₂ ss; 0.34 (Er, Y, Pu, Zr)O ₂ mid; 0.32 (Y, Pu, U, Zr)O ₂ mid; 0.31 (Er, Y, Pu, Zr)O ₂ mad; 0.31 (Y, Pu, U, Zr)O ₂ mad; 0.30
Instrumentation ^b	TF's and PF's in all targets, EF in two targets	TF's and PF's in all targets, EF in two targets	TF in three targets
Reactor	HBWR	HBWR	HFR
Fuel stack length (mm)	500	500	67
Fuel outer diameter (mm)	8.19	8.19	8.00
Max. coolant temperature (K)	506	506	553 (in average)
Target burnup (kW d cm ⁻³)	500	400–450	200

^a ss = solid solution; mid = micro-dispersion of IMF particles < 25 μm in MgAl₂O₄; mad = macro-dispersion of IMF particles 200–250 μm in MgAl₂O₄.

^b TF = central fuel thermocouple (or central elongation thermometer in case of 2nd rod of IFA-651), PF = pressure transducer, EF = fuel stack elongation sensor.

- The observed fuel temperatures are significantly higher than what can be seen of standard UO₂ for a given heat rate but in the expected range due to the lower thermal conductivity of YSZ-based IMF. The thermal conductivity (see Fig. 2) was derived from out-of-pile tests and was used in fuel performance calculations, where the measured temperatures could be satisfactorily reproduced. No dependence from burnup was observed. Trends of fuel time constants calculated from scram data confirm the steady state trends.
- A strong densification took place at beginning of irradiation, determined by the decrease of rod inner pressure and decrease in fuel column length. It led to completely dense material within a burnup of 40 kW d cm⁻³ and 100 kW d cm⁻³ (depending on the materials microstructure). Faster sintering was found for the material with very small grain size. The densification did not lead to increased fuel central temperatures, therefore the gap size was assumed to be maintained and the sintering was assumed to proceed from the centre to the rim [32].
- A later increase of rod inner pressure in combination with high power periods indicated FGR comparable to what would be expected for UO₂ fuel at these fuel temperatures. However, micro-sections of the rod subjected to PIE reveal significant grain growth in the inner part of the fuel and slight decoration of grain boundaries with small bubbles. Therefore, the driving force for FGR in this fuel was the grain boundary movement. This conclusion is also supported by fuel performance calculations, at least for the fuel with very small grain size.
- Fuel swelling of roughly 0.4% per 100 kW d cm⁻³ was derived from fuel elongation measurements. PIE of the attrition milled material revealed considerable fuel swelling in the hot centre. A density decrease was also found for this fuel in re-sinter tests. It was assumed that this effect was driven by impurities in the feedstock plutonia [33]. Micro-sections of the examined rod reveal large, well-distributed, round shaped pores but only slight grain boundary decoration, indicating that the gas swelling is driven by impurities.

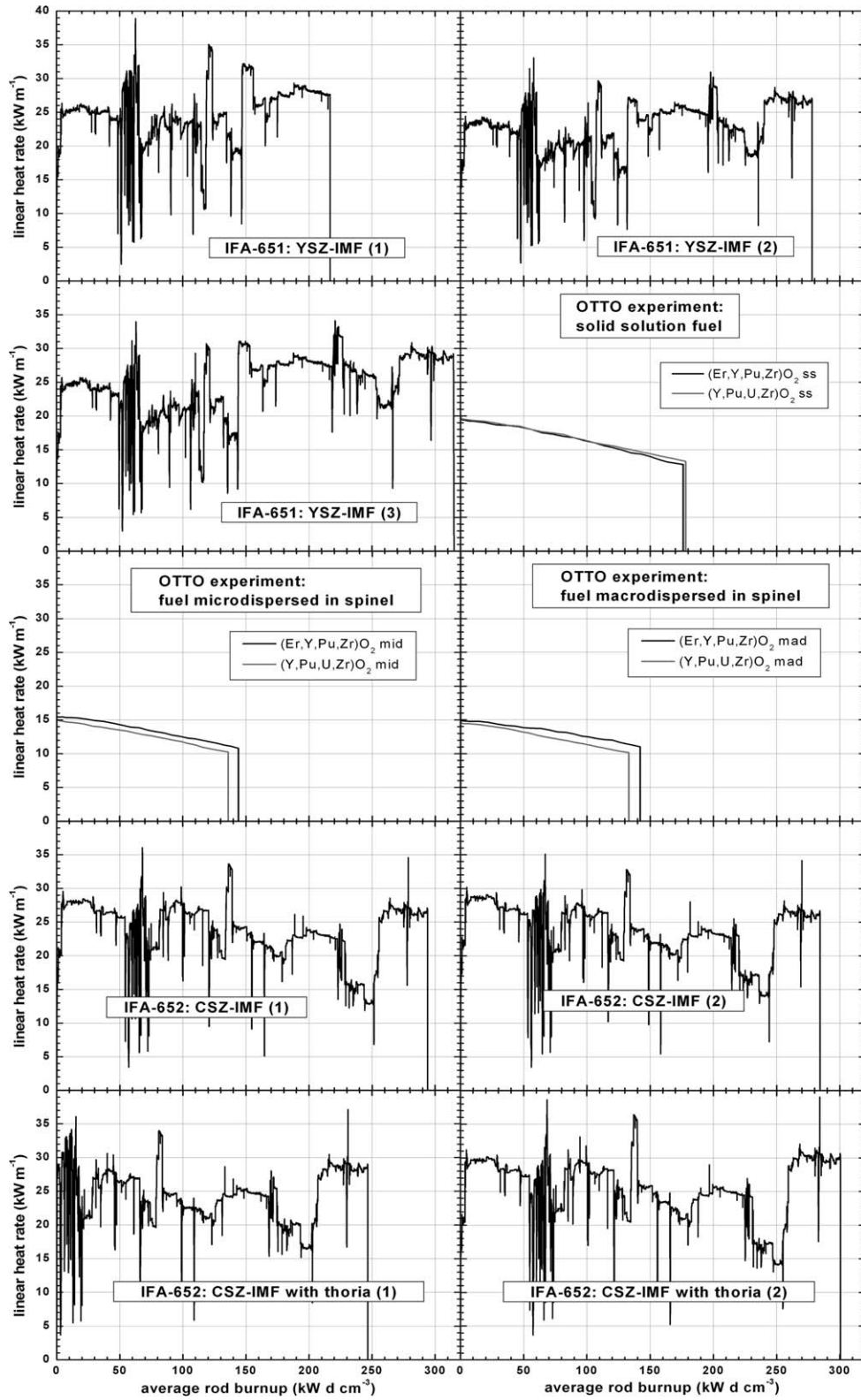


Fig. 1. Recorded power histories of the IMF rods from the three experiments. Ramps are mainly excluded from the plots for clarity reasons.

Table 2
Irradiation results in an overview

Fuel	Type ^a	Density (%TD)	Max. linear heat rate (kW m ⁻¹)	Fuel temperature		Burnup achieved (kW d cm ⁻³)	FGR (%)
				Estimated peak (K)	Measured (K)		
<i>IFA-651</i>							
(Er, Y, Pu, Zr)O ₂	ss	95.2	38.9	2175	1860	217	18
(Er, Y, Pu, Zr)O ₂	ss	95.4	34.1	1945 ^b	1912 ^c	315	32
(Er, Y, Pu, Zr)O ₂	ss	85.6–90.7	32.2	2285	2052	278	34
<i>IFA-652</i>							
(Ca, U, Zr)O ₂	ss	90.0	36.1	1905	1877	294	14 ^d
(Ca, U, Zr)O ₂	ss	90.0	35.1	2095	1870	284	13 ^d
(Ca, U, Th, Zr)O ₂	ss	93.0	36.1	2020	1771	247	22 ^d
(Ca, U, Th, Zr)O ₂	ss	93.0	38.6	2085	1839	300	25 ^d
<i>OTTO</i>							
(Er, Y, Pu, Zr)O ₂	ss	91.0	19.8	1790	–	176	9.2
(Y, Pu, U, Zr)O ₂	ss	92.7	20.0	1775	–	178	6.1
(Er, Y, Pu, Zr)O ₂	mid	94.5	15.6	1075	1226	144	0.1
(Y, Pu, U, Zr)O ₂	mid	93.8	15.2	1040	1032	136	–
(Er, Y, Pu, Zr)O ₂	mad	85.6	15.0	1135	1345	142	4.3
(Y, Pu, U, Zr)O ₂	mad	88.2	14.8	1085	–	133	6.0

^a ss = solid solution; mid = micro-dispersion of IMF particles < 25 μm in MgAl₂O₄; mad = macro-dispersion of IMF particles 200–250 μm in MgAl₂O₄.

^b Calculated for a pellet with central hole.

^c Measured with an extension thermometer through the whole fuel column.

^d Determined for an average assembly burnup of 240 kW d cm⁻³.

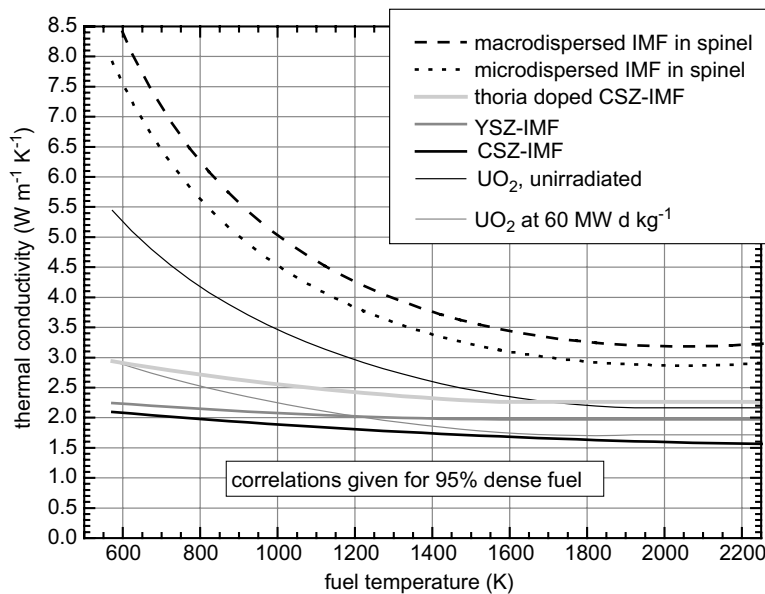


Fig. 2. Estimated thermal conductivity of the different fuel types.

3.2. The IFA-652 experiment in Halden

For the IFA-652 experiment the following main observations can be made:

- The CSZ-based IMF rods with and without thoria show stable fuel temperatures consistently higher than what can be seen of standard UO₂ for a given heat rate, due to the lower thermal

conductivity of these fuels (see Fig. 2). The thoria content seems not to affect the thermal behaviour significantly. Trends of fuel time constants calculated from scram data show a slight tendency of decrease with burnup, probably related to fuel swelling and gap closure.

- For the CSZ-based IMF, a densification of some 2.0% was determined by rod inner pressure measurements while for the IMF with thoria roughly 1.3% was deduced. The maximal densification was reached after a burnup of some 60 kW d cm⁻³. The comparison with the fuel elongation signal indicates that the densification is more distinct in the hot inner part, while the colder shoulders of the dished pellets determining the fuel stack elongation show less densification.
- Significant FGR associated with high power periods is seen in the rods of both fuel types. The fuel temperatures leading to significant FGR can be loosely correlated with the crossing of the Halden threshold for 1% FGR in UO₂. FGR in the thoria containing rods are higher although the temperatures seem to be comparable.
- Swelling of some 0.3% per 100 kW d cm⁻³ was derived from the fuel elongation data. A higher swelling in the hot pellet centre can neither be excluded nor proved by the data currently available.

3.3. The OTTO experiment in Petten

The general findings are:

- During irradiation, rod 4 with micro-dispersed fuel and urania additions failed. The failure is related to high radial swelling, i.e. most probably gaseous swelling. This is supported by the other micro-dispersed rod which stayed intact but showed also signs of high radial fuel swelling and very low fission gas release. The results of the homogeneous YSZ-based IMF are in line with the IFA-651 results.
- Temperature measurements were only performed in the two micro-dispersed fuel rods and one macro-dispersed fuel rod. They showed stable fuel temperatures and no tendency of decrease with burnup. Peak temperature estimations with the thermal conductivity correlations shown in Fig. 2 show a clear under prediction of the fuel temperatures (as the temperatures in the annular pellets with the thermocouples should be signifi-

cantly below the peak temperatures). This indicates that the thermal conductivity of the heterogeneous fuel is lower than anticipated. Unfortunately, the rod with the lowest fuel temperatures failed, i.e. micro-dispersed (Y,Pu,U,Zr)O₂.

- Densification and swelling behaviour was observed by means of regular neutron radiographs. The stack length of the solid solution fuel showed slight shrinkage, which indicates densification. The macro-dispersed spinel based samples show small axial swelling while the micro-dispersed samples show large swelling. The cladding of the rod with micro-dispersed (Y,Pu,U,Zr)O₂ in spinel was damaged, very probably due to high radial fuel swelling.
- FGR was measured at end of irradiation (EOI). The FGR shows a clear dependence on fuel temperatures. Furthermore, a certain correlation between fission gas retention and swelling can be observed.

4. Attempt of an evaluation of the experiments

The IMF-types described in this overview showed clearly their potential in Pu destruction. The final burnup of the OTTO experiment correspond to a Pu-depletion of 50–60%. Today, the burnup values for IFA-651 and IFA-652 corresponds to a Pu-depletion between 45% and 50% for IFA-651 (for the pins still under irradiation) and between 30% and 35% for the thoria-free IFA-652 rods. The target burnups for these two experiments correspond to a Pu-depletion of roughly 75% and 50%.

For all tested fuel types, the normalized temperatures are comparably stable with burnup. The measured fuel temperatures are in the expected range for the solid solution fuels, i.e. they correspond roughly with the thermal conductivity correlations used for the design of the experiments.

Principally, irradiation stability is good for the solid solution fuels. Considerable densification at beginning of irradiation was observed for several rods. However, this unfavourable behaviour did not lead to an increase in the fuel central temperature (IFA-651) or only to a very moderate increase (IFA-652). Contrary to the long development history of UO₂, no efforts were made so far for IMF to produce a stable pore size distribution and thus further development of the fabrication process could address the densification behaviour. Later in

life strong central swelling was observed at least for the IFA-651 rod that was subjected to PIE. As this swelling was related to impurities and clearly not to fission gas, an improvement of the fabrication process would also help to avoid this type of swelling in future irradiations. Furthermore, round-shaped pores observed during PIE indicate soft behaviour at elevated temperatures, which is of advantage to mitigate Pellet–Clad Mechanical Interaction.

The heterogeneous concepts irradiated in the OTTO experiment suffer in two aspects: (i) the fuel temperatures are higher than anticipated and thus the concept seems not to take full advantage of the thermal conductivity of spinel; (ii) the observed high swelling, at least for the micro-dispersed fuels, are not acceptable for commercial use.

The fission gas retention is clearly correlated with the fuel temperatures. In the case of the homogeneous fuel concepts, the FGR data are roughly comparable with what could be expected for UO₂ operating at the same temperatures. The fission gas retention potential for YSZ-based IMF could not be clearly identified as the small grain size had a crucial effect on FGR.

5. Conclusion and outlook

Different types of IMF showed good irradiation stability, namely YSZ- and CSZ-based IMF as well as the heterogeneous concept with macro-dispersed fuel particles. The heterogeneous concept with micro-dispersed fuel particles failed. Given the more complicated fabrication and the more difficult quality control of the heterogeneous concept, together with the limited operation temperature of the spinel matrix, the disadvantages may level out the advantages of lower fuel temperatures.

Stabilized zirconia seems to be the most promising option for future research. Its behaviour under irradiation and at the back end of the fuel cycle is outstanding [34]. Additions like yttria, calcia, erbia, and thoria are dissolved in the matrix and do not alter the irradiation behaviour significantly. Their addition can be decided upon the neutronic requirements. Temperature effects of additions seem to be small.

Special emphasis should be laid on the fabrication process and quality control for the next test fuel. Both fuels used for the irradiation tests, YSZ- and CSZ-based IMF, suffered from low density and/or unexpected density changes under irradiation. It is expected that the as-fabricated

density can be increased by using purified starting material and higher sintering temperatures.

The recent political developments (especially the GIF activities) have shifted the focus away from IMF employment in an LWR environment. Additionally the call for fast and efficient Pu-destruction is diminishing with new Fast Reactors being a future option. At the same time, the possible application area for IMF has widened: In new reactor types, the low thermal conductivity may be better acceptable, the high proliferation resistance and the softer mechanical properties may be of importance. Today's IMF research results form therefore a valuable input for future nuclear fuel development.

Acknowledgements

We gratefully acknowledge the fuel production work of F. Ingold, P. Heimgartner and R. Zubler at PSI, R. van der Laan at NRG and K. Lunde, B.C. Oberländer, G. Iversen and M. Espeland at IFE. The fuel fabrication for IFA-651 became possible by the initiative of G. Ledergerber and a collaboration with KAERI (D.S. Sohn, Y-W. Lee, H.S. Kim). We would like to thank H. Thesingh at HFR and Y. Minagawa and his group of experimentalists in Halden for their support and patience. Thanks also to U. Kasemeyer, G. Rossiter, J. Wright, and K. Bakker for taking care of the experiments. We are grateful for the PIE performed in Petten by F. van den Berg, W. Tams and R. Schuring and in Kjeller by H.K. Jenssen. The actinide and fuel research work at PSI was financially supported by swissnuclear (Swiss NPP's). The work performed at NRG was financially supported by JAERI and the Dutch Ministry of Economic Affairs.

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