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Yttrium stabilised zirconia inert matrix fuel irradiation at an international research reactor

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Abstract

Different concepts have been developed during the last decade to transmute transuranium elements (TRU) using uranium-free inert matrix fuels (IMF) in a once-through-cycle to reduce the amount of TRU in the nuclear waste. For today's LWRs yttrium stabilised zirconia (YSZ) and other oxides like alumina, spinel or ceria have been proposed as inert matrix materials. By employing IMF, a larger fraction of plutonium can potentially be consumed in comparison with MOX fuels without breeding new plutonium. The aim of the presented study is to measure the general thermal behaviour of YSZ-based IMF under irradiation conditions similar to those in current LWRs in direct comparison to standard MOX fuel. Of particular interest are the fuel thermal conductivity (and its degradation with burnup), fission gas release (FGR), fuel densification and fuel swelling. A secondary aim is the direct comparison of the fuel performance between YSZ-based IMF and MOX fuel. The irradiation is performed under HBWR conditions and has reached an average assembly burnup of $\sim$300 kW d cm^{-3} until the end of 2004, which is equivalent to $\sim$29 MW d kg^{-1} for the MOX fuel.

1. Introduction

The present large stockpiles of reactor and weapons grade plutonium worldwide are of major concern from non-proliferation and economic aspects. To reduce these stockpiles, several options are under discussion. The plutonium could be treated as waste and put in a final disposal, thus loosing the energy potential of the material, which is of economic interest. Reprocessing the plutonium and closing the fuel cycle with the use of fast reactors would optimise the utilisation of nuclear fuel, but this development is delayed due to political reasons. However, today's recycling is focused on producing uranium plutonium oxide fuel, the so-called mixed oxide fuels (MOX). MOX is already in commercial use in light water reactors (LWRs), which is a reasonable possibility to use the plutonium energy source. Unfortunately, the total amount of plutonium is not significantly reduced due to the breeding of plutonium in the uranium matrix. In addition, recycling of second or third generation plutonium as

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MOX in LWRs seems to be economically unfavourable. To reduce the plutonium stockpile and also the amount of other transuranium elements (TRU) in the nuclear waste, concepts have been developed during the last decade to transmute TRU using uranium-free inert matrix fuels (IMF) in a once-through-cycle. For today's LWRs yttrium stabilised zirconia (YSZ) and other oxides like alumina, spinel or ceria have been proposed as inert matrix materials [1–4]. By using IMF, a larger fraction of plutonium can potentially be consumed in comparison with MOX fuels without breeding new plutonium.

The aim of the presented study is to measure the general thermal behaviour of YSZ-based IMF under irradiation conditions similar to those in current LWRs in direct comparison to standard MOX fuel. Of particular interest are the fuel thermal conductivity (and its degradation with burnup), fission gas release (FGR), fuel densification and fuel swelling. A secondary aim is the direct comparison of the fuel performance between YSZ-based IMF and MOX fuel.

The presented study is a joint experiment of the Korean Atomic Energy Research Institute (KAERI), British Nuclear Fuels plc (BNFL), Paul Scherrer Institute (PSI) and the OECD-Halden-Reactor-Project (HRP).

The irradiation is performed under Halden Boiling Water Reactor (HBWR) conditions (i.e., ~34 bar D_2O at ~510 K) and has reached an average assembly burnup of ~300 kW d cm^{-3} until the end of 2004, which is equivalent to ~29 MW d kg^{-1} for the MOX fuel with a target burnup of ~500 kW d cm^{-3}, which is equivalent to ~48 MW d kg^{-1} for the MOX fuel. This report summarizes the results after six cycles of irradiation (671 days) and gives the first post irradiation examination (PIE) results of an IMF rod after a burnup of ~210 kW d cm^{-3}.

2. Experimental setup

The irradiation experiment (IFA-651) contains one rig (Fig. 1) holding a cluster of six highly instrumented rods (Fig. 2). Two rods with IMF pellets and two rods with MOX pellets were manufactured at PSI using a dry attrition milling process (ATT) developed at KAERI. Another IMF rod delivered by PSI was manufactured via a co-precipitation route (CO) using the well known PSI internal gelation method. The last rod contains BNFL standard MOX manufactured by using the short binderless route (SBR) [5,6]. The fuel compositions were determined such that all rods have comparable linear heat rates.

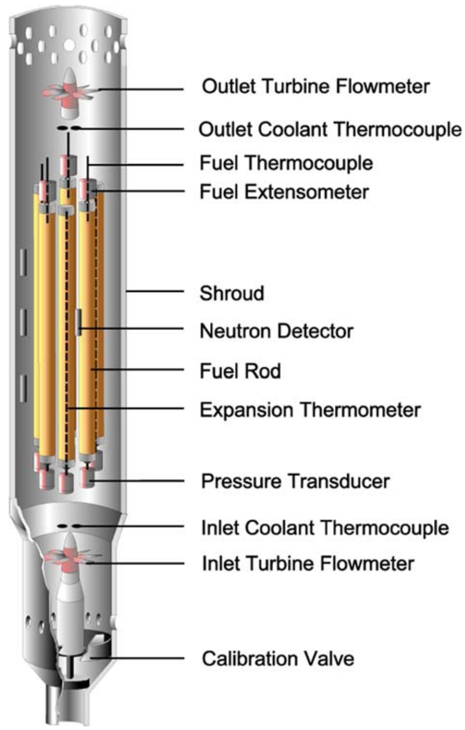

Fig. 1. Schematic view of IFA-651 rig.

Two of the six test rods are instrumented with expansion thermometers (ET) while each of the other four rods have a thermocouple (TF) at the top end. All rods have pressure transducers (PF) at the bottom end. In addition, three rods, two IMF and one MOX, are instrumented with stack elongation detectors at the top of the fuel stack.

In order to obtain an accurate record of the axial flux distribution, the rig is instrumented with three co-linear neutron detectors (NDs 1, 2 and 5) at three different elevations. The radial flux distribution is measured by three axisymmetric neutron detectors (NDs 2, 3 and 4), which are placed at the central elevation corresponding to the fuel stack midpoint. This is also the axial position of maximum flux in the HBWR. An overview of the rod characteristics is given in Table 1.

3. Irradiation history

The rig is irradiated under HBWR conditions. Irradiation of the first loading began at the end of

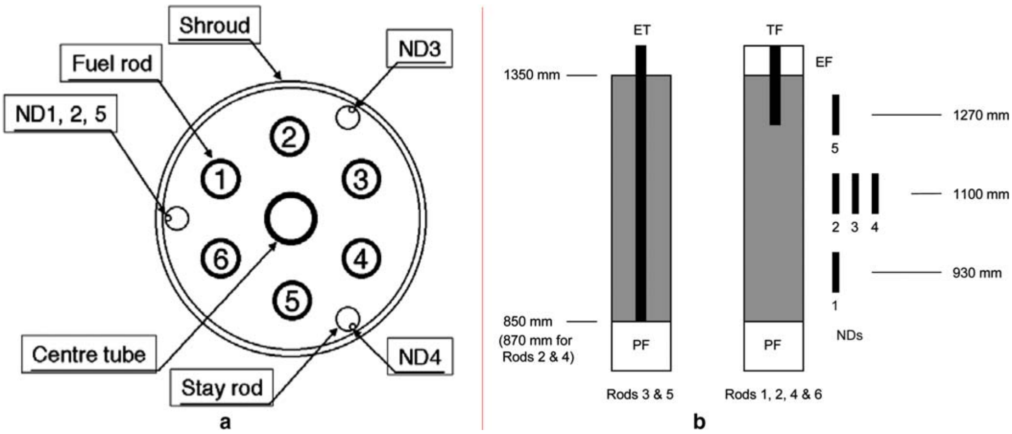

Fig. 2. Radial and axial geometry of IFA-651 rig.

Table 1
Summary of rod fabrication data in IFA-651

Rod number	1	2[b]	3	4	5	6
Fuel pellets						
Fuel type	MOX-SBR	IMF-ATT	MOX-ATT	IMF-CO	IMF-ATT	MOX-ATT
Fuel supplier	BNFL	PSI	PSI	PSI	PSI	PSI
Fissile Pu[a], wt% metal	5.98	14.2	5.85	13.6	14.2	5.85
Density[a], g/cm^3	10.401	6.294	10.404	5.833	6.298	10.434
Density, %TD	94.6	95.5	94.6	88.4	95.5	94.9
Outer diameter, mm	8.2					
Hollow pellet ID, mm	1.8	1.8	2.0	1.8	2.0	1.8
Pellet length, mm	12.1	9.8	9.8	9.8	9.8	9.8
Dishing	Both ends					
Cladding						
Material	Low tin Zircaloy-4					
Outer diameter, mm	9.5					
Inner diameter, mm	8.3					
Fuel rod						
Diametral gap[a], mic	158 ± 4.3	168 ± 2.4	157 ± 4.6	166 ± 4.0	173 ± 3.5	162 ± 4.2
Fill gas and pressure	Helium, 10 bar g					
Free volume[a], cm^3	8.4	9.4	8.3	10.4	8.4	8.5
Active fuel length[a], mm	498.7	477.4	500.5	479.9	497.8	500.5
Active fuel mass[a], kg	0.2681	0.1542	0.2532	0.1421	0.1519	0.2686
Instrumentation	TF, PF	TF, PF, EF	ET, PF	TF, PF, EF	ET, PF	TF, PF, EF

[a] Measured values (mean ± one standard deviation, where applicable).
[b] Discharged at end of loading 1.

June 2000 and the assembly average burnup at mid of May 2003 (end of fourth cycle and of first loading) was $\sim$210 kW d cm^{-3} (equivalent to $\sim$21 MW d kg^{-1} for the MOX fuel). IMF rod number 2 was discharged for PIE and irradiation of the second loading began at the beginning of February 2004. The assembly average burnup at the end of 2004 (end of sixth cycle) was $\sim$300 kW d cm^{-3}. The target burnup is $\sim$500 kW d cm^{-3}, which should be achieved over a period of $\sim$7 calendar years. The rig was first in core position 6–29 during the first loading and was then moved to 6–32 with a rig rotation of $\sim$180° during the shutdown in July/August 2001 ($\sim$58.7 kW d cm^{-3}). The rig has then been moved to 4–19 in the September–December 2002 shutdown ($\sim$140 kW d cm^{-3}) were it has stayed also in the second loading. Both changes in core position were performed to allow higher linear heat rates (LHRs) to be achieved in an attempt to induce a large amount of FGR in the IMF rods.

After the first movement, LHRs up to 35 kW m^{-1} were achieved at temperatures of 1500–1600 °C measured in the IMF rods and 1575–1675 K in the MOX rods. These temperatures were also achieved in the second loading with LHRs of around 30 kW m^{-1}. Fig. 3 shows the irradiation history and the burnups of rig IFA 651 during the first six irradiation cycles. The burnup is given in kW d cm^{-3} to make IMF and MOX fuel comparable.

Due to handling damage during the June–August 2001 shutdown (~58.6 kW d cm^{-3}), the thermocouple cables for rod 2 (IMF-ATT) had to be respliced. The temperature values from IMF rod 2 after this point should therefore be interpreted with some special recalculation. A comparison with the temperature in IMF rod 5 (Fig. 5) shows that the recalculated values seem reasonable for this thermocouple after the damage.

3.1. Post irradiation examination

Rod 2 (IMF-ATT) was discharged from the rig during the May–July 2003 shutdown for interim PIE at Kjeller. The average rig burnup at this time was ~210 kW d cm^{-3}, whereas the burnup of rod 2 was ~217 kW d cm^{-3}. The ongoing PIE includes dimensional and density measurements, axial gamma scanning, neutron radiography, ceramography, rod puncturing and fission gas analysis. The complete results of PIE are to be published. Some preliminary results are already mentioned in this report.

Diameter profiles measurements along the rod axis at 0°, 45°, 90° and 135° orientations were performed in a vertical profilometer bench. The axial positions coincide with the scale given under neutron radiography, i.e., the positions are given relative to the PF tip at the lower end of the rod. Length measurements were performed between the upper and lower *v*-grooves at 0°, 45°, 90° and 135° orientations.

The main objective of neutron radiography is to produce images for examination of the fuel stack and detailed information about continuity of the fuel stack column, instrumentation details, possible moisture inside the rod and cladding corrosion. The neutron radiography images also provide informa-

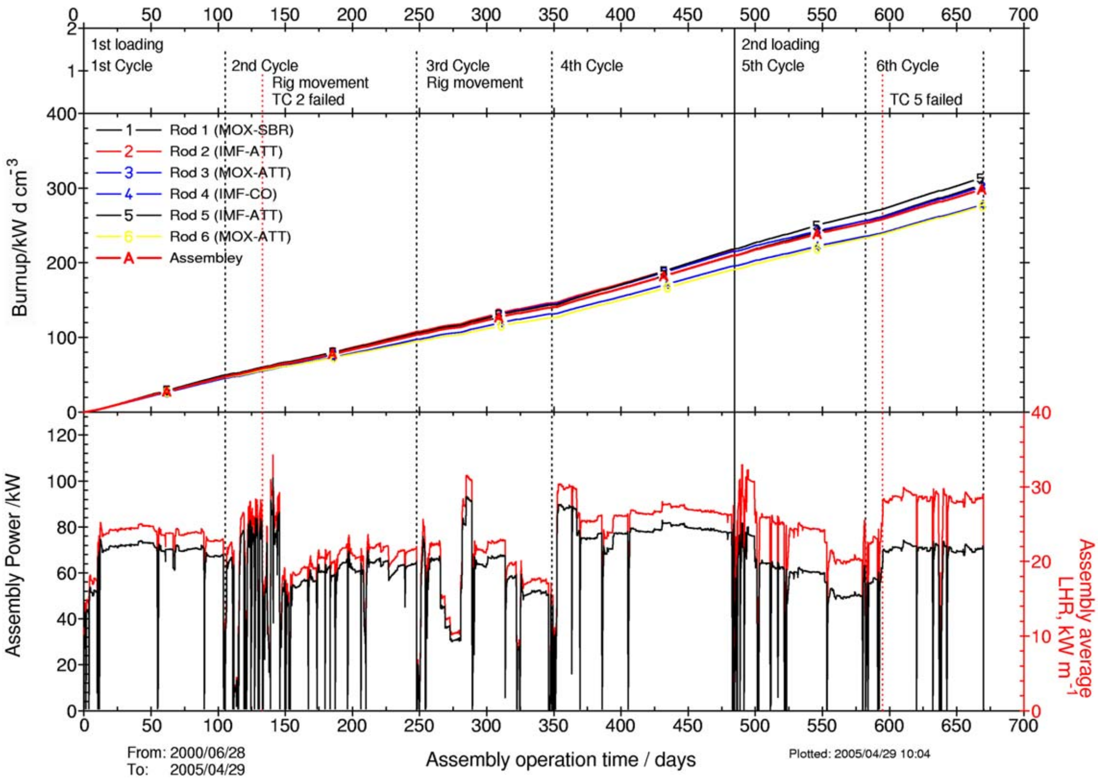

Fig. 3. Irradiation history of IFA-651 first and second loading.

tion that is used to define cutting positions to obtain samples for destructive examinations.

The neutron radiography facility at the JEEP II reactor consists of a 10-inch horizontal beam channel into which a conical collimator has been inserted. The aperture at the inner, narrow end of the collimator is 1 cm in diameter, and the distance from the inner end to the position of the converter to be activated is 390 cm. A cassette loaded with Dy-foil is brought into contact with the aluminum tube into which the fuel rod is inserted. Control of the irradiation time is achieved by shutting a concrete door at the exit of the collimator. The activation image formed in the Dy-foil is transferred to a single coated X-ray film by placing the foil and film in contact and allowing the decay radiation from the foil to produce the latent image on the film. An irradiation time of 4.5 min was used for the observation of the cladding and instrumentation as well as for the fuel stack column, whereas the following film exposure time was about 20 h.

To determine the amount of fission gas released and the free volume, fuel rod 2 was punctured in the lower end (PF region). Expansion pressure became stable immediately after puncturing, i.e., no problems with degraded gas communication in the rod. Gas samples taken after the puncturing were analysed using mass spectrometry.

4. Results and discussion

The operating temperatures in the IMF rods were considerably higher than those in the MOX fuel due to the lower thermal conductivity of the IMF (Fig. 4). The temperature behaviour of the MOX rods were stable throughout the third and fourth cycles with the exception of two periods for rod 1 (MOX-SBR). This seems to be due to either FGR or to the rapid power changes. An examination of the normalised temperatures shows that the IMF rods consistently had higher temperatures than the MOX rods. A large drop in the normalised temperatures for IMF-ATT rod 5 at the beginning of the fourth cycle was observed which can be explained by a change in the temperature-LHR relationship. The MOX rods and IMF-CO rod 4 shows little change in the temperature-LHR behaviour.

The fuel stack elongation signals were examined for both hot stand-by and steady state conditions. The signals for the IMF rods (2 and 4) indicate that the sensors became stuck. The thermal response to scrams was examined for rods 1, 4 and 6. Rod 4 exhibits a slower response due to it's lower density and therefore lower thermal conductivity. All rods exhibit a slight degradation in the thermal response in the period during which fission gas is released, possibly due to gap poisoning. A strong densification took place at the beginning of irradiation, determined by the decrease of the rod internal

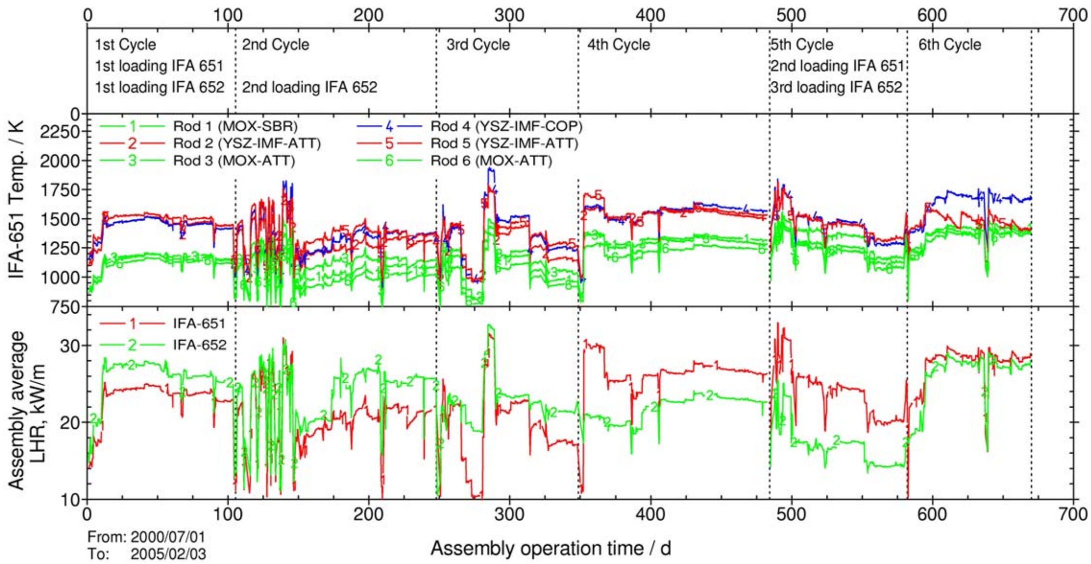

Fig. 4. Fuel temperature (measured) of IFA-651.

pressure and fuel stack length. Faster sintering was found for the IMF-ATT fuel with very small grain size. Since the densification did not lead to increased fuel centerline temperatures, the gap size was assumed to be maintained and the sintering was assumed to proceed from centre to the rim [7].

The rod internal pressures were examined and normalised to hot stand-by conditions (zero power and a coolant temperature of 510 K). The normalised pressures indicate that significant fission gas release occurred during the third and fourth cycles for all rods except rod 3 (MOX-ATT), which remained below the FGR threshold. All the other rods, both IMF and MOX, showed significant FGR upon crossing the FGR threshold for UO_2-fuel, also known as the Halden threshold [8], indicating that it may be applicable to both MOX fuels and IMF. However, caution must be applied since the threshold was rapidly crossed in most instances rather than by a slow stepwise increase in the LHR's. The threshold was also exceeded by a large amount in most cases and therefore it is difficult to exactly ascertain the position of the FGR thresholds for IMF. Furthermore, micro-sections of the rod subjected to PIE revealed significant grain growth in the inner part of the fuel. Suggesting the driving force for FGR in this fuel was the grain boundary movement. This conclusion for the fuel with very small grain size is also supported by fuel performance calculations. Therefore, the fission gas retention potential for YSZ-based IMF could not yet be clearly identified.

At the end of the fourth cycle the fission gas release rate (without swelling) was estimated from the normalised pressures at ∼19%, ∼20% and 18% for rods 2, 4 and 5, respectively. For the MOX rods, the estimated gas release was ∼10%, ∼3% and 7% for rods 1, 3 and 6. At the end of the sixth cycle, FGR for the IMF rods of 34% (rod 4) and 32% (rod 5) were estimated. The three different MOX rods 1, 3 and 6 show FGR of 32%, 8% and 25%, respectively.

During PIE of the discharged rod 2, the cladding of the fuel rod showed no defected areas. The diameter was found as fabricated, ∼9.5 mm, and the length measurements in all measured orientations resulted in a length of ∼619.4 mm. The axial gamma scanning results show predominately distributions

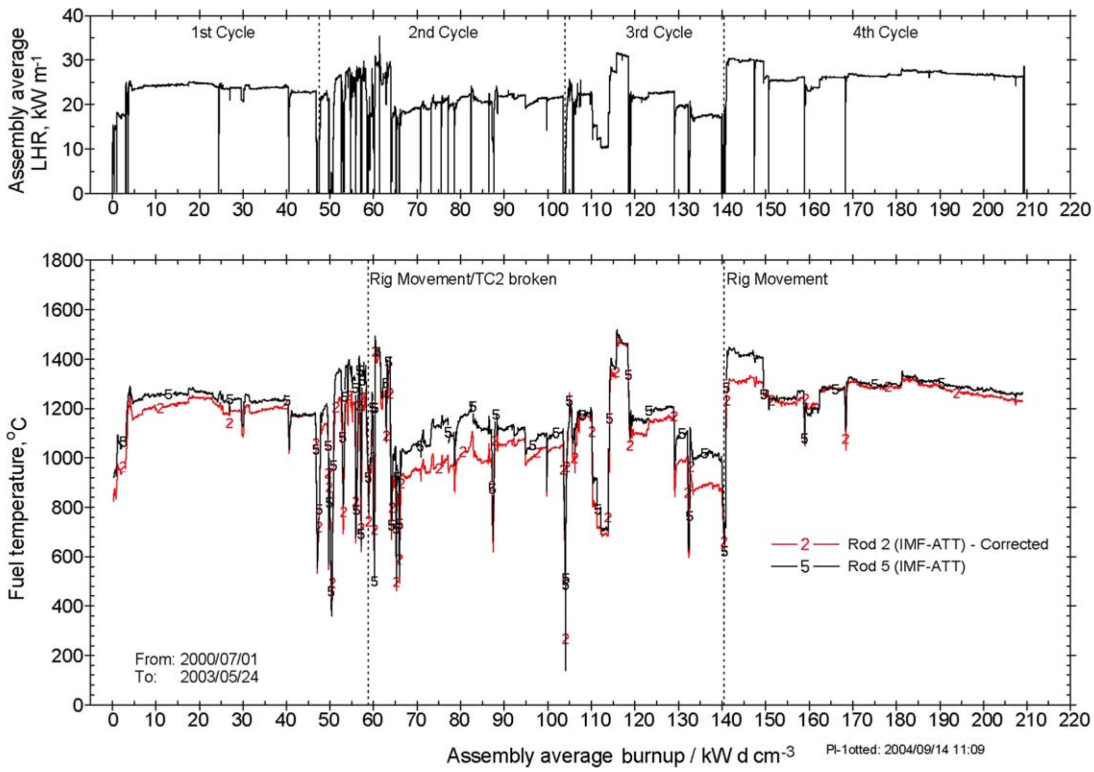

Fig. 5. Recovered signal of thermocouple 2.

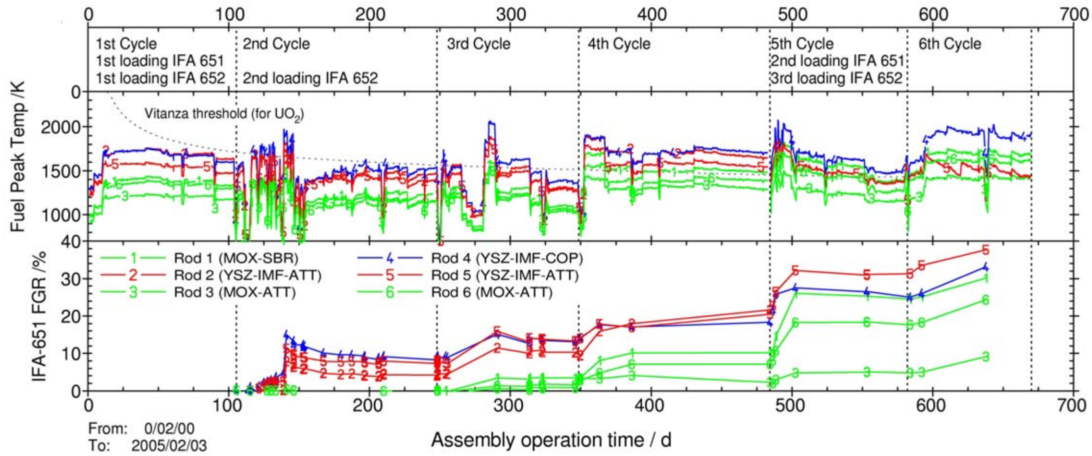

Fig. 6. Fuel peak temperature and FGR against burnup of IFA-651.

of gross gamma of ^{95}Nb and ^{137}Cs. Due to the rather short irradiation and cooling time, as ^{95}Nb and ^{95}Zr, with a half-life of 35 and 65 days, respectively, dominate. The burnup distribution was relatively flat. The annular pellets, which hold the thermocouple, had slightly lower fuel temperatures. Transversal crackings, indicated by mid-dips, were found in the solid pellets (Fig. 6).

From the neutron radiography given in Fig. 7, it was observed that the fuel stack length (474 mm) is shorter compared to the as fabricated length (477.4 mm). The densification in length is therefore approximately 3–4 mm or about 8%.

A total gas amount of 107.48 cm^3 at a pressure of 1.262 MPa was found during puncturing. The free volume of the rod was found to be 8.63 cm^3, while the initial free volume of the rod was 9.4 cm^3, which is in good agreement with 4.6% swelling (given by density measurements) of the fuel volume. The total gas amount found from puncturing test and mass

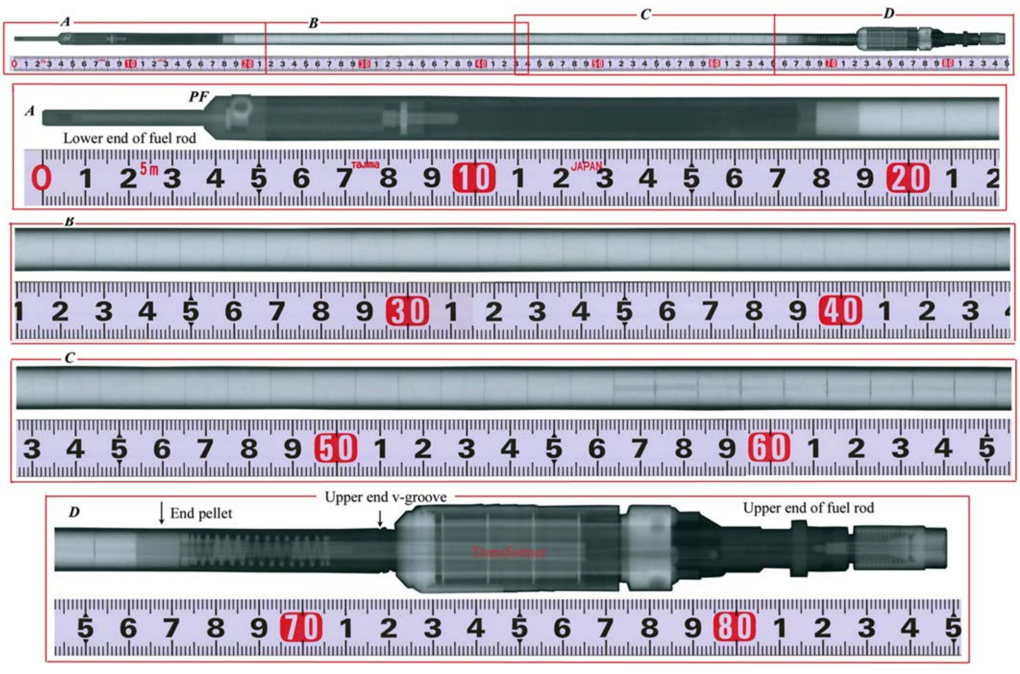

Fig. 7. Neutron radiography of IFA-651, rod 2.

Table 2
Results of mass spectrometric analysis of gas contents of IFA-651, rod 2

	He	Ar	Kr	Xe	N_2/CO	O_2	Total	Punct.	
Mass, mg	14.46	0.02	5.84	167.13	0.89	0.18			
Volume, cm^3	81.01	0.01	1.56	28.54	0.71	0.13	111.96	107.48	Average
				Released fission gas (cm^3):			30.95	26.47	28.71

spectrometric measurements given in Table 2 are in good agreement with each other, i.e., ~3% deviation on the two results. The amount of He found was 81.01 cm^3, which corresponds with the initial He content. The amount of fission gas released could be calculated by subtracting the He volume from the total volume. Fractional fission gas release is defined as the ratio of the released to the produced amount of fission gas. Assuming a production of about 30 cm^3 of fission gas per MWd burnup, the produced amount of fission gas was calculated to be 159.13 cm^3, using a fuel mass of 0.1542 kg and a average rod burnup of 217 $kW\,d\,cm^{-3}$ (34.4 $MW\,d\,kg^{-1}$). The fraction of fission gas released is then about 18%, which is in good agreement with the predicted 19%.

5. Conclusions

The irradiation experiment is ongoing and currently shows no major problems, except for two failed stack elongation detectors. All IMF rods showed stable irradiation behaviour, but consistently had higher temperatures compared to the MOX rods due to the lower thermal conductivity. However, normalised fuel temperatures did not show any dependency on burnup. Even the strong densification that took place at the beginning of the irradiation did not lead to higher central temperatures. Therefore, the gap size was assumed to be maintained and the sintering was assumed to proceed from the centre to the rim.

The IMF rods released fission gas earlier than the MOX rods due to the higher fuel temperatures. The FGR of 18% in IMF rod 2 measured by puncturing is in good agreement with the predicted 19%. FGR was found to be comparable to what would be expected for UO_2 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. Therefore, the driving force for FGR in this fuel was the grain boundary movement. For the fuel with very small grain size, this conclusion is also supported by fuel performance calculations.

PIE of IMF-ATT 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 plutonia feedstock.

Higher burnup will be accumulated in the future as the irradiation is still ongoing.

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