Performing Organisation	Document no.:	Date
Institutt for energiteknikk Halden	IFE/HR/E-2010/002	2010/02/02

Project/Contract no. and name	Client/Sponsor Organisation and reference:

Title and subtitle

Comparative study of inert matrix fuels concepts from IFA-652 experiment

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Abstract

Key Words

ISSN	ISBN	Numbers of Pages
0807-5514	978-82-7017-805-6 (printed) 978-82-7017-806-3 (electronic)	8

Supplementary Data

Presented at the 17th International Conference on Nuclear Engineering ICONE 17, July 12-16, 2009, Brussels, Belgium

Proceedings of the 17th International Conference on Nuclear Engineering
ICONE17
July 12-16, 2009, Brussels, Belgium

ICONE17-75151

COMPARATIVE STUDY OF INERT MATRIX FUELS CONCEPTS FROM IFA-652 EXPERIMENT

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ABSTRACT – *Notwithstanding Mixed OXide (MOX) fuel is considered the most probable option to reduce the separated plutonium stockpiles, in advance on a significant deployment of Fast Breeder Reactors (FBR), inert matrix fuels (IMF) remain a possible way to accomplish this task in nowadays Light Water Reactors (LWR) fleet. A high burning efficiency achieved by preventing new plutonium build-up under irradiation (U-free fuel), a proved high radiation damage and leaching resistance are fundamental requirements when a once-through fuel cycle strategy is planned. Amongst other options, both calcia-stabilised zirconia (csz) and thoria fulfill these criteria standing as the most promising matrices to host plutonium. The investigation of the under-irradiation performance of the calcia-stabilised zirconia fuels is underway, with this regard the thermal conductivity, lower than Uranium OXide (UOX) and MOX fuels, has proved to play a fundamental role.*

For this reason, ENEA has conceived a comparative in-pile testing (IFA-652 experiment) of viable U-free inert matrix fuel concepts that has been performed in the Halden Heavy Boiling Water Reactor (HBWR) of the Organisation for Economic Co-operation and Development (OECD). The accumulated burnup, under typical LWR conditions, at the end of the irradiation history, was about 90% of the foreseen 45 MWd/kgU_{eq}. The test-rig is a six-rod bundle loaded with IM, IMT and T innovative fuels. IM and T fuels have, respectively, csz and thoria matrix, the fissile phase is High Enriched Uranium (HEU) oxide (UO_2 93% ^{235}U enriched). IMT is a ternary fuel composed of csz+thoria matrix and HEU oxide as fissile phase. Thoria is added in IMT fuel to improve the low IM reactivity feedback coefficients. Pins are instrumented providing fuel centreline temperature, pin inner pressure and fuel stack elongation measurements.

This paper is focused on the csz-based IMF under irradiation performance, especially regarding the Fission Gas Release (FGR) topic. Low FGR is fundamental to achieve a high burnup performance and, in the case of IMF, an effective plutonium incineration. Relying on the IFA-652 experimental dataset, especially regarding the ramping-up to the peak power, and the built-in FGR modelling of TRANSURANUS (TU), a well-known nuclear fuel performance code, we expect to provide a contribution on related issues such as the existence of a correlation of IMF FGR onset with the Vitanza threshold and the main processes driving the FGR in these innovative fuels.

KEYWORDS:
Inert Matrix Fuel, FGR, Fuel Rod Performance Code

1 INTRODUCTION

One of the key issues dealing with the public concern on nuclear energy is the plutonium management due to the radiotoxicity and the misuse risk associated to it. To this purpose, worldwide efforts have been devoted at finding efficient solutions to reduce both the radiotoxicity and the amount of separated plutonium stockpiles [1, 2]. One of most challenging ENEA Research and Development (R&D) efforts has been focused on the concept of a U-free inert matrix fuel aiming at burning separated plutonium both weapon and reactor grade in Light Water Reactors (LWR). The proved advantage of these innovative fuels, compared with the commercial Mixed OXide (MOX) fuel, is the great improvement in the proliferation resistance issue by avoiding new plutonium build-up under irradiation meanwhile the fissile plutonium undergoes fission. In this case, a once-through fuel cycle strategy becomes viable provided that a

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careful choice of the matrix material is made. ENEA has envisaged that calcia-stabilised zirconia and thoria could fulfil required specifications, in particular three inert matrix fuel concepts have been conceived for an in-pile test. The irradiation experiment, so-called IFA-652, has been carried out in the Halden Heavy Boiling Water Reactor (HBWR) in the context of the ENEA membership in the Halden Reactor Project (HRP), the largest joint project of the Organisation for Economic Co-operation and Development/Nuclear Energy Agency (OECD/NEA) [3-5]. The experimental data from the instrumented test rig, loaded with IM (Calcia-Stabilised Zirconia- csz - matrix), T (thoria matrix) and IMT (csz + thoria matrix) fuels, has been collected in a specialised environment for the off-line analysis. The fissile phase of investigated fuels is High Enriched Uranium (HEU) oxide (93% ^{235}U enriched) instead of PuO_2 for practical reasons [6, 7]. In this paper, aiming at discussing the Inert Matrix Fuels (IMF) Fission Gas Release (FGR) topic, the responses of rods 1, 2 (IM) and 3, 6 (IMT) in IFA-652 under ramping power have been studied in three different timeframes. The investigated burnup domain extends up to 25 MWd/kgUO_2. The correlation of the IMF FGR onset with the Vitanza threshold curve is discussed on the basis of the IFA-652 experimental dataset. Afterwards a simulation by the TRANSURANUS code (TU) has been performed in related irradiation histories, aiming at investigating the deviation of FGR predictions in calcia-stabilised based fuels with respect to the FGR modelling of UO_2 fuel reference [8, 9]. We expect to give a contribution to the ongoing studies regarding the IMF modelling, being aware that amongst related topics, FGR is the most challenging and troublesome feature in the nuclear fuel performance modelling.

2 IFA-652 HALDEN EXPERIMENT

The IFA-652 test rig is a six-pin fuel bundle, where rod 1 (all hollow pellets) and 2 are loaded with IM fuel, 3 and 6 with IMT fuel, 4 and 5 with T fuel (see Table 1 and Fig. 1) [6, 7]. The fuel rods have been fabricated at the Institutt For Energiteknikk (IFE), Kjeller (Norway). On the basis that the fissile material, irrespective of the adopted nuclide, should have a negligible effect on the performance of the inert matrices under irradiation, in this experiment, for practical reasons, the fissile phase was HEU oxide instead of PuO_2. IFA-652 was however considered a first experiment to be followed by a second one, fully representative with plutonium. The fabrication has been based on dry powders mixing route [10, 11]. All pins were equipped with fuel temperature thermocouple (TF) and internal pressure transducer (PF). Three pins (2, 4, 6), house a fuel stack elongation sensor (EF). The rig was also equipped with co-linear neutron detectors (ND) to accurately record the neutron flux along the axial direction and at the mid-plane section (Fig. 1). A 900 s acquisition period was assumed in storing the in-pile measurements in the Test Fuel Data Bank (TFDB) [12].

This experiment has been focused on the IMF performance under irradiation especially regarding thermal conductivity and its degradation with burnup, the densification-swelling behaviour and the FGR. In Table 2, the nominal fuel pin design parameters, representative of a typical LWR fuel rod, are resumed. The in-pile test (2000-2005) succeeded in accomplishing, for the fuels under study over than 90% of the 45 MWd/kgU_{eq} target burnup. Rods instrumentation performed well but the PF1, PF2, PF6 (inner pressure gauge) and the TF1, TF6 (fuel centreline temperature thermocouple) showed an unreliable response in the last part of the irradiation history.

Table 1. IMF matrix test: IFA-652

Rods	*1-2*	*3-6*	*4-5*
density, g·cm^{-3}	5.637	6.995	8.180
density, %TD	89.9	93.2	82.1
csz, wt%	81	45	-
thoria, wt%	-	39.2	88.3
HEU, wt%	19	15.8	11.7

The TFDB system is the reference source of the IFA-652 experimental data [12].

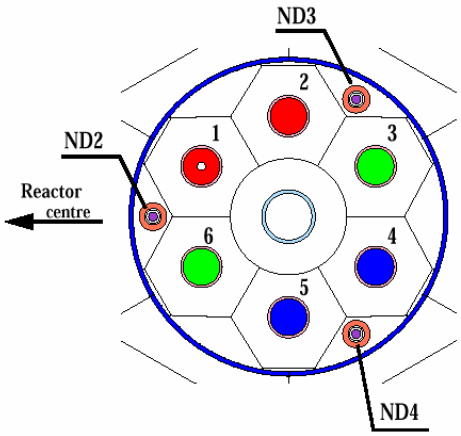

Figure 1. IFA-652 test rig (cross section)

Table 2. IFA-652 pin design

	Nominal values
Thermal shield pellets	UO_2
Cladding material	Zircaloy 4
Fill gas	He
Pre-pressurisation, MPa	1.0
Pin active length, mm	500
Cladding outer diameter, mm	9.50
Fuel pellet outer diameter, mm	8.19
Fuel pellet inner diameter, mm	1.80
Fuel-cladding gap, μm	170

In this paper we focused on the performance of rods 1, 2, 3 and 6 in three different timeframes at increasing burnup up to 25 MWd/kgUO_2. The peak heat ratings of each rod are resumed in Table 3. In Fig. 2 the heat rating at each identified time interval is shown (rod 2).

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3 POWER RAMPS EXPERIMENTAL DATA

In this analysis we aim at investigating the FGR performance of the IFA-652 inert matrix fuels. Three timeframes have been identified in which power ramps occur at different burnup (< 25 MWd/kgUO_2). The fuel temperature at the FGR onset and the related burnup are presented in Table 4.

Table 3. Peak heat rate (kW/m) at each time interval

Rods	*TimeFrame 1*	*TimeFrame 2*	*TimeFrame 3*
1	36.1	33.7	28.6
2	35.1	32.1	28.2
3	36.1	34.0	30.5
6	38.7	36.4	32.0

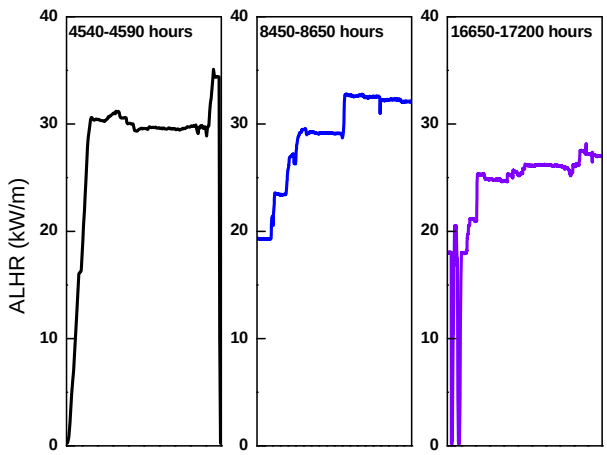

Figure 2. Heat ratings in investigated timeframes (rod2)

In Fig. 3-6 the experimental inner pressure versus the pin average linear heat rate, from the IFA-652 database, are shown. The time windows are identified through their burnup also resumed in Table 4.

In general it is noticed that the inner pressure grows up at higher burnup proving that FGR occurred meanwhile, provided that low fuel swelling is expected, furthermore a clear deviation is envisaged from a roughly proportional response on heat rating suggesting a burst fission gas release. These events occur at lower rating moving through the irradiation timeframes confirming, in the case of calcia-stabilized inert matrix fuels, the impact of burnup on FGR. In particular in rods 1, 2 the increase at the selected ramps adds up to get the final inner pressure level while in rods 3, 6 the final pressure is not fully explained by investigated burst releases.

In the first time interval, rod 3 experimental data indicate that no FGR occurs while, in the second, FGR onset is envisaged with better accuracy in comparison with presented results, see Fig. 5.

noting that the IMF we are dealing have comparable thermo-physical properties and under similar irradiation conditions, the experimental data suggests that FGR in rods 3, 6 is higher than in rods 1, 2.

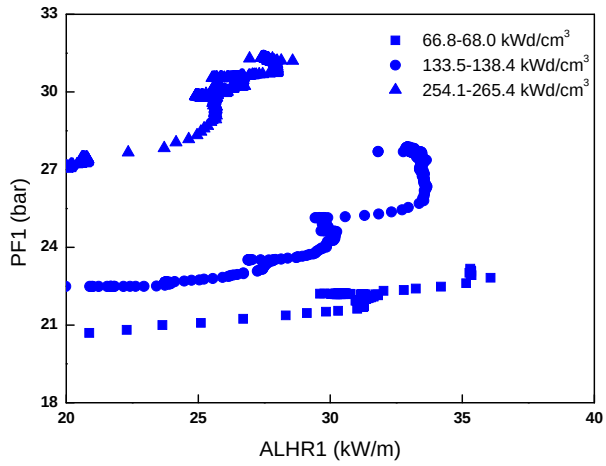

Figure 3. Inner pressure vs. average linear heat rate (rod 1)

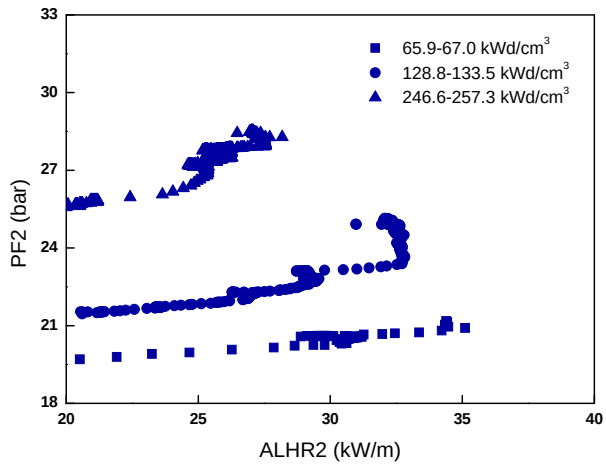

Figure 4. Inner pressure vs. average linear heat rate (rod 2)

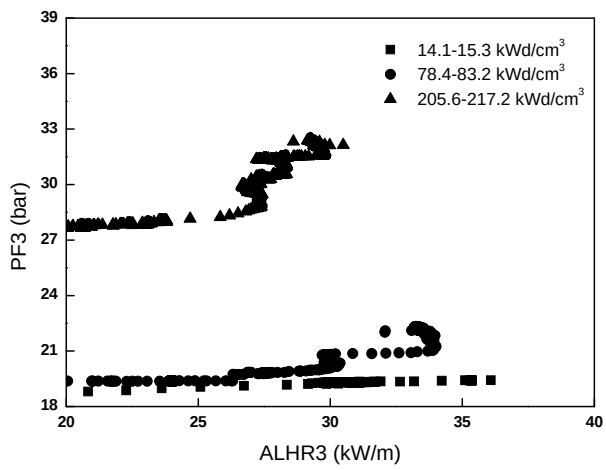

Figure 5. Inner pressure vs. average linear heat rate (rod 3)

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Table 4 Power ramps description

	Timeframe 1 4540-4590 hours		Timeframe 2 8450-8650 hours		Timeframe 3 16650-17200 hours	
rods	burnup kWd/cm^3	T_{fuel} °C	burnup kWd/cm^3	T_{fuel} °C	burnup kWd/cm^3	T_{fuel} °C
1	66.8-68.0	1430-1450*	133.5-138.4	1340-1360*	254.1-265.4	1180-1200*
2	65.9-67.0	1420-1440*	128.8-133.5	1330-1360*	246.6-257.3	1160-1180*
3	14.1-15.3	1500^	78.4-83.2	1330-1350*	205.6-217.2	1240-1260*
6	67.3-68.5	1370-1390*	134.3-139.4	failed	257.8-269.8	failed

* FGR expected
^ no FGR expected

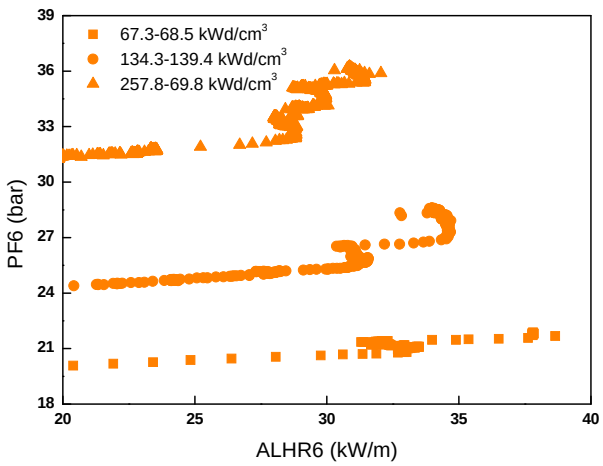

Figure 6. Inner pressure vs. average linear heat rate (rod 6)

The fuel temperatures measured in coincidence with the suspected FGR events have been qualitatively identified (on inner pressure versus fuel temperature graphs-not shown) and reported in Table 3. At increasing burnup, fuel temperatures decrease consistently with a previous remark. In rod 6 the fuel temperature thermocouple failed so that the comparison is not complete. After converting the IMF volumetric burnup to standard unit (10.4 g·cm^{-3} adopted for UO_2 density) a comparison with UO_2 FGR curve by Vitanza is presented in Fig.7. This curve predicts, in the case of UO_2 fuel, the burnup at which the FGR onset (>1 %) occurs at a certain maximum fuel temperature. In csz-based fuels we are dealing with, the experimental data is in good agreement with the reference threshold. In particular, at low burnup, rod 3 data proved to be in the no-FGR region and nicely at the boundary as in our previous comment.
Moreover is to be taken into account that maximum fuel temperatures are higher than the measured ones except in the case of rod 1 (hollow pellets) whose data is anyhow consistent with the Halden curve. These results confirm for the fuels under study the impact of burnup on FGR. As shown in Fig. 7, this effect could be milder in rod 3, 6, while rod 1, 2 experimental data could suggest that Vitanza curve may overestimate the time at which FGR onset occurs at high burnup. A possible linear correlation in rods 1, 2 fuel is envisaged. The stepwise heat rating in the second timeframe is generally required in FGR experimental investigations, moreover the typical Halden irradiation history is not consistent with a constant temperature irradiation. Further features to be taken into account are the lower accuracy of the Vitanza curve at low burnup and the high fission gas release in IFA-652 pins that is well above the typical UO_2 FGR.
Provided previous remarks, shown data, both in IM and IMT pins, is fairly in agreement with a UO_2 Vitanza threshold.

4 OUTLINE OF IMF MODELLING

IM and IMT fuels have a two-phase microstructure, in the former the fissile phase is generally globular and separated from the matrix, in the latter, the fissile phase is constituted of urania together with thoria in a "cellular" arrangement. Concerning the thermal performance it is assumed that fresh fuel thermal conductivity is kept unchanged under irradiation, being about 30% of reference UO_2 (at 1000 °C). It is to be recalled that this gap is partly diminished at increasing burnup as a consequence of the well-investigated degrading of UO_2 thermal conductivity under irradiation. Regarding the dimensional behaviour, previous analyses have suggested that a strong densification may occur in the central region of solid pellets during the early stage of irradiation while a swelling lower or comparable with typical LWR fuels was suggested [13-15].

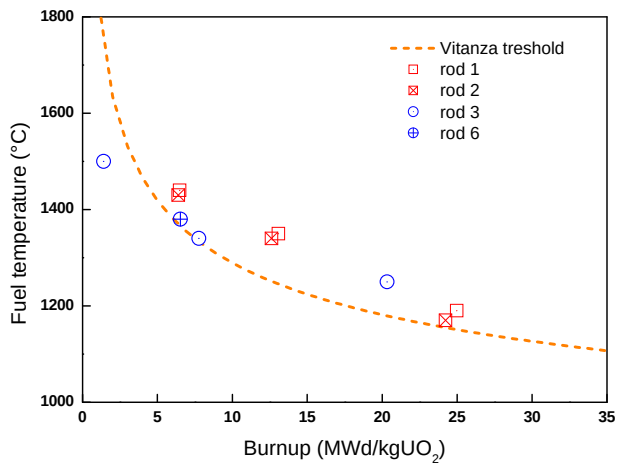

Figure 7. IMF experimental data comparison with Vitanza threshold

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5 POWER RAMP PREDICTIONS THROUGH TU CODE

The TU analysis of IMF is mainly focused on the code predictions in the timeframes resumed in Table 4. The objective is to investigate the FGR modelling of these advanced fuels, especially the deviation of IMF gas release if a UO_2-like FGR modelling is adopted in the calculations. Except this topic, the MF modelling is consistent with [14].

In the adopted FGR modelling, the diffusion equations are solved through URGAS algorithm and a thermal diffusion coefficient according to Matzke (intra-granular modelling). A constant grains boundary saturation limit ($1 \cdot 10^{-4}$ $mol \cdot m^{-2}$) is assumed (inter-granular modelling). For the grains growth the model by Ainscough is applied in simulations [9]. These models are fully validated in the case of UO_2 for commercial applications in the burnup domain we are dealing with.

The irradiation histories have been condensed through a proper code, moreover the experimental dataset has been fully exploited in the investigated timeframes to improve the accuracy of calculations [16].

The predicted and experimental fuel temperature of rods 1, 2 and 3, 6 are shown in Fig. 8 and 10. In Fig. 9 and 11, the predicted and measured inner pressure are presented.

Fuel temperature range is similar in all pins confirming that no noticeable improvement has been achieved through the thoria addition in IMT fuel. As general comment, the code overestimated the fuel temperature and underestimated the pin inner pressure at higher burnup. This behaviour is not consistent with standard fuel performance as an overestimation in fuel temperature should lead to higher FGR and related inner pressure predictions.

The fuel thermocouple of rod 6 failed preventing from a deeper investigation on the rod with the most pronounced inner pressure. The maximum deviation of the fuel temperature and the inner pressure code predictions in the investigated time intervals are resumed in Table 5.

Table 5. Temperature-inner pressure: code predictions deviations (max values)

Rods	*TimeFrame 1* %	*TimeFrame 2* %	*TimeFrame 3* %
1	11.1-7.2	10.0-8.7	16.7-11.7
2	8.7-6.9	6.4-2.5	16.1-5.1
3	9.1-8.9	6.3-4.7	23.4-39.6
6	10.4-6.7	failure-18.6	failure-29.1

Regarding rods 1, 2, 6 fuel temperature predictions, the worst performance is around 15%, being 10% the commonly accepted accuracy in TU fuel temperature calculations [17]. In rods 1 and 2 inner pressure is underestimated by about 10% up to the burnup investigated. In rod 6 the underestimation grows up to about 30% while in rod 3 this trend is even more pronounced up to 40 % as clearly shown in Fig. 11.

Considering that in rod 3 fuel temperature is also underestimated by about 20%, see Fig. 10, it is expected that, in this case, the fuel densification is greatly overestimated in the last timeframe with a massive calculated disappearance of as-fabricated porosity in the inner region of solid pellet leading to an improvement in thermal conductivity. This could also not be excluded in the code results pertaining rod 6 but, as consequence of the temperature transducer failure is not possible to ascertain if spurious densification has been predicted. Investigations are underway, at the moment it is assumed that the inner pressure calculations for rods 3, 6 have a greater deviation with respect to rods 1 and 2.

6 DISCUSSION

This paper is focused on the FGR performance of IMF. These fuels have been conceived aiming at burning in LWR the separated plutonium inventories in the context of a once-through fuel cycle strategy. In particular we are dealing with calcia-stabilised zirconia that, together with yttria-stabilised zirconia, are, at the moment, the most promising matrices amongst several candidates investigated to the purpose in the last decade. They have been studied by various organizations, both research and industry, and in-pile irradiations data is available [15].

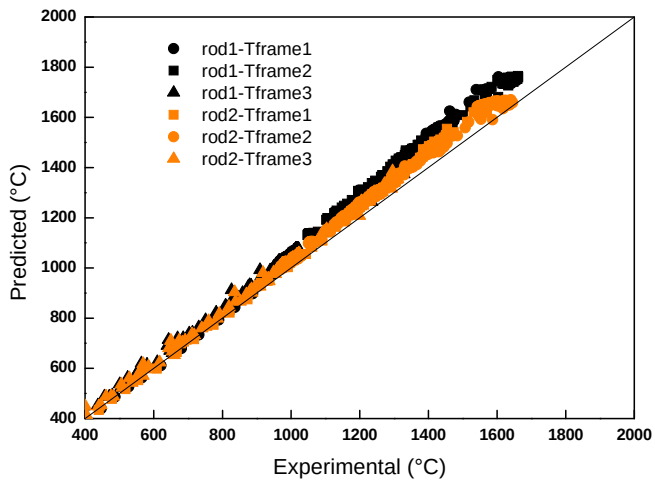

Figure 8. Fuel centreline temperature (IM pins): predictions vs. experimental

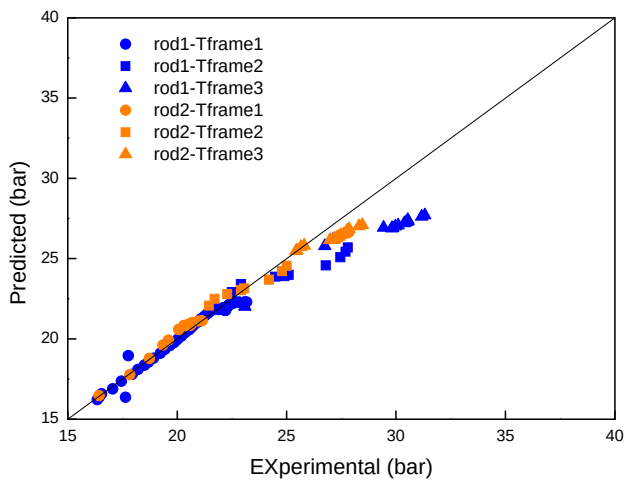

Figure 9. Inner pressure (IM pins): predictions vs. experimental

In these fuels, in principle, fissile and inert matrix form an homogeneous solid solution. In this case, published literature

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reports a proved low swelling while fuel temperature and FGR are well above UO_2 fuel if similar rating conditions are applied [15, 18].
Various features of IMF FGR are under investigation, amongst them the accuracy of the Halden Vitanza threshold in predicting the FGR onset and the understanding of the physical phenomena driving the release of fission gases to the plenum.
For UO_2 fuel, the Halden Vitanza curve predicts that the onset of an appreciable release (>1%) occurs at lower fuel centreline temperature with increasing burnup. Regarding the IMF, various authors consider that a similar correlation may hold, moreover the experimental conditions and the amount of gas released are quite different from the experimental conditions on which the Vitanza correlation was assessed [15, 18-20].

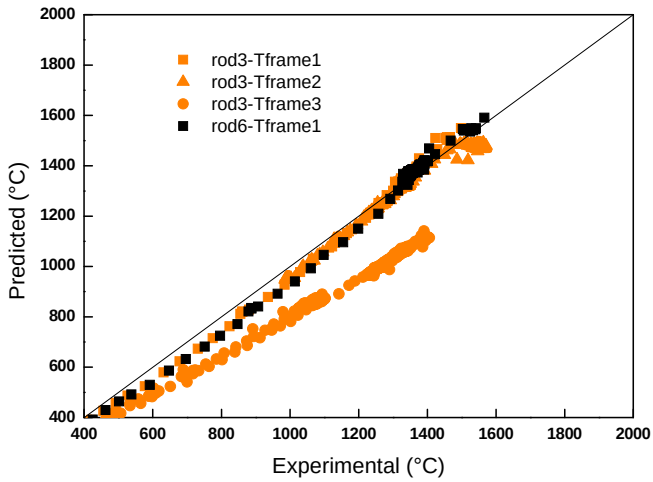

Figure 10. Fuel centreline temperature (IMT pins): predictions vs. experimental

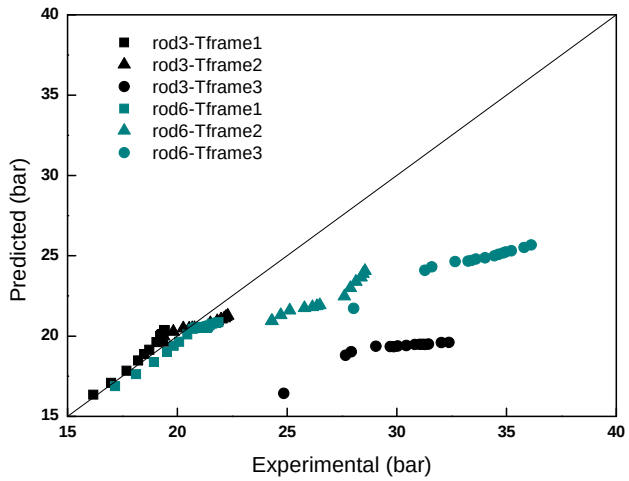

Figure 11 Inner pressure (IMT pins): predictions vs. experimental

Concerning the second topic, notwithstanding that xenon diffusion coefficient in zirconia matrix is estimated to be low, a possible increasing when solid solution with actinides oxides is formed could be expected. On the basis that the sweeping by grains boundaries movements is the driving force of the high fission gas release, it has also been proposed a review of the grains growth model. The high fuel temperature is commonly accepted as the primary driving force in FGR being well above the typical values in LWR commercial fuels [15, 18].
Finally it is worth to report a result from Japan Atomic Energy Research Institute (JAERI), where solid solution yttria stabilized zirconia fuel at a heat rating and burnup comparable with IFA-652 test, showed low FGR (5 %) and central hole formation [21].
In the IFA-652 irradiation history, three timeframes featured by power ramps were identified and discussed in this paper. The burnup domain extends up to 25 MWd/kgUO_2.
In the first part of the analysis the experimental dataset was investigated suggesting that FGR events may occur at lower rating with increasing burnup. Assuming that the fuel thermal conductivity does not degrade under irradiation, the FGR events occur at lower fuel temperature with increasing burnup [19]. If the experimental data investigated is compared with the Vitanza threshold a correlation apparently exists as previously discussed. In rods 1, 2 FGR events are distinguished and add up to get the final inner pressure. In rod 3, 6 contributions to the inner pressure are lacking as a consequence of FGR events not taken into account either possible drift of inner pressure gauge measurements under irradiation.
Afterwards, it has been attempted to predict the FGR response, focusing on the selected time intervals, by the TRANSURANUS fuel performance code. The main purpose of this analysis is to discuss the hypothesis that the low IMF thermal conductivity plays the key role in determining a FGR higher than UO_2, assuming that the same heat rating conditions occur [18].
To this purpose, the FGR modelling recommended for the UO_2 and LWR fuels has been adopted in the code calculations [9]. Our results suggested that the assumed intra and inter-granular fission gases release modelling under-predict the pin inner pressure of calcia-stabilised based fuels tested in IFA-652.
In particular while in rods 1 and 2 the underestimation is acceptable in rod 3 and 6, moving towards some 50%, is troublesome especially at high burnup, moreover these latter results may be biased by the spurious triggering of the developed densification model [14]. On this issue further investigations are needed moreover it is expected that this reason is not sufficient to cause the enhanced degrading in accuracy of the inner pressure predictions moving from rods 1, 2 to 3, 6. Furthermore the same conclusions could be argued considering that the fuel temperature ranges are fairly similar but a higher FGR has been evaluated in IMT rods [19].
As final remarks, these analyses support the hypothesis that a correlation may exist between the onset of FGR in IMF and the experimental Vitanza curve, moreover the quantitative evaluation of release is underestimated by a UO_2 standard model addressing that notwithstanding the fuel temperature is a key factor, other processes could play a significant role. Already evidenced in heterogeneous inert matrix fuels, micro-cracks and mechanical interactions between phases (e.g. differential thermal expansion) could be considered also in IFA-652 inert matrix fuels especially in the thoria-doped pins. The code calculations predict the closure of the fuel-cladding

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gap at the investigated timeframes, this mechanical interaction could take part in the gas release process coupling both the high temperature and following thermal expansion and the mechanical properties of the investigated fuels. Relying on this hypothesis, the herein showed results may suggest that the thoria addition to the pure calcia-stabilised zirconia matrix could degrade the capability in fission gas retention due to the accompanied change in phases distribution. Interesting indications could come from results in [15, 21].

Although adopting the standard UO_2 FGR modelling, the inner pressure predictions by the TU code are still acceptable, especially in rod 1, 2 of IFA-652, these results further emphasize the need of a proper modelling for IMF fission gas release where the most complex processes occurring in the nuclear fuel under irradiation are involved.

The performance of IMF has been predicted with good accuracy regarding the low fuel thermal conductivity, not degrading with burnup, the low swelling and in general the stable performance under irradiation. Concerning the adopted densification model further investigation is needed and improvements are underway.

7 CONCLUSIONS

Together with MOX, inert matrix fuels (IMF), could give a possible contribution to solve the problem of the separated plutonium inventories, a key aspect in nuclear power acceptance by public opinion. Notwithstanding that in the nuclear energy sustainability studies MOX is forecasted to be widely used as a consequence of the future deployment of Fast Breeder Reactors, considering the nowadays LWR fleet, IMF maintain competitive advantages concerning the beneficial economical figures and the high plutonium burning efficiency, as plutonium breeding in ^{238}U is avoided. These attractive aspects make worth to further investigate their performance under irradiation.

In this field, ENEA has investigated the calcia-stabilised zirconia and thoria inert matrices. Within the Italy membership in HRP, an experiment regarding the in-pile performance of these innovative fuels has been performed in the OECD Halden HBWR. In this experiment, so-called IFA-652, these innovative fuels have been tested up to about 45 MWd/kgU_{eq} under typical LWR conditions. The IMF showed good irradiation and dimensional stability, low swelling while the main drawback is the low thermal conductivity and the following high fuel temperatures that are accompanied with a pronounced FGR.

This latter feature is investigated through the paper by using the extensive IFA-652 irradiation database and the precious information from the TRANSURANUS fuel performance code. In particular we have selected three timeframes in the irradiation history in which the IFA-652 test rig has been subjected to power ramps. The code simulations have been performed investigating the provided predictions in the same time intervals.

The analysis of the experimental data confirmed the key role of the fuel temperature and burnup in the FGR of IMF. A correlation with the Vitanza threshold may exist but caution is to be taken on the basis that some aspects of the experimental conditions of IFA-652 are not overlapping with those in which the Vitanza curve has been validated.

TU code predictions proved a noticeable deviation of the inner pressure calculations questioning the hypothesis that the high temperature due to the low IMF thermal conductivity is the only factor driving the gaseous fission products release process. Provided that a proper model for IMF FGR is not available, possible contributions to FGR, especially for the thoria-doped IMF, could come from micro-cracks due to a pellet-cladding contact, predicted by the code at the investigated ramps, or possible mechanical interactions of the fuel phases.

In the fuel temperature predictions good agreement was seen with the experimental data, while dealing with the dimensional features of under irradiation response, further investigation is envisaged on densification modelling.

Herein presented results are fairly in accordance with main findings published in the open literature and we expect to have given a contribution to the investigation of IMF fission gas release being aware that studies are in progress. In this paper, it is stressed the fundamental role of coupling the in-pile irradiation tests data with the detailed description of a deeply validated fuel performance code. With this regard Post Irradiation Examination (PIE) will be precious as well.

NOMENCLATURE

CSZ	Calcia-Stabilised Zirconia
FBR	Fast Breeder Reactors
FGR	Fission Gas Release
HBWR	Heavy Boiling Water Reactor
HEU	High Enriched Uranium
HRP	Halden Reactor Project
IFE	Institutt For Energiteknikk
IMF	Inert Matrix Fuels
JAERI	Japan Atomic Energy Research Institute
LWR	Light Water Reactors
MOX	Mixed OXide
NEA	Nuclear Energy Agency
OECD	Organisation for Economic Co-operation and Development
PIE	Post Irradiation Examination
R&D	Research and Development
TD	Theoretical Density
TFDB	Test Fuel Data Bank
TU	TRANSURANUS code
UOX	Uranium OXide

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