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INERT MATRIX FUELS ANALYSIS BY MEANS OF THE TRANSURANUS CODE: THE HALDEN IFA-652 IN-PILE TEST

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ABSTRACT - *Inert matrix fuels are a possible option to reduce separated plutonium stockpiles by burning it in LWR fleet. A high burning efficiency targeted 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. Among other options, both calcia-stabilised zirconia (csz) and thoria fulfill these criteria standing as the most promising matrices to host plutonium. While several in-pile tests concerning thoria fuels are found, calcia-stabilised zirconia under-irradiation performance is still to be fully assessed, with this regard the thermal conductivity, markedly lower than UOX and MOX cases, plays a fundamental role.*

For this reason, ENEA has conceived a comparative in-pile testing of three different U-free inert matrix fuel concepts, that have been performed in the OECD Halden HBWR (IFA-652 experiment). The discharge burnup accomplished about 90-97% of the 45 MWd/kgU$_{eq}$ target under typical LWR irradiation conditions. 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 as matrix, the fissile phase being HEU oxide (UO$_2$ 93% ^{235}U enriched). IMT is a ternary fuel composed by 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 centerline temperature, pin inner pressure and fuel stack elongation measurements.

Our purpose is to investigate the key processes of IMF under-irradiation behaviour by means of the TRANSURANUS code. Thermal conductivity and its degradation with burnup, densification-swelling response and FGR are tentatively modelled in the burnup domain of IFA-652. In particular it is pointed out the effects of pellet geometry and fuel microstructures in the IM and IMT cases. The consistency of our results is discussed aiming at understanding the in-pile response, as a fundamental step, in the perspective of future deployment of the nuclear fuels we are dealing with. Notwithstanding this ambitious objective, it is clear, however, that these results rely on a limited data set and that, as TRANSURANUS is a semi-empirical code mostly tailored for commercial fuels, the modelling of the IMF is still a work in progress.

KEYWORDS: Inert Matrix Fuel, 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 plutonium stockpiles [1, 2]. One main ENEA recent R&D effort has been focused on the U-free inert matrix fuel concept aimed at burning separated plutonium (both weapon and reactor grade) in LWRs. One prominent feature of these innovative fuels is the pronounced proliferation resistance if compared with commercial MOX. In fact, in the former, while most of the fissile plutonium undergoes fission, no new plutonium is generated under irradiation. Thanks to this achievement, a once-through fuel cycle strategy becomes viable provided a careful choice of the matrix material is made. ENEA has selected calcia-stabilised zirconia and thoria as matrices to be used in three U-free fuel concepts for testing in the OECD Halden HBWR [3-5].

The irradiation test, labelled IFA-652, has been performed in the Halden reactor in the context of the ENEA membership to the HRP. Instrumented pins of IM (csz matrix), T (thoria matrix) and IMT (csz + thoria matrix) fuels have been irradiated and monitored storing the collected data in a specialised data base environment for the off-line analysis. HEU oxide (93% ^{235}U enriched) is the fissile phase of investigated fuels [6, 7].

In this paper, the thermo-mechanical response of IM (pin 1 and 2) and IMT (pin 6) is studied up to the discharge burnup (about 40 MWd/kgU_{eq}) by using the TRANSURANUS code [8]. To this purpose, starting from TU V1M2J00 standard version, fuel library has been enlarged to include the un-irradiated IM and IMT thermophysical properties, and the newly developed under-irradiation modelling introduced.

2 IFA-652 HALDEN EXPERIMENT

The IFA-652 test rig is a six-pin fuel bundle, in particular rod 1 (all hollow pellets) and 2 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 IFE-Kjeller (Norway). On the basis that the choice of the fissile material should have a negligible effect on the matrices under-irradiation behaviour, for practical reasons, in this experiment the fissile phase is HEU oxide instead of PuO_2. IFA-652 was however intended to be a first experiment to be followed by a second one, fully representative with plutonium. The fabrication has been based on dry powders mixing standard route. After pressing at 440 MPa, the green pellets have been sintered at 1700 °C (4 hours) in a reducing atmosphere (hydrogen) [9, 10].

All pins are 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 is also equipped with co-linear neutron detectors (ND) to accurately record the neutron flux along the axial direction and on the mid-plane section (Fig. 1). The acquisition period is· 900 s, data is then stored in the TFDB system [11].

Table 1. IFA-652 fuels

Fuel	*IM*	*IMT*	*T*
density, $g \cdot cm^{-3}$	5.637	6.995	8.180
csz, wt%	81	45	-
thoria, wt%	-	39.2	88.3
HEU, wt%	19	15.8	11.7

The purpose of this experiment is to investigate the thermal conductivity and its degradation with burnup, the densification-swelling behaviour and the FGR of the fuels under study. This paper deals with IM and IMT fuel, in particular with IFA-652 rod 1, 2, 6 whose design parameters are resumed in Table 2.

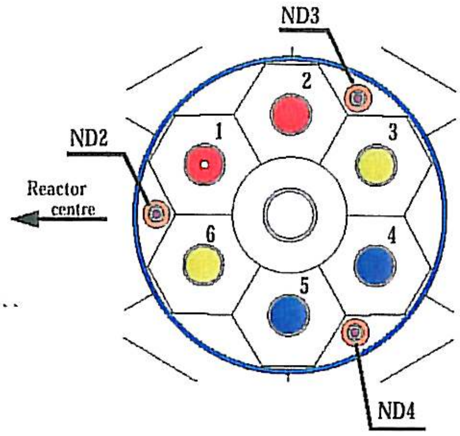

Figure 1. IFA-652 Test Rig (cross section)

Table 2. IFA-652 pins design

	Rod 1 (2)	*Rod 6*
Fuel	IM	IMT
Fuel density, %TD	89.900	93.155
O/M	2.0	2.0
Thermal shield pellets	UO_2	UO_2
Cladding	Zircaloy 4	Zircaloy 4
Fill gas	He	He
Pre-pressurisation, MPa	1.0	1.0
Free volume, cm^3	9.3 (8.4)	8.7
UO_2 pellets length, mm	10	10
Pin active length, mm	499.7 (499.7)	499.2
Hollow pellets, mm	499.7 (97.3)	97.3
Cladding outer diameter, mm	9.50	9.50
Fuel pellet outer diameter, mm	8.19	8.19
Fuel pellet inner diameter, mm	1.80	1.80
Fuel-cladding gap, µm	170	170
Fraction of dish volume, %	1	1

The in-pile test (June 28, 2000 - October 22, 2005) succeeded in accomplishing, for analysed cases, about 90-97% of the 45 MWd/kgU_{eq} target burnup. In Table 3, burnup is detailed in each loading of the IFA-652 experiment. Rod 1, 2, 6 instrumentation performed well but in the third loading PF1, PF2, PF6 (inner pressure gauge) and the TF1, TF6 (fuel centerline temperature thermocouple) showed a partly unreliable response.

Table 3. IFA-652 accumulated burnup (MWd/kgU_{eq})

Rod	*1*	*2*	*6*
Loading1	5.9	5.8	6.0
Loading 2	24.3	23.4	24.5
Loading 3	41.9	40.5	43.5

The TFDB system is the reference source of the IFA-652 experiment data [11]. In Fig. 2 we show the average linear heat rate and power of IFA-652 assembly.

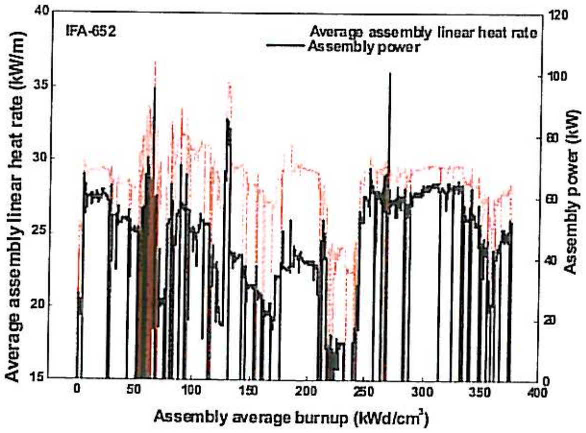

Figure 2. Average linear heat rate and power of IFA-652 rig

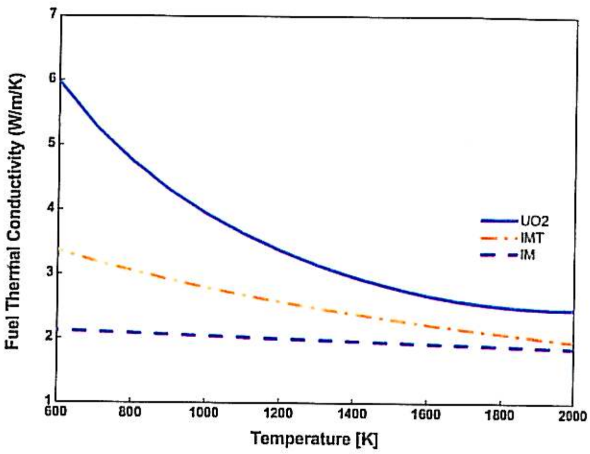

Figure 4. Fully dense thermal conductivity [8,12]

3 TRANSURANUS MODELLING

3.1. IMF thermophysical properties

The new TU version embeds the IM thermophysical characterisation of un-irradiated sample fuel pellets carried out at the JRC/ITU, Karlsruhe (Germany) [12].

A two-phase microstructure fuel was revealed for both IM and IMT. In the former, fissile islands (*blue*) are separated from the matrix (*red*), in the latter, a "cellular" structure is found with urania dispersed in a single phase together with thoria (*green*) while the second phase is still csz (Fig. 3). The thermal diffusivity, measured by laser-flash method, and the specific heat, by DSC measurement, allowed us to evaluate the IMF thermal conductivity as shown in Fig. 4. In the same figure it is possible to appreciate the comparison with fully dense UO_2 thermal conductivity [8, 12, 13].

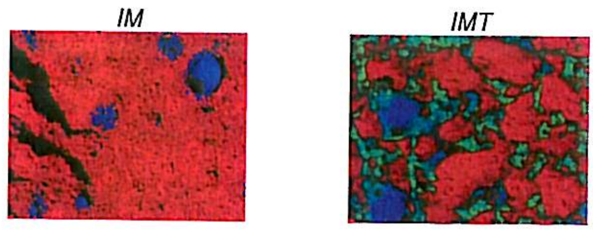

Figure 3. IMF Energy Dispersive-X surface mapping

Besides these experimental results, from vendors, further parameters values were determined (Young modulus, yield stress, emissivity). In this analysis, the IMF thermal conductivities do not degrade with burnup [14, 15]. Finally a TU confidential MOX fuel creep correlation has been adopted (implicit treatment).

3.2. IMF complex behaviour

Concerning the fuel complex behaviour, we have reviewed and simplified our previous hypotheses [16] aiming at introducing as few as possible changes in the standard TU code version. The modelling has then been lumped in the following issues:

Densification

Assuming that no central hole is formed at these thermal conditions alike in similar experiment [14, 17], it has then been attempted to predict the initial densification by a modified MATPRO-11 re-sintering model [8]. We assume that a re-sintering may take place when fuel temperature is above 1600 °C (actually lower than the true sintering temperature of the fabrication process). The re-sintering volume is added to the inner relocation volume of pre-existing cracks or it is accommodated in newly formed circumferential cracks.

Swelling

A 0.5 %/at % burnup solid swelling has been assumed.

Fission Gas Release

The diffusion equation is solved by URGAS algorithm with constant grain boundary saturation limit ($1\cdot10^{-4}$ mol·m^{-2}), while pertaining the thermal diffusion coefficient, the Matzke relationship has been modified by an exponential factor only depending on the local rating (W/gU$_{eq}$).

TU-standard model, has been assumed for the other processes as the Ainscough model for grains growth, the URGAP model for the gap conductance and the FRAPCON-3 model for relocation [8].

4 IRRADIATION HISTORY AND TU INPUT

We adopted a seven-slice axial meshing in the modelling of the single fuel rod (five slices for the fuel active length and two for the shield pellets), for the radial mesh four *coarse* zones are assumed for the fuel region and one for the cladding. The axial meshing is consistent with the four-fold average linear heat rate record as defined in the TFDB. The experimental fuel centerline temperature is compared with the TU code predictions pertaining the fifth slice.

The average linear heat rate record and the moderator temperature have been imported at each acquisition time (about 10^5 steps) and condensed by the Power Condense code [18]. The moderator pressure is 3.36 MPa. A common radial power profile (HELIOS calculations), is assumed not depending on burnup [6].

5 RESULTS

We grouped the results of calculations as follows:

- Unbounded comparison of. the fuel centerline temperature and inner pin pressure code predictions with the experimental data (Fig. 5-10);
- Analysis of the HSB pin inner pressure (zero power and moderator temperature 220-222 °C) (Fig. 11-13);
- Analysis of the HSB fuel stack elongation (rod 2, 6) (Fig. 14, 15);
- FGR and HSB gap width (Fig. 16, 17).

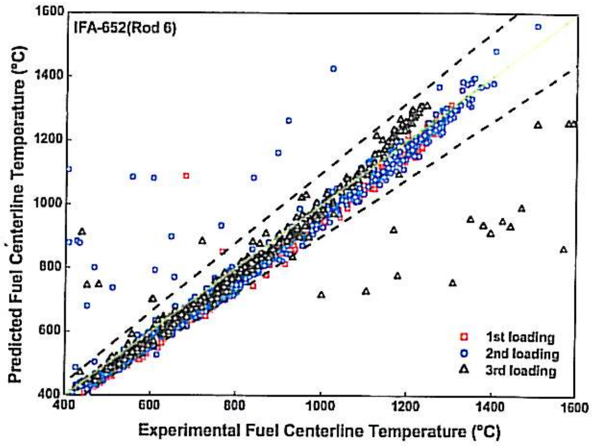

Figure 7. Fuel centerline temperature – rod 6 (±10% error band)

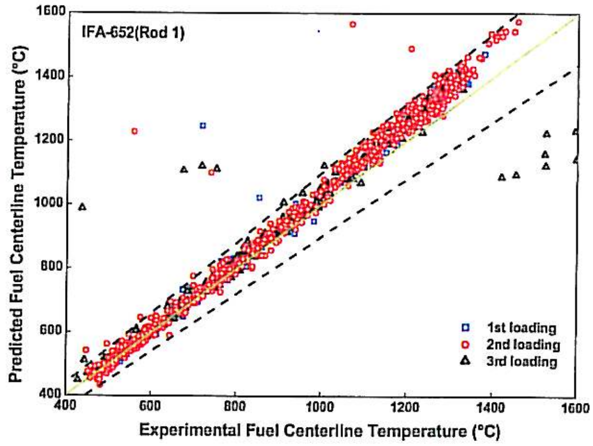

Figure 5. Fuel centerline temperature – rod 1 (±10% error band)

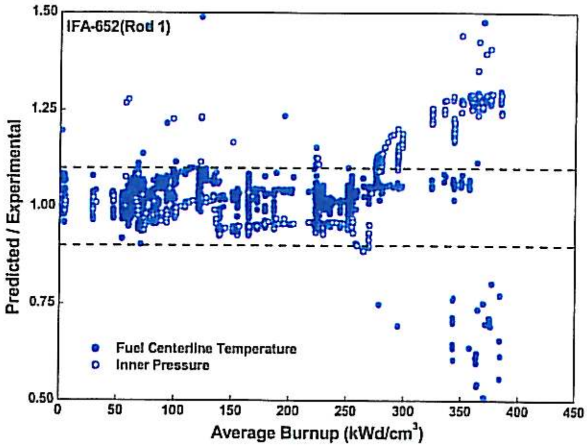

Figure 8. Ratio of predicted fuel centerline temperature and inner pressure to experimental temperature and inner pressure – rod 1 (±10% error band)

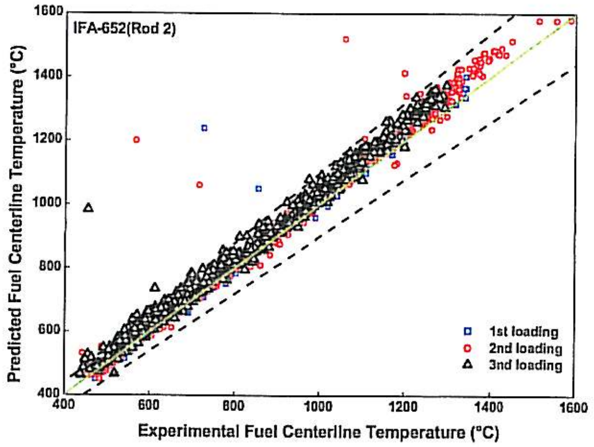

Figure 6. Fuel centerline temperature – rod 2 (±10% error band)

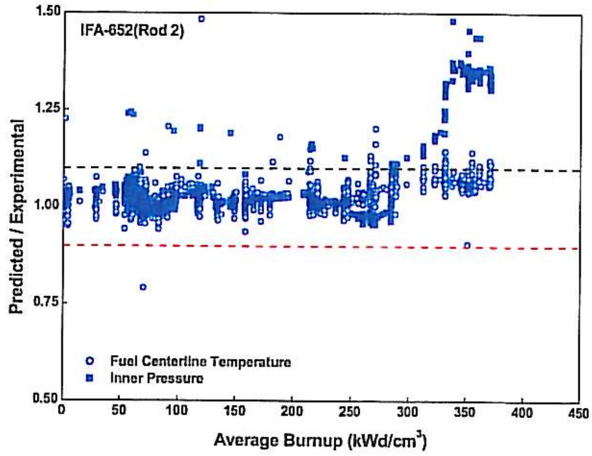

Figure 9. Ratio of predicted fuel centerline temperature and inner pressure to experimental temperature and inner pressure – rod 2 (±10% error band)

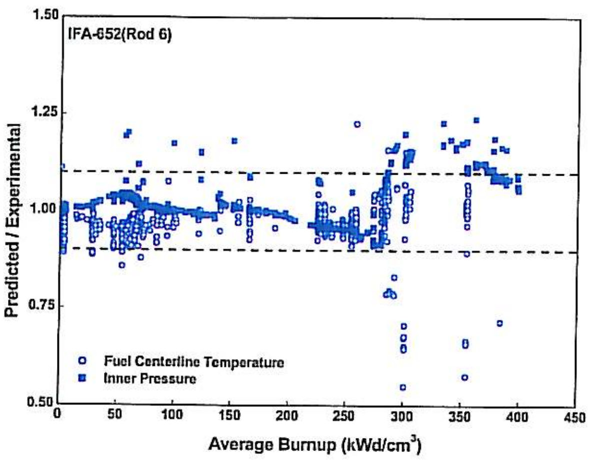

Figure 10. Ratio of predicted fuel centerline temperature and inner pressure to experimental temperature and inner pressure – rod 6 (±10% error band)

6 DISCUSSION

The code fuel temperature predictions have a 10% accuracy for all the analyzed pins and the ratio of the theoretical to experimental fuel temperature ratio does not show bias or regular trend till the discharge burnup (Fig. 5-7). Similar statements hold also for the inner pin pressure, moreover in the concluding part of the irradiation test reported failures of instrumentation are confirmed by the enhanced deviation of ratios from the band of acceptance (Fig. 8-10). Concerning the fuel temperature response (Fig. 5-7), in rod 1 and 2 data is consistently (IM fuel) overestimated, while for rod 6 (IMT fuel) TU code proves an underestimation even if a 13% reduction (with respect to ITU measurement) of the fuel thermal conductivity has been assumed.

Relying on these considerations, further investigation to determine the fuel thermal conductivity especially for IMT fuel together with a deeper understanding of the contribution to our results of each single TU modelling assumptions (e.g. fuel power profile) are required, moreover, at this stage, the final deviations we are discussing are of the same order of the experimental uncertainties and further refinement are inconsistent with given data [19]. Nevertheless, in our opinion, an uncertainties analysis of the post-processing parameters of the raw to experimental data conversion could add useful information to improve the accuracy of our analysis.

Regarding the HSB predictions (Fig. 11-15), a high accuracy is found as to the inner pressure of all pins, while the detected EOL failures are confirmed. In the evaluations of the HSB fuel stack elongation (rod 2 and 6), it is noted that accuracy is degrading in the second and third loading when TU fuel stack length evaluations grows up consistently with the assumed fuel swelling rate. Testing the fuel stack elongation sensor measurements at power (24-25 kW/m at the second slice) we see a sharp shift in the code predictions not followed by the in-pile measurement, we could then argue that stack elongation measurement pertain the cold pellet shoulders, in this case, it should be expected that pellet swelling is mostly developed in the inner hot region of fuel pellets. It is not excluded a possible "sticking" failure of the fuel stack sensors. To ascertain these hypotheses PIE will be fundamental.

Our last results regard predicted FGR and HSB gap width history (Fig. 16, 17). FGR takes place for our pins at the peak power ramps with burst-like release, TU code evaluations are in good agreement with the timing of the gas release possibly proving that IMF gas release is mainly a diffusion-driven process. In our TU modelling the gas diffusion coefficient has been increased questioning the IMF compliance of the experimental Halden Vitanza curve. Rod 1 and 2 FGR predictions are in good agreement suggesting that hollow fuel pin design has a minor impact on fuel temperature with respect to a solid pellet design due to a possible re-sintering of the inner region of solid pellets (rod 2) that increases the fuel thermal conductivity. The predicted FGR, at the same thermal loading, is significantly higher for IMT than IM fuel suggesting an important impact of the different microstructures of fuels under study. With this respect, the assumed strong re-sintering and the fissile "islands" macrostructure, as described in the IM fuel, it are expected to decrease the FGR. These results, in accordance with literature [20], may prove that IMF FGR figure is degraded with respect to the standard UOX playing the lower thermal conductivity a prominent role (Fig. .4) with a minor effect of the microstructure-related fabrication options (e.g. as-fabricated porosity, thoria content). Finally the gap width at HSB is foreseen to decrease by about 35 μm during in-reactor residence time causing the gap closure at power in the third loading (not shown).

As final remarks we may confirm that IMF proved to reach a valuable percentage of the target burnup with a limited degradation of under-irradiation performance, in particular a good dimensional stability and a not degrading fuel thermal conductivity are faced by an enhanced FGR, that could be a limiting factor for the future deployment of IMF under investigation. The thermal conductivity seems standing as a bottle-neck in the IMF in-pile performance as it turned out to be difficult to get significant improvement by acting on fabrication parameters [12]. Above statements are certainly not ultimate, it is easily recognised, in fact, the urgent need of further experimental investigations to let the collected data interpretation be unaffected by the statistics of the specific fuel fabrication process (e.g. densification model).

From the point of view of the TU code as a tool of investigation, it is important to note that with minor changes it was possible the fitting of in-pile IFA-652 measurements with an accuracy of the same order of the experimental uncertainties. This empirical approach is a sort of a glance into the key features of the IMF under-irradiation processes without claiming to be a theoretical modelling of fuels we are dealing with. These innovative fuels together with the fuels for the next generation nuclear power systems, are expected to be investigated by the so-called multi-time scale approach that aims at a deep description of under-irradiation behaviour starting from the first-principle analysis at electronic structure scale [21]. These new techniques are very promising but still at an early stage, so far the traditional codes for the thermo-mechanical analysis still get good results without the tremendous amount of computational resources that are coupled with a finely detailed description as in the MTS approach.

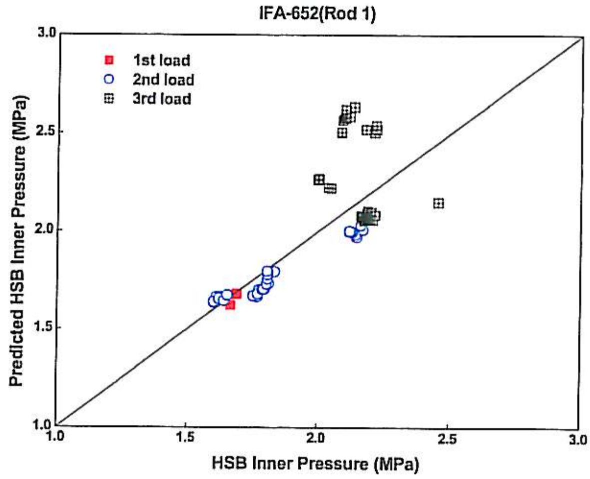

Figure 11. HSB inner pressure – rod 1

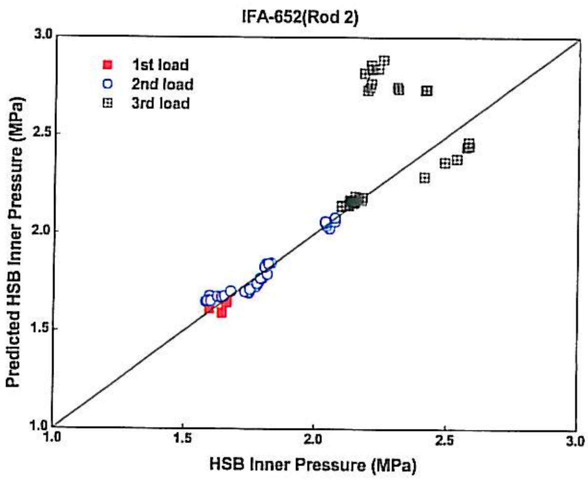

Figure 12. HSB inner pressure – rod 2

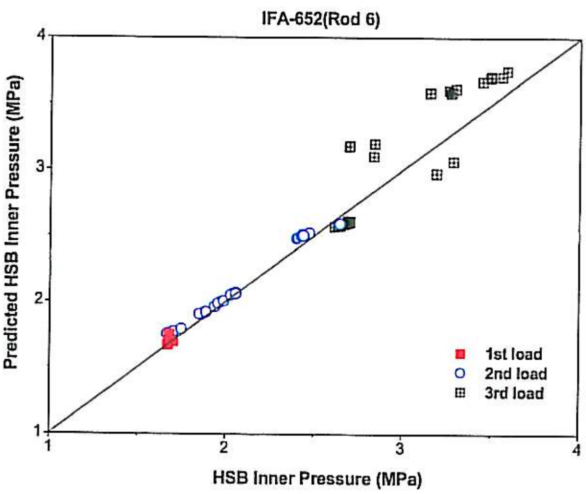

Figure 13. HSB inner pressure – rod 6

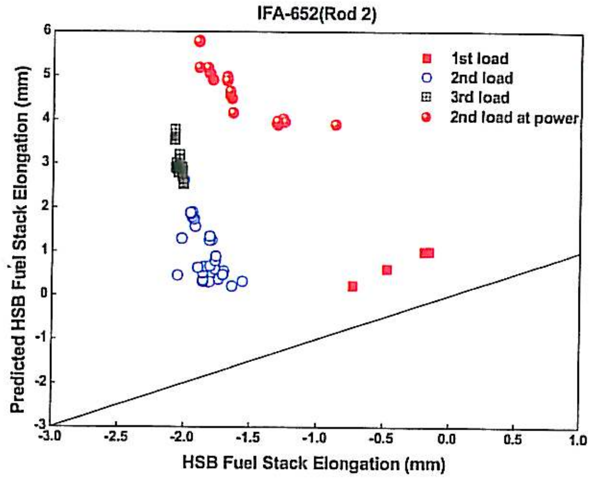

Figure 14. HSB fuel stack elongation – rod 2

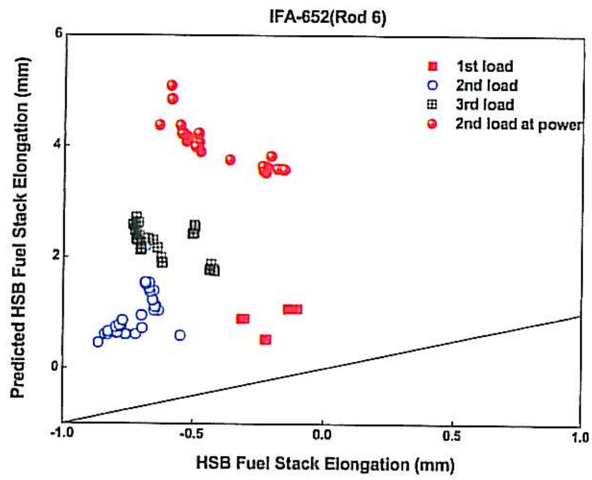

Figure 15. HSB fuel stack elongation – rod 6

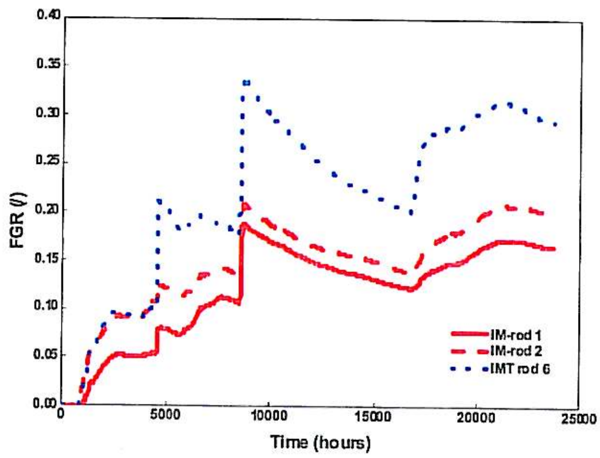

Figure 16. FGR

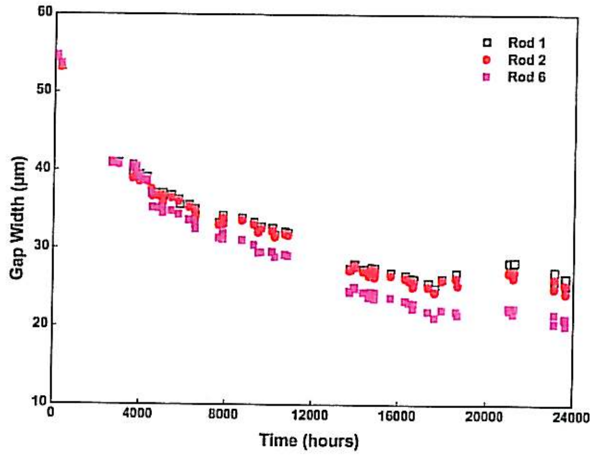

Figure 17. HSB gap width (slice 5)

7 CONCLUSIONS

In this paper we have described the results of the thermo-mechanical analysis of the IM (rod 1, 2) and IMT fuel (rod 6) irradiated in the OECD Halden reactor (IFA-652 experiment). The calculations have been performed by using a newly developed TU code version, extended to cover the IMF thermophysical properties and the herein proposed under-irradiation modelling. The accuracy of performed calculations is within the experimental uncertainties till the discharge burnup for both fuel temperature at thermocouple and inner pin pressure. Relying on these results we may suggest some preliminary conclusions.

A good IMF dimensional stability is revealed in the IFA-652 fuels even if it is suspected that an intense re-sintering occurs in the hot region of solid pellets with an un-expected overlapping of hollow and solid pellets in-pile response. If confirmed, a deeper investigation on fabrication process is required. The low accuracy of the fuel stack elongation predictions could support the hypothesis that the fuel swelling takes place in the inner region of pellets. As to the thermal performance, a limited degrading of IMF thermal conductivity with good stability of fuel temperature with burnup was shown.

Regarding the FGR, the TRANSURANUS description by the diffusion model may hold even for the IMF case. In our calculations the TU modelling of the Halden Vitanza curve has been modified (by increasing the thermal diffusion coefficient), so that it is still a debated issue if this commonly accepted reference for UOX FGR, could still be applied to our case. As concluding remarks, concerning the higher IMF FGR compared with reference UOX, it seems that fuel suffered from low density. It is expected that the as-fabricated density can be strongly improved by using purified starting material and higher sintering temperatures. The TU evaluation of the HSB gap width is about 20 μm at EOL with closure at power in the third loading, in this case, as the IMF creep correlations are unknown, further considerations could be not realistic at moment.

The TU code proved to be a fundamental tool in investigating the IFA-652 experimental data by a limited set of key parameters in which the IMF under-irradiation performance could be lumped, but caution is to be taken due to the single-experiment data set investigated and the strictly empirical approach adopted. In the long term, complementary experimental data and MTS approach applications are expected to give an answer to many questions risen regarding the IMF in-pile response. In the near term PIE and deeper investigation of assumed TU input parameters are a viable route to try to fix the milestones of possible future extensive modelling of these innovative fuels.

NOMENCLATURE

CSZ	Calcia-Stabilised Zirconia
DSC	Differential Scanning Calorimetry
EOL	End Of Life
FGR	Fission Gas Release
HBWR	Heavy Boiling Water Reactor
HEU	High Enriched Uranium
HRP	Halden Reactor Project
HSB	Hot Stand-By
IFE	Institutt For Energiteknikk
IMF	Inert Matrix Fuels
ITU	Institute for TransUranium elements
JRC	Joint Research Centre
LWR	Light Water Reactor
MOX	Mixed OXide
MTS	Multi Time-Scale
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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