Progress and Advancements in Fuel Performance Modelling for MYRRHA/XT-ADS

Sergei E. Lemehov, Vitaly Sobolev, Kevin Govers, Marc Verwerft, H.Aït Abderrahim

Belgian Nuclear Research Centre SCK·CEN, Boeretang 200, 2400 Mol, Belgium

Tel. +32 014 33 31 97, Fax. +32 014 33-3097, E-mail: slemehov@sckcen.be

ABSTRACT: Transmutation of the minor actinides (neptunium, plutonium, americium and curium) in either a fast reactor (FR) or a fast accelerator-driven system (ADS) is currently considered as one of the promising options. The current reference fuels for a European ADS-transmuter are oxides such as MOX, *(Cm,Am,Pu)O_2* diluted in *MgO*, *ZrO_2*, and *Mo* matrices. In the framework of the number of EC projects and its own project MYRRHA, the Belgian Nuclear Research Centre SCK·CEN started few years ago the studies on fuels for transmutation in ADS. To evaluate basic in-pile features of fuel design the full-scale fuel performance code MACROS has been developed and is currently used. This paper will show the main results of calculation of the in-pile behaviour of two inert matrix oxide fuels dedicated to transmutation of Am in a fast spectrum ADS performed with MACROS. The results of the modelling studies show that designs of the fuel/target rods considered in FUTURE and MYRRHA projects (European projects for ADS transmuters) are quite suitable for long term operation under representative ADS conditions.

KEYWORDS: *Oxide fuel, modelling, minor actinides, fuel performance.*

1. Introduction

Transmutation of the minor actinides (neptunium, plutonium, americium and curium) in either a fast reactor (FR) or a fast accelerator-driven system (ADS) is currently considered as one of the promising options. The current reference fuels for a European ADS-transmuter are oxides such as MOX (driver fuel), *(Cm,Am,Pu)O_2* diluted in *MgO*, *ZrO_2*, and *Mo* matrices. In the framework of the EC FP5 and FP6 projects (FUTURE, EUROTRANS IP) and its own project MYRRHA [1,2], the Belgian Nuclear Research Centre SCK·CEN started few years ago the studies on fuels for transmutation in ADS. These studies are focused on the evaluation of the safety limits, reliability margins and in-pile flexibility of the proposed designs. To evaluate basic in-pile features of fuel design the SCK-CEN full-scale fuel performance code MACROS has been developed and is currently being tested and used.

The present communication summarises a SCK-CEN preliminary design study of the inert matrix fuel targets for MYRRHA ADS and fuel pins for the EFIT core, the conceptual industrial burner designed in EUROTRANS IP. Among different potentially attractive oxide inert matrixes, ZrO_2 and MgO seem to be the most promising compositions. The first one represents fully homogeneous composition for transmutation of minor actinides in the form of (Pu,Am, Cm,Zr)O_2 dioxide, while MgO introduces possibility of deep transmutation of MA target oxides keeping heterogeneous arrangement of (Pu,Am,Cm,)$O_{1.88}$ fuel in high conductive MgO matrix.

The purpose of the present study was to apply the SCK-CEN fuel performance code MACROS for analysis of in-reactor behaviour of two inert matrix compositions under assumed in-pile conditions. The pin designs have been proposed based on previous feasibility studies performed in the framework of the EC FP5 FUTURE project. The use of lead-bismuth liquid metal coolant dictates consideration of T91 martensitic steel for cladding, the properties of which are far from being complete. Because of the difficulties related to the production and handling of fuel with minor actinides, only few measurements have been made of their important properties, such as heat capacity, thermal conductivity and thermal expansion. Most of the physical properties were estimated with analytical models based on known properties and recommendations for ZrO_2, MgO, PuO_2, AmO_2, CmO_2 and mixed oxides [3].

Experimentally measured properties have been used where possible. Furthermore, in-pile properties of truly heterogeneous fuel targets such as MgO+(Pu,Am,Cm)O_{2-x} are not well known as well. Therefore, the fuel study concludes to a preliminary design only, raising at the same time questions on in-pile behaviour of candidate inert matrixes.

2. Design Specification and Irradiation Conditions

2.1. Fuel rod specifications

The behaviour of two fuel pins was modelled in the hottest irradiation position of the inner zone of MYRRHA/XT-ADS – one with homogeneous (Pu,Am,Cm,Zr)O_{2-x} pellets (69 vol. % ZrO_2) and another with heterogeneous pellets containing 43 vol. % of *(Np,Pu,Am,Cm)$O_{1.88}$* fuel particles dispersed in 57 vol. % of MgO matrix. It was assumed that both have 10 % porosity at start. The isotopic vectors of actinides were chosen on the basis of the recommendation for a double strategy where a fast spectrum ADS performs the transmutation of plutonium and americium coming from spent MOX of LWR. The isotopic vectors of zirconium, magnesium and oxygen were those for the natural elements. The cooling conditions at nominal power level are kept constant over all irradiation period.

TABLE 1 summarizes the main design parameters of fuel pins and cooling conditions.

TABLE 1. EFIT FUEL ROD DESIGN DATA

Parameter	Unit	Value	**Plutonium**	Vector
Fuel rod length	mm	2100	Pu^{238}	5.05
Active length	mm	900	Pu^{239}	37.91
Clad outer diameter	mm	8.72	Pu^{240}	30.31
Clad inner diameter	mm	7.52	Pu^{241}	13.21
Pellet outer diameter	mm	7.20	Pu^{242}	13.51
Pellet inner diameter	mm	0.00	**Americium**	
Pellet height	mm	10.00	Am^{241}	66.67
Mean roughness of fuel	μm	3	Am^{243}	33.33
Mean roughness of clad	μm	3	**Curium**	
Fuel composition:			Cm^{244}	90.00
$(Pu_{0.4}Am_{0.5}Cm_{0.1})O_{1.88}$: ZrO_2	vol.%	31:69	Cm^{245}	10.00
$(Pu_{0.4}Am_{0.5}Cm_{0.1})O_{1.88}$: MgO	vol.%	40:60	Cm^{246}	0.00
Total fuel porosity	%	10		
Open porosity	%	0.05		
Upper plenum length	mm	274		
Lower plenum length	mm	854		
Rod lattice pitch (triangular)	mm	13.63		
Inlet coolant temperature	°C	300		
Outlet coolant temeprature	°C	450		
Coolant pressure	MPa	0.6		
Helium pressure	MPa	0.2		

2.2. Irradiation conditions

Irradiation conditions pre-calculated earlier in neutronic modelling [4] for the hottest positions of the EFIT core were used. Three cycles of operation with cycle duration of 255 EFPD for homogeneous (ZrO2) fuel and 245 EFPD for heterogeneous (MgO) fuel and shutdown period of 30 days in between cycles were considered. The linear heating rate was kept constant during

the irradiation periods expecting that the reactivity losses can be compensated. As it has been mentioned above, the calculations were performed for the fuel pins sustaining the maximum power and burnup. These pins operate at maximum linear heating rate (LHR) of 260 W/cm (ZrO_2) and 333 W/cm (MgO) (the axial power form-factors are 1.26 and 1.22 respectively).

The neutron spectrum is fast with a majority of neutrons (72 %) in a low-fast group with $E_n >$ 25 keV. This leads to a practically flat radial power and burnup profiles and relatively effective transmutation of americium.

3. Calculation Results

The in-pile behaviour of the two fuel pins is characterized hereafter on the basis of results obtained in the calculations with the code MACROS [5]. The presented results mainly cover and take into account fuel temperatures, fuel restructuring and volumetric changes due to solid and gas swelling; irradiation-induced degradation effects on fuel thermal conductivity; and evolution of initial minor actinide vectors (MA burning out). All calculation results are preliminary (in view of limited verification database) and intend to outline potentially important problems.

3.1. Fuel Temperature

In-pile evolution of the fuel surface and centreline temperatures at the peak power location and at the location representing (in terms of burnup) "rod average node" for homogeneous IMF(ZrO_2) and heterogeneous IMF(MgO) are shown in Fig. 1a and 1b respectively.

It can be seen, that in the peak node of the heterogeneous (MgO) rod the fuel surface temperature remains in the range of $600 \pm 50\ ^0$C and is approximately 50-100 °C higher than in the homogeneous rod (ZrO_2) rod. This difference is associated with a higher LHR and a different state of gap. Gap closure in the homogeneous rod occurs after ≈ 200 days of operation due to higher linear thermal expansion and fuel relocation.

The temperature of cladding slowly increases with time because of the clad corrosion in LBE coolant (a simplified model was developed using the available data and recommendations). According to these calculations the clad corrosion is limited to about 80 μm and shall not lead to considerable influence on the fuel surface temperature which is mostly affected by contamination of free volume by fission gases (Xe, Kr). Gap closing was not revealed in the heterogeneous (MgO) fuel rod and, yet, fuel surface temperature showed stable behaviour at nominal power with irradiation time. This is due to the fact that gap conductance was not affected to considerable degree by release of Xe. Fuel containing minor actinides (Pu, Am and Cm) generates and releases α-decay helium (He) in quantities which excess generation and release of fission Xe. As a consequence fraction of Xe in a free volume gas tends to saturate as well as a gap contamination.

The centreline temperature of the homogeneous rod is not very sensitive to irradiation due to a very low thermal conductivity of the ZrO_2 matrix. Contrary to MgO it shows a slight gradual decrease. Understanding of such behaviour comes from the consideration of specifics of the fuel restructuring.

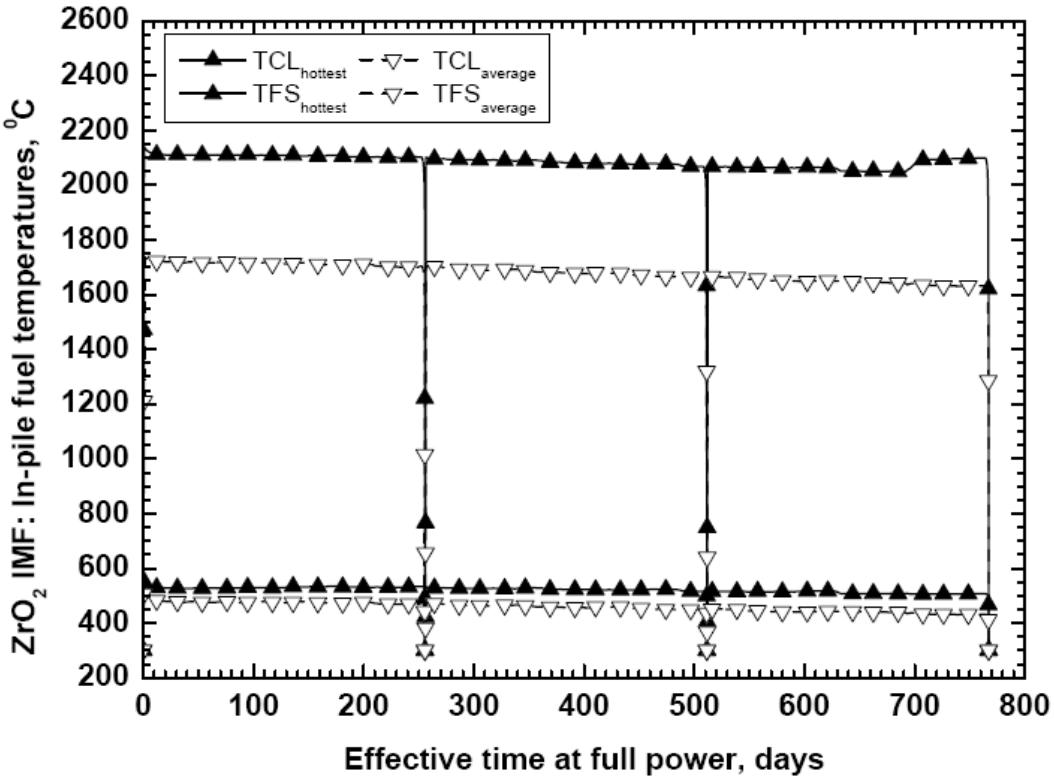

Fig. 1a. In-pile evolution of the fuel centreline (TCL) and the fuel surface (TFS) temperatures in ZrO_2 based IMF.

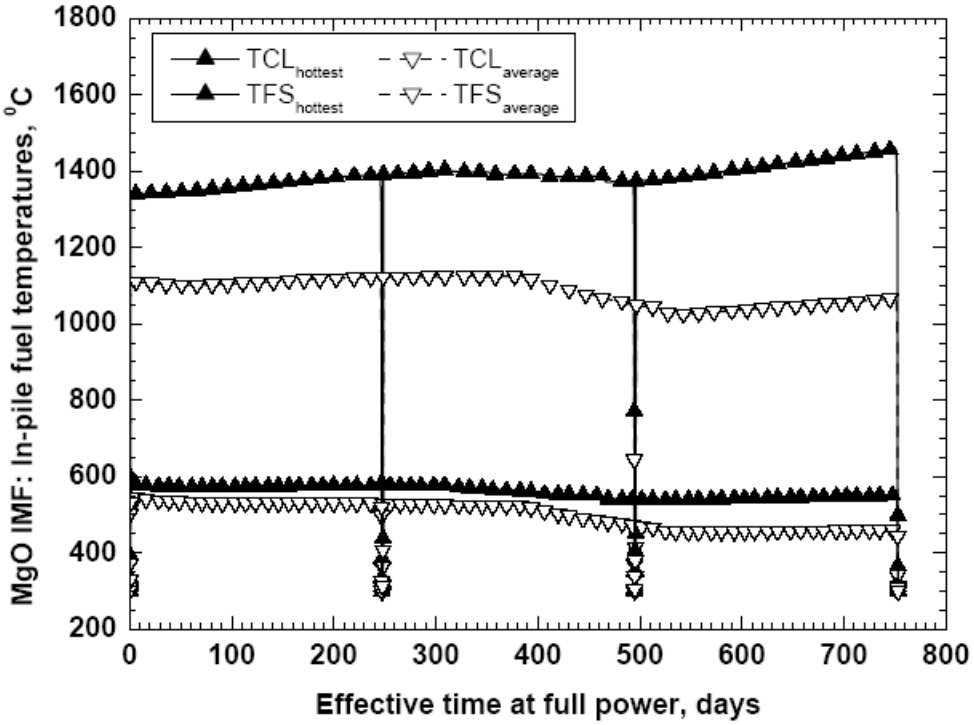

Fig. 1b. In-pile evolution of the fuel centreline (TCL) and the fuel surface (TFS) temperatures in MgO based IMF.

The in-pile centreline temperature in the heterogeneous MgO based IMF shows a nonlinear increase in the third cycle which is due to thermal conductivity degradation and porosity development. Response to power changes is shown in Fig 2.

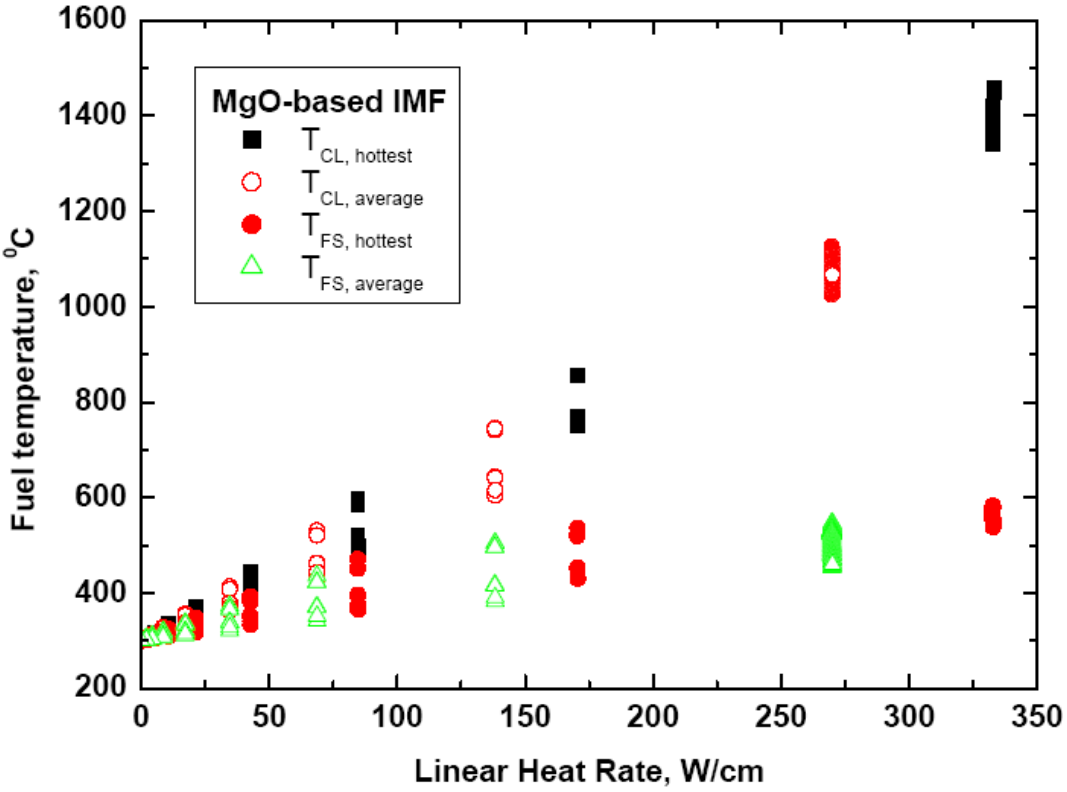

Fig. 2. Centre-line temperature vs Linear Heat Rate in MgO-base IMF.

3.2. Fuel Restructuring

At the beginning of irradiation, the radial distribution of porosity is constant and equal to as-fabricated value, which was assumed to be 10 % of theoretical density (TD). According to the models of the code MACROS, shortly after reaching the nominal power, a rapid restructuring in the hottest part of ZrO_2 type IMF occurs and the columnar grain structure is formed with the dramatic influence on the local fuel density. Propagation of this structure change is shown in Fig. 3. At the end of 3rd cycle columnar grain zone extends up to about mid-radius position. The local density of fuel in the columnar grain zone is considerably higher than that in the intermediate (still hot) zone with equiaxed grain growth. At the very rim, where the local fuel temperature is not high enough to sustain influence of burnup, a high burnup structure formation was detected by the model calculations.

Due to considerably better thermal conductivity of MgO restructuring in MgO-IMF fuel rod is limited with growth of equiaxed grains in hottest parts and high burnup structure formation in cold radial parts of fuel as shown in Fig. 3.

In both IMF rods, the gap closure did not influence fuel restructuring. Plastic deformations of clad mainly occur after the complete gap closure, which was not revealed. It certainly had influence on more rapid (non-restrained) development of inter-granular gas porosity, since hot pressing did not play any role over irradiation time of about 800 effective full power days.

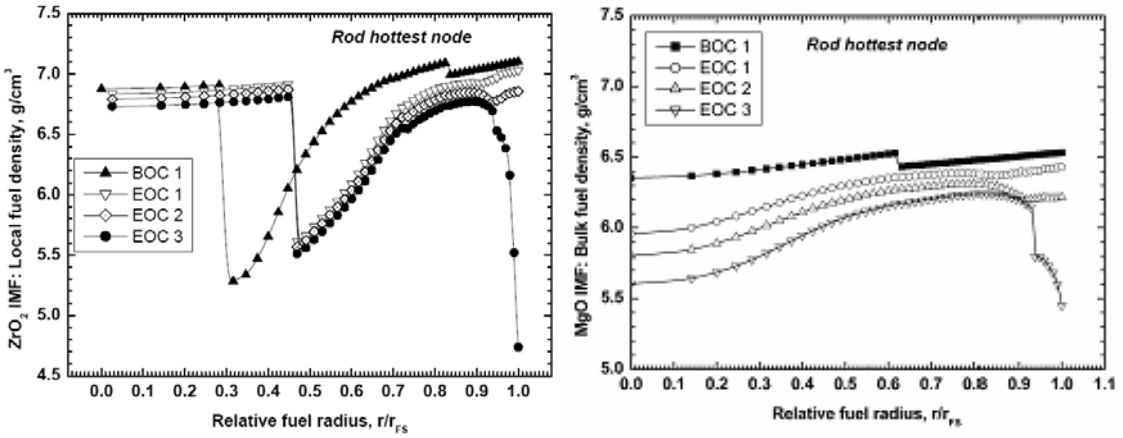

Fig. 3. Radial density profiles and their evolution with irradiation in IMF rods

Main concerns associated with the fuel restructuring are obvious for ZrO_2 based IMF rod. It influences radial power profiles and leads to a considerable degradation of the fuel thermal conductivity due to macroscopic porosity. As a consequence, the temperature profiles in restructured fuel deviate from a parabolic shape. Higher temperature gradients and thermal barriers facilitate further fuel restructuring.

3.3. Fission and Decay Gas Release

Due to high accumulation rate (Fig. 4) and temperatures, gas release starts early in life and continuously increases with burnup. Specifics of minor actinide fuel is that helium, which originates from α-decay and fast neutron reactions, rapidly takes over the release of fission gases, such as Xe and Kr.

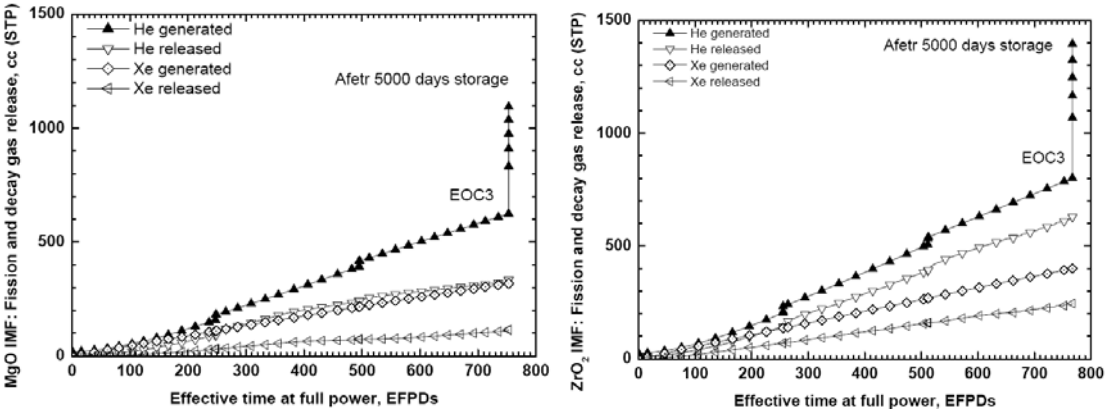

Fig. 4. Fission and decay gas generation and release in inert matrix fuels

A low initial filling pressure facilitates non-restrained He release to the rod free volume. The rate of this increase slows down with time in both IMF rods due to progressive gas starvation in the fuel zones with restructured equiaxed grains (Fig. 5).

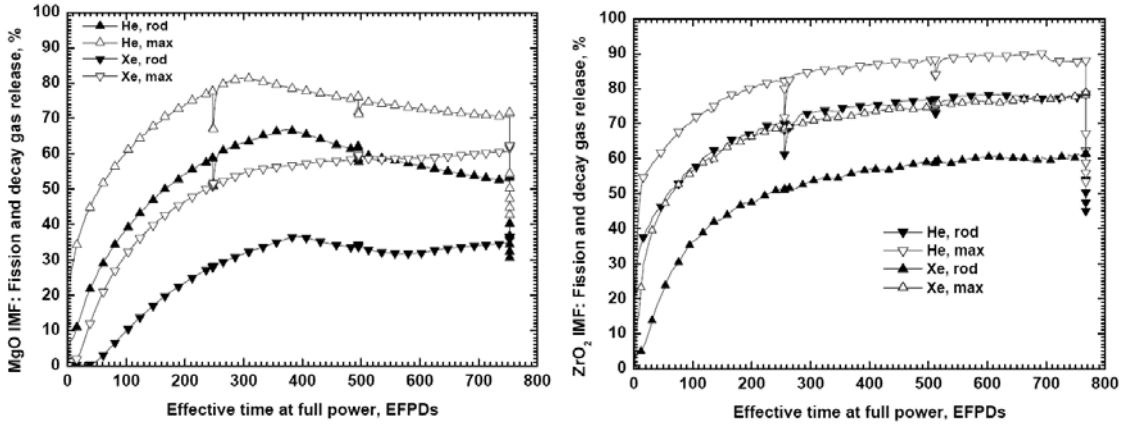

Fig. 5. Fraction of the released fission and decay gases during the operational period.

The resulting rod inner pressure increases up to 10 to 12 MPa in hot conditions after 800 EFPD (Fig. 6).

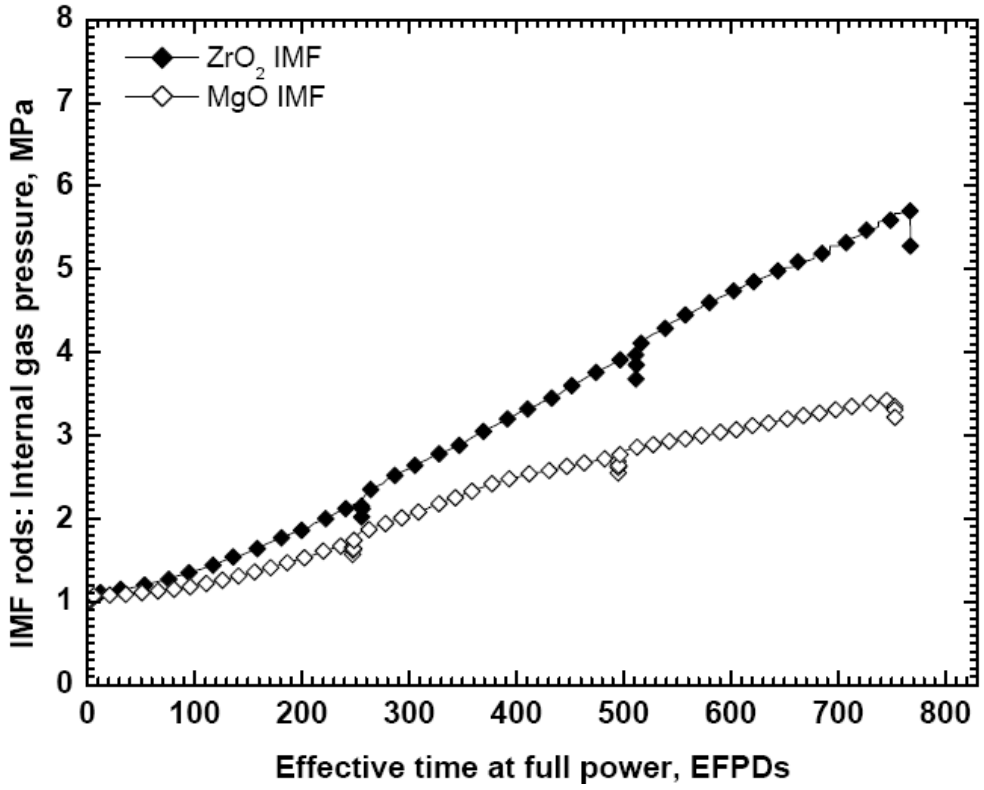

Fig. 6. Inner rod pressure evolution.

The inner pressure increase due to gas release may have an important impact on cladding creep. The T91 material is creep resistant at temperatures below 450 $^{\circ}$C. Under considered conditions it makes the cladding thermal creep almost negligible. However, the time-dependent clad oxidation may lead to the local clad temperatures in excess of 450 ^{0}C where thermal creep is already important. The inner gas pressure increase due to the fission and decay gas release up to 10 to 12 MPa will generate an important hoop stress and may cause a driving force for appreciable clad thermal creep.

4. Conclusion Remarks

Safety design criteria impose absence of the fuel melting. For the considered IMF rods it means that peak fuel temperature must not exceed 2500 $^{\circ}$C as lowest estimate for melting. Calculations give a maximum fuel centreline temperature in ZrO_2 based IMF rod of 2200 °C and a much lower value for MgO-IMF rod. The value of the peak temperature obtained for ZrO_2 based IMF rod at nominal power conditions shows that safety margin is only 300 ^{0}C. In reality it can be even smaller because few non-conservative assumptions (options) and uncertainly in the code models. More conservative calculations still need to be performed.
Safety criteria also impose requirement of clad integrity, which limits cladding strain accumulated by in-pile creep. For FBR rods strain criterion reads 0.2. In the ADT-800 conditions, it seems to be difficult to avoid in-pile thermal creep of cladding. The coolant temperature is such that activation limit of 450 $^{\circ}$C can be exceeded (at least after certain time of operation). It makes the release of fission gasses (Xe, Kr and volatile species) and decay He a potential issue for the fuel design. Complementary conservative and non-conservative analysis of fission gas release and T91 cladding in-pile creep problem has to be performed.
It might also be advised to re-assess power histories. Potentially, MgO based fuel rods could operate higher nominal powers. Contrary to that, nominal linear heat rate for ZrO_2 based IMF seems to be reduced.
In view of restructuring problems, the only feasible way to increase safety margins is to make technological advancements in fuel itself in order to ensure its stability, integrity and better resistance to burnup-induced degradation effects.
The present code calculations are performed using a best-estimate approach. Many properties are today insufficiently known. As a result, a conservative approach is not possible yet. One may hope that irradiation effect on thermal conductivity is fairly understood. Unfortunately, the verification database is too limited to address properly the mechanical and diffusion properties of the considered inert matrix fuels. Direct instrumented in-pile measurements of temperature, inner pressure and clad elongation in the designed fuel rods will be necessary both for testing of fuel and for verification of the fuel performance codes.

REFERENCES

1. H. Aït Abderrahim et al., Nucl. Inst. Meth. Phys. Res. **A 463** (2001) 487.
2. E. Malambu, et al., *MA and LLFP Transmutation Performance Assessment in the MYRRHA small-scale ADS*, Proc.: 8th International Exchange Meeting on P&T, Las Vegas, Nevada, USA, November 8-11 (2004).
3. M A Mignanelli, R Thetford, In: *"The Second Workshop Proceedings of the International Conference on Advanced Reactors with Innovative Fuels* (ARWIF - 2001)", Chester, UK, 2001.
4. W. Maschek et al., Safety Analyses for Accelerator Driven Transmuters (ADTs) with CERCER and CERMET Fuels, DEL-2 Report, EURATOM FP5 FUTURE project, August (2004).
5. S. Lemehov, M. Verwerft, V. Sobolev, **"Thermomechanical modeling of prototypic targets containing high concentrationsof minor actinides"**, Chapter 5, Editor 2004. Reactor Safety Division, OECD TECDOC 402.