

Under irradiation issues of a heterogeneous fuel bearing minor actinides: Fuel codes results

R. Calabrese, F. Vettraino, C. Artioli

Agenzia nazionale per le nuove tecnologie, l'energia e lo sviluppo economico sostenibile (ENEA),
Italy

V. Sobolev

SCK•CEN, Belgian Nuclear Research Centre, Belgium

R. Thetford

Serco Technical Consulting Services, United Kingdom

Abstract

Provided that plutonium is re-cycled in fast reactors of new-generation, the radiotoxicity of long-term nuclear waste inventory could be decreased by two orders of magnitude, through the transmutation of minor actinides (MA). This objective could be achieved in a double-strata scenario via purpose-built accelerator-driven systems (ADS). The projects FUTURE (Fuels for Transmutation of Transuranium Elements) and EUROTRANS (European Research Programme for the Transmutation of High-level Nuclear Waste in Accelerator-driven Systems) funded by EURATOM Framework Programmes identified two composite fuel systems: a ceramic-ceramic (cercer), where fuel particles are dispersed in a MgO matrix, and a ceramic-metallic (cermet), with molybdenum in the place of magnesia matrix. The fissile phase is a solid solution of plutonium and MA oxides coming from the reprocessing of LWR fuel discharged at 45 MWd/kg_{HM} and stored for 30 years. The peculiar features of designed MA-bearing fuels require significant R&D efforts to meet safety and technological performance typical of mature fuel systems. While a considerable knowledge was attained on the thermophysical properties driving the response of such innovative fuels at the beginning of their in-reactor life, key under irradiation issues such as production/release of helium, swelling and degrading of matrix thermal conductivity are still open. On this topic, code developments introduced in FEMALE, TRAFIC and TRANSURANUS are discussed and applied to the hottest fuel rods of a European Facility for Industrial Transmutation core (EFIT) loaded with cercer fuel, both pre-designed within EUROTRANS project.

Introduction

Various options are envisaged in Europe to manage spent fuel from nuclear power plants. In the 6th Framework Programme of EURATOM, within the integrated project EUROTRANS, it was developed the design of an European Facility for Industrial Transmutation (EFIT) of minor actinides (MA), such as neptunium (Np), americium (Am) and curium (Cm), accumulated in the spent fuel of LWR park [1].

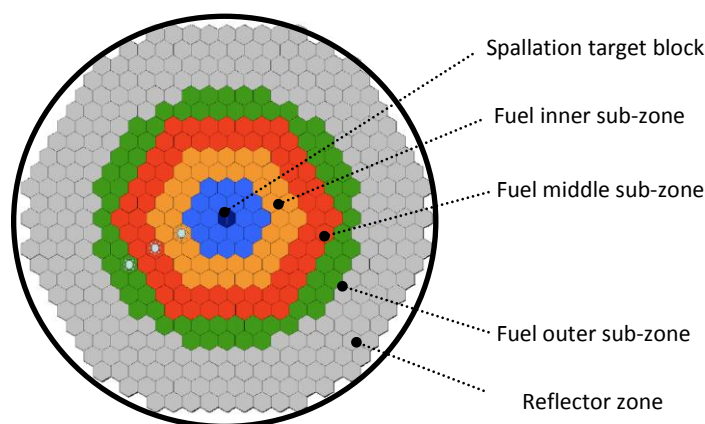
Based on the analysis of different fuel candidates performed at an earlier stage of the EUROTRANS project (in Domain 3 AFTRA “Advanced Fuels for Transmutation systems”), two composite fuel systems were selected for detailed studies and optimisation: one with the (Pu, Np, Am, Cm)O_{2-x} fuel particles incorporated in the ceramic MgO matrix (cercer) and another with the metallic Mo matrix (cermet) [2]. The simulation of the behaviour and the prognosis of performances of the candidate fuels under representative operation conditions of EFIT is a very important step in the optimisation of such advanced fuels.

This paper presents progress in modelling the under irradiation thermomechanical behaviour of the hottest cercer fuel rods of EFIT with the fuel performance codes FEMALE, TRAFIC and TRANSURANUS.

EFIT fuel candidates

A reference core of EFIT consists of the central spallation target surrounded by three zones loaded with hexagonal fuel assemblies (FA) with different matrix fraction in composite fuel or/and with different diameters of the fuel pins, in order to flatten the radial power distribution (Figure 1) [3]. The core is cooled by liquid lead with inlet temperature of 400°C and pressure of 0.6 MPa. The total coolant mass-flow rate is normalised to obtain the coolant average temperature of 480°C at the core outlet.

Figure 1: Cross-section of the three-zone EFIT core [3]



The fuel rod schematic outlook is given in Figure 2. The FA geometry is the same in all zones. Each FA contains one dummy pin in the central position and 168 fuel pins. The main parameters of the cercer fuel pin geometry are presented in Table 1.

Accordingly to AFTRA specifications, in cercer pellets, (Pu_{0.4570}Np_{0.0211}Am_{0.4986}Cm_{0.0233})O_{1.88} ceramic fuel particles are uniformly distributed within the matrix, 90% TD MgO. The mean diameter of fissile inclusions, also 90% TD, is 0.1 mm [4]. The isotopic vectors of Pu, Np, Am and Cm were recommended by the EUROTRANS project [5]. The matrix volume fraction is 57% and 50% respectively in the inner and remaining zones. The fuel rod cladding was assumed to be T91 ferritic-martensitic steel [6]. The fuel rod free volume is filled with helium (He) at 0.2 MPa inner pressure (at T = 291.15 K).

Figure 2: Geometrical schematics of a fuel pin of EFIT [4]

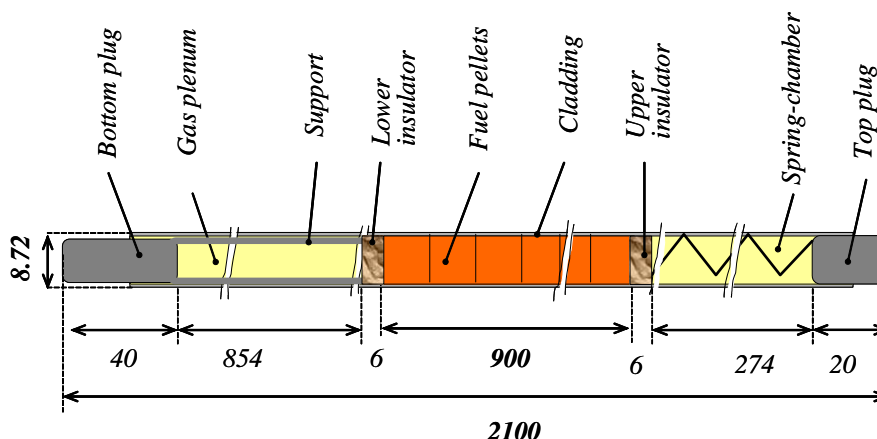


Table 1: Main geometrical dimensions of cercher fuel pins (EFIT)

Fuel pin element, mm	All core zones (outer)
Pellet diameter	7.20 (8.00)
Pellet height	10.0
Clad outer diameter	8.72 (9.52)
Clad inner diameter	7.52 (8.52)
Fuel column height	900
Insulation pellet diameter	7.2 (8.00)
Insulation pellet height	6.0
Gas plenum length	854
Spring chamber	274
Bottom plug	40
Top plug	20
Fuel pin length (total)	2 100

Computational tools

The SCK•CEN performance code FEMALE was developed mainly for the preliminary design modelling of the mixed oxide fuel behaviour in the fast spectrum systems cooled by liquid lead and lead-bismuth eutectic. For the development of this code, the JAERI code FEMAXI-V.1 [7] adapted to the fast neutron spectrum was used. The properties database was extended to the Pb-Bi and Pb coolants, to T91 steel cladding and to MOX and minor actinide fuels taking into account supplementary recommendations developed in the FP6 IP EUROTRANS and ELSY projects [8-12]. The classical parabolic law was used for growth of the oxidation layer on the cladding outer surface due to oxygen dissolved in the coolant ($\sim 10^{-6}$ wt.%). The corrosion rate was normalised to a value of 25 μm after one year of exposure in Pb-flow at 500°C. This value was used as a conservative estimation for the effect of protective layers deposited on T91 steel cladding.

The fuel performance code TRAFIC was used by Serco to model the EFIT fuel behaviour. TRAFIC was originally developed to model homogeneous fuels of sodium cooled fast reactors [13]. The code includes sophisticated mechanistic models for the thermal and mechanical behaviour of the pin, including fission gas release and detailed chemical modelling of fuel. The model of the behaviour of gas inside the fuel grains has been updated to separate helium from fission gas. The models of heat flow, mechanical interaction and (recently) fission gas behaviour have been extended to handle heterogeneous inert-matrix fuels. A library of correlations recommended by EUROTRANS for properties of fuel, cladding and Pb-coolant was used in these studies [8].

TRANSURANUS is a computer program that permits the thermal and mechanical analysis of fuel rod in various nuclear reactors [14]. Its development is ongoing at the ITU Joint Research Centre, Karlsruhe (Germany). As in TRAFIC, the code is 1 ½ dimensional. The geometric representation considers a number of axial slices each one divided in annular regions with constant and isotropic mechanical parameters known as “coarse zones”. Each coarse zone is divided into a fine mesh. The thermal analysis is performed under steady-state and transient conditions taking into account phase changes. The mechanical analysis relies on the solution of the constitutive equations coupled with the equilibrium and compatibility relations (semi-analytical solution). A carefully validated set of models was developed to describe thermal and irradiation induced densification of fuel, swelling due to solid and gaseous fission products, creep, plasticity, pellet cracking and relocation, oxygen and plutonium redistribution, volume changes during phase transitions, formation and closure of central void and treatment of axial friction forces.

Results of modelling

The irradiation conditions and power distribution at the beginning of fuel life (BOL) for the hottest fuel rods of the three-zone EFIT cercer core was calculated with MCNPX and ERANOS codes [15] and these results were used as input for the fuel performance codes FEMALE, TRAFIC and TRANSURANUS. The preliminary results of modelling of the thermomechanical state of the hottest rods at BOL are presented in [4,16].

In this paper, preliminary calculations are presented, under EFIT conditions, extending the BOL conditions up to 360 EFPD. A peak linear heat rate of between 23.0 and 23.2 kW/m was assumed in presented analyses.

Fuel temperature and gap performance

The performances of the hottest fuel rods of each of three zones of the EFIT cercer core described above was modelled with FEMALE, TRAFIC and TRANSURANUS code during operation time up to about one year.

The evolution of the maximum fuel temperature and the gap width are shown in Figures 3 and 4. For TRANSURANUS two runs were carried out assuming in fission gas release modelling a standard grains boundary saturation limit (10^{-4} mol/m²) and a reduced one (10^{-5} mol/m²).

Figure 3: Fuel centreline temperature (cercer core, hottest section)

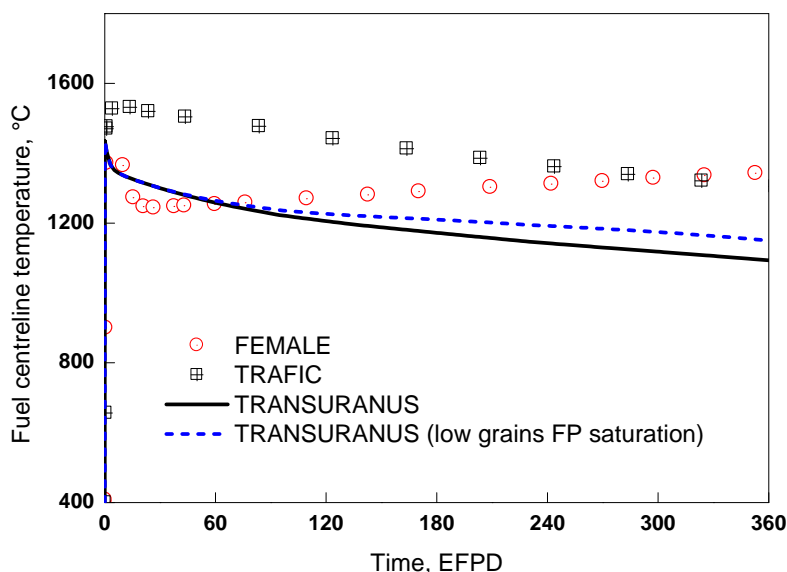
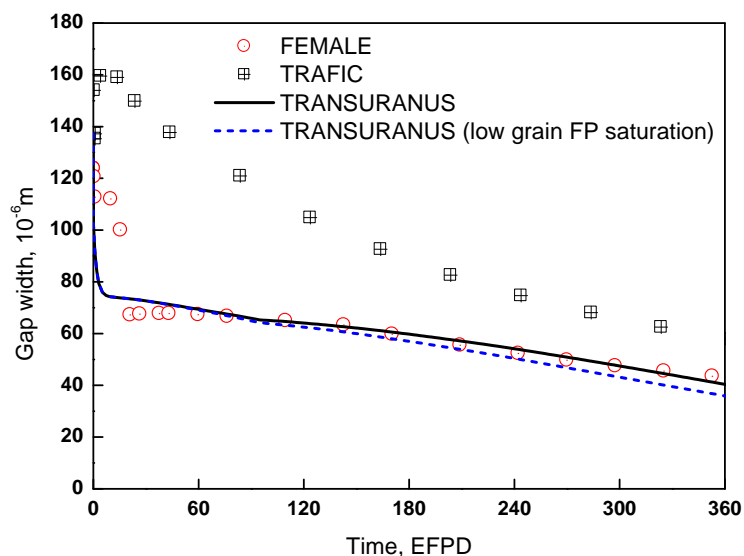
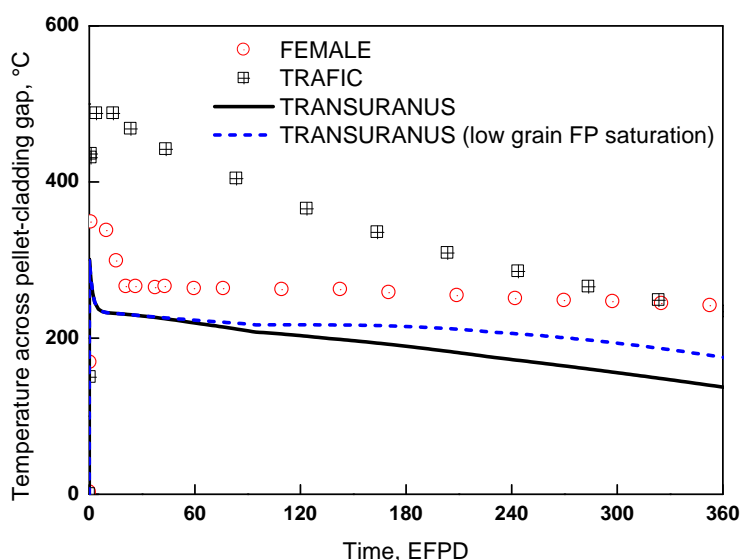


Figure 4: Gap width (cercer core, hottest section)



A fairly good agreement is seen between codes predictions, especially for FEMALE and TRANSURANUS, while a higher deviation is noted in TRAFIC gap width predictions up to about 100 EFPD. Regarding the fuel centreline temperature, FEMALE predictions point out a dominant role of the irradiation-induced degradation of the cercer composite thermal conductivity. This statement is confirmed considering that TRANSURANUS and FEMALE show a nice agreement of predictions for gap width as well as the temperature jump across the fuel-cladding gap. As to this latter parameter, a better agreement is shown if a higher release of gaseous fission products is assumed in TRANSURANUS calculations, see Figure 5. TRAFIC shows a more rapid decrease in the gap width than the other two codes, as shown in Figure 4, but starting from higher values. The larger gap drives the discrepancy found in the fuel centreline temperature prediction with values higher by 200-300°C than those from FEMALE and TRANSURANUS, that is mostly explained by the temperature jump across the gap predicted by TRAFIC, see Figure 5. Applied codes predict that peak centre temperature occurs at the start of in-reactor life, in all cases calculated values are well below the limit of 2 130 K where MgO is expected to begin to vaporise.

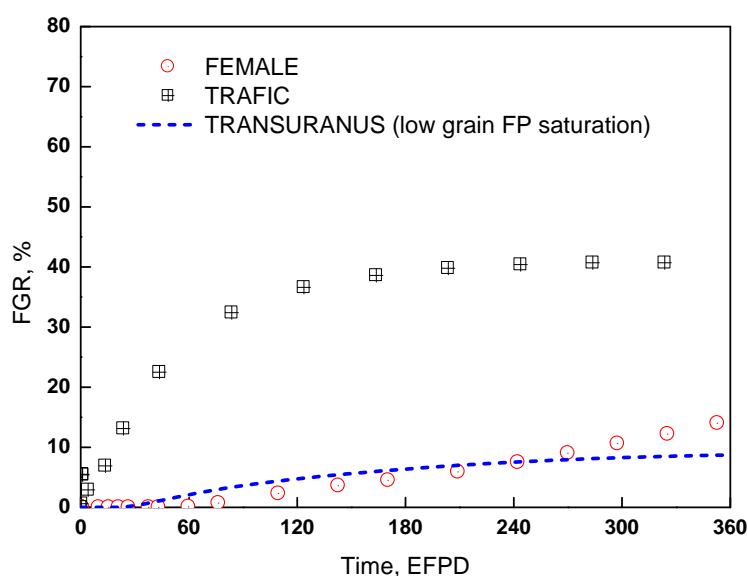
Figure 5: Temperature jump across the fuel-cladding gap (cercer core, hottest section)



Fission gas release

FGR calculated by different codes are presented in Figure 6. In FEMALE, due to a rather high fuel temperature, see Figure 3, the gaseous fission products (Xe, Kr, He) and the α -decay gas (He) accumulated within fuel grains start to be released to the fuel rod free volume at the early stage of irradiation.

Figure 6: Fission gas release calculated by applied codes (cercer core, hottest section)



In FEMALE the production rate assumed for helium is nearly four times higher than gaseous fission products. Due to a higher mobility within TRU-oxide fuel and also within MgO matrix helium release starts at lower fuel temperature and at a larger fraction in comparison with Xe and Kr, although all through a diffusion-based pathway.

In TRAFIC the produced gas is assumed to be completely released when fuel particle boundaries are reached (as would occur if the inert matrix were strongly cracked) so that the model is somehow a burst release mitigated by the diffusion that, due to the high fuel temperature in the first 100 EFPD, leads to a noticeable predicted release within the first year of irradiation. This result markedly deviates from the other codes predictions but this discrepancy could be mainly explained through gap size at the early stage of irradiation. A version of TRAFIC that included a preliminary model of the hold-up of fission gas in the inert matrix was produced later in the EUROTRANS project.

TRANSURANUS prediction presented in Figure 6 assumes a diffusion-based fission products drift to the fuel grains boundaries where a low saturation limit was assumed (10^{-5} mol/m³). TRANSURANUS prediction is fairly in good agreement with FEMALE provided that helium release is not accounted in Figure 6. In these calculations produced helium is fully released.

Conclusions

The analysis, up to 360 EFPD, of the thermomechanical behaviour of the composite cercer fuel rods under expected conditions of EFIT was performed with the fuel performance codes FEMALE (SCK•CEN), TRAFIC (Serco), and TRANSURANUS (ENEA) updated to the purpose.

The predictions of applied codes proved to be fairly consistent, especially regarding FEMALE and TRANSURANUS. Discrepancies of TRAFIC predictions were envisaged particularly regarding the early stage values of gap size and hence fuel temperature and fission gas release. These results, in general, confirm the agreement assessed at BOL conditions.

The fuel rod sited in the inner core region proves higher fuel temperature that is, in the investigated residence time, below the assumed limit. While the lower FGR predicted by FEMALE and TRANSURANUS coupled with a limited degrading of fuel thermal conductivity keeps nearly constant the peak fuel centreline temperature, in TRAFIC higher FGR seems not impacting fuel temperature as gap decrease does.

However, it should be taken into account that the present code calculations are performed applying certer fuel and T91 cladding properties where irradiation-induced effects are still not sufficiently known. In particular the degrading of thermal conductivity of MgO matrix could change noticeably the presented predictions. FGR modelling and matrix capability of retaining the helium produced under irradiation are still open issues. Supplementary benchmarks of the code models with the adequate experimental data have still to be performed.

Acknowledgements

This work was performed in the framework of the EURATOM FP6 Integrated Project EUROTRANS. The authors would like to express special thanks to the UK National Nuclear Laboratory for supporting Serco in this work and to the SCK•CEN MYRRHA project.

References

- [1] *Integrated Project EUROTRANS, Annex 1 – Description of Work*, Contract No. FI6W-CT-2004-516520, EURATOM FP6, March (2005).
- [2] Wallenius, J., et al., *Selection of the Preliminary Fuel Candidate for DM1*, Deliverable D3.7, FP6 IP EUROTRANS, Contract No. FI6W-CT-2004-516520, EURATOM FP6, February (2006).
- [3] Artioli, C., *Specification for the EFIT Core and Fuel Element*, Deliverable D1.6, FP6 IP EUROTRANS, Contract No. FI6W-CT-2004-516520, EURATOM FP6, November (2006).
- [4] Sobolev, V., et al., *Estimation of the Thermomechanical Performances of the Fuel Rods of EFIT-400(AFTRA), Loaded with the Selected TRU-fuels, at the Beginning of Operation (Intermediate Technical Report)*, Deliverable D3.5, EURATOM FP6, IP EUROTRANS, Contract No. FI6W-CT-2004-516520, March (2008).
- [5] Rimpault, G., et al., *Definition of the Detailed Missions of Both the Pb-Bi Cooled XT-ADS and Pb Cooled EFIT and Its Gas Back-up Option*, Deliverable D1.1, FP6 IP EUROTRANS, Contract No. FI6W-CT-2004-516520, EURATOM FP6, December (2005).
- [6] Haarmann, K., et al., *The T91/P91 Book*, Vallourec & Mannesmann Tubes (1999).
- [7] Suzuki, M., *Light Water Reactor Fuel Analysis Code FEMAXI-V*, JAERI Report, Tokai-Mura, Japan, January (2001).
- [8] Thetford, R., V. Sobolev, *Recommended Properties of Fuel, Cladding and Coolant for EFIT*, Deliverable D3.4, EURATOM FP6 IP EUROTRANS, Contract No. FI6W-CT-2004-516520, May (2006).
- [9] Sobolev, V., "Thermophysical Properties of Lead and Lead-bismuth Eutectic", *J. Nucl. Mater.*, 362, 235-247 (2007).

- [10] Toloczko, M.B., F.A. Garner, C.R. Eiholzer, "Irradiation Creep and Swelling of the US Fusion Heats of HT-9 and 9Cr-1Mo to 208 dpa at ~400°C", *J. Nucl. Mater.*, 212-215, 604-607 (1999).
- [11] Carbajo, J.J., et al., "A Review of the Thermophysical Properties of MOX and UO₂ Fuels", *J. Nucl. Mater.*, 299, 181-198 (2001).
- [12] Sobolev, V., "Thermophysical Properties of NpO₂, AmO₂ and CmO₂", *J. Nucl. Mater.*, 389, 45-51 (2009).
- [13] Cameron, R.F., et al., "Development and Application of the TRAFIC Code for the Analysis of Fast Reactor Safety Experiments", *Nucl. Energy*, 29, 141-147 (1990).
- [14] Lassmann, K., "TRANSURANUS: A Fuel Rod Analysis Code Ready for Use", *J. Nucl. Mater.*, 188, 295-302 (1992).
- [15] Rineiski, A., et al., *Comparison of the Neutronic Performances of the ETD Reference Core Loaded with the Selected TRU-fuels from BOL to EOL*, Deliverable D3.3, EURATOM FP6 IP EUROTRANS, Contract No. FI6W-CT-2004-516520, March (2010).
- [16] Calabrese, R., et al., "Heterogeneous Fuels for Minor Actinides Transmutation: Fuel Performance Codes Predictions in the EFIT Case Study", *Ann. Nuc. Energy*, 37, 867-874 (2010).