Current status and future developments of the TRANSURANUS code: Oxide fuels for the homogeneous recycling of minor actinides

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Abstract

Nuclear energy is playing a significant role in the global energy market thanks to its competitiveness and low carbon technology. While nowadays nuclear power plants (NPPs) are almost entirely based on thermal reactor technology, a new generation of fast reactors is investigated pursuing an improvement of the effectiveness in the use of natural uranium resources in parallel with an increase in proliferation resistance and a reduction in radiotoxicity of nuclear high-level waste, objectives achieved through the recycling of plutonium and the transmutation of minor actinides (MA). The deployment of fast NPPs conceived in the framework of the Generation-IV International Forum is foreseen to take place towards the middle of the century.

The transmutation of minor actinides in oxide fuels is achieved through two main options: homogeneous recycling, where small quantities of MA are diluted in the MOX driver fuel, and heterogeneous recycling, where MA are dispersed in high concentration in a UO_2 matrix and irradiated at the periphery of the core. The addition of minor actinides in oxide fuels has an impact on various aspects of in-pile performance such as the degrading of thermal conductivity and melting temperature, a pronounced helium generation leading either to high release or high fuel swelling. Radial redistribution of actinides and changes in fuel microstructures occurring under irradiation are also of concern. These aspects need to be carefully investigated to fulfil safety criteria under normal and accident conditions as a key requirement to support the sustainability of the nuclear business.

The description and modelling of MA-bearing fuels for Generation-IV systems is challenging and investigations are on-going. In parallel with the conventional methodology for fuel modelling the multi-scale approach, where the description of fuel behaviour relies on an atomistic, up to a macroscopic scale modelling, is gaining higher and higher importance. In addition, the TRANSURANUS fuel performance code benefits of this approach and research is on-going. In this paper the current status of the TRANSURANUS code is briefly reviewed and future developments are outlined with particular attention to the homogeneous strategy for MA transmutation where ENEA is actively involved and contributing to the verification and validation of new models as in the recently launched PELGRIMM Project.

Introduction

As a consequence of the Fukushima-Daiichi accident, some countries have announced their decision to phase out from nuclear business in the next two decades (Belgium, Germany and Switzerland) or to abandon any plans to reintroduce nuclear energy in the near term (Italy), others, while confirming their nuclear option, have revised the projected rate of development [1]. Notwithstanding the decrease in the projections of development, nuclear energy stands as one of the energy sources with the highest potential in tackling climate change through a reduction of greenhouse gas emissions, in particular CO_2 [2].

Management of radioactive waste arising from nuclear power production is a crucial aspect due to its political, economic and social implications. The development and implementation of a strategy to safely incinerate fissile plutonium and transmute MA is an important issue pursuing waste minimisation and reduced proliferation risks. A continuous recycling of actinides, implemented by combining reprocessing technologies with advanced reactor concepts, allows multiplication of the energy extracted per tonne of mined uranium by a factor between 30 and 100 [3]. Homogeneous recycling – where the reactor core is loaded with a driver fuel having a MA content between 2 and 5 wt.% – would therefore support the long-term sustainability of nuclear energy development and would respect non-proliferation issues [4] [5].

The potential benefits of MA transmutation have been strong drivers and several R&D initiatives have been launched worldwide (e.g. the Omega Project, AFCI – Advanced Fuel Cycle Initiatives, EURATOM FPs, etc.). International organisations such as the OECD/NEA, through its working groups, and the IAEA, through its projects, have been promoting R&D on partitioning and transmutation. These concepts were well-fitted in more general strategies such as those developed in the framework of the SNETP (Sustainable Nuclear Energy Technological Platform), Generation-IV, GNEP (the Global Nuclear Energy Partnership) [6].

In this paper the status of the TRANSURANUS fuel performance code for modelling MA-bearing oxide fuels is briefly presented and future perspectives are discussed. The main focus of the paper is on the homogeneous strategy where ENEA will actively contribute to the code verification and validation within the FP7 PELGRIMM project [7] [8].

Fuel requirements and modelling approaches for the deployment of fuel cycle closure

Competitiveness and high MA transmutation rate are fundamental objectives of next generation fast reactors to achieve these goals the average fuel burn-up should be in these innovative NPPs of the order of 150 MWd/kg_{HM} with peak values of 200-250 MWd/kg_{HM}, This requirement is demanding for the performance of fuel in terms of thermal conductivity degradation, fission gases and helium production/release, swelling, chemical and mechanical interaction with cladding. The integrity of cladding should be assured for values of fast neutron damage up to 250 dpa. The outlet coolant temperature and the fuel centreline temperature in the design of these innovative systems are of the order of 550°C and 2 350 K, respectively [8]. The fabrication process of MA-bearing fuel is very demanding in all its stages from reprocessing through conversion up to the shaping into suitable form for irradiation (e.g. regarding the risk of high occupational doses). For these reasons, the deployment of such an advanced fuel cycle goes far beyond the state of the art and significant R&D efforts are required [8].

The development and licensing of a new fuel needs typically 20–25 years accounting for the necessary irradiation testing and post-irradiation examinations. In this process, fundamental issues are the definition of fresh fuel properties and burn-up impact

through integral or separate-effects testing of the following properties: thermophysical properties (e.g. thermal conductivity, heat capacity), physical and mechanical properties (e.g. density and hardness), phase equilibria or stability characteristics, (e.g. solidus-liquidus and dissociation temperatures) and inter-diffusion and compatibility of fuel constituents [9]. According to this methodology, modelling was also greatly based on costly and long-lasting experimental campaigns. In the meantime, physics-based models have been developing and are expected to gain a relevant role for the description of innovative fuels under investigation. Analytical tools and models with different spatial and time scales (multi-scale approach) are available and helpful in supporting the analysis of experimental data.

Main experimental findings on MA-bearing oxide fuels

As mentioned, the homogeneous recycling strategy to safely incinerate fissile plutonium and transmute MA (Np, Am, Cm) considers the dilution in MOX driver fuel of MA at low concentration (<5 wt.%) to cope with reactivity coefficient requirements. In this regard, MA group extraction from irradiated fuel in a reprocessing facility integrated with the process of re-fabrication makes homogeneous recycling to be in general preferable from the point of view of non-proliferation [5].

The milestone experiment that demonstrated the technical feasibility of Np and Am transmutation is SUPERFACT [8]. In this experiment, four of eight rods (SF7 and SF13 containing 2% Np, SF4 and SF16 containing 2% Am) were aimed at investigating the homogeneous recycling option. The test was performed in the Phénix reactor, reaching a burn-up of 6.4 FIMA%. The elemental compositions of SF7-13 and 4-16 fuels were respectively $(U_{0.74}Pu_{0.24}Np_{0.02})O_{1.973}$ and $(U_{0.74}Pu_{0.24}\ Am_{0.02})O_{1.957}$. The fabrication route (sol-gel) proved to achieve high-quality pellets [8]. At given rating (BOL ~380 $W{\cdot}cm^{-1}$, EOL ~350 $W{\cdot}cm^{-1}$) a high-fission gas release was noted with values around 70% [10]. Helium release proved to be four times higher than reference MOX [11]. The SUPERFACT experiment showed that the radial distribution of americium was quite flat with a slight increase in the inner region of fuel pellet while for neptunium no redistribution was observed [10]. Regarding the radial distribution of plutonium, an increase in the central region of SF13 pin was noted[10].

Recent results on Np- and Am-bearing fuels irradiated at high linear heat rate for 24 hours revealed that in general the effects on measured un-irradiated properties and irradiation performance were slightly affected by the addition of a few percent of minor actinides [12]. The addition to MOX of americium and neptunium, the latter up to 12 wt.%, proved to have a limited impact on the values of thermal conductivity in a range of fuel temperature up to 1 770 K [13] [14]. Melting temperature is an important physical property in the evaluation of the thermal performance of nuclear fuel. In this regard, the effect of americium content on the solidus temperature was predicted to be about 4 K per wt.% [15]. Moreover, special attention was paid to the radial redistribution of actinides in MA-MOX fuel during the early stages of irradiation having significant effects on fuel power density and on thermal properties such as melting temperature and thermal conductivity. The results achieved in [16] showed that Pu and Am redistribution can be significant already for short-term operation where actinide migration is mainly driven by the pore migration mechanism.

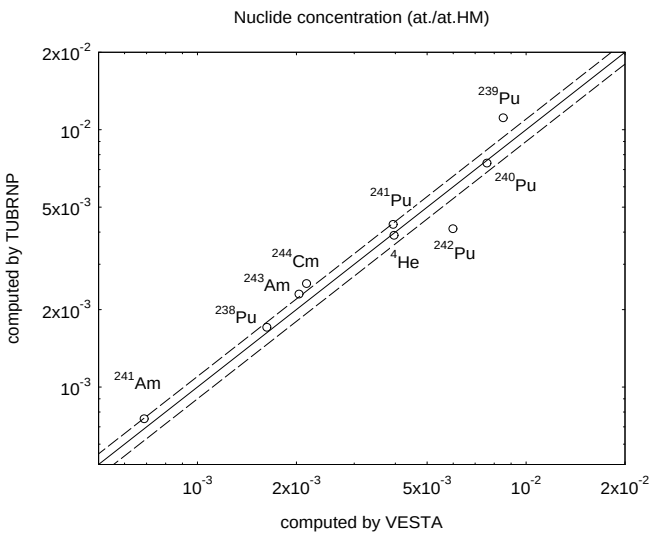
Knowledge of the amount of redistribution is very important for the design and integrity of fuel pins. This can be a limiting factor affecting the maximum allowable linear heat rating, eventually reducing the margin to the fuel melting point. Thus, the assessment of the two competing contributions (pore migration and solid-state diffusion) must be further analysed and it will be done in the framework of the PELGRIMM Project.

Current status of the TRANSURANUS code

Helium production under thermal neutron spectrum

The first step in the improvement of the TRANSURANUS code predicting capability is described in [17]. Helium is of great importance in MA-bearing fuels, where its production and release play an important role both under irradiation and after shut-down. Helium is produced in the fuel rod by means of α decays, (n, α) reactions and ternary fissions. The TRANSURANUS burn-up model (TUBRNP) was refined by extending the simulation of $^{238-242}$Pu, ^{241}Am, ^{243}Am and $^{242-245}$Cm, i.e. of nuclides relevant for power and helium generation in UO_2 and MOX fuels for thermal reactors. In particular, the contribution of ternary fissions, the (n, α) reaction in ^{16}O and an improved description of ^{241}Am branching ratios dealing with its neutron capture and following decay of ^{242}Am to ^{242}Pu was introduced in the code. The revised model for the production of helium proved to be in good agreement with VESTA code results up to a value of burn-up of 100 MWd/kg$_{HM}$, (see Figure 1).

Figure 1: Comparison between the average concentrations at the EOL in the MOX fuel predicted by VESTA and by TUBRNP for $^{238-242}$Pu, ^{241}Am, ^{243}Am, ^{244}Cm, ^{4}He [17]



The dashed lines represent a ±10% deviation from the bisector.

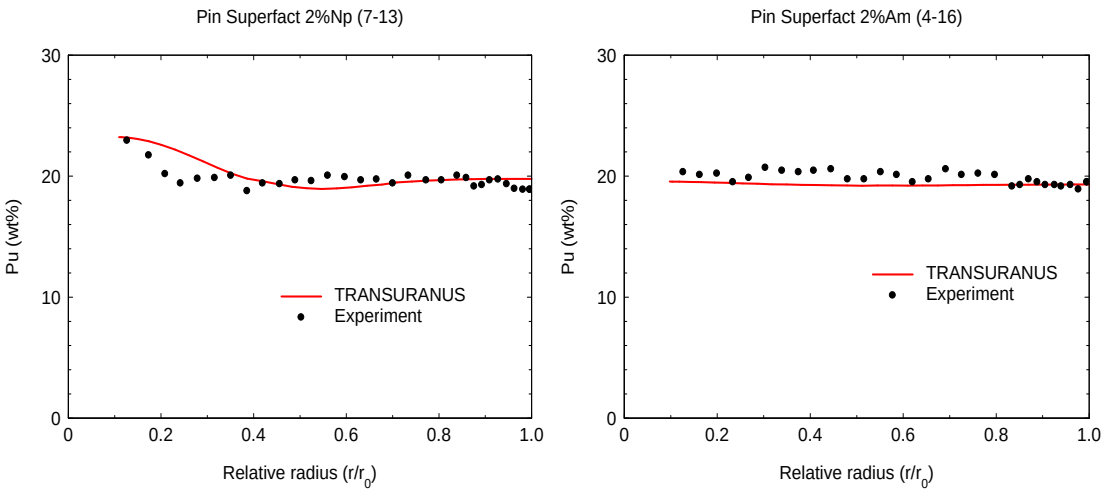
Fast neutron spectrum: plutonium and oxygen redistribution models

The TRANSURANUS model for the calculation of local burn-up (TUBRNP) was updated to extend code applicability to fast reactor analyses. One-energy group cross-sections for fast neutron spectrum were embedded in the TUBRNP Programme. A database of fission yields including Cs, Nd, Kr and Xe isotopes was built-in considering fission reactions for ^{235}U, ^{238}U, ^{238}Pu, ^{239}Pu, ^{241}Pu, ^{241}Am and ^{243}Cm nuclides [18]. These modifications enable TRANSURANUS to accurately calculate the radial power density profile and the evolution of MA in fast reactors. On this sound basis, plutonium and oxygen redistribution models (called PUREDI and OXIRED, respectively) were discussed [18]. The plutonium redistribution model, which is based on solid-state thermal diffusion, was extended by establishing the coupling with burn-up calculations. Model assumptions and the numerical stability of the extended version of PUREDI have been carefully verified by means of a code-to-code comparison with the finite element based COMSOL code [19]. In addition, diffusion coefficients for Pu and oxygen in MOX fuels have been revised, taking into account the reciprocal effect of the Pu concentration and the oxygen-to-metal ratio

on the mobility of such fuel elements. TUBRNP, PUREDI and OXIRED are coupled within the TRANSURANUS structure, constituting a robust and numerically stable package essential for fast reactors simulations. It should be noted that further investigations are needed for high fuel temperatures (above 2000 K), where a lack of data and large uncertainties have been identified concerning the Pu diffusion coefficient [19].

A preliminary application of the extended version of TRANSURANUS was carried out in the case of SF13 and SF16 pins irradiated in the SUPERFACT experiment. Figure 2 presents the comparison of the code results with the EPMA measurements carried out at ITU [10]. Predictions are in a reasonably good agreement with measurements suggesting that the increase of plutonium concentration in the SF13 rod is well described by a solid-state diffusion model in the case of long-term irradiation.

Figure 2: Plutonium redistribution: TRANSURANUS predictions in SUPERFACT SF13, SF16 [19]



Helium release model

A model for helium release was implemented as described in [18]. Concerning the intra-granular model it should be noted that trapping and resolution are in equilibrium and can be described by an effective diffusion coefficient while the grain boundary acts as a perfect sink [7]. The treatment of the inter-granular behaviour is based on the experimental evidence that the diffusivity of helium at the grain boundary is accelerated at temperatures above 800°C. As regards the absorption of helium, it is assumed that helium can infuse if its partial pressure in the inter-granular cavities is lower than that in the free volume, and that these cavities are already partially filled with the helium trapped at the grain boundaries [18].

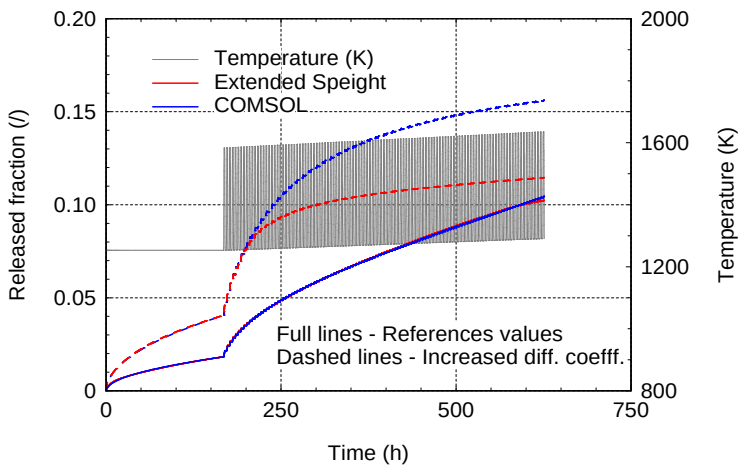
Future developments of the TRANSURANUS code

In addition to this well-assessed approach to fuel behaviour modelling, new methodologies in a multi-scale approach have been applied. Multi-scale modelling becomes more important and supports the conventional fuel performance codes by improving the description of material properties and models. The TRANSURANUS code also benefits from such an approach. In this perspective, current multi-scale developments for the TRANSURANUS code are mainly focused on fast reactor fuels under investigation. First principle and finite element calculations are underway to assess for example mechanical (e.g. elastic constants) as well as heat transport properties (e.g. heat capacity) or in order to assess the contribution of solid fission products to fuel swelling

[20]. First principle-based predictions of the migration energies for point defects in UN have recently been introduced in the thermal creep model.

Within FUMEX-III, a project on LWR fuel modelling at high burn-up co-ordinated by the IAEA, and F-BRIDGE, an EURATOM Project devoted to developing an approach to fuel modelling based on a fundamental understanding of its behaviour at atomic to the macroscopic scale, a new model for the description of fission gas in UOX fuel has been proposed. In particular, the intra-granular fission gas behaviour model was extended to account for gas bubbles motion relying on both molecular dynamics calculations for the evaluation of irradiation-induced resolution and finite element simulations for the simultaneous evolution of gas atoms and bubbles in the grains [20]. Standard operative as well as transient conditions were addressed accounting for, in the first case, the contribution of bubble motion to the effective gas diffusion coefficient and, in the second case, besides this, accounting for the precipitation rate of gas atoms in bubbles during ramps. These objectives were achieved, retaining the advantages of the existing formulation of Speight in terms of mathematical-numerical treatment. This extension of the Speight model was tested in a code-to-code comparison with COMSOL code as well as versus an experimental dataset referring, in particular, to UO_2 rodlets pre-irradiated up to 21 GWd/tU in an advanced gas cooled reactor (AGR) and ramped in the OECD/NEA Halden Reactor. The effect of power cycling on the predicted FGR fraction by means of the extended Speight model is shown in Figure 3.

Figure 3: Predicted FGR in a single grain submitted to a power cycling test by means of COMSOL (blue colour) and the extended Speight model implemented in the TRANSURANUS code (red colour), applying reference values of the diffusion coefficients (full lines), as well as diffusion coefficients multiplied by a factor 5 (broken lines) [20]

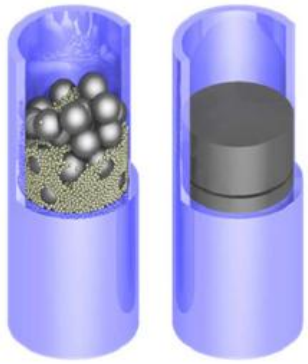


The results in Figure 3 reveal that at operating temperatures below about 1 900 K, the approximations in the extended Speight model provide good predictions for FGR in a spherical grain in comparison with the COMSOL predictions, when taking into account uncertainties caused by the physical parameters such as diffusion coefficients (prone to uncertainties by a factor of 5 up or down). Above this temperature, the smearing out of the trapping rate or bubble concentration in the grains causes errors on predicted FGR fractions during power variations which become of the same order of magnitude as those caused by uncertainties on the physical parameters. Nevertheless, it should be pointed out that at those temperature levels other physical phenomena must be taken into account, such as grain growth, which is prone to very large uncertainties as well. Furthermore, when applying the extended Speight model in a fuel performance code, it should be noted that the behaviour of the fission gas at the grain boundaries is also subject to uncertainties during power ramps, for example when grain boundary cracking

occurs. For this reason, the more detailed physical model for fission gas behaviour is still under development.

The development and validation of the TRANSURANUS code, on the basis of a multi-scale modelling, is extended in the frame of the PELGRIMM Project. This is a 4-year project where 12 partners from research institutions, education establishments and industries are addressing MA-bearing fuel developments for Generation-IV fast reactors. The TRANSURANUS code will be developed for both the heterogeneous and the homogeneous recycling options. In this framework, the current PUREDI model will be extended to include MA redistribution through a pore migration model. ENEA will collaborate with the code modelling group, focusing mainly on the description of the homogenous option and standard fabrication process (pellet). In this regard, the key activity is the modelling of the SPHERE semi-integral irradiation of americium containing MOX performed within the FAIRFUELS Project and on-going at the HFR of Petten (The Netherlands). This experiment will, for the first time, assess the in-pile performance of sphere-pac fuels compared with conventional pellet fuel, both fabricated at the JRC-ITU. Sphere-pac fuel allows for a large amount of gaseous fission products and helium to be accommodated without high internal stress, swelling and pore formation. A possible disadvantage is low-thermal conductivity and related high-central temperatures, an aspect that could be of concern. The SPHERE irradiation (2009/2013) will extend for nearly 300 full power days. The irradiation experimental data and PIE will be used for modelling purposes by the participants of PELGRIMM.

Figure 4: SPHERE experiment schematics, comparing sphere-pac and pellet type fuel



Conclusions

The development and qualification of fuel have been so far a successful but long and expensive process essentially based on an empirical approach. This knowledge is embedded in fuel performance codes deeply validated for thermal reactor conditions. Generation-IV fuels having prerequisites such as actinide recycling and high burn-up require an extension of the capabilities of current analytical tools. An intense activity by modellers is on-going to refine the TRANSURANUS fuel performance code to extend its applicability to the description of innovative fuels for fast reactors. In particular, significant efforts have been devoted to improving the models for the calculation of burn-up, helium production/release, redistribution of plutonium and oxygen for oxide fuels aimed at MA transmutation. In addition to this, a multi-scale approach is being applied, aiming at tackling the modelling of thermo-physical properties as well as fission gas release and swelling behaviour. Further developments and validation of improved models are expected from the PELGRIMM Project, where investigations will be performed on the promising concepts of sphere-pac fuel, in this framework the ENEA will contribute to the analysis of the SPHERE reference fuel.

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