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# Modelling actinide redistribution in mixed oxide fuel for sodium fast reactors

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ABSTRACT

In this paper, a model for the actinide redistribution mechanism in mixed oxide fuel for a sodium fast reactor is presented and compared with measured data collected from post irradiation examination on fuel from the JOYO experimental reactor. The model considers that solid-state thermal diffusion and vapour transport can simultaneously contribute to the plutonium and americium radial profiles. The effect of fuel non-stoichiometry on actinide transport as well as on pore velocity is taken into account. The model is embedded into the TRANSURANUS fuel rod performance code and calculates the evolution of the Am and Pu concentrations as a function of the radial temperature profile. The calculated actinide distribution is in good agreement with the experimental data. The results confirm that under normal operation conditions with a decreasing fuel power during its life-time, redistribution via pore migration is extremely important only during the central hole formation at the beginning of life, whereas thermal diffusion represents the dominant effect for long-term irradiation.



## 1. Introduction

In hypostoichiometric MOX (Mixed OXides) fuel for fast breeder reactors (FBRs), Pu and eventually Am redistribute and migrate to the central, high temperature pellet region. As a consequence, fuel thermal properties such as the thermal conductivity and the melting temperature could be strongly affected, resulting in a restriction of the safety margins for power uprating in commercial fast reactors (Olander, 1976). Accurate and reliable modelling of the redistribution effect is therefore essential for fuel performance codes in order to provide quantitative information to be used for design improvements and safety analysis for Generation IV reactors with MOX.

Several post irradiation examinations and out-of-pile experiments indicated that actinide migration is promoted by two main mechanisms (Bober and Schumacher, 1973): a) Solid-state thermal diffusion; b) Vapour transport by migrating pores. Besides diffusion, which is thought to be the most relevant, since it constitutes a continuously growing effect (Welland, 2011), it was recognized that pore migration could give a non-negligible contribution to the actinide concentration in the central part of fuel pellet (Guarro and Olander, 1975; Meyer, 1974). Unlike diffusion, pore migration towards the pellet centre usually occurs during the first hours of irradiation determining the central void formation and therefore, this represents a limited mechanism in time (Clement and Finnis, 1978; Olander, 1973a,b). There was a strong debate in the past on this subject, and no definitive conclusion was drawn up to now, especially because of the lack of experimental data and because the involved empirical parameters used for modelling are prone to large uncertainties (Guerin, 2012).

More recent short irradiation experiments carried out in Japan (Maeda et al., 2009, 2011) brought again the attention on this phenomenon, showing that redistribution via pore migration could be relevant at the beginning of irradiation. Nevertheless, modelling attempts were carried out by adopting an analytic solution, which is actually over-simplified and only valid for a very short time. In order to obtain reliable predictions, accurate calculations of fuel temperatures and hence of fuel restructuring must be carried out in the same simulation environment because of the strong coupling between thermal and transport related phenomena.

For this purpose, we have developed a comprehensive model for actinide redistribution (Am and Pu), which has been implemented

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in the TRANSURANUS fuel rod performance code with the aim to give a quantitative estimation of the modelling parameters involved and of the relative importance of the two mechanisms, given the available experimental data. In particular, the TRANSURANUS model for Pu redistribution (PUREDI), described in Lassmann (1992) and Lassmann et al. (2013), and recently refined in order to include the effects of oxygen-to-metal ratio, burn-up and their feedback (Di Marcello et al., 2012a), is further extended in this work in order to take into account Am redistribution and the effect of vapour transport. The latter is discussed in detail, because of the implications it may have on fuel restructuring and central void formation, which are essentially driven by pore migration.

After the description of the TRANSURANUS actinide redistribution model, the parameters found in the literature are reviewed and discussed. The code is then applied to the simulation of the B11 and B14 short-term irradiation experiments carried out in the JOYO nuclear reactor facility, consisting of 7 fuel pins containing few percentages of minor actinides. Post irradiation examinations (PIE) data have been used as validation database for the TRANSURANUS code. Finally, a specific sensitivity study is carried out in order to assess the importance of the vapour transport redistribution mechanism on the final predicted Am and Pu radial profiles.

## 2. Actinide redistribution model

### 2.1. Main equations and boundary conditions

In the present work, the Pu and Am migration is modelled considering that thermal diffusion occurs simultaneously with vapour transport via pores in the fuel. Vapour transport can occur either via pores or cracks which cross the fuel from the central void to the periphery. However, cracks heal relatively fast during irradiation due to the high temperature, and this contribution was identified as negligible for long-term operation (Clement and Finnis, 1978; Olander, 1973a). Therefore, only pores are taken into account for vapour transport modelling.

The actinide migration model is based on the work of Ishii and Asaga (2001). The time-dependent radial evolution of the actinide concentration is given according to the Fick's second law:

$$\frac{\partial c}{\partial t} = -\nabla \cdot J \tag{1}$$

where $J$ is the vector flux of actinide atoms (Pu and Am) per unit area and unit time; $c$ is the actinide concentration; and $t$ is time. Since migration occurs via two different paths, the transport flux $J$ must take into account both the solid-state diffusion ($J_{\mathrm{Diff}}$) and pore migration ($J_{\mathrm{Pore}}$) contributions as follows:

$$J = J_{\mathrm{Diff}} + J_{\mathrm{Pore}} \tag{2}$$

In the TRANSURANUS fuel performance code (Lassmann, 1992), the Pu migration model has been recently revised by the authors (Di Marcello et al., 2012a,b, 2011) in order to include the effects of oxygen-to-metal ratio, burn-up and their feedback. Based on these studies, the actinide redistribution occurring via thermal diffusion can be calculated according to Bober et al. (1973):

$$J_{\mathrm{Diff}} = -D_{\mathrm{Diff}}\left(\nabla c + c(1-c)\frac{Q}{RT^2}\nabla T\right) \tag{3}$$

where $J_{\mathrm{Diff}}$ is the diffusive flux of actinides; $R$ is the universal gas constant; $T$ the absolute temperature; $Q$ is the effective molar heat of transport; and $D_{\mathrm{Diff}}$ is the thermal diffusion coefficient.

The second mechanism, represented by pore migration, is based on the work of Clement and Finnis (1978). The model considers that a macroscopic flux of actinide atoms, $J_{\mathrm{Pore}}$, is established due to the movement of lenticular pores which have in front of them a layer of enhanced Pu or Am concentration:

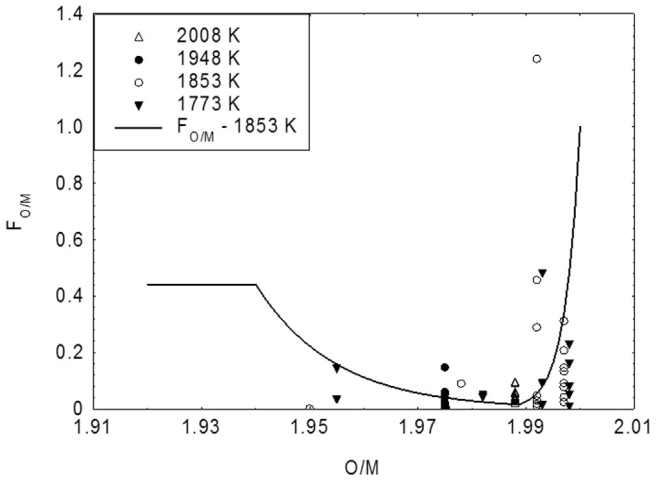


**Fig. 1.** Correction factor for the diffusion coefficient as a function of the oxygen-to-metal ratio. Measured data refer to Glesser-Leme and Matzke (1982).

$$J_{\mathrm{pore}} = \frac{PD_{\mathrm{Pore}}}{d}\frac{\vec{v}}{v}\delta c \exp\left(-\frac{D_{\mathrm{Pore}}}{lv}\right) \tag{4}$$

where $l$ and $d$ are the pore diameter and thickness, respectively, $P$ is the fuel porosity, $D_{\mathrm{Pore}}$ is the diffusion coefficient in the enhanced layer and $v$ is the pore velocity, which is always directed towards the pellet centre. The expression for the actinide enhancement across the pore is the following:

$$\delta c = Alc|\nabla T| \tag{5}$$

where $A$ is a constant. Assuming axial symmetry and neglecting axial concentration gradients, actinides can migrate only along the radial coordinate ($r$), so that the following equation is obtained by combining Eqs. (1)–(5):

$$\frac{\partial c}{\partial t} = \frac{1}{r}\frac{\partial}{\partial r}\left\{r\left[D_{\mathrm{Diff}}\left(\frac{\partial c}{\partial r} + c(1-c)\frac{Q}{RT^2}\frac{\partial T}{\partial r}\right) - D_{\mathrm{pore}}\left(APc\frac{l}{d}\frac{\partial T}{\partial r}\exp\left(-\frac{D_{\mathrm{pore}}}{lv}\right)\right)\right]\right\} \tag{6}$$

The boundary conditions at the fuel outer $R_{\mathrm{o}}$ and inner $R_{\mathrm{i}}$ radius for Eq. (6) must ensure that the mass balance of Pu or Am is preserved during migration:

$$J(r = R_{\mathrm{o}}, t) = J(r = R_{\mathrm{i}}, t) = 0 \tag{7}$$

The solution of Eq. (6) with boundary conditions (7) is obtained by means of the finite difference scheme described in Di Marcello et al. (2012a). The model applies for the different Pu isotopes contained in the fuel (Pu-238, Pu-239, Pu-240, Pu-241, Pu-242) and to Am as well (only Am-241 is considered for the sake of simplicity). In order to reduce the computational burden in the Pu case, the concentration of the different isotopes after redistribution is calculated on the basis of their relative concentration at the previous time step. This allows solving Eq. (6) only for the total Pu concentration and keeping, at the same time, good accuracy (for details see Di Marcello et al., 2012a).