Journal of Alloys and Compounds 271–273 (1998) 632–635

The influence of americium oxide inclusions on the thermal conductivity of inert-matrix fuel

K. Bakker*, R.J.M. Konings

Netherlands Energy Research Foundation ECN, P.O. Box 1, 1755 ZG Petten, The Netherlands

Abstract

The thermal conductivities of heterogeneous mixtures of americium oxides and inert-matrix materials are computed. In the case of americium-oxide inclusions with a random shape the Finite Element Method (FEM) has been used and in the case of inclusions with a spherical shape an analytical equation has been used. The influence of fission product-induced damage to the inert matrix on the temperature distribution and on the overall thermal conductivity has been modelled for various sizes of the fissile particles. © 1998 Elsevier Science S.A.

Keywords: Thermal conductivity; Americium; Transmutation; CERCER

1. Introduction

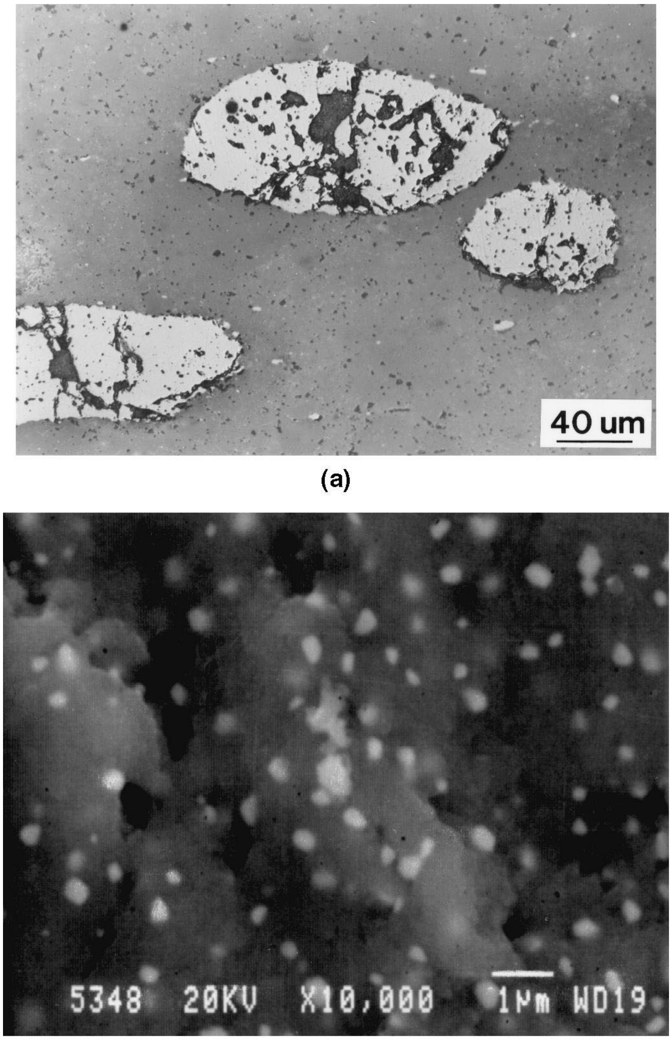
Partitioning and transmutation of actinides is presently being studied as a complementary option in the management of nuclear waste. It involves the separation of the actinides from spent fuel and subsequent re-radiation in nuclear reactors. Heterogeneous ceramic–ceramic (CERCER) mixtures of americium oxide in a support material, such as $MgAl_2O_4$ or MgO, are fuel concepts that are studied extensively at present [1]. The thermal conductivity of CERCER mixtures is an important property since it determines the power that can be generated in a fuel rod that contains americium oxide. At present no data is available on the thermal conductivity of CERCER fuels that contain americium oxide. Also, data for the pure americium oxides are limited. The only experimental thermal conductivity (λ) data are reported by Schmidt [2]. However, these data for freshly annealed AmO_2 (λ(333 K)=0.69 W/mK) and Am_2O_3 (λ(333 K)=0.82 W/mK) are extremely low compared to the other actinide dioxides. Schmidt et al. [3] observed that the conductivity of freshly annealed AmO_2 and Am_2O_3, strongly decreases due to α-decay. Moreover, the uncertainty of the thermal conductivity of AmO_x is enlarged by burn-up effects. Hence, the thermal conductivity of AmO_x is treated as a variable in the present approach, in which the upper and lower limits of the overall thermal conductivity of heterogeneous fuels are estimated.

The thermal conductivity of CERCER fuels decreases during irradiation, which is partially due to the influence of fission products on the microstructure of the inert-matrix material. Fission products, that are formed by fission of americium or its transmutation products are likely to cause a decrease of the thermal conductivity of the fuel material due to two reasons. Firstly, they induce irradiation damage in the material, such as dislocations and eventually polygonization. Secondly, they form gas bubbles, precipitates or are incorporated in the lattice of the AmO_x particles or the matrix. Irradiation in a nuclear reactor of $MgAl_2O_4$ [4] or ZrO_2/CaO matrices [5] containing UO_2 particles showed that fission products induce damage to a layer (thickness several microns) of the inert matrix around the UO_2 particles. As a result of this damage the thermal conductivity locally decreases, which influences the temperature distribution in the fuel. The lower thermal conductivity of the damaged layer increases the temperature difference between the centre of the fissile particle and the outside of the damaged layer. Moreover, the overall conductivity of the fuel decreases and consequently the central temperature of the fuel increases. Since the size of the AmO_x-inclusions influences the distribution of the damaged matrix layers, it is an important parameter. The size of the actinide-oxide particles in a matrix depends strongly on the preparation technique used, as has been shown for mixtures of $MgAl_2O_4$ and UO_2 [6]. The coprecipitation results in spherical UO_2 particles with a

---

*Corresponding author. Tel.: (31-224) 564386; fax: (31-224) 563608.

PII: S0925-8388(98)00175-3



(a)

(b)

Fig. 1. Micrographs showing the microstructure of a UO_2 pellet prepared with the (a) powder mixing technique, and (b) coprecipitation.

diameter smaller than 1 μm, while powder mixing yields spherical UO_2 particles with a diameter of up to 150 μm (Fig. 1).

In this paper the influence of the shape and the size of AmO_x-inclusions and the influence of fission products on the thermal conductivity of the fuel and the central temperature of the fuel are discussed.

2. Inclusion shape

The Finite Element Method (FEM) is a useful technique to compute a lower limit and a best estimate value of the thermal conductivity of heterogenous materials. The FEM has been used to compute the thermal conductivity of a $MgO–AmO_x$ mixture [7], using a micrograph of this material [8] to model the microstructure. Details of these computations can be found in [7].

Spherical inclusions have the smallest influence on the overall thermal conductivity of all inclusion shapes [9]. Eq. (1) describes the overall thermal conductivity of a material containing randomly distributed spherical inclusions [9].

$$1 - c_D = \frac{\lambda_D - \lambda_C}{\lambda_D - \lambda_M}\left(\frac{\lambda_M}{\lambda_C}\right)^{1/3} \tag{1}$$

where λ_M, λ_D and λ_C represent the thermal conductivity of the matrix material, the actinide oxide and the mixture, respectively. c_D ($0 \le c_D \le 1$) represents the concentration of inclusions.

Fig. 2 shows the results of FEM thermal conductivity computations on an $AmO_x–MgO$ mixture containing 14.4 vol% AmO_x and 2% porosity, as described in [7]. The thermal conductivity of the $AmO_x–MgO$ mixture is computed for various ratios of the thermal conductivity of AmO_x and MgO. The thermal conductivity of a AmO_x–

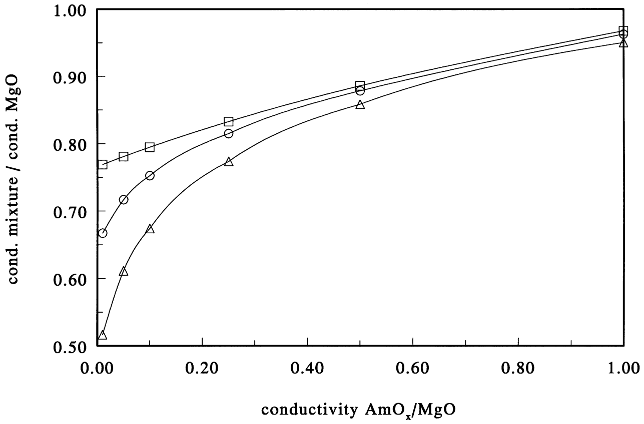


Fig. 2. A lower limit (△) and a best estimate value (○) of the thermal conductivity of AmO_x inclusions in a MgO-matrix, using the microstructure described in [7], computed with the FEM for various ratio's of the thermal conductivity of AmO_x and MgO. The thermal conductivity (□) of spherical AmO_x inclusions in a MgO-matrix, as computed with Eq. (1), is shown as a comparison. The AmO_x content (14.4 vol%) and the porosity (2%) for all three curves are equal. The lines serve as a guide to the eye.

MgO mixture containing spherical (3D) AmO_x inclusions (14.4 vol%) and spherical pores (2%) in a MgO-matrix (Eq. (1)) is shown as a comparison. Fig. 2 shows that spherical inclusions induce a considerably smaller decrease of the overall conductivity compared with the inclusions in the MgO–AmO_x mixture used in the FEM computations. It should be taken into account that the porosity in the AmO_x/MgO material is 2%, which causes the conductivity ratio of the mixture to be slightly less than 1.0 for a conductivity ratio AmO_x/MgO of 1.0.

3. Inclusion size

The influence of the size of the AmO_x particles on the temperature distribution in a $MgAl_2O_4$ matrix is discussed in this section. In the present computations the thermal conductivity of $MgAl_2O_4$ is considered constant (7 W/mK) in the temperature range of the present study (1150–1750 K). It is further assumed that the thermal conductivity of the matrix is not changed by β-, γ- and neutron-irradiation. In order to approximate the influence of fission products, it is assumed that in the volume of the $MgAl_2O_4$ matrix that is within 10 μm of an AmO_x-inclusion, the thermal conductivity is decreased by a factor of 2 compared to unirradiated $MgAl_2O_4$. This is a rough estimation, since the amount of damage varies over this 10 μm layer and the actual decrease of the thermal conductivity due to fission products is unknown and will depend on temperature and burn-up. The thermal conductivity of AmO_x is assumed to be 0.6 W/mK, also independent of burn-up and temperature. The influence of the temperature gradient that exists between the centre and the edge of the fuel is not taken into account in these calculations. The temperature calculations were performed for a pellet diameter of 7.14 mm, a linear heat generation rate of 20 kW/m, an AmO_x content of 14.4 vol% (independent of inclusion size) and an outer temperature of the pellet of 900°C.

The temperature difference (ΔT_{sphere} between the centre and the edge of the AmO_x particles, shown in Table 1, is obtained from the following equation [10]:

$$\Delta T_{\text{sphere}} = \frac{q'''r^2}{6\lambda} \tag{2}$$

where q''' is the power density and r is the radius of the AmO_x particle. ΔT_{sphere} is added to the temperature difference over the boundary layer of damaged $MgAl_2O_4$ and this sum (ΔT_{sum}) is shown in Table 1. The results show that the temperature difference between the outside of the damaged $MgAl_2O_4$ layer and the centre of the AmO_x particle increases approximately quadratically with the radius of the inclusion and is 10 K for a radius of 200 μm.

Next, the Finite Element Method (FEM) has been used to compute the overall conductivity of the fuel for four inclusion sizes (20 μm, 50 μm, 100 μm and 200 μm). The FEM computations have been performed in a cylindrical symmetry. Finally, the overall thermal conductivity has been used to calculate the central temperature of the fuel (in a non-damaged $MgAl_2O_4$ region), which is shown in Table 1 together with the vol% of $MgAl_2O_4$ that is damaged by fission products. These computations were also performed for the case when no damage is induced in the $MgAl_2O_4$ lattice, allowing a comparison of the results.

The percentage of damaged $MgAl_2O_4$ depends strongly on the size of the inclusion, which is reflected in the overall thermal conductivity and the central temperature of

Table 1
The influence of inclusion size on various parameters and properties

Inclusion (μm)	ΔT_{sphere}	ΔT_{sum}	Overall therm. cond. W/mK	Damaged $MgAl_2O_4$ (%)	Fuel central temperature (K)
20	0.1	0.1	2.9	100	1720
50	0.6	0.7	4.7	29	1514
100	2.4	2.6	5.3	12	1477
200	9.6	9.9	5.5	6	1464
No damage	[a]	[a]	5.7	0	1453

[a] This depends on the sphere size, which has not defined for this case.

the fuel. ΔT_{sum} increases and the central temperature decreases with particle size. Since the differences in the central temperatures are more significant than the differences in ΔT_{sum} large inclusions are preferred in order to keep the fuel temperature relatively low. The amount of damaged material is smaller in the case of spherical inclusions, compared to inclusions of other shapes but comparable size. This is due to the smaller surface/volume ratio of spherical particles. Thus, both from the point of view of the extent of damage of the inert-matrix material and from the influence of the inclusion shape on the thermal conductivity as discussed in Section 2, spherical inclusions are favourable.

4. Conclusions

The thermal conductivity of a heterogeneous mixture containing spherical AmO_x inclusions is higher than that of a mixture containing randomly shaped inclusions due to the following two reasons. Firstly, spherical inclusions have the smallest effect on the decrease of the overall conductivity of all inclusion shapes. Secondly, the influence of fission products on the thermal conductivity of the inert matrix is smaller in the case of spherical inclusions.

The temperature difference between the centre of an AmO_x-particle and the edge of the damaged $MgAl_2O_4$ layer around this particle is relatively small and will probably not have a large influence on the fuel behaviour.

Changes of the $MgAl_2O_4$ structure induced by fission products have only a small influence on the overall thermal conductivity and the central temperature when the size of the AmO_x particles is larger than ±100 μm.

Acknowledgements

Hj. Matzke and S. Casalta are acknowledged for supplying us with the photograph of the $MgO–AmO_x$ microstructure. This work was financially supported by the Dutch Ministry of Economic Affairs (Mefisnumber 60 437).

References

[1] J.F. Babelot, R. Conrad, W.M.P. Franken, J. van Geel, H. Gruppelaar, G. Mühling, C. Prunier, M. Rome, M. Salvatores, Proc. GLOBAL'95 Versailles, France (11–14 Sept., 1995) 524.

[2] H.E. Schmidt, Proc. 4th Journées d' Actinides, Harwell, UK (June 26, 1974), Atomic Energy Research Establishment Report AERE-R-7961 (1975).

[3] H.E. Schmidt, J. Richter, Hj. Matzke, J. van Geel, Proc. 22nd Intl. Conf. on Thermal Conductivity, Tempe, AZ (1993) 920.

[4] Ph. Dehaudt, A. Mocellin, G. Eminet, L. Caillot, G. Delette, M. Bauer, I. Viallard, Proc. IAEA Technical Committee Meeting on Research of Fuel Aimed at Low Fission Gas Release, Moscow (1–4 October 1996) IAEA-TECDOC-970, p. 23.

[5] R.M. Berman, M.L. Bleiberg, W. Yeniscavich, J. Nucl. Mater. 2 (1960) 129.

[6] R.J.M. Konings, K. Bakker, J.G. Boshoven, H. Hein, M.E. Huntelaar, J.D. Meeldijk, C.F. Woensdregt, H. Zhang, Proc. GLOBAL '97, p. 1044.

[7] K. Bakker, R.J.M. Konings, J. Nucl. Mater., in press.

[8] S. Casalta, Etudes des proprietes du systeme Am–O en vue de la transmutation de l'americium 241 en reacteur a neutrons rapides, Thesis, Université d'Aix–Marseille I (1996).

[9] B. Schulz, High Temp.–High Press. 13 (1981) 649.

[10] C.T. Walker, W. Goll, T. Matsumura, J. Nucl. Mater. 245 (1997) 169.