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Journal of Nuclear Materials 275 (1999) 112–114

journal of nuclear materials

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Reply to the comments by J.H. Evans about two papers 'Thermal treatment of UO_2 irradiated in a pressurized water reactor: swelling and release of fission gases' [1] and 'Microstructural analysis and modelling of intergranular swelling of an irradiated UO_2 fuel treated at high temperature' [2] by I. Zacharie, S. Lansiart, P. Combette, M. Trotabas, M. Coster and M. Groos

P. Combette *, I. Zacharie

STXN, 31-33 Rue de la Fédération, 75752 Paris cedex 15, France

Received 29 April 1999; accepted 29 April 1999

The comments made by J.H. Evans call for the following remarks.

1. Xenon diffusion coefficient and activation energy

Measurements performed during thermal treatment (1130–1715°C) led to an empirical xenon diffusion coefficient expressed as (see Fig. 1):

$$D = 3.3 \times 10^4 \ \exp\left(\frac{-4.6\ \text{eV/atom}}{kT}\right)$$
$$= 32887 \ \exp\left(\frac{-53527}{T}\right).$$

The experimentally determined activation energy is 4.6 eV/atom. In Ref. [1] Matzke reports a value of 4.7 eV/atom for the diffusion of the most abundant fission gas, i.e., Xe, in a damaged UO_2 lattice. This value is in good agreement with the above result. Matzke's value is valid for damaged UO_2. His value for undisturbed Xe-diffusion of 3.9 eV/atom (very short irradiation) increases by 0.8 eV/atom to 4.7 eV/atom for the case that gas atoms interact and get temporarily trapped due to radiation damage. This damage saturates rather early during the reactor irradiation. The value of 4.7 eV/atom is thus thought to be valid for the burner in the work of Zacharie et al. as well.

2. Work presented by Zacharie [5]

Measurements made by Zacharie et al. using UO_2 fuel irradiated to 25 GWd/tU, then annealed between 1130°C and 1715°C, showed the following:

An initial phase of less than 30 min during which oxide swelling and xenon release are interrelated. This phase is characterized by:

- *fast xenon release* due to interlinking of intergranular bubbles present at grain boundaries and formation of 'tunnels' that emerge at the free surfaces of UO_2, thus enabling massive release of fission gases to the environment,
- a predominant *intergranular swelling* effect that increases rapidly with time. This is due to interlinking of bubbles present at the grain boundaries,
- *intergranular swelling* of lesser intensity, which also increases with time. The present study does not cover

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* Corresponding author. Tel.: +33-1 40 56 18 32; fax: +33-1 40 56 13 07.

[1] J. Nucl. Mater 255 (1999) 85–91.

[2] J. Nucl. Mater 255 (1999) 92–104.

PII: S0022-3115(99)00135-X

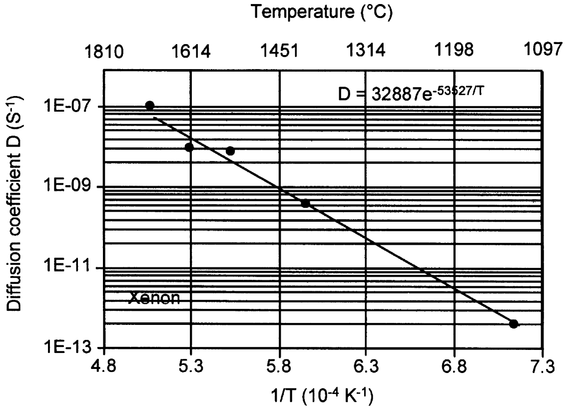

Fig. 1. Empirical xenon diffusion coefficient D as a function of the inverse temperature [5].

the intragranular phenomenon, which could not be investigated for lack of a transmission electron microscope (TEM).

A second phase that occurs during the interval from 30 to 600 min after start of irradiation. This phase is characterized by:

- slow release of thermally activated xenon with square root of time,
- intergranular swelling that increases slowly with time and is due to coalescence of the intergranular bubbles,
- intragranular swelling that remains nearly stationary over time.

As evidenced in Fig. 2, intergranular swelling is far more significant than intragranular swelling regardless of annealing temperature.

The model developed by Zacharie et al. is only concerned with intergranular swelling, which is well adapted to the experiment. This model does not explain the intragranular swelling phenomenon. Gas release observed during phase two correlates well with a Booth-type atom diffusion model with square root of time where the xenon diffusion coefficient, as depicted in Fig. 1, accounts for only one mechanism over the temperature range of interest (1130–1715°C).

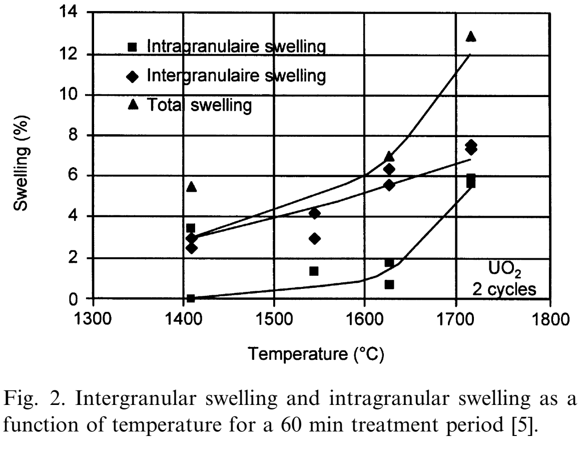

Fig. 2. Intergranular swelling and intragranular swelling as a function of temperature for a 60 min treatment period [5].

Fission gas atom transport to the grain boundaries takes place by atomic diffusion and/or intergranular bubble migration. According to the relevant literature [2–4], the latter is only activated at temperatures exceeding 1545°C.

Consequently, a bubble migration mechanism was not included in our study of xenon release; and despite the fact that the mechanism proposed by Evans [6–8] is known to the authors and quoted in Ref. [5].

3. Evans' model based on bubble migration in a concentration gradient of vacancies from the grain boundaries

The model does not explain the observed intergranular swelling, which is always the predominating factor in swelling measured between 1130°C and 1715°C.

The model does not suitably account for the observed intragranular swelling, either in quantitative or in qualitative terms. Measurements made by Zacharie et al. (Fig. 8 of Zacharie et al.'s first paper) show little swelling up to 1600°C; then over the range from 1630°C to 1715°C this phenomenon 'takes off' (see Fig. 2). By contrast, the Evans model (curve of Fig. 2) shows monotonous swelling between 1130°C and 1715°C.

Moreover, the asymptotic phenomenon of intragranular swelling measured by Zacharie et al. with an asymptotic dependence on temperature (Fig. 8 of Zacharie et al.'s first paper) is not replicated by Evans. All of his curves seem to tend toward a common limit.

A study of the sensitivity of results to the model input parameters remains to be carried out.

4. Conclusions

Theoretical work conducted by Zacharie et al. on the basis of Brownian bubble movement at the grain boundaries provides a good explanation for the intergranular swelling observed in experiments. It also explains why xenon is transported by diffusion from the matrix to the boundaries and consequently accounts for the also observed fission gas release. It is valid over a range from 1000°C to 1700°C for fuel irradiated to 25 GWd/tU, which contains little fission gas.

The mechanism proposed by Evans is nevertheless interesting, since it associates 'Xe release' and 'intragranular swelling', unfortunately without providing an explanation for the predominant intergranular swelling phenomenon. Nor does it explain the asymptotic change in intragranular swelling with annealing temperature.

The two studies are complementary, since one (Zacharie et al.'s model) explains the observed intergranular swelling and the other (Evans model) described intragranular swelling as correlated with xenon gas release. An analysis based on the use of a TEM (similar to that performed by Zacharie et al.) would enable the correlation of intragranular swelling values and fission gas release data obtained experimentally with the Evans model.

References

[1] Hj. Matzke, J. Chem. Soc. Faraday Trans. II 83 (1987) 1121.

[2] C. Baker, Europ. Appl. Res. Rep. 1 (1979) 35.

[3] S. Kashide, K. Une, J. Nucl. Sci. Tech. 28 (1991) 1090.

[4] C. Ronchi, Hj. Matzke, Europ. Appl. Res. Rep. 5 (1984) 1105.

[5] I. Zacharie, PhD Thesis, Ecole Centrale de Paris, 1997.

[6] J.H. Evans, J. Nucl. Mater. 210 (1994) 21.

[7] J.H. Evans, J. Nucl. Mater. 225 (1995) 302.

[8] J.H. Evans, J. Nucl. Mater. 238 (1996) 175.