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Journal of Nuclear Materials 265 (1999) 112–116

journal of nuclear materials

New model of equiaxed grain growth in irradiated UO_2

O.V. Khoruzhii *, S.Yu. Kourtchatov, V.V. Likhanskii

National Research Center of Russian Federation 'TRINITI', Troitsk Institute for Innovation and Fusion Research, Troitsk, Moscow Region 142092, Russian Federation

Received 26 June 1998; accepted 18 September 1998

Abstract

Analysis of available models of equiaxed grain growth in polycrystalline UO_2 nuclear fuel demonstrates the discrepancy between their predictions and experiments under irradiation conditions. As the discrepancy is a systematic overprediction it could be explained if an additional retardation effect exists under irradiation, which is not taken into account by the correlations. As a possible reason of the effect we consider the defect areas arising on the grain faces as a result of interaction with fission tracks. An additional retarding force grows with the irradiation power. Predictions of the model based on the above hypothesis are in qualitative and quantitative agreement with existing experimental data in a wide range of temperatures and intensities of irradiation as well as of fuel burnup.

1. Introduction

The grain size is one of the most important characteristics of polycrystalline reactor fuel affecting significantly its behaviour under nominal and accident conditions. The highest uncertainty in this parameter is caused by the grain growth process, which accelerates as the fuel temperature grow. Besides the change of grain size, the propagation of grain faces in course of grain growth results in sweeping of fission products from the fuel matrix, that could be the most effective channel of fission product release from grain interior to grain faces. For this reason a lot of efforts were spent to investigate grain growth kinetics in polycrystalline reactor fuel in detail. Two types of grain growth may be distinguished – equiaxed and columnar. In this work we restrict our consideration to the equiaxed grain growth only.

Currently several models and correlations describing equiaxed grain growth are available [1–4] which are widely used in various codes for modelling the fuel behaviour [5–7]. The effect of principal importance which was revealed from the very beginning of the problem experimental analysis is the retardation of the grain growth under irradiation (see Ref. [8] and discussion therein). For the most part this effect is attributed to an influence of fission products accumulation at the grain surfaces in form of small inclusions and bubbles [2,3]. The respective correlations take into account this effect by diminishing of the grain growth rate with burnup [1].

Despite the fact that correlation suggested in Ref. [1] is generally considered to be the best available it has some fundamental shortcomings. First, available data on burnup which would be sufficient for appreciable retardation of grain growth are essentially different [8]. This indicates that correlations of such type are incomplete. Second, rapid grain growth was also observed during out-of-reactor annealing of fuel with high burnup in presence of great amount of intergranular bubbles [9,10]. Hence, it may be concluded the retardation effect to be a consequence of the irradiation itself rather than the burnup.

Recent experimental results of Risø project [11] also compel to take a critical look at models similar to those in Ref. [1]. The work [11] reports on observed grain growth under irradiation in a wide temperature range. The comparison of these data with predictions of the correlation [1] demonstrates that the use of burnup dependence of grain growth rate leads to qualitative

* Corresponding author. Tel.: +7-095 334 6182; fax: +7-095 334 5158; e-mail: okhor@fly.triniti.troitsk.ru.

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disagreement, whereas without taking into account the factor of burnup the correlation [1] considerably overestimates grain growth as compared with observations. For example at starting grain size 5 μm and temperature 1500°C the observed growth is approximately 4.7 μm whereas the predicted one is about 20 μm.

To resolve the contradiction authors of Ref. [11] suggest a significant change of model parameters – appreciable decrease of growth rate and limiting grain size. Although such an approach eliminates the disagreement between the model predictions and particular set of experimental data it is undesirable from the view point of practice. The reason is that the parameters proposed in Ref. [1] were validated on a wide set of out-of-pile experiments and therefore properly describe grain growth without irradiation.

The aim of the present paper is the development of a model that would properly describe available experimental data on grain growth under annealing conditions as well as under irradiation and would be reasonable from the physical point of view.

2. Qualitative analysis

The physical cause of equiaxed grain growth lies in impossibility of thermodynamical equilibrium of boundary between grains of different size. If conditions of dynamic equilibrium in the grain corners are fulfilled then boundary is curved and convexity is turned to grain of greater size. Trying to get flat the boundary will migrate and large grain will grow at the expense of the small one. According to existing conceptions [12] the described process forms self-similar size distribution of grains and results in auto-model growth of characteristic grain size. The latter means that the characteristic growth rate in any moment of time depends only upon characteristic grain size a at a given moment. This may be written as follows:

$$V_a = \frac{\mathrm{d}a}{\mathrm{d}t} = K(T)F_{\mathrm{dr}}(a), \tag{1}$$

where V_a is characteristic rate of grain boundary migration under action of driving force F_{dr}, which depends on mean grain size a, K is mobility, which is function of temperature T. Characteristic value of driving force acting on the unit surface area is of the order of σ/a, where σ is surface tension.

In presence of any retarding effect one has to add to the right-hand side of Eq. (1) a corresponding retarding force F_{ret}

$$\frac{\mathrm{d}a}{\mathrm{d}t} = K(T)(F_{\mathrm{dr}}(a) - F_{\mathrm{ret}}). \tag{2}$$

According to Ref. [1], equiaxed grain growth without irradiation is well described by the following relation

$$\frac{\mathrm{d}a}{\mathrm{d}t} = K\left(\frac{1}{a} - \frac{1}{a_{\max}}\right). \tag{3}$$

Here

$$K = 5.24 \times 10^7 \exp(-32\,100/\mathrm{T})\ \mu\mathrm{m}^2/\mathrm{h}, \tag{4}$$

and retarding force is expressed through limiting grain size

$$a_{\max} = 2.23 \times 10^3 \exp(-7620/T)\ \mu\mathrm{m}. \tag{5}$$

It was proposed in Ref. [1] to introduce additional retarding force proportional to fuel burnup in order to account for grain growth retardation, which is caused by fission products accumulation on grain faces during irradiation. With the use of experimental data [13] proportionality factor was determined and the modification of relation (2) was constructed, which takes into account the dependence of grain growth rate upon burnup:

$$\frac{\mathrm{d}a}{\mathrm{d}t} = K\left(\frac{1}{a} - \frac{1 + 0.002B}{a_{\max}}\right), \tag{6}$$

where B is fuel burnup in [MW d/tU]. Relation (6) properly describes experiments [13] but leads to qualitative disagreement with results of some other experiments, for instance Refs. [8,11].

As mentioned above, available experimental data could be brought into correspondence if one assumes that irradiation may immediately give rise to additional retarding effect on grain growth. The most natural is an idea of some inclusions similar to gas bubbles situated on grain boundary as a physical cause of retardation. It can be presumed the retarding effect to be associated with short-lived 'inclusions' which are generated when fission fragments intercept grain face. For instance, such inclusions could appear as areas with intermediate orientation of crystalline lattice and/or with high concentration of defects which arise in course of track annealing owing to counter-recrystallisation on a boundary between two grains. The peculiarities of interaction of fission fragments with grain boundaries could be also assumed on the basis of the observations of fission tracks in small-grain UO_2 films [14].

As a consequence of nonequilibrium of their state, these defect areas will disappear due to diffusion as pores do, but until ultimate disappearance they may act on grain boundary like inclusions of the secondary phase. It means, they will retard boundary migration with a force proportional to their radius R and surface concentration c. To determine the force, simple expression proposed by Zener [15] can be used, based on the assumption of 'rigid' grain boundary

$$F_{\mathrm{retr}} \cong \pi\sigma Rc. \tag{7}$$

One may estimate radius of 'inclusions' as a track size, i.e. 1–5 nm [14]. Surface concentration is determined by the production rate w (per unit area) and the characteristic time of annealing τ

$$c \cong w\tau. \tag{8}$$

The latter depends upon a type of process, which accounts for inclusion disappearance. If it is grain boundary diffusion then according to Ref. [16],

$$\tau^{(\mathrm{gb})} \cong C^{(\mathrm{gb})} \frac{R^4 kT}{D^{(\mathrm{gb})} W \sigma \Omega}, \tag{9}$$

where $C^{(\mathrm{gb})}$ is a numerical factor, k Boltzmann constant, $D^{(\mathrm{gb})}$ grain boundary diffusivity, W ($\cong$1 nm) boundary width, Ω atomic volume (volume per atom of uranium in crystal lattice UO_2).

If grain boundary diffusion does not work for annealing of inclusions, their time of existence may be determined by volume diffusion. Then [16]

$$\tau^{(\mathrm{vol})} \cong C^{(\mathrm{vol})} \frac{R^3 kT}{D^{(\mathrm{vol})} \sigma \Omega}. \tag{10}$$

The rate of inclusions production is determined by the number of fission fragments that intercept unit area of boundary in a unit time

$$w \cong 2\dot{F} l_{\mathrm{tr}}, \tag{11}$$

where $\dot{F}$ is fission rate per unit volume, l_{tr} track length. Eventually we get an expression for retarding force

$$F_{\mathrm{ret}}^{(\mathrm{gb})} \cong \frac{2\pi C^{(\mathrm{gb})} R^5 l_{\mathrm{tr}} kT}{D^{(\mathrm{gb})} W \Omega} \dot{F} \tag{12}$$

in case of grain boundary diffusion and

$$F_{\mathrm{ret}}^{(\mathrm{vol})} \cong \frac{2\pi C^{(\mathrm{vol})} R^4 l_{\mathrm{tr}} kT}{D^{(\mathrm{vol})} \Omega} \dot{F} \tag{13}$$

in case of volume diffusion.

In both cases retarding force per unit area of boundary does not depend upon grain size and can stop grain growth when the limiting size is attained.

$$a_{\mathrm{ir}} \cong \sigma / F_{\mathrm{ret}}. \tag{14}$$

As a consequence of strong dependence of a retardation force on radius of inclusions two possibilities exist, which could be responsible for the effect observed. First possibility is large-sized inclusions with small lifetime determined by grain boundary diffusion. Second one is small-sized and relatively long-lived inclusions which are annealed due to volume diffusion.

Let us estimate limiting grain size for both cases. In the first case after assuming $R_{\mathrm{in}} \cong 10$ nm, $D^{(\mathrm{gb})} \cong 10^{-12}$ m^2/s, $T \cong 2000$ K, $\dot{F} \cong 10^{19}$ 1/(m^3 s), $C^{(\mathrm{gb})} \cong 1$ and using values $l_{\mathrm{tr}} \cong 6$ μm, $\Omega = 4.1 \times 10^{-29}$ m^3, $\sigma = 1$ J/m^2 [17] we come to

$$a_{\mathrm{ir}}^{(\mathrm{gb})} \cong 40\,\mu\mathrm{m}. \tag{15}$$

In the second case assuming $R_{\mathrm{in}} \cong 1$ nm, $D^{(\mathrm{vol})} \cong 10^{-18}$ m^2/s, $T \cong 2000$ K, $\dot{F} \cong 10^{19}$ 1/(m^3 s), $C^{(\mathrm{vol})} \cong 0.2$ we obtain

$$a_{\mathrm{ir}}^{\mathrm{vol}} \cong 21\,\mu\mathrm{m}. \tag{16}$$

Both evaluations in order of magnitude properly describe additional retarding effect observed under irradiation. Of course, simple considerations stated above bear only qualitative and schematic features. Nevertheless, they provide quite natural explanation of all basic properties observed. Therefore relations similar to Eqs. (12) and (13) may be used for correlation. Remarkable that in both cases the dependences on temperature and fission rate may be put in the form

$$F_{\mathrm{ret}} = CT\dot{F}\,\exp(E/T), \tag{17}$$

where C is proportionality factor and E corresponding activation energy.

Consequently, at the frame of the present model the grain growth rate is described by expression (3) with additional retarding force:

$$\frac{\mathrm{d}a}{\mathrm{d}t} = K\left(\frac{1}{a} - \frac{1}{a_{\max}} - \frac{1}{a_{\mathrm{ir}}}\right), \tag{18}$$

where

$$a_{\mathrm{ir}} = \left[\frac{\dot{F}_0 \cdot T_0}{\dot{F} \cdot T}\right] A\,\exp(-E/T). \tag{19}$$

Here $\dot{F}_0 = 50$ MW/tU and $T_0 = 1400$ K are characteristic values of fission rate and temperature, chosen for making variables dimensionless, and as for model parameters A and E they should be found on the basis of available experiments.

3. Analysis of experiments

Table 1 shows basic characteristics of experiments which were used for choosing model parameters. Given experimental set covers wide range of temperatures and fission rates and also allows to compare observed grain growth for fuels of substantially different burnup.

It is remarkable that the only experiment in this set with no grain growth observed differs from others merely in considerably higher intensity of irradiation. This may be naturally explained on the basis of above stated concept and serves as an important argument in favour of the proposed model. Although this experiment cannot be used for quantitative comparison with model predictions, it establishes an important qualitative restriction on the model parameters.

Shown in Table 1 data of experiments on out-of-pile fuel annealing (including data [9] for fuel with 2% burnup) well agree with correlation (3) proposed in Ref. [1] with no burnup dependence. This suggests a weak influence of burnup on grain growth. Taking into account a wide range of experiments lying in the base of correlation (3), it tells about necessity to use parameters K and $a_{\max}$ from Ref. [1] in absence of irradiation.

Table 1
Basic characteristics of experiments used to find and validate the model parameters

Experiment, reference	Intensity of irradiation (MW/tU)	Temperature, (K)	Fuel burnup (%)	Duration of experiment	Initial grain size (μm)	Final grain size (max) (μm)
Risø [11]	50	1300–2100	4	62 h	6	12
Turnbull [8]	21	2023	0–0.4 [a]	2–6 months	7	18
	0	1973	0	72 h	7	40
Small [9]	0	2073	2	2000 s	5.6 ± 0.3	7.14 ± 3.65
Zimmermann [18]	200–1000	1250–2000	0–10 [a]	1–10 months	10	10[b]
Hargreaves and Newbigging [13]	14–17	1000–1900	0–1.5	830 days	5	25
	20–21	1000–1900	0–0.7	320 days	5	25
Ainscough et al. [1]	0	1273–2123	0	168 days	3–15	10–50

[a] Experiment started for fuel with no burnup. Grains grew as burnup was getting higher.
[b] No grain growth was observed.

Available to date correlations on grain growth properly describe experimental data they are founded upon, but lead to essential contradictions under conditions of other experiments. In order to avoid such drawbacks as far as possible, the following scheme was used in the present work to find the correlation parameters for Eq. (19). At first, correlation parameters A and E were determined merely on the basis of Risø project data on grain growth at different temperatures [11]. Next, the model was validated. For this purpose its predictions were compared with data of other experiments with out change in parameters.

Fig. 1 shows experimental dependencies of final grain size on temperature [11] in comparison with predictions of the model with parameters $A = 326.5$ μm and $E = 5620$ K. These parameters were selected by minimisation of mean square deviation of computed grain size from the measured one in experiment [11]. Fig. 2 shows the ratio between computed and measured grain size for the present model as well as for model [1] both with taking burnup into account (relation (6)) and without granting it (relation (3)).

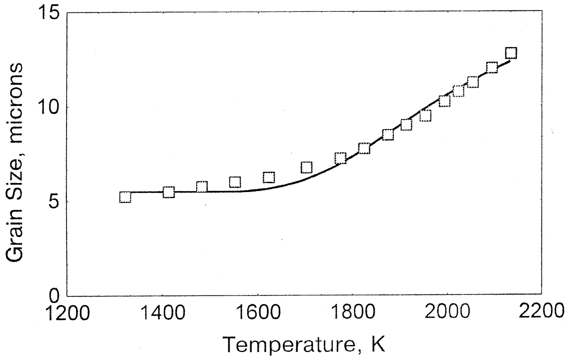


Fig. 1. Temperature dependence of experimentally observed [11] final grain size (□) and model (18) predictions (—–).

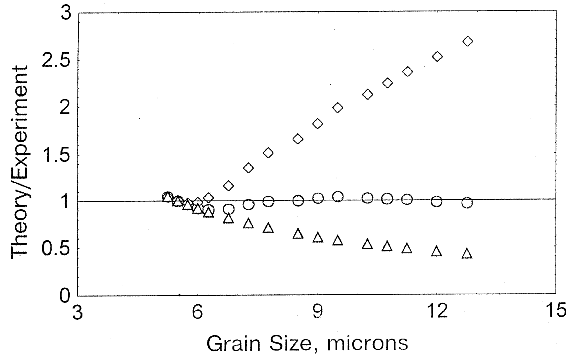
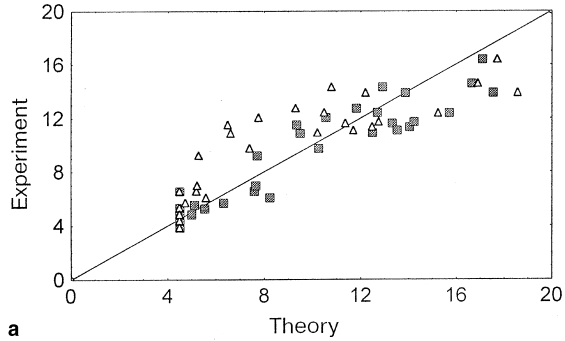


Fig. 2. Ratio between calculated and observed final grain size under conditions of experiment [11]. ○ – presented model, □ – correlation (3) from Ref. [1], △ – correlation (6) from Ref. [1] which takes burnup into account.

Predictions of the model and of correlation (6) are compared in Fig. 3 with experimental results [13] for two different intensities of irradiation. It is worth noting that predictions of the present model have lesser mean square deviation from the measurements, despite the fact that exactly these data were used to choose parameters of correlation (6).

Fig. 3 (b) also shows the comparison of computations with data of Turnbull experiment [8] which was performed at approximately the same intensity of irradiation. Predictions based on dependence Eq. (18) are in a satisfactory agreement with measurements [8], whereas calculations according to Eq. (6) lead to substantially greater grain sizes than experimentally observed.

For conditions of Zimmermann experiment [18] and chosen parameters of the model, the calculated value of maximum grain size is less than initial grain size of a sample in the whole temperature range. This result is in a qualitative concordance with the fact that grain growth was not observed in this experiment.



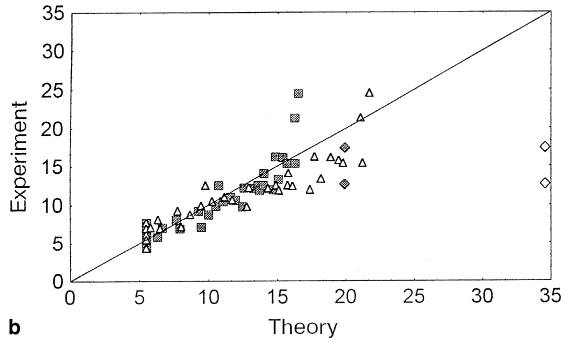


Fig. 3. Grain size (μm) predicted by the presented model (■) and by correlation (6) (△) in comparison with experimental data [13]. (a) For intensity of irradiation 14–17 MW/tU. (b) For intensity of irradiation 20–21 MW/tU. In (b) the model predictions are compared also with measurements [8]. ♦ – present model, ◇ – correlation (6).

4. Conclusions

1. Available experimental data on grain growth under in-reactor conditions show that irradiation leads to retardation of equiaxed grain growth.

2. Similar kinetics of grain growth in experiments with substantially different level of burnup suggests that the account for burnup alone cannot explain the observed retardation of the grain growth.

3. Available experimental data could be brought into correspondence if one assumes that irradiation may immediately give rise to an additional retarding effect on grain growth. The most natural is an idea to associate the effect with short-lived defect arrears arising on the grain faces as a result of interaction with fission tracks.

4. The model based on such a hypothesis is proposed, which gives qualitatively and quantitatively proper predictions of grain growth dynamics in wide range of temperatures, intensities of irradiation and level of fuel burnup.

Acknowledgements

The work was carried out under the support of the Russian Foundation of Basic Researches, grant N 96-02-18686.

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