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Growth mechanisms of interstitial loops in α-doped UO_2 samples

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Abstract

New experimental size distributions are presented for interstitial-type dislocation loops in $(U,Pu)O_2$ samples after 4 and 7 years of self-irradiation along with a new model. For this model, the work of Hayns has been extended to doped materials and to take into account additional phenomena, namely re-solution and coalescence as applied to gas bubble precipitation. The calculations are compared to the experimental size distributions obtained by means of Transmission Electron Microscopy for two different storage times. The role of re-solution and coalescence is discussed based on this model. This constitutes a basis for modelling the high burn-up structure (HBS) formation which is a consequence of the accumulation of radiation damage in nuclear fuels.

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1. Introduction

An increase of the discharge burn-up of UO_2 nuclear fuels in light water reactors results in the emergence of a microstructural change. This structure, called HBS for "high burn-up structure", is characterised by fission gas depletion from the matrix, appearance of a micrometric porosity, and a sub-division of the original grains. Although well characterised experimentally, important points have still to be clarified, among which are HBS formation mechanisms. In order to answer this issue, a study of the contribution of dislocation-type defects was conducted [1]. Since irradiated fuel is very complex, a simpler system reproducing some of the features observed in the HBS, was chosen in the form of UO_2 sintered pellets doped with ~10 wt% ^{238}Pu and stored for different times at room temperature, during which α-damage resulting from the decay of ^{238}Pu was produced. In this way, the single growth mechanisms of interstitial-type dislocation loops can be studied as these loops are directly formed by aggregation of interstitials, generated by the self-irradiation.

The objective of the present paper is to clarify the role of two basic mechanisms, namely irradiation-induced re-solution and migration and coalescence coarsening, involved in the damage evolution in a simplified system being self-irradiated fuel on the basis of a comparison with experimental results. It therefore contributes to the understanding of defect evolution in UO_2, rather than proposing a comprehensive model for the simulation of the behaviour of UO_2 during its irradiation in a reactor.

2. Experiments

The present samples were produced by incorporating 10 wt% $^{238}PuO_2$ (half-life = 87 years) to natural UO_2 by a sol–gel process [2], producing a homogeneous solid solution.

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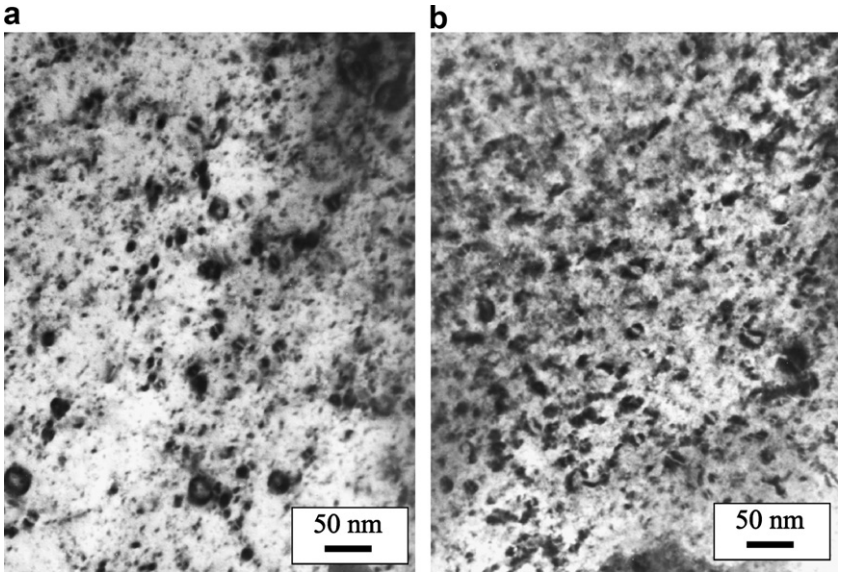

Fig. 1. TEM bright field image evidencing the formation of dislocation loops at $t = 4$ years (a) and $t = 7$ years (b), respectively.

The α-activity of the doped material was calculated to be 3.76×10^{10} Bq/g, from which the damage rate was found to be 9.06×10^{-9} dpa/s. TEM examinations were carried out on samples after storage times of 4 and 7 years at room temperature, which exhibit a dpa value of 1.1 and 2, respectively. Fragments of a few milligrams of the original $(U_{0.9}, Pu_{0.1})O_2$ pellets were crushed in ethanol. The resulting suspension was dropped on a copper grid filmed with carbon. The TEM used in this study is a Hitachi H700 HST operating at 200 kV and modified to handle radioactive materials. Fig. 1 shows two bright field pictures of the sample after 4 and 7 years, respectively.

The main defect that was observed consists of interstitial-type dislocation loops, whose contrast is similar to the ones simulated by Marian et al. [3] in α-Fe, homogeneously distributed within the material (Fig. 1). The dislocation loop size and density have been determined by manual image analysis. The loops size-distributions were obtained and are shown in Fig. 2.

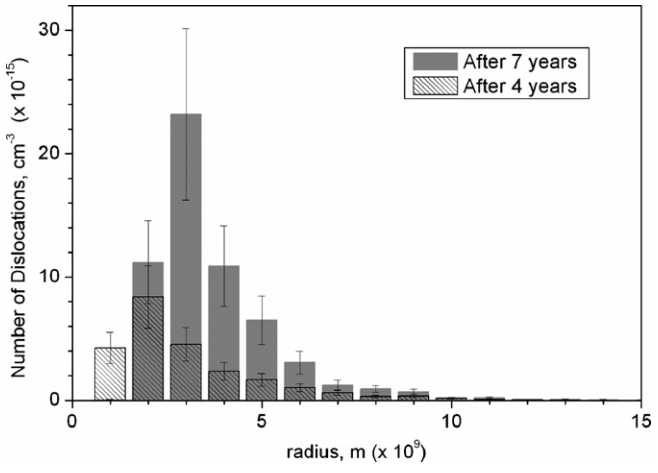

Fig. 2. Loop size histogram from the sample observed at $t = 4$ years and $t = 7$ years.

In order to enable a comparison with the computations in Section 4, the experimental distributions are described in terms of statistical values, more precisely the first four moments: the loop mean radius, standard deviation, skewness and normalised kurtosis (Table 1).

3. A comprehensive model of defects in α-doped UO_2

In the following, the diffusion of uranium interstitials and vacancies is only considered. Because of its low mobility, uranium diffusion in UO_2 is rate-controlling [4].

The most straightforward manner to model the behavior of uranium interstitials is by using the chemical rate theory [5]. Hayns [6] proposed a set of equations describing the nucleation and growth of a dislocation loop distribution in irradiated graphite and steel:

$$\frac{dc_v(t)}{dt} = K - \alpha_r c_v(t) c_i(t) - \sum_{n=2}^{n_{eq}} L_n c_{ni}(t) c_v(t) - D_v \rho_d c_v(t), \quad (1)$$

$$\frac{dc_i(t)}{dt} = K - \alpha_r c_v(t) c_i(t) - \sum_{n=2}^{n_{eq}} R_n c_{ni}(t) c_i(t) + L_2 c_{2i}(t) c_v(t), \quad (2)$$

$$\frac{dc_{2i}(t)}{dt} = \frac{1}{2} K_2 c_i(t) c_i(t) + L_3 c_{3i}(t) c_v(t) - L_2 c_{2i}(t) c_v(t) - R_2 c_{2i}(t) c_i(t), \quad (3)$$

$$\frac{dc_{3i}(t)}{dt} = R_2 c_{2i}(t) c_i(t) + L_4 c_{4i}(t) c_v(t) - R_3 c_{3i}(t) c_i(t) - L_3 c_{3i}(t) c_v(t), \quad (4)$$

$$\cdots$$

$$\frac{dc_{ni}(t)}{dt} = R_{n-1} c_{(n-1)i}(t) c_i(t) + L_{n+1} c_{(n+1)i}(t) c_v(t) - R_n c_{ni}(t) c_i(t) - L_n c_{ni}(t) c_v(t). \quad (5)$$