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Irradiation growth analysis by net defect flux in polycrystalline zirconium

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1. Introduction

Irradiation effect on metals has been a great concern since commercial nuclear reactors are constructed. In order to predict radiation degradation, various approach methods have been adopted by using computation simulation. Recently, rate theory, kinetic Monte Carlo (kMC), molecular dynamic (MD) and phase field are major computer simulation methods. Among these various methods, rate theory is most fundamental approach to understand reaction between mobile defects and immobile defects.

In early research stage, mean field approximation was applied in rate theory in order to simplify modeling [1-2]. In this method, two major assumptions were adopted to predict number density and size of extended defects. First, initial defect generation was assumed that defect will be homogenously distributed. Second, the reaction probability of mobile defects and immobile defects reaction is directly proportion with net defect flux to immobile defect. From this simplification approach, general phenomenon of irradiation degradation could be explained with fast calculation time. However, within this assumption, precise prediction was impossible because heterogeneous distribution of defect generation and fluctuation of net defect flux to extended defects were not considered. In the early 1990s, Semenov and Woo consider these heterogeneous defect generation and fluctuation of net defect flux in order to predict the radiation swelling [3].

This approach was also extended to zirconium alloy to understand irradiation growth behavior of cold worked zircaloy [4-7]. Zirconium alloys are widely used as fuel cladding materials in commercial reactors. Because these materials have reasonably good mechanical properties at commercial reactor condition and fairly low scattering neutron cross section. In order to enhance the safety of cladding materials, it is necessary to understand irradiation degradation of microstructures in materials in view of the safety enhancement and the life prediction of zirconium and its alloys.

However, annealed polycrystalline was not predicted by advanced rate theory. The aim of this study is to understand the fundamental behavior of zircaloy under irradiation. Therefore, in this work, irradiation growth of annealed polycrystalline will be predicted by using advanced rate theory. Specifically, extended defects will be quantified and then irradiation growth will be calculated.

2. Literature study of experimental results

Defects are generated, interacted each other and finally become extended defects, such as dislocation loops and grain boundaries, by diffusion. In zirconium and its alloy, cluster defects were eventually developed by dislocation loops because they are thermodynamically most stable state.

<a> dislocation loops:

Point defect consist of <a> dislocation loops either of vacancy or interstitial nature with Burgers Vector $1/3\langle 11\bar{2}0\rangle$ situated in the prismatic planes, which is perpendicular with <a> - axis.

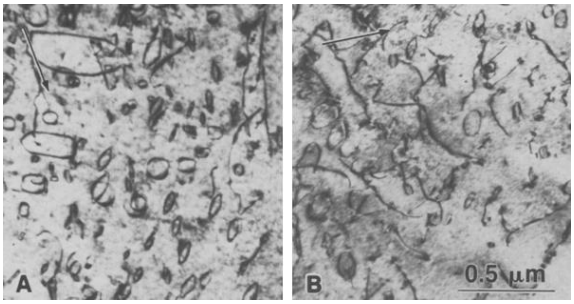

Fig.1. <a> type dislocation in an annealed zirconium microstructure at 700 K: (a) 1.1×10^{25} n/m^2; (b) 1.5×10^{25} n/m^2. Diffracting vector g = 1011 [8].

<c> dislocation loops:

The <c> dislocation loops have been analyzed as the vacancy type. They are located in the basal plane with a Burgers vector $1/6\ \langle 20\bar{2}3\rangle$ having a component parallel to the <c> axis.

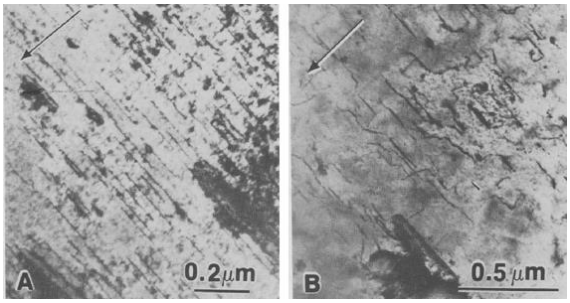

Fig.2. <c> dislocations loops in recrystallized annealed zircaloy-4 at 560K: (a) 8.5×10^{25} n/m^2 in BWR, (b) 7×10^{25} n/m^2 in a PHWR. Diffracting vectors are g = 0002 [8].

3. Classical rate theory

For the simplification, mean field approximation was adopted in rate theory, which was called as mean field rate theory (MFRT). The concentration of Frenkel pair defects was calculated by master equation of MFRT. In this equation, the reaction rate of mobile and immobile defects was calculated. Also it was assume that only point defect is generated and cluster defect is directly developed as dislocation loops. Since cluster defects were neglected and dislocation number density was obtained from experiment database, master equation could be composed of only two equation. Therefore, density change of dislocation loops could be analyzed by simple reaction probability. Defect rate equations are expressed by Eqs. (1) and (2).

$$\frac{dC_v}{dt} = K_o - K_{iv}C_iC_v - Z_v\rho C_vD_v \quad (1)$$

$$\frac{dC_i}{dt} = K_o - K_{iv}C_iC_v - Z_i\rho C_iD_i \quad (2)$$

K_0; Defect production rate
K_{iv}; Vacancy–interstitial recombination
ρ; Total sink density of aluminum matrix
Z_i and Z_v; Interstitial and Vacancy capture efficiency
C_i and C_v; Interstitial and Vacancy concentration
D_i and D_v; Interstitial and Vacancy diffusivity

Where, ρ is sink density of major sink i.e., dislocation loops and grain boundary in zirconium ($\rho = \rho_{dl} + \rho_{gb}$). Sink density of dislocation loops could be analyzed by number density and radius. Since radius could be analyzed by reaction probability, which could be calculated from Eq. (1) and (2). However number density is could not be calculated in this modeling because cluster defect concentration equation was neglected and Fokker – Planck equation was not adopted. Therefore, experimental result is used for analysis of density change of sink,

$$r_v = \sqrt{(S_v / \pi b N_v)} \quad (3)$$

$$\dot{S}_v = 2\pi r_v N_v (D_vC_v - D_iC_i) \quad (4)$$

N_v; Number density of dislocation loops
S_v; Total number vacancy in the dislocation loops
r_v; Radius of dislocation loops

Change of the total number of vacancy in <c> dislocation loops could be obtained from defect flux because reaction probability is directly proportion with circumference of dislocation loops.

4. Results and Discussion

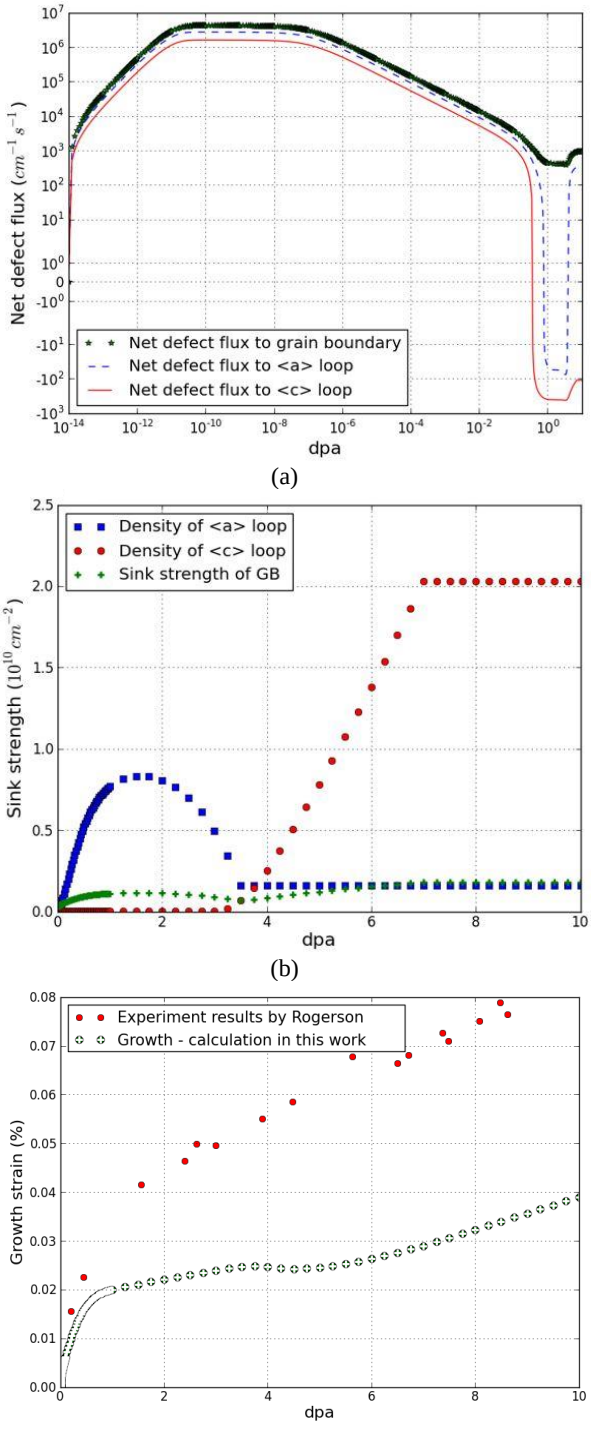

Fig.3. (a) net defect flux to dislocation loops and grain boundary; (b) sink strength of dislocation loops and grain boundary; (c) Modeled and experimental irradiation growth strain in annealed polycrystal zirconium at 553 K

For the behavior of the net defect fluxes to sinks, three types of defects are also analyzed because the major sinks are dislocation loops and grain boundaries. Fig. 3 (a) shows the net defect fluxes to sinks.

Major sinks are <a> and <c> dislocation loops and grain boundaries. Figure 3(b) shows the dislocation loop density and grain boundary sink

strength versus the dpa. The grain boundary sink strength increases with increasing dpa because it depends on the dislocation line and loop density. The <a> dislocation loop density has the shape of a semicircle before 4 dpa, after which <a> dislocation loops are saturated. The increase in the <c> loop density resembles an exponential model. However, <c> dislocations have a much higher sink strength than <a> dislocation loops.

This result does not match the experimental results well. Therefore, this advanced model clearly cannot be adopted for annealed polycrystal zirconium. Consequently, the assumptions for annealed polycrystal zirconium should be modified. The strength model for grain boundaries is based on the dislocation line and loop densities. The dislocation lines are assumed to be constant with respect to the dpa.

Therefore, the primary reason for the disagreement in the case of annealed polycrystal zirconium is dislocation loops. In this model, the sink strength of dislocation loops is calculated using experimental data on the number density. The behavior of the sink strength depends strongly on the number density.

However, as in the case of a single crystal, there are no precise experimental data in the given dpa region. In the other cases, a new mechanism for the grain boundary strength should be suggested. However, there is currently no general theory or simulation result for the grain boundary sink strength.

5. Conclusion

Irradiation growth was analyzed by net defect flux. <a> and <c> dislocation loops sink strength are depend on number density of dislocation loops. Initial irradiation growth was similar with experiment result. After initial stage, modeling results show discrepancy with experiment results.

Acknowledgement

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