Radiation induced growth of zirconium alloy in research reactor

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ABSTRACT

Zirconium alloys are widely used as reactor core materials because they have excellent corrosion- and radiation-resistant properties, while they are subjected to high neutron fluence, causing radiation-induced growth. Until now, several studies of radiation induced growth modeling have been conducted for the application to commercial reactor environments. However, the growth modeling for the application to research reactor condition has not been established well. To examine radiation induced growth behavior, this study is focused on radiation induced defect behavior at relatively low temperature by adopting radiation damage theory. For fundamental understanding, radiation damage theory is also briefly reviewed in this study.

1. Introduction

Zirconium is one of the most favorable core materials because of its excellent properties (radiation and corrosion resistant) compared to other structure materials in light water reactor. Hence vast amount of experiment and theoretical performance test has been done in various points of view such as corrosion and change of mechanical properties and dimension.

In commercial reactor core, structure materials are located in high temperature and high pressure environment. Therefore, main concern of structure materials is corrosion and mechanical properties change than radiation effects on materials.

However, radiation effects on materials become more important phenomena because research reactor condition is different from commercial reactor. The temperature is lower than 100 ℃ and radiation dose is much higher than that of commercial reactor.

Among the radiation effect on zirconium based metal, radiation induced growth (RIG), known as volume conservative distortion, is one of the most important phenomena.

Recently, theoretical RIG modeling based on radiation damage theory (RDT) and balance equation are developed. However, these growth modeling have limited framework of single crystal and high temperature.

To model theoretical RIG in research reactor, qualitative mechanism must be set up. Therefore, this paper intent is establishing defect flux mechanism of zirconium base metal in research reactor for RIG modeling. After than theoretical RIG work will be expanded to research reactor condition.

2. Radiation damage theory

RDT is the mathematical expression of radiation induced defect quantity and quality.

Hence quantification of radiation induced defect (RID) concentration is the essential part of theoretical RIG modeling. RDT is must needed to RIG modeling.

Because RDT is the basic part of radiation induced phenomena, it is researched from uranium to structure materials. The early stage RDT, the basic assumption about the RID was considered only one type that point defect (frenkel pair) [1, 2]. However, in that framework, theoretical modeling of radiation effects on metal was not well fit experimental result.

These problems were solved by adopting the concept of radiation damage morphologies. In the 1990s, Woo and Singh modified the incorrect framework by adopting the cascade concept that radiation damage produced not only point defects but also clusters [3, 4].

2-1. primary radiation damage

The RID generation sequence could be classified by two parts. The first part is composed with cascade generation and relaxation phenomena. In these phenomena, atom behavior was explained by kinetics and atomic fluctuation. This phenomena occurs so quickly (< 10-11). Therefore, in this time region, atom diffusion dose not considering. The mathematical expression is

$$G_{NRT} = N \int_{\tilde{E}}^{\hat{E}} \Phi(E_i)\sigma_D(E_i)dE_i$$

Where $\mathbf{N}$ is the lattice atom density, $\Phi(\mathbf{E_i})$ is the energy-dependent particle flux, $\sigma_{\mathbf{D}}(\mathbf{E_i})$ is the energy-dependent displacement cross section.

2-2 Rate Theory & balance equation

After cascade generation and relaxation time region, atom diffusion is main effect of RID concentration. These phenomena could be mathematically expressed by balance equation from chemistry. From balance equation it could know defect concentration at any given time.

$$\frac{dC_v}{dt} = K_o - K_{iv}C_iC_v - K_{vs}C_vC_s^T$$

$$\frac{dC_i}{dt} = K_o - K_{iv}C_iC_v - K_{is}C_iC_s^T$$

Where $\mathbf{K_0}$ is defect production rate, $\mathbf{K_{iv}}$ is vacancy–interstitial recombination, $\mathbf{C_i}$ is interstitial concentration, $\mathbf{K_{vs}}$ is vacancy–sink reaction, $\mathbf{K_{is}}$ is interstitial–sink reaction, $\mathbf{C_s^T}$ is the total sink strengths of all the extended defects in the material.

3. Radiation induced growth mechanism

The fundamental mechanism of RIG is atom rearrangement by defect flux behavior of RID. Historically, defect flux behavior established at 1979 by Holt [5]. Most recently, Woo expanded Holt work by establishing defect flux mechanism on zircaloy [6]. However until now, sink (grain boundary, dislocation loop, dislocation line, precipitate, defect cluster) effect on

zircaloy is not unclear.

Therefore, this paper intends to reveal defect flux behavior at zirconium alloy in research reactor. Fortunately defect flux mechanism of single crystal and zircaloy was briefly revealed by many researchers [7, 8].

Prior to modeling defect flux in research reactor condition, these model need to be reviewed. After that, defect flux to sink will be researched at low temperature.

3.1 Growth mechanism of single crystal mechanism

In case of single crystal, major sink is only dislocation and dislocation loop. Therefore, RIG begins from developing of dislocation and dislocation loop. Before breakaway region, interstitial loop in prism plane is main reason of <a> -axis elongation. However after breakaway region, vacancy loop on basal plane is also main reason of <c> -axis construction.

At first stage of RIG, dislocation and dislocation loop are bias sink because interstitial diffusion coefficient is much higher than vacancy diffusion coefficient i.e. DiC1i >> DvC1v (where Di >> Dv). Therefore interstitial loop generated and growth.

At middle stage, dislocation and dislocation loop became neutral sink because vacancy concentration is rapidly increasing. While interstitial, which have to recombination with vacancy, disappear at matrix i.e. DiC1i = DvC1v (where C1v >> C1i). Therefore growth strain will be saturation.

At last stage, <c> dislocation loop is formed on the basal plane. Therefore, dislocation loop and dislocation on prism plane once again absorb interstitial. While interstitial concentration is increasing by vacancy loop. Therefore, RIG increases proportionately with radiation dose.

3.2 Growth mechanism of recrystallization zircaloy

Annealed zircaloy have low dislocation density cause grain boundary play major role of sink. Therefore, RIG behavior is controlled by defect flux to grain boundary before dislocation loop generated.

At first stage, grain boundaries perpendicular to the basal plane are preferential sinks for SIAs. In contrast, grain boundaries parallel to the basal plane constitute preferential sinks for vacancies. Therefore, first growth occurs by grain boundary

At middle stage, dislocation loops are generated but interstitial is already absorbed by grain boundary. It will make dislocation loop become neutrally sink. Also, growth shows saturation tendency.

At last stage, same tendency of single crystal case, vacancy loops are generated and growth behavior shows linear increasing with radiation dose.

3.3 Growth mechanism of cold worked zircaloy

In case of cold worked zircaloy, dislocation density is sufficiently high. Therefore dislocation as sink is dominant effect of defect flux on matrix. Growth strain increases linearly in proportion to the radiation dose.

4. Theoretical radiation induced growth modeling of single crystal

Until the present, theoretical RIG was developed to the single crystal. Most recently two researchers (Golubov and Christien) model independently theoretical RIG with RDT and defect flux mechanism. Before, to expand the theoretical RIG model, it is needed that to understanding and analysis of the prior RIG model.

Despite of both cases have same limit framework of single crystal and commercial reactor condition, those model can give clue to fundamental understanding of RIG. And two case show big different assumption about self interstitial atom (SIA) cluster's mobility. However,

both case fit well experiment RIG data.

Both of RIG modeling has same starting point of RIG modeling process. This starting point, is concept of defect flux behavior, base on Holt's paper which is published 1979. Holt paper is also first work is about to defect flux behavior in zirconium. This defect flux could be expressed by mathematical form.

$$\varepsilon_d = (Niy - Nvy)\Omega Ady = (Niy - Nvy)\Omega \sum_{\omega} Xy\,(\omega)cos2\omega$$

Where Niy, Nvy is number of interstitials and vacancies respectively arriving at the type of sink denoted by subscript 'y', Ω is atomic volume, Ady is anisotropy factor resolving strain in the direction of interest 'd', Xy is the fraction of sinks of type 'y' oriented as defined by ω.

Golubov not only adopt this concept but also developed by using differential form to express defect flux by time. he derived RIG strain by expressing the dislocation density in the materials. The mathematical expression of <a>-axis elongation is

$$\frac{d\varepsilon_a}{dt} = \sum_m \rho_m V_m b_m cos^2\varphi_m = \sum_m [nD_{cl}C_{cl}^m k_m^2 - \rho_m(D_v C_v - D_i C_i)]cos^2\varphi_m,$$

φ_m Is the angle between a and b_m, ρ_m is the dislocation density with m (a1, a2, a3 in H.C.P structure) direction, b_m is the bugers vector with m direction.

In his work, to solving defect flux to sink (dislocation line, dislocation loop), he use simplest RDT modeling. In this RDT, the sink term is only one that dislocation density. Therefore, if it assumed that defect concentration is constant by time, defect flux term could be expressed simply by defect generation and dislocation density term.

$$\frac{dC_v}{dt} = G_{NRT}(1-\varepsilon_r) - D_v C_v \sum_j \rho_j = 0 \rightarrow D_v C_v = \frac{G_{NRT}(1-\varepsilon_r)}{\rho}$$

$$\frac{dC_i}{dt} = G_{NRT}(1-\varepsilon_r)(1-\varepsilon_i^g) - D_v C_v \sum_j \rho_j = 0 \rightarrow D_i C_i = \frac{G_{NRT}(1-\varepsilon_r)(1-\varepsilon_i^g)}{\rho}$$

$$\frac{dC_{cl}^m}{dt} = G_{NRT}(1-\varepsilon_r)\frac{\varepsilon_i^g}{3n} - D_{cl}C_{cl}^m k_m^2 = 0 \rightarrow D_{cl}C_{cl}^m = \frac{2}{3n}\frac{G_{NRT}(1-\varepsilon_r)\varepsilon_i^g}{\pi^2 r_0^2 \rho_m^2}$$

$G = G_{nrt}(1-\varepsilon_r)$ is the defect production rate, G_{nrt} being the NRT standard rate, ε_r the fraction of defects recombining in cascades, ε_i^g is the fraction of SIAs clustered in cascades; n is the mean number of SIAs in a cluster; $D_{v,i}$ are the vacancy or SIA diffusion coefficients, k_m^2 is the sink strength for SIA clusters migrating along m-th direction.

5. Advanced RIG modeling for research reactor condition

There is two big limitation of recent theoretical RIG modeling. The first limitation is

assumption that defect concentration in the materials is steady state. However, in the real matrix in the materials, defect concentration is variable with time. In the prior works, to solve defect flux term, defect concentration in the matrix is assumed steady state.

The second limitation is RIG modeling is based single crystal. Therefore, grain boundary sink effects on defect flux did not considering. To extend model from single crystal to zirconium based metal, grain boundary sink strength must be considered for RIG modeling.

The first limitation could be solved by RDT. Defect flux term could be separated by two term that diffusion coefficient and defect concentration. Diffusion coefficient term could be derived by atomic computer simulation and defect concentration also could be derived by RDT.

The second limitation is needed the qualitative analysis of growth mechanism of zirconium base metal. However, present RID mechanism is not unclear to explain grain boundary effects on defect flux. Therefore, to expand RIG model from single crystal zirconium to zirconium base metal, grain boundary sink term must be established. After that, grain boundary sink strength term will be added in RDT model.

6. Reference

[1] R. Sizmann, The effect of radiation upon diffusion in metals. Journal of Nuclear Materials, 69-70(C), pp. 386-412 (1978)

[2] L.K. Mansur, Theory and experimental background on dimensional changes in irradiated alloys. Journal of Nuclear Materials, 216(C), pp. 97-123 (1994)

[3] C.H. Woo, B.N. Singh, and H. Heinisch, A diffusion approach to modelling of irradiation-induced cascades. Journal of Nuclear Materials, 174(2-3), pp. 190-195 (1990)

[4] C.H. Woo, B.N. Singh, and H.L. Heinisch, Diffusion-based evaluation of defect processes in cascade zones. Journal of Nuclear Materials, 179-181(PART 2), pp. 951-953 (1991)

[5] R.A. Holt, and E.F. Ibrahim, Factors affecting the anisotropy of irradiation creep and growth of zirconium alloys. Acta Metallurgica, 27(8) pp. 1319-1328 (1979)

[6] C.H. Woo, Theory of irradiation deformation in nono-cubic metals: effects of anisotropy diffusion, Journal of Nuclear Materials, 159, pp. 237-256 (1988)

[7] F. Christien and A. Barbu, Effect of self-interstitial diffusion anisotropy in electron-irradiated zirconium: A cluster dynamics modeling. Journal of Nuclear Materials, 346(2-3), pp. 272-281 (2005)

[8] S.I. Golubov, A.V. Barashev, and R.E. Stoller, Radiation growth of HCP metals under cascade damage conditions, Material Research Society, 1383, pp 55-60 (2012)

[9] F. Onimus and J.L. Bechade, radiation effects in zirconium alloys, comprehensive nuclear materials, Vol 4, pp.1-31 (2012)