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Irradiation creep analysis base on rate theory in iron based cladding materials

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1. Introduction

In the nuclear history, there were several cladding candidate materials: aluminium, zirconium, and iron. Specifically, the first modern type cladding was aluminium alloy in Chicago pile-3 (CP-3), however, poor corrosion properties of aluminium cause a necessity of its replacement. Stainless steel was chosen as cladding material because chrome element could increase corrosion resistant.

From the experimental result of CPs, researchers conformed that neutron chain reaction is sustainable and controllable. And then, research focuses moved on development of power reactor, which could consistently generate its power resource i.e., fissile materials. As result concept of Fast Breed Reactor (FBR) was developed.

Since one of the most important characteristic of FBR is using the fast neutron, cladding material is exposure in the harsh environment condition. Subsequently advanced iron based alloy is developed and adopted in FBR. Cladding materials, in this condition, should maintain reasonable mechanical strength. Therefore, stainless steel was chosen for cladding material and various type of stainless steel (HT-9 and D9) were tested in Experiment Breeder Reactor-II (EBR-II) [1].

However early in 1970s navy admire Rickover design Pressurized Water Reactor (PWR) system to prevent sodium and water reaction in seawater. Simultaneously, Kroll process was developed and nuclear grade pure zirconium could be generated.

Hence zirconium based alloys were chosen for PWR, because excellent corrosion resistant property and high neutron economy. After 1970s, FBR program loosed its driving force and then PWR became most dominant nuclear reactor, hence, zirconium cladding materials was researched more than 4 decade.

Until now, irradiation degradation mechanism of zirconium are well developed including growth, hardening, and creep [2]. However, in the same times, irradiation behaviour of iron based materials was not systemically organised and mechanism is not well established when it is compared with zirconium based alloy.

Therefore in this paper, our research goal is development the prediction model of irradiation behaviour of iron based cladding materials in SFR condition.

Recently there are several research groups such as UNIST or Tera-power, try to establish the nuclear reactor of long cycle fuel design. Those reactors commonly have more extreme and harsh radiation condition, so that irradiation degradation behaviour is much severer then normal FBR. HT-9 and D-9 show great irradiation resistant properties until 200 dpa. However, there is no experiment and theoretical evidence that these materials can be sustainable over 200 dpa. The preliminary result of this study could be theoretical base for safety analysis.

2. Rate theory

Basics of rate theory, which is including specific approach and methodology, are well-described in previous works [2]. However, in case of the irradiation creep and swelling in applied stress condition, dislocation or void formation and growth affected by external stress. This stress plays a major role in irradiation degradation [3]. Brailsford already show stress effect on creep and in his model dislocation loop growth was govern by Stress Preferential Inducted Attraction (SPIA). However in his model dislocation number loop and void density were assumed constant. Therefore in this paper irradiation degradation will be more realistic described. Master equations of rate theory is given Eqs (1) ~ (4).

$$\begin{aligned}\frac{dC_i}{dt} &= K_o - K_{iv}C_iC_v - \sum_n \rho_n Z_n^i C_i D_i \\ &+\left(E_i^2+\beta_v^2\right)C_{2i} + E_i^3 C_3 + E_i^4 C_{4i}\end{aligned} \tag{1}$$

$$\begin{aligned}\frac{dC_{2i}}{dt} &= \eta G_{dpa}\frac{f_{icl}^2}{2} + \beta_i^1 C_i/2 + \left(\beta_v^3+E_i^3\right)C_{3i} \\ &\quad -\left(\beta_v^2+\beta_i^1+E_i^1\right)C_{2i}\end{aligned} \tag{2}$$

$$\begin{aligned}\frac{dC_{3i}}{dt} &= \eta G_{dpa}\frac{f_{icl}^3}{3} + \beta_i^2 C_2 + \left(\beta_v^4+E_i^4\right)C_{4i} \\ &\quad -\left(\beta_v^3+\beta_i^3+E_i^3\right)C_{3i}\end{aligned} \tag{3}$$

$$\frac{dC_{4i}}{dt} = \eta G_{dpa} \frac{f_{icl}^4}{4} + \beta_i^3 C_{3i} - \left(\beta_v^4 + \beta_i^4 + E_i^4\right) C_{4i} \quad (4)$$

Where $C_{v\ or\ i}$ is vacancy or interstitial concentration in the iron matrix (cm^{-3}), $C_{xv\ or\ \mathrm{x}i}$ is vacancy or interstitial cluster concentration in the iron matrix (cm^{-3}), K_0 is the defect generation rate ($\mathrm{cm}^{-3}s^{-1}$), which means vacancy and interstitial are combined to be a perfect lattice atom. G_{dpa} is the cluster defect generation rate ($\mathrm{cm}^{-3}s^{-1}$), f_{cl}^{x} is the fraction of cluster, η is the cascade efficiency, K_{iv} is the recombination rate (cm^3s^{-1}), ρ_n is the density of sink of n type in the iron matrix (cm^{-2}), $Z_n^{v\ or\ i}$ is the vacancy or interstitial bias factor of sink on n type in the iron matrix, which is a dimensionless number, and $D_{v\ or\ i}$ is the diffusion coefficient of vacancy or interstitial in the matrix (cm^2s^{-1}). β is the point defect absorption constant, E is the point defect emission constant, ρ_n is the density of a specific sink such as dislocation line, dislocation loop, void, and precipitation in the iron matrix (cm^{-2}). The detail constant is given in Table 1.

The physical meaning of the first term on the right hand of Eq (1) is defect generation rate; the second is recombination rate; the third is the vacancy absorption rate of any sink in the iron matrix; the rest term is about cluster growth and dissolve. In Eq (2) ~ (4), first term show cluster generation rate and the rest term represent cluster behaviour. The rate theory of vacancies follow the same method as that of interstitials in Eq (1) ~ (4). It was assumed that the density of dislocation line does not change because dislocation loops or voids absorb all defect fluxes. In Eqs, both of interstitial and vacancy defect generation rate, recombination rate, and sink absorption rate were calculated simultaneously in each time step in numerical integration. Among these three terms, sink absorption rate is the most important factor because it determines the accumulation rate of defect on the sinks. The accumulating defects make changes in the sink size and number density and then the properties of iron cladding materials are degraded by microstructural change. In case of defect generation rate and recombination radius, such are independent from dpa.

The terms of defect absorption rate at sinks in Eqs (1) are derived by a model of diffusion's limited reactions. In this model, the reaction between irradiation defect and sink is calculated by the gradient of defect concentration at a given capture volume of sink. In this study, it was assumed that iron had three major sinks in the matrix based on experiment result [3]. In case of dislocation loop, it had a two-dimensional shape such as disk while other sinks had sphere morphology such as void [4]. Therefore, defect abortion rates were expressed differently by using the density of each sink type, which was classified by disk and sphere.

$$\rho_{disk} = 2\pi r_{disk} N_{disk} \quad (4)$$

$$\rho_{void} = \rho_{ppt} = N_{sph} \quad (5)$$

Here, $r_{disk}\ and\ N_{disk}$ are the radius of sink and number density, as a sink has disk morphology. Finally, from these density equations, defect accumulation can be calculated by using defect flux and bias factor by using total number defect sink [5, 6].

$$\frac{dS_x}{dt} = \rho_x \left(Z_x^i D_i C_i - Z_x^v D_v C_v\right) \quad (6)$$

Here, S_x is the total number of defects in sink, $Z_x^{i\ or\ v}$ is the vacancy and interstitial bias factor of any given sink. Owing to interstitial and vacancy recombination at sink, net defect flux ($Z_x^i D_i C_i - Z_x^v D_v C_v$) is used for defect accumulation rate (In Equation 6, interstitial net flux is used. However, the type of net defect flux is decided by sink type; i.e., void will be expressed by vacancy net defect flux because void is composed by vacancy). Finally, sink morphology is changed as can be expressed by the radius of the sink. The total number of defects has the same physical meaning as that of volume change of sink by adopting defect volume. Therefore, the radius of sphere type of sink can be derived by dividing the volume of sphere [7]. The equation of radius is:

$$\frac{dr_{sph}}{dt} = \frac{\Omega}{r_{sph}} \left(Z_{sph}^i D_v C_v - Z_{sph}^v D_i C_i\right) \quad (7)$$

Here, r_{sph} is the radius of sink and has sphere type while Ω is the defect volume in the sink and $Z_x^{i\ or\ v}$ is vacancy and interstitial bias factor of sphere type sink. In case of disk type, however, sink volume could not be calculated because disk has zero volume. Therefore, the total defect number of disk type sinks is used and this method is well-described in a previous work [2].

$$S_{dis} = \pi r_{disk}^2 b N_{disk} \quad (8)$$

In this equation, S_{disk} is the total number of defers in disk type sink, r_{disk} is radius of disk type sink, b is Burgers vector of iron, and N_{disk} is number density of disk type sink. In Equation 8, S_{disk} can be calculated by integrating Equation 6. The radius of four major sinks is derived by diffusion limited model. In order to calculate the density of sink, the number density of sink should be obtained.

3. Irradiation creep model.

Radiation induced dimensional change in cladding materials is mainly caused by defect and sink reaction mechanism. Irradiation growth could be fully explained with defect flux behavior with dislocation loop growth and nucleation behavior. However in case of irradiation creep, stress induced dislocation loop nucleation also should be considered. Therefore irradiation creep and growth will be explain with two mechanism.

2-1 Stress-induced preferential nucleation (SIPN)

In the stress applied situation, atomic plane which is perpendicular with applied stress take tensile potential field whilst compressive potential filed is applied to atomic plane which is parallel with applied stress. Hence preferential nucleation is different portion with dislocation loop orientation. Figure.1 show the schematic of stress induced dislocation loop formation.

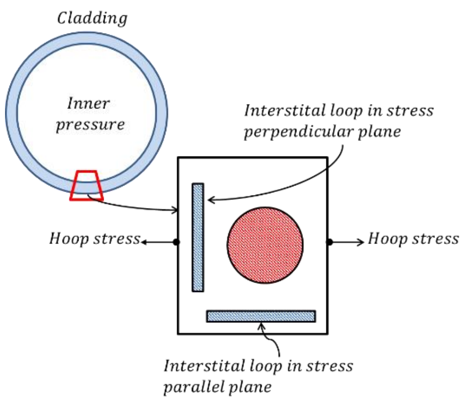


Fig. 1. Schematic of stress preferential induced nucleation.

Brailsford simply express the excess dislocation loop concentration with excess fraction of *f*, total dislocation concentration N_L and aligned dislocation loop concentration N_{AL} [3].

$$N_{AL} = 1/3(1-f)N_L + fN_L \tag{9}$$

$$N_{NL} = 2/3(1-f)N_L \tag{10}$$

Dislocation loops nucleation fraction could be expressed with arrenious function

$$f = \exp(\sigma n\Omega / kT - 1) / \exp(\sigma n\Omega / kT + 2) \tag{11}$$

where σ is applied stress (MPa), Ω is atomic volume (cm^3), k is the Boltzmann constant and n is total defect number in dislocation loop.

2-2 Stress-induced preferential absorption (SIPA)

Not only dislocation loop nucleation but also interstitial diffusion inducted loop growth is the reason of irradiation creep. Bulk defect concentration play the major role since bulk defect concentration is decreased by applied stress. Therefore, defect flux could be expressed by stress term.

$$J = \rho_j (z_i^j D_i C_i - z_v^j D_v C_v + z_v^j D_v C_v^j) \tag{12}$$

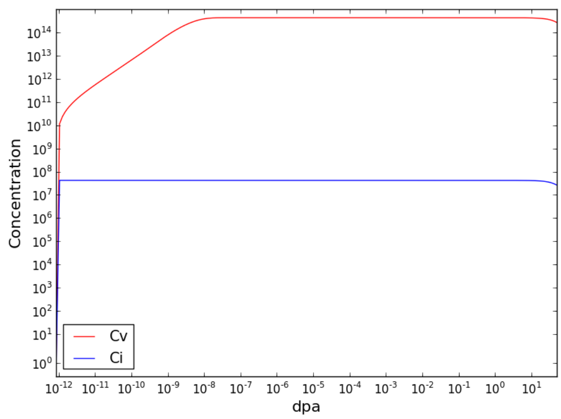
From defect flux behavior, irradiation creep could be calculated. The general rate of strain and dislocation climb velocity can be described by the simple equations

$$\frac{d\varepsilon}{dt} = \rho b V \tag{13}$$

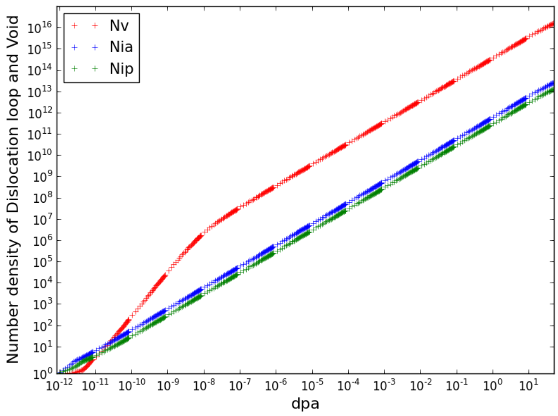
$$V = \frac{1}{b}(z_i^j D_i C_i - z_v^j D_v C_v + z_v^j D_v C_v^j) \tag{14}$$

where ϵ is the elongation (cm), ρ is the dislocation density (cm^{-2}), b is the Burgers vector (cm), v is the dislocation velocity ($cm^{-2}s^{-1}$), and z_i and zv are the interstitial and vacancy bias factors of dislocations, respectively. In the Eq (12), defect concentration and dislocation density is most important part in this modeling.

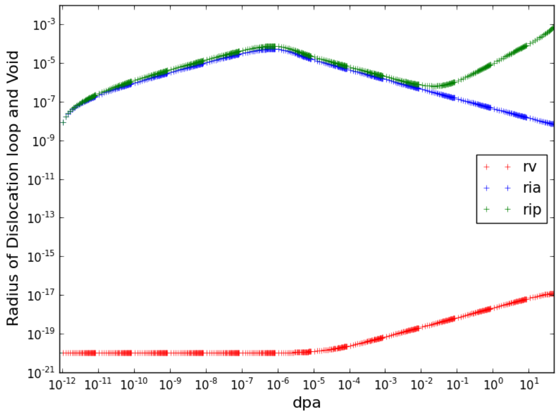
4. Result & Discussion



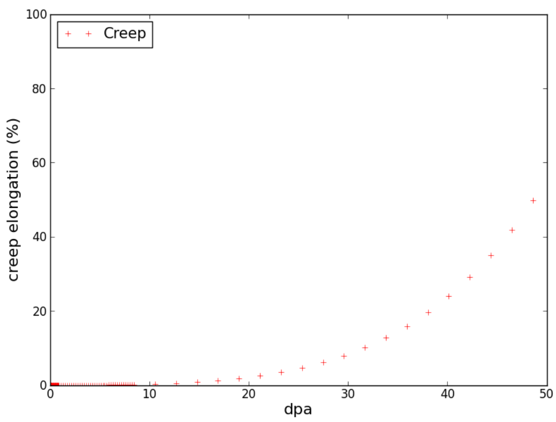
(a)



(b)



(c)



(d)

Fig.2. (a) defect concentration of point defect in iron matrix; (b) dislocation loop and void number density; (c) dislocation loop and void average radius; (d) irradiation creep elongation

In order to calculate irradiation creep, point defect, cluster number density, and radius was calculated. From point defect concentration, it could be recognized that steady state behavior of defect flux. However irradiation creep behavior show exponent tendency because cluster number density is not saturated.

The radius of each type of sink was calculated using defect concentration and cluster number density. In the engineering point of view, radius is most important parameter because it can be compared with experimental result. Dislocation loop show similar behavior of experimental result, however, void has high discrepancy with experimental result. It is too law to compare with vacancy defect concentration

Both of problems such as non-saturation of cluster number density and low void size will be solved by reconsidering of the frame of rate theory. In this model, only up to tri-cluster was considered as seed of extended defect. In the next research step, cluster effect will be considered up to 10^6 by using grouping method.

5. Summary

The aim of this study is understanding of stress and radiation effect on irradiation creep. From the simplified assumption, dislocation loops nucleation and growth mechanism, point defect concentration and irradiation creep could be analyzed. Moreover, cluster behavior, which is generated by cascade, could be confirmed. In order to confirm mobile cluster effect on dislocation loop number density, cluster behavior will be more specifically demonstrated by grouping method in next research step.

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