

Embrittlement of Nickel-based Alloys by Helium Bubbles under Neutron Irradiation

Junhyun Kwon^{a*}, Dong-Hyun Ahn^a, Gyeong-Geun Lee^a, Hyung-Ha Jin^a

^aKorea Atomic Energy Research Institute, 989-111 Daedeok-daero, Daejeon, 34057, KOREA

*Corresponding author: jhkwon@kaeri.re.kr

1. Introduction

The formation of bubbles in structural materials under neutron irradiation leads to the degradation of mechanical properties. In particular, stainless steels and nickel-based alloys lose their ductility due to the growth of helium bubbles on the grain boundaries [1]. Helium is likely to cause the embrittlement because of its insolubility and tendency to form internal gas bubbles. Also, irradiation creep of steels can arise as a result of growth of gas bubbles on the boundaries orthogonal to the applied tensile load [2]. The transmutation gas and point defects, which are created by neutron irradiation, are the source of materials degradation.

Ni-based alloys are used as spacers in the CANDU fuel channel assemblies. Spacers are tight fitting springs which provide support to the pressure tube, separating it from the calandria tube. This spacer is made of Inconel X-750, one of Ni-based alloys. Post-irradiation and testing of the removed X-750 spacer show that the ductility is reduced compared to the as-installed condition. Also the fracture behavior was found to be totally intergranular after about ten years of operation [3]. The material was embrittled following neutron irradiation. In this paper, we present the helium embrittlement model based on the bubble growth mechanism. Further, mechanical parameters will be derived along with the measurement of bubble distribution.

2. Theory

In this section, we first consider the stability criterion for gas-filled bubbles in mechanical equilibrium followed by the development of bubble growth models.

2.1 Mechanical Equilibrium of Gas Bubbles

For a gas-filled bubble of radius R embedded in a solid medium, the equilibrium condition for a bubble is expressed by the force balance, in which the force due to the outward pressure of the gas is balanced by the inward force due to surface tension. When a tensile stress σ is applied and the van der Waals equation of state is used to describe the thermodynamic state of gas in a bubble, the new equilibrium radius of the bubble is given by:

$$\sigma = \frac{2\gamma}{R} - \frac{mkT}{V - mB} \quad (1)$$

where γ = surface tension, k = Boltzmann constant, B = van der Waals constant, T = temperature, and V = bubble volume ($4\pi R^3/3$). Assuming an initial bubble of radius R_0 contains m helium atoms, the van der Waals equation of state gives the following relation.

$$m = \frac{(4\pi R_0^3/3)}{B + (kT/2\gamma)R_0} \quad (2)$$

By plotting Eq. (1) for given R_0 , the bubble stability can be described in terms of the applied stress and the bubble size. For bubbles of size R_0 , Eq. (1) gives the critical stress σ_c and the critical bubble radius R_c for stability. For a solid with bubbles of size $R_0 < R_c$, the application of tensile stress σ_c will cause the bubble to grow to size R_c . If $R_0 > R_c$ or if the applied stress is greater than σ_c , then the bubble will grow without upper bound.

2.2 Gas Bubble Growth Model

Bubbles are readily nucleated on grain boundaries where stress concentrations take place. Once nucleated, bubbles tend to grow by absorbing vacancies and gas atoms. Vacancies probably flow to the bubbles via grain boundary because grain boundary diffusion is more rapid than the lattice diffusion at the modest temperature. Since transmutation gas atoms, such as helium, are created with a certain amount of kinetic energy from the neutron absorption reactions, they can migrate up to the range on average.

The growth law for the helium-filled bubble is formulated in a manner similar to that applied by Hull and Rimmer to grain boundary voids [4]. It is assumed that initially N_{gb} bubbles of radius R_0 have been nucleated per unit area of grain boundary perpendicular to the tensile stress. The bubble population is divided into a series of unit cells, which are surrounded by grain boundary area with a central bubble. The extent of grain boundary Q from which the bubble draws its vacancies is determined by:

$$(\pi Q^2) N_{gb} = 1 \quad (3)$$

The helium atoms created by Ni two-step reactions come to rest in a material as they lose energy by means of interactions with the lattice atoms. The distance traveled by the atom is called the range μ . Since this is a random process, the range has a normal distribution

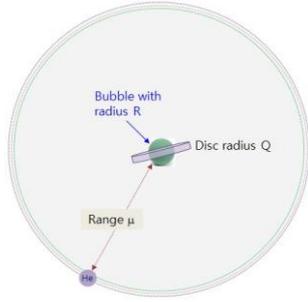


Fig. 1. A model for the growth of helium bubbles on grain boundaries by means of vacancy diffusion in the grain boundary and helium migration in the matrix

with a standard deviation $\Delta\mu$. It is likely that helium atoms newly created by transmutation reactions are involved in the growth of the initial bubbles after traveling the range. In this framework shown in Fig.1, the growth rate of a bubble is given by:

$$\frac{dR}{dt} = \frac{w\Omega P_v}{4} \left(\frac{Q^2}{R^2} - 1 \right) + 2\pi\Delta\mu G_{He} B \quad (4)$$

where w = grain boundary width, Ω = atomic volume of alloys, P_v = vacancy production rate, $\Delta\mu$ = range straggling, and G_{He} = helium generation rate.

2.3 Estimation of Mechanical Properties

Creep rupture may occur when two adjacent bubbles touch. If the spherical bubbles are disposed on a regular square array, linkage occurs when the bubble radius reaches $R_F = (\pi/4)^{1/2}Q$ which corresponds to the radius at fracture. Accordingly, the time to rupture t_R is obtained by integrating Eq. (4) from R_o to R_F such as:

$$t_R = \int_{R_o}^{R_F} \frac{dR}{(dR/dt)} \quad (5)$$

3. Sample Calculations

In order to examine the effects of the applied load to the behavior of a bubble, we investigated the condition of mechanical equilibrium of a helium bubble by plotting the Eq. (1) above. Then, the stress distribution of the CANDU spacers was evaluated using the finite element method (FEM) code, which is important to determine the critical bubble size for the growth.

3.1 Stability of Gas Bubbles

The condition of mechanical equilibrium of a helium bubble in a solid subject to tensile stress is given by Eq. (1). A bubble will tend to grow if its initial radius is

greater than the critical radius or if the applied tensile stress is greater than the critical stress. The bubble stability is accounted for in terms of the applied stress and the bubble size by plotting Eq. (1) for given values of various R_o . Eq. (1) is displayed in Fig. 2 for three values of R_o . For a given initial bubble with $R_o = 2.5$ nm, if the applied stress is less than the critical one of 1 GPa, the bubble grows from R_o to a final value that satisfies Eq. (1) at a rate given by Eq. (4).

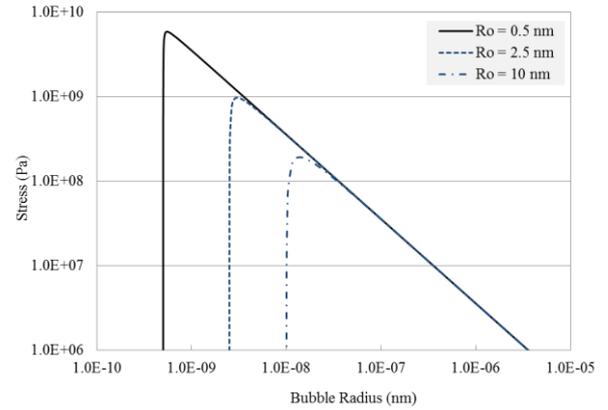


Fig. 2. Critical stress for stress-induced growth of equilibrium bubbles for given initial bubble size R_o

3.2 Stress Analysis of Annulus Spacers

There are four spacers over the 6 meter span of the fuel channel, which are required to remain tight on the pressure tubes until the tubes have sagged. The spacers are made using X-750 wire, which has a square cross-section and is to form a coiled spring. Each spacer coil has an outside diameter of 4.8 mm and there are 15 coils per 25 mm of spacer length. The springs become pinched between the hot pressure tubes and relatively cold calandria tube at the bottom position. Accordingly, an applied load causes bending stress on some parts of the spacers during the reactor operation. We performed the FEM calculation to estimate the stress distribution on the single coil. The basic assumption is that one segment of spacers is suppressed slowly between two flat plates with a certain load.

Fig. 3(a) presents the local contour plot of the simulated stress for a single spacer coil. It is seen that the maximum tensile stress occurs at the 12 and 6 o'clock position of the inner coil, while the maximum compressive stress arises in the outward radial directions. For the applied load of 35 N, designated by black arrows in Fig. 3(a), the maximum tensile stress can reach up to 220 MPa. The stress profile obtained from the inner coil ring is shown in Fig. 3(b). The stress was taken from the value of the nodal point in the FEM solution. We can anticipate the potential location of spacers, which is prone to the failure.

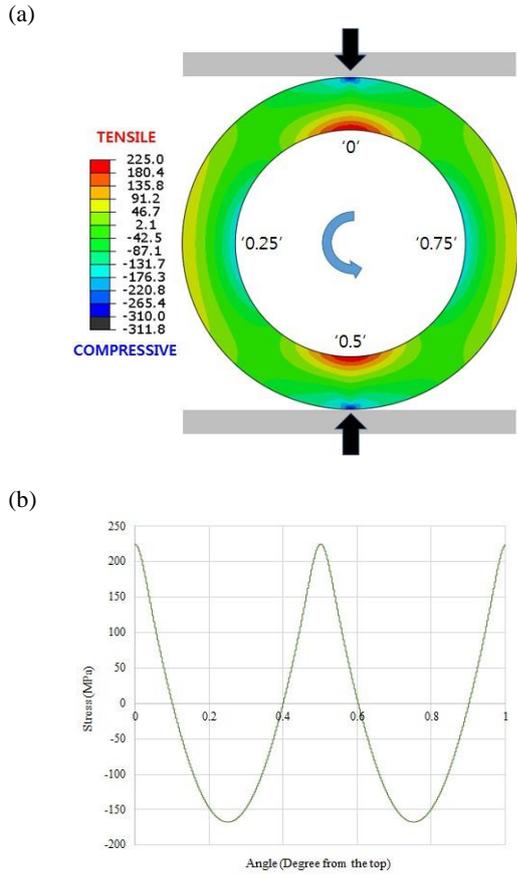


Fig. 3. (a) Contour plot of the simulated stress at the spacer coil for the applied load of 35 N. The inner coil diameter is 3.28 mm and its thickness is 0.76 mm. (b) Stress profile for a coil depending on the angle – 0.25 corresponds to 90° from the top in the counterclockwise direction.

4. Conclusions

The embrittlement model of neutron-irradiated Ni alloys was suggested based on the growth mechanism of helium bubbles. This model is being developed in order to apply to predicting the behavior of annulus spacers in a CANDU reactor. Since the spacers, made of Inconel X-750, are located in the reactor core, they are exposed to neutron irradiation. In addition to the generation of point defects, the helium gas production by means of Ni two-step reactions is critical to the degradation of materials. Considering these effects, we develop the helium bubble growth model which is used to predict the material property changes.

As a preliminary step in this work, we estimated the initial bubble stability which provides the information on the possibility of bubble growth depending on the environmental conditions. Also, the stress analysis was performed on the spacers using the FEM codes. It is found that the maximum tensile stress occurs at the top and bottom positions of the inner coil, which are believed to be prone to failure. Within this framework,

we will predict the mechanical behavior of the spacers by developing the current model.

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