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EARLY SWELLING BEHAVIOR OF NEUTRON-IRRADIATED STAINLESS STEEL

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Abstract

Stainless steel in fusion and fast breeder reactors is exposed to high neutron flux, with subsequent dimensional changes. This work is a theoretical investigation of the behavior of type-316 stainless steel during the first 100 sec. of neutron irradiation. Unlike most previous work, vacancy loops were considered. They were found to cause an initial rapid decrease in void radius and percentage swelling. This is followed by a slow increase in both quantities. Such transient behavior can be interpreted in terms of physical parameters as the gas pressure and surface tension of the voids. A mathematical treatment is given which supports the physical interpretation.

Introduction

One of the most serious problems that faces the development of fusion reactors is the swelling of the first wall and blanket materials due to neutron irradiation. Similar phenomenon has been known for quite sometime in the fuel cladding of fast breeder reactors. In principle, all metals swell when exposed to a high flux of energetic particles. Swelling is caused by the formation and growth of voids over a temperature range from 0.3 to 0.5 of the melting temperature [1]. Swelling of type-316 stainless steel was considered in this work since it is widely used as structure and cladding in fast breeder reactors. It is also the prime candidate for the first wall in fusion reactors [2].

It is very difficult to study actual neutron-induced swelling experimentally. A damage state in stainless steel which normally requires more than a year of fast nuclear reactor irradiation, can be produced in a matter of hours using electron or particle accelerators. Therefore, most studies of metal swelling are simulations using electrons or charged particles. However, microstructure and swelling behaviour was found to be different at identical operating temperature, and simulation was not a perfect tool. The discrepancy in behaviour has been attributed to the difference in damage rate (dpa/sec) [3]. Theoretical treatment of neutron-induced swelling, based on realistic materials and irradiation parameters, is an alternative method.

Investigation of early stage swelling of type-316 stainless steel in this work is based on the steady state rate theory [4-6]. The first 100 sec. of void growth are particularly important since they contain rapid dimensional changes. A detaild physical account of this period is given in this paper. References [7] through [9] give a detailed description of the theory as well as the material and irradiation parameters used in the calculations.

Theory

According to the rate theory, the study of void growth requires simultanious solution of the equations,

$$\frac{dq_v}{d\Delta} = \varepsilon - \frac{1}{K}\left(\frac{4q_v N_{v1}}{b}\right)^{\frac{1}{2}} \left[Z_i D_i C_i - Z_v D_v C_v + Z_v D_v C_v^e \exp\left(\frac{[\gamma_{SF} + F_{e1}(\bar{r}_{v1})]\, b^2}{KT}\right)\right], \qquad (1)$$

$$\frac{dN_{v1}}{d\Delta} = \frac{n_{v1}}{b^3 K} - \frac{N_{v1}}{r_{v1}(0)bk} \; Z_i D_i C_i - Z_v D_v C_v + Z_v D_v C_v^e \exp\left(\frac{[\gamma_{SF}\, F_{el}\, r_{vl}(0)]\, b^2}{KT}\right)], \qquad (2)$$

$$\frac{dr_c}{d\Delta} = \frac{1}{Kr_c} \left[D_v C_v - D_i C_i - D_c C'_v(r_c)\right], \qquad (3)$$

and

$$\frac{dr_{il}}{d\Delta} = \frac{1}{Kb} \left[Z_i D_i C_i - Z_v D_v C_v - Z_v D_v C'_v(r_{il})\right],$$

together, with the coupled algabraic equations,

$$C_i = (D_v k_v^2/2\alpha) \left[- (1 + \mu) + \left[(1 + \mu)^2 + \eta\right]^{\frac{1}{2}}\right], \qquad (5)$$

$$C_v = (D_i k_i^2 / 2\alpha) \, [-(1-\mu) + [(1+\mu)^2 + \eta]^{\frac{1}{2}}] \; . \quad (6)$$

Where,

Δ = Kt is the irradiation dose (dpa) at time t,

dq_v/d is the rate of change in the total number of vacancies present in vacancy loops,

dN_{v1}/d is the rate of change of the number of vacancy loops per unit volume,

dr_c/d is the rate of change of the average void radius,

dr_{il}/d is the rate of change of the average interstitial loop radius.

All other terms have their usual meaning [7].

Because of the vacancy loop rate equation, the system of differential equations becomes very sensitive to time variations (dose variations) in the early stage of growth. This is mainly because of the rapid change in swelling rate. Consequently, the integration step size, h, should be reduced to assure the stability of the solution and to minimize the per-step error. For neutron irradiation at a temperature of 600 C, a value of 10^{-8} was found to be suitable for h. Therefore, 10,000 steps of calculations were executed to cover the 100 seconds irradiation time which corresponds to a total dose of 10^{-4} dpa.

Swelling occurs due to void formation and growth. Macroscopic swelling can be determined by just multiplying the number of voids per cm^3 by the average void volume. Therefore,

$$SW\ \% = \frac{\Delta V}{V} = \frac{4\pi}{3} r_c^3(t) N_c \qquad (7)$$

Hence, the problem of determining the swelling rate becomes the determination of $r_c(t)$. The rate of change in $r_c(t)$ is determined by calculating the total flux of point defects to the voids. This requires the calculation of the point defect concentrations (C_i and C_v) by solving equations (5) and (6), after determining the rates of change of q_v, N_{v1}, r_c, and r_{i1} from equations (1) through (4).

Results

The swelling versus time variation of irradiated type-316 stainless steel, at a dose rate of 10^{-6} pda/sec and temperature of 600 C, is shown in Fig. 1. The average void radius $r_c(t)$ is also plotted. It is clearly shown that the swelling decreases rapidly during the first 20 seconds. Thereafter, it starts to increase slowly with time. Similar behavior is shown for r_c. It is expected that both quantities will continue to increase up to high doses.

The net flux of point defects to the voids is partially responsible for the decrease in radius. In other words, the void absorb more interstitials than the net vacancies (absorbed - emitted).

Considering only the diffusion of point defects to the voids, we can define the interstitial and void fluxes as,

$$\varphi_v = D_v C_v \qquad (8)$$

and

$$\varphi_i = D_i C_i \tag{9}$$

When φ_i is greater than φ_v, voids apparently shrink. Fig. 2 shows the plot of φ_v and φ_i versus time. The interstitial flux, φ_i, is greater than the vacancy flux, φ_v, during the first 8.5 sec. The vacancy flux becomes greater at larger times. Therefore, the difference between interstitial and vacancy fluxes shares in decreasing the average void radius only during the first 8.5 seconds.

In addition to φ_i and φ_v, the emission flux plays an effective role in the change in void radius. The rate of change in the average void radius can be expressed as,

$$\frac{dr_c}{dt} = \frac{1}{r_c}(\varphi_b - \varphi_e) , \tag{10}$$

where φ_b is the bias flux and is given by,

$$\varphi_b = \varphi_v - \varphi_i \tag{11}$$

and φ_e is the emission flux. Clearly, the difference between φ_b and φ_e determines whether the void radius increases or decreases.

The effects of bias flux and emission flux can better be shown by plotting the ratios φ_b/r_c and φ_e/r_c. This is shown in Fig. 3. The emission flux ratio has a sharp peak which is completely recovered at about 20 seconds. Then, it starts to increase slowly. The bias flux ratio starts with negative value which means greater insterstitial flux than vacancy flux. The bias flux ratio has a small peak coresponding to the peak of the emission flux. This is due to the very

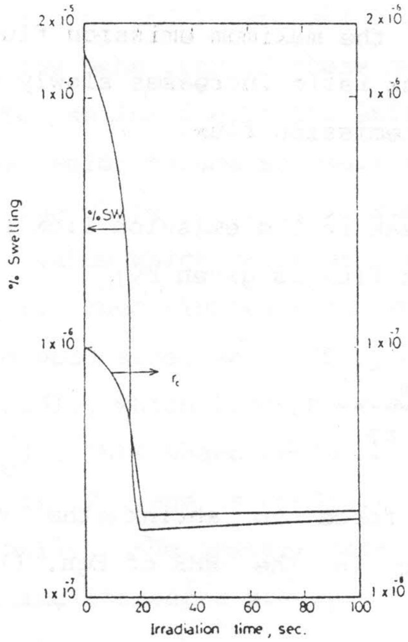

Fig. 1. Swelling and average void radius variations versus time for neutron irradiated type-316 stainless steel during the first 100 sec. at 600°C and 1×10^{-6} dpa/sec. Vacancy locps are considered.

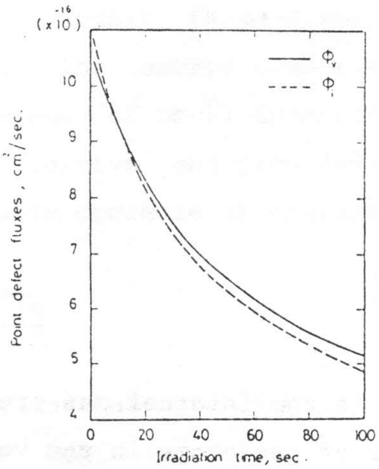

Fig. 2. Interstitial and Vacancy flux variations versus time for neutron irradiated type-316 stainless steel at 600°C and 1×10^{-6} dpa/sec.Vicancy loops are considered.

small void radius at the maximum emission flux. After the peack is reccvered, the bias flux ratio increases slowly with a value slightly higher than that of the emission flux.

The existance of the peak in the emission flux ratio can be explained as follows. The emission flux is given by;

$$\varphi_e(t) = D_v C_v^e \exp\left(\frac{F_m(t) b^3}{KT} \right), \qquad (12)$$

where the mechanical force to shrink the void F_m is the only time-dependent variable in the RHS of Eqn. (12), and is given by,

$$F_m(t) = \frac{2\gamma}{r_c(t)} - p_g(t) \qquad (13)$$

where,

$$p_g(t) = \frac{3 n_g(t)\, KT}{4\pi r_c^3(t)}, \qquad (14)$$

and

$$n_g(t) = \frac{k_g t}{N_c b^3} + n_g^o \qquad (15)$$

In these equations, p_g is the internal gas pressure of the voids, n_g and n_g^o are the number of gas atoms in the voids at time t and in the void nucleus, respectively. k_g is the transmutation gas generation rate in at/at. sec. So that, the time dependance of $\varphi_e(t)$ comes from $F_m(t)$ through $r_c(t)$, $P_g(t)$ and $n_g(t)$.

The values of $2\gamma/r_c$, p_g and F_m are all plotted versus time in Fig. 4. Based on the behaviour of these quantities one can see that the voids start to shrink due to the emission of vacancies to form vacancy loops. The voids become so small that the value of $2\gamma/r_c$ reaches a high value while p_g is still small. As a result $F_m(t)$ reaches a maximum value which produces a peak in $\varphi_e(t)$ due to the exponential function. Such increase in the emission term leads to a more decrease in the void size. As $\varphi_e(t)$ goes to its peak, the void radius decreases rapidly, which leads to a sharp increase in the value of p_g ($p_g = fn\ (1/r_c^3)$). This sharp increase in p_g leads to a rapid decrease in F_m and accordingly a rapid decrease in the emission flux. Finally, the average void radius starts to increase, and percentage swelling increases slowly.

The preceeding paragraph clarifies, also, the observation by many workers in the field that the presence of gas in the void nuclei is essential to ensure their three-dimensional morphology. In other words, collapse of the embryo void into a vacancy loop is probably impeded by the presence of small quantities of helium gas in the void and thus it may survive and grow [10]. The rapid increases in gas pressure prevents more decrease or collapse of the void.

Mathematical Analysis

An important question that comes to mind is; why does the void radius decreases at the beginning when vacancy loops are taken into consideration? such question can be explained by examining the flux equations for both vacancies and interstitials which are;

$$\varphi_v = D_v C_v = (D_i D_v k_i^2/2\alpha)[-(1-\mu) + (1+\mu)^2 + \eta]^{\frac{1}{2}}]$$

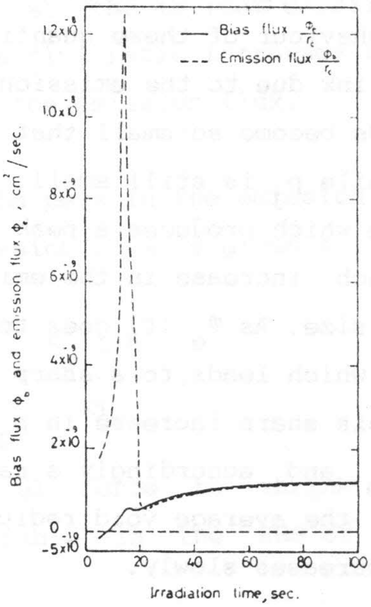

Fig. 3. Bias flux/ r_c and Emission flux/ r_c variation versus-time for neutron irradiated type-316 stainless steel at 600°C and 1×10^{-6} dpa/sec. Vacancy loops are considered.

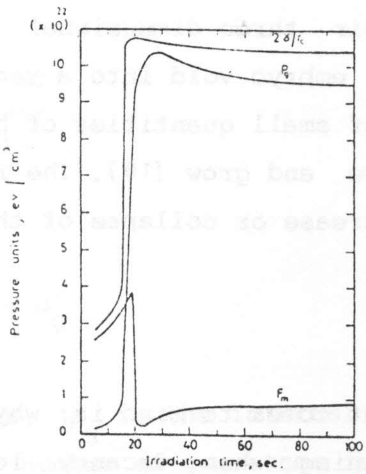

Fig. 4. Mechanical force Fm, surface tension $2\gamma/r_c$ and internal gas pressure Pg, variations with time for neutron irradiated type-316 SS.

$$\varphi_i = D_i C_i = (D_i D_v k_i^2 / 2\alpha)[-(1-\mu) + (1+\mu)^2 + \eta]^{\frac{1}{2}}]$$

where,

$$\eta = 4\alpha K / D_i D_v k_i^2 k_v^2 , \qquad (18)$$

$$\mu = (K' - K)\eta / 4K, \qquad (19)$$

All quantities have their usual meaning as in reference [7].

Equations (16) and (17) are similar except for k_i^2 and $[-(1-\mu)]$ in the first, which are replaced with K_v^2 and $[-(1+\mu)]$ in the second equation. Let us consider, first, the values of the overall vacancy and interstitial sink strengths k_v^2 and k_i^2. For simplicity, they are given, without considering the grain boundries, by,

$$k_v^2 = Z_v \rho_d + 4\pi r_c N_c + 4\pi r_p N_p \qquad (20)$$

and

$$k_i^2 = Z_i \rho_d + 4\pi r_c N_c + 4\pi r_p N_p [1 + \frac{(Z_i - Z_v)\rho_d}{Z_v \rho_d + \pi 4 r_c N_c}]$$

The interstitial bias Z_i is known to be greater than the vacancy bias Z_v. Accordingly, the interstitial sink strength k_i^2 is greater than the vacancy sink strength k_v^2. This results in a higher vacancy flux than interstitial flux as they appear in equations (16) and (17), respectively. In other words, the difference between k_v^2 and k_i^2 does not explain the initial decrease in void radius.

The other difference between the vacancy flux and interstitial flux is $-(1-\mu)$ versus $-(1+\mu)$. The value of μ as given by equation (19), is much greater than unity. Consequently, $-(1-\mu)$ and $-(1+\mu)$ can be approximated as μ and $-\mu$, respectively. In addition, μ itself can be either positive or negative. The effective part of μ which determines its sign is the value of $(K' - K)$.

In the absence of vacancy loops, $(K' - K)$ becomes,

$$K' - K = K^{e} \tag{22}$$

where, K^{e} is the rate of thermal emission of vacancies from all sinks that emit vacancies. K^{e} is always positive and so is μ in this case. Accordingly, the vacancy flux φ_v is always greater than the interstitial flux φ_i. Therefore, in absence of vacancy loops, void radius is increasing from the start.

Taking vacancy loops into consideration, the value of $(K' - K)$ is given by,

$$K' - K = K^{e} - \varepsilon K$$

where, K is the fractional rate at which vacancies are removed from the matrix to form vacancy loops. K was found to be greater than K^{e} at early stage of swelling. Hence, μ is negative, and its value is added to φ_i and subtracted from φ_v. This means a higher interstitial flux than vacancy flux to voids. Accordingly, an initial reduction of void radius.

The vacancy flux begins to increase rapidly; mainly because the dislocation density ρ_d increases, which gives higher value for k_i^2. Also, the vacancy emission rate K^e increases and reverses the sign of μ. Now μ is added to the vacancy flux and subtracted from the interstitial flux. The final result is that φ_v becomes greater than φ_i and voids begin to grow.

Conclusion

Vacancy loops cause the initial rapid decrease in void radius and percentage swelling during neutron irradiation of type-316 stainless steel. After about $2x10^{-5}$ dpa, shrinkage stops and voids start to grow. This phenomenon can be explained in terms internal gas pressure and surface tension of voids.

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