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HELIUM BUBBLE FORMATION AND SWELLING IN METALS

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The effect of a continuous production of helium on swelling is analyzed using an improved equation of state for gaseous helium that incorporates both a gas-kinetic as well as a static pressure for densely packed helium. In the absence of a bias, bubble swelling is characterized by two temperature regimes. At temperatures below about 550°C, bubble swelling is due to thermally assisted interstitial emission and is of a magnitude determined by the volume occupied by densely packed helium. At higher temperatures, bubbles grow by thermal vacancy emission at dislocations and their absorption at cavities. For high He/dpa ratios, bubbles remain overpressurized, and swelling is less than obtained from the equilibrium assumption. When a modest bias exists, bias-driven swelling or void growth in the low temperature regime is always larger than bubble swelling, and helium has no effect on the swelling rate.

1. INTRODUCTION

The production of helium in structural materials by (n,α) reactions is considered to be one of the major factors which influences void formation. Many experimental and theoretical studies, reviewed recently by Farrell [1] and by Hayns [2], have clearly shown that helium, either pre-injected or continuously implanted or produced, does increase the void nucleation rate. However, this does not always imply that the terminal void number density at a given irradiation temperature is also increased with helium. In fact, the review in section 2 of experimental data on void number densities indicates that for He/dpa ratios of interest to fusion reactors, the terminal void number density in neutron irradiated austenitic stainless steels is independent of the helium concentration, but the time to reach it is dependent on the He production rate. As a result, swelling in excess of a few percent may not be affected by increased helium production at irradiation temperatures below about 0.5 T_m (T_m is the melting temperature). At higher temperatures, however, helium bubbles are expected to form and thereby extend the temperature range of swelling.

This high-temperature swelling by helium bubbles can be easily computed provided the bubble density, the gas law, and the surface energy γ are known, and provided all the helium can be assumed to reside in the bubbles. Furthermore, the most important assumption is, of course, that the bubbles are in thermodynamic equilibrium, i.e. $2\gamma/r = p$, where r is the bubble radius and p the gas pressure. Strictly speaking, the equilibrium assumption is never completely satisfied, particularly when the He/dpa ratio is large. The reason is that gas-driven swelling requires an overpressure, i.e. $p > 2\gamma/r$, for a finite bubble growth rate. As a result, the equilibrium assumption leads to the maximum possible bubble swelling when bias-driven growth is negligible.

The purpose of the present paper is to evaluate the effect of helium on swelling without adopting the equilibrium assumption and to compare it with bias-driven swelling and swelling by equilibrium bubbles. Important features of the present swelling model are an improved equation of state for helium and several new contributions to the driving force for gas-driven swelling. The latter consist of the static pressure of densely packed He atoms, and the possibility for interstitial generation by overpressurized bubbles. The model is applied to type 316 stainless steel irradiated in either HFIR or EBR-II after the terminal void number density has been reached.

2. TERMINAL VOID NUMBER DENSITIES

The evolution of the void number density in neutron-irradiated AISI 316 has been found to depend on a number of variables [3]. In general, however, it follows for isothermal irradiation two major regimes, transient and saturation, followed by a minor stage of void coalescence at high values of swelling. Most variables exert their strongest influence on the transient regime where the void nucleation takes place. These variables are temperature, displacement rate, cold-work level, prior thermal history, He/dpa ratio, and applied stress. The cold-work level is perhaps the dominant determinant for the duration of the transient regime.

The saturation regime, defined by a dose-independent void number density, has been found to

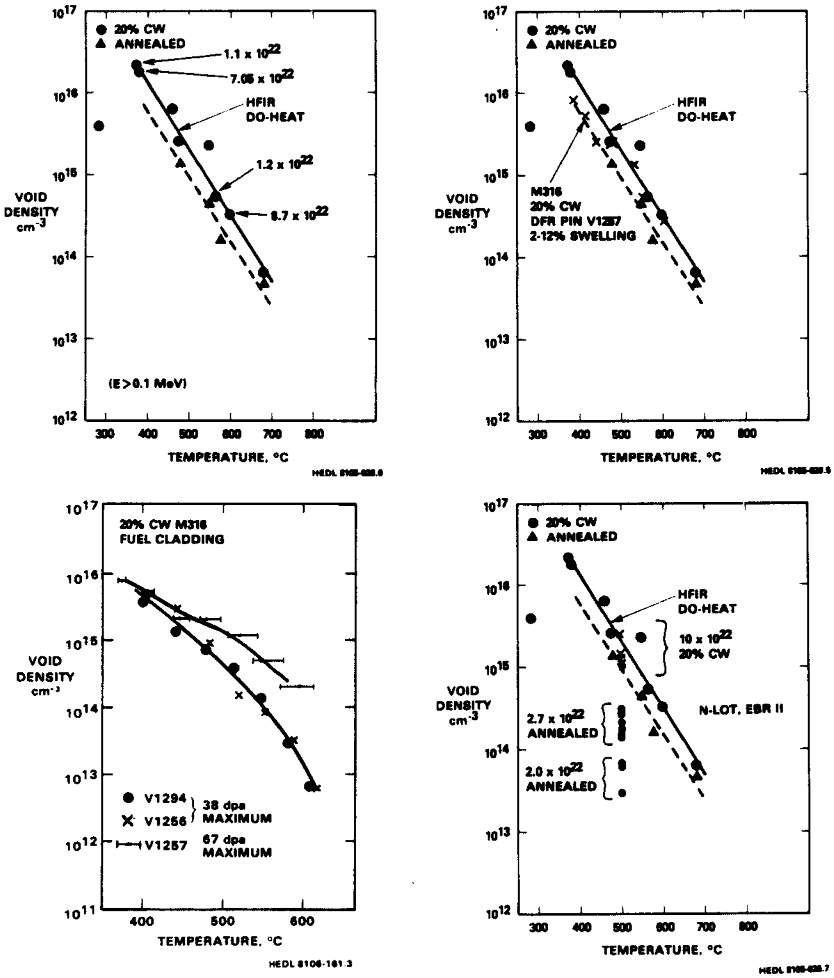

Figure 1. Void number densities in stainless steels.

be much less sensitive to these variables than originally anticipated with the exception of temperature and displacement rate. It is important to note that the action of these two variables on void number densities is very often coupled in fission reactor irradiations. For instance, the very high γ-heating rates in HFIR required to obtain the highest irradiation temperatures are obtained at the core positions of peak displacement rate. In EBR-II, on the other hand, the highest and lowest irradiation temperatures are usually obtained at the top and bottom of the core where the displacement rates are the lowest. Only in special test assemblies can other flux-temperature combinations be achieved. As a result, great care is required when comparisons are made between data from various fission reactors. When this is done, the following findings emerge.

Figure 1(a) shows the void number densities developed in a particular heat of AISI 316 [4] during HFIR irradiation. Note that the displacement rate at 380°C is roughly one-third of that at 680°C. There are two temperatures, 380°C and around 575°C, where void number densities for different fluences can be compared. It is seen that the void number densities have not increased beyond their values observed at the low fluence of 1×10^{26} n/m^2 ($E > 0.1$ MeV). This means that void nucleation is over before about 300 appm of helium has accumulated. Since the He/dpa ratio increases in HFIR substantially after a fluence of 1×10^{26} n/m^2, the large He/dpa ratios quoted for HFIR at higher fluence are of little consequence to void nucleation in this material.

In Figure 1(b), the void number densities in a DFR [6] breeder fuel pin cladding are added to the HFIR data. It is seen that the DFR data are comparable to the HFIR data, in spite of the lower helium concentration produced in the DFR. Figure 1(c) shows additional DFR data from two fuel pins with lower dose. It is seen that the void number densities from different fuel pins are reproducible, and that the approach to the terminal void number densities at low He/dpa ratios is slowest at higher temperatures.

This build-up of the void number density towards saturation is further illustrated in Figure 1(d). The two data sets added are for the N-lot heat irradiated at 500°C in EBR-II. Due to the lower He/dpa ratio in EBR-II the void number densities have not yet reached the saturation levels already attained in comparable HFIR irradiations. The intermediate void number densities in each fluence subset of data also increase with increasing stress [6,7].

3. EQUATION OF STATE FOR HELIUM GAS

The helium density in small gas bubbles is sufficiently high so that neither the ideal nor the van der Waals gas law is an appropriate equation of state. Therefore, Wolfer [8] has recently developed a gas law which can be written in the form

$$z(y_0,T) = p_k/(N_{He}kT) \qquad (1)$$

where p_k is the kinetic gas pressure, N_{He} the number of helium atoms per cavity volume, k is the Boltzmann constant, and T the absolute temperature. Furthermore

$$y_0 = \frac{\pi}{6} N_{He}\, d_0^3 \qquad (2)$$

is the "hard-sphere" packing fraction with $d_0 = 0.2637$ nm as the "diameter" of the hard-core potential of helium. Equation (1) as obtained earlier [8] has been compared more recently with the empirical gas law of Mills, Liebenberg, and Bronson [9] developed from their data in the temperature range from 75 to 300 K. This comparison makes it necessary to slightly modify the earlier theoretical gas law in addition to correcting an error. These changes mainly affect the compressibility factor z for temperatures below 400 K and packing fractions $y_0 \geq 0.7$. The newly revised compressibility factor z is shown in Figure 2 together with the empirical results of Mills et al. [9] at 200 K; the agreement between the empirical and theoretical correlations is excellent. When the hard-sphere packing fraction becomes of the order of one or larger, the densely packed helium acts more like a compressible medium than a gas. As a result, a static pressure p_s exists even at 0 K. Baskes and Holbrook [10] have obtained this static pressure from computer simulations of helium-vacancy clusters in copper. From their work it is found that

$$p_s \cong 30400(y_0 - 1) \text{ MPa}$$

for $y_0 > 1$ and $p_s = 0$ for $y_0 < 1$.

The static pressure builds up rapidly as y_0 exceeds one, whereas the kinetic gas pressure p_k rises much less. Accordingly, we have made the assumption that the total helium pressure for $y_0 > 1$ is equal to the sum of p_s and p_k, where p_k is obtained from Eq. (1) for $y_0 = 1$. For packing fractions $y_0 < 1$, only the kinetic pressure p_k contributes to the total pressure. This particular matching avoids a discontinuity in pressure at $y_0 = 1$.

4. BUBBLE AND VOID GROWTH

After the terminal void number density N_v has been reached, the swelling rate $\dot{S}$ can be evaluated with the equation

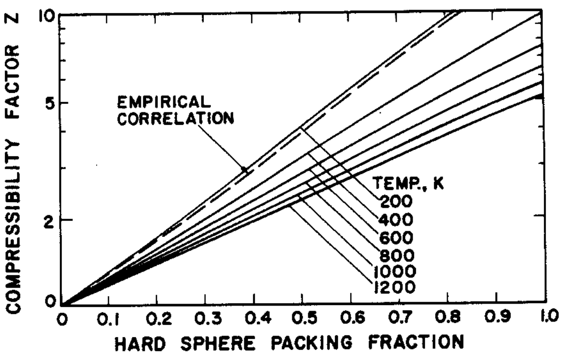

Figure 2. Compressibility factor for gaseous helium.

$$\dot{S} = \Omega N_v 4\pi r\{D_v(\Delta C_v + \bar{C}_v - C_v^o) - D_i(\Delta C_i + \bar{C}_i - C_i^o)\} \quad . \tag{3}$$

Here, Ω is the atomic volume, D_v and D_i are the diffusion coefficients for vacancy and interstitial migration, and ΔC_v and ΔC_i are the corresponding concentrations as produced by displacement damage. The concentrations of vacancies and interstitials in thermodynamic equilibrium with voids of radius r are given by

$$C_v^o = C_v^{eq}\exp(-p_o\Omega/kT), \; C_i^o = C_i^{eq}\exp(p_o\Omega/kT), \tag{4}$$

where:

$$p_o = p - 2\gamma/r \tag{5}$$

is the net pressure in the void and C_v^{eq} and C_i^{eq} are the equilibrium concentrations in an ideal and stress-free crystal. Although C_i^{eq} is very small, C_i^o may be substantial for very large overpressures p_o in the voids. The equilibrium concentrations in the real crystal are given by

$$\bar{C}_v = C_v^{eq}\exp(\sigma_H\Omega/kT), \; \bar{C}_i = C_i^{eq}(-\sigma_H\Omega/kT). \tag{6}$$

Since the hydrostatic stress σ_H is no larger than the yield stress, the exponential in Eqs. (6) is never large enough to give $\bar{C}_i$ a substantial value in contrast to C_i^o; hence, we may assume that $\bar{C}_i \simeq 0$.

5. RESULTS

Swelling was computed by numerical integration of Eq. (3), assuming a dose-independent void density given by

$$\log N_v = 26.94 - 7.54 \times 10^{-3} * T(K) \tag{7}$$

which represents the terminal values given in Figure 1.

The dislocation density at saturation is assumed to be equal to 6×10^{14} m^{-2} according to experimental observations [7], and the displacement rate is 10^{-6} dpa/sec. Furthermore, it is assumed that the helium concentration increases linearly with dose at a specified rate. Other materials parameters are those given in Ref. [11], and the net bias for void growth is varied with temperature according to theoretical results as $B_o * 873/T$ where $B_o = 0.2$ is the net bias at 773 K [12].

In addition to computing the swelling with Eq. (3), bubble swelling is evaluated under the assumption that $2\gamma/r = p$ and that all helium atoms are contained in cavities. This results in a swelling depending only on the helium concentration C_{He} [atoms m^{-3}] but not the dose, and it is given by

$$S = [3/(4\pi N_v)]^{1/2}(zC_{He}kT/2\gamma)^{3/2} \quad . \tag{8}$$

Note that $C_{He} = N_{He}S$. The surface energy in Eqs. (8) and (5) is taken to be temperature dependent, $\gamma = 1 + 0.00173(773 - T)[J/m^2]$, with the temperature coefficient as measured for 304 SS [13].

Figures 3, 4, and 5 show computed swelling values for helium contents of 100, 1000, and 10,000 appm. Curves a are obtained from Eq. (8), whereas curves b and c are numerical integrations of Eq. (3) for a He/dpa ratio of 70 and for a net bias of zero and 0.2, respectively. Comparing the curves a and b, we find that Eq. (8) overpredicts the bubble swelling by a factor of 3 to 4 at high temperatures. This is due to the fact that a finite bubble swelling rate requires an overpressure, i.e. a positive value of p_o, as a driving force for swelling. The higher the He/dpa ratio, the greater is the overpressure and the difference between the equilibrium bubble size and the dynamically growing bubble size. To confirm this, curve d in Figure 3 gives the swelling for a He/dpa ratio of one, being an upper value for breeder reactor irradiations. At temperatures greater than 750°C, bias-driven void growth is negligible, and swelling is entirely gas-driven. It is seen that for the low values of He/dpa in breeder reactors, the assumption of equilibrium bubble swelling is indeed appropriate at high temperatures. Note that the displacement damage for case d in Figure 3 is 100 dpa, whereas for case a it is only about 1.4 dpa.

The difference between equilibrium and the dynamic bubble swelling depends also on the surface energy. For example, if the surface energy is assumed to be $\gamma = 3[J/m^2]$, this difference becomes very small even for the He/dpa ratio of 70. This is mainly due to the reduction in the equilibrium bubble swelling, since $S \sim \gamma^{-3/2}$.

At temperatures below 550°C, the dynamic bubble swelling without bias (curves b) becomes nearly independent of temperature. This is explained by the fact that the bubble growth by vacancy absorption, being proportional to $D_v(\bar{C}_v - C_v^o) \lesssim D_v\bar{C}_v$, becomes negligible no matter how large the overpressure p_o is. In fact, the helium packing fraction y_o becomes larger than one, resulting in a dramatic increase of the helium pressure from the kinetic to the static pressure regime. As a result, the interstitial emission term in Eq. (3) is activated, i.e. the bubble grows by the thermally assisted generation of Frenkel pairs with retention of the vacancy and thereby relieves the static helium pressure. This mechanism is analogous to the spontaneous Frenkel pair formation discovered by

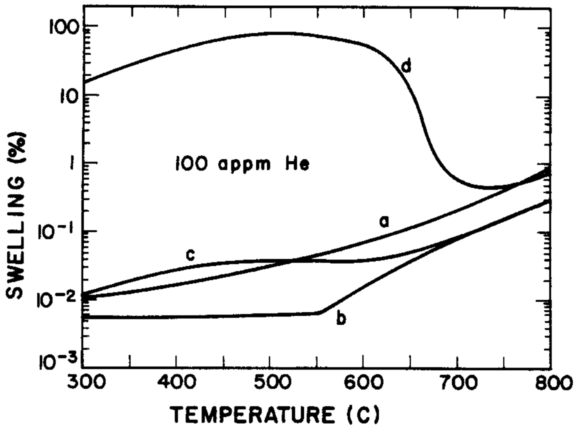

Figure 3. Swelling for He/dpa ratios of 70 (a,b,c) and 1 (d) after 100 appm He is produced; (a) for equilibrium bubble swelling; (b) dynamic bubble swelling without bias; (c,d) dynamic bubble swelling with bias.

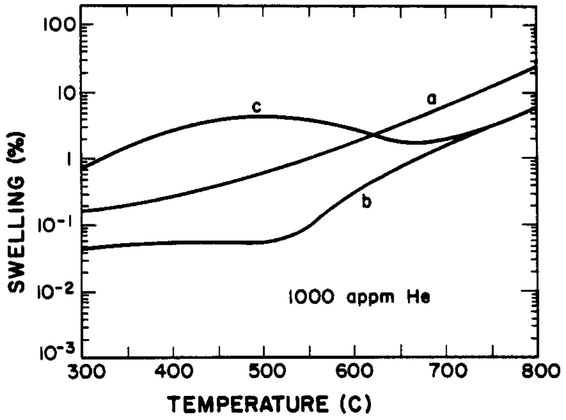

Figure 4. Swelling for He/dpa ratio of 70 after 1000 appm He is produced.

Bisson and Wilson [14] in computer simulation studies. They found that (at 0 K) a cluster of five helium interstitial atoms in a fcc metal will spontaneously create a Frenkel pair and then occupy the vacancy.

The relief of the static helium pressure by thermally-assisted Frenkel pair creation keeps the helium to vacancy ratio in these overpressurized bubbles close to one. Swelling produced by this densely packed helium is then simply given by $S \cong 0.6\, C_{He}\Omega$ where the factor 0.6 is based on the results of Baskes and Holbrook [10].

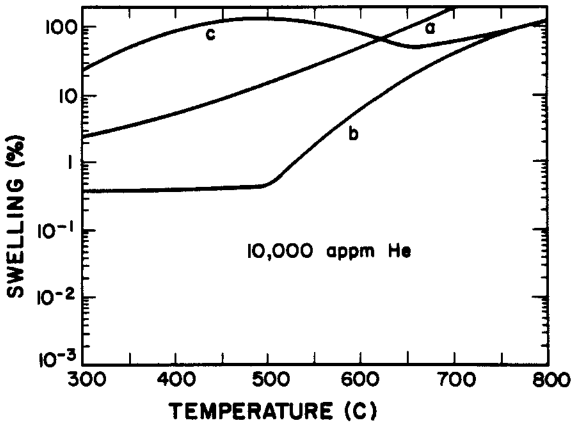

Figure 5. Swelling for He/dpa ratio of 70 after 10,000 appm He is produced.

With a net bias of $B_0 = 0.2$, void swelling remains in excess of bubble swelling for temperatures below 600 to 650°C. Even if the net bias is reduced to 0.02, bubble swelling still remains negligible compared to void swelling below about 600°C.

6. DISCUSSION AND CONCLUSIONS

Although helium accelerates the void nucleation rate at all temperatures it does not contribute to the bias-driven growth of voids below irradiation temperatures of about 600°C and for displacement rates of 10^{-6} dpa/sec. It is often argued that higher helium production may create a higher void number density, and thereby alter void swelling. The present review of void number densities in AISI 316 irradiated in fission reactors suggests, however, that differences in void number densities for irradiations carried out at the same temperature and dose rate will not persist indefinitely. Similar observations have been made with regard to the dislocation density [7]. Consequently, one must expect that the steady-state swelling rate is nearly independent of the helium producton rate at low temperatures. Since small void nuclei, even if present at low doses, do not produce large swelling values, early termination of the void nucleation by higher helium generation rates may not result in drastically different swelling behavior, particularly when microchemical evolution is a major variable for the transient regime [3].

It is possible to produce a higher void number density than the saturation value by helium preinjection or dual-ion bombardment. Experiments demonstrating this have been reported by Agarwal et al. [15]. These experiments were carried out at a dose rate of 3×10^{-3} dpa and at a correspondingly higher irradiation temperature of 700°C. In spite of the likelihood for bubble growth at this temperature, the void number densities produced for different He/dpa ratios show the tendency for converging to a saturation value both from higher and lower values. Unfortunately, the irradiations were not carried out to sufficiently large displacement doses to achieve complete saturation in all cases.

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