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A COMPARISON OF THE RELATIVE IMPORTANCE OF HELIUM AND VACANCY
ACCUMULATION IN VOID NUCLEATION*

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ABSTRACT

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Void nucleation in irradiated austenitic stainless steels generally requires the presence of either residual or transmutant gases. Classical nucleation rates are much too low to account for the number of voids observed at temperatures greater than about 450°C. An alternate path is generally believed to be responsible for void formation; viz. the growth of gas-stabilized bubbles until they reach a critical size beyond which further gas accumulation is not required to promote growth. Two limiting paths can be envisioned for void nucleation on a population of sub-critical helium/vacancy clusters; one is limited to growth by helium accumulation alone and the other to growth by stochastic fluctuations in the vacancy accumulation. As bubbles approach the critical size, stochastic processes could begin to contribute to the void nucleation rate. A comparison is made of nucleation rates along these two limiting paths as a function of the gas content of the clusters. The calculations indicate that the gas

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accumulation path is generally dominant, particularly at higher temperatures and for lower gas contents. The fraction of the critical size required for the vacancy path to contribute to the total nucleation rate increases with temperature. The results confirm the important role of transmutant helium in promoting void swelling.

KEY WORDS: cavities, helium effects, radiation damage, void nucleation, void swelling

INTRODUCTION

From the time that the phenomenon of void swelling in neutron irradiated structural materials was first discovered, transmutant and residual gases have been suspected of playing an important role in void formation (1,2). Subsequently, a considerable amount of theoretical and experimental research has confirmed those initial suspicions. Most of this work has focused on the effects of the helium which is produced by transmutation reactions between both fast and thermal neutrons and the various atomic species that comprise the material. Because helium is chemically inert and insoluble in most metals, it has been assumed that it would be a more potent aid to void nucleation than either transmutant hydrogen or residual gases such as nitrogen or oxygen. While recent work has suggested that residual oxygen can have a significant influence on void nucleation in an austenitic alloy during heavy ion irradiation in the absence of helium (3), when the same alloy is co-implanted with helium during the ion bombardment the effect of the helium appears to swamp that of the oxygen (4). Hence, the following discussion explicitly considers only the influence of transmutant helium on void nucleation. The method developed to demonstrate the importance of this gas should also be applicable for other gases that are chemically inert with respect to alloy constituents.

Classical homogeneous nucleation theory was applied to the problem of void formation in austenitic stainless steels very early (5-8). Russell and coworkers have continued to develop this theory over the past 10 years to include the effects of helium and heterogeneous nucleation (9-13).

Wolfer and coworkers have developed a Fokker-Planck formulation of the void nucleation problem and have explored the effects of mobile di-vacancies and solute segregation to void surfaces (14-16). Despite these refinements, the classical theory fails to predict the experimentally observed void densities in the intermediate to high temperature range ($450 < T < 700^{\circ}\text{C}$) where measurable void swelling occurs in these steels (12,16,17). An alternate mechanism has been proposed to cause void formation at these temperatures and to promote void formation at low temperatures. This mechanism involves the growth of small gas-stabilized bubbles until they reach a critical size beyond which further gas accumulation is not required to promote growth. Theoretical and recent experimental work has shown that the time required for these small bubbles to reach the critical size correlates well with observed void swelling nucleation times (18-23). Accordingly, one can envision two limiting paths for void formation on a population of subcritical helium/vacancy clusters; one is limited to growth by helium accumulation alone and the other to growth by stochastic fluctuations in the vacancy cluster population. A recent discussion concerning the relative magnitudes of these two processes provided some of the impetus for this work (24). A mathematical description of the two paths is given below and their magnitudes are compared for irradiation conditions typical of fast reactors.

Models of Void Formation

The two methods discussed below attempt to compute a characteristic time for nucleation or the nucleation rate per cluster for a helium/vacancy

cluster with a given number of helium atoms. The number of vacancies in this cluster or bubble is computed assuming that the bubble radius is that of a stable bubble in an irradiation environment characterized by a vacancy supersaturation S at a temperature T where:

$$S = \frac{D_v C_v - D_i C_i}{D_v C_v^e} \quad (1)$$

This stable bubble radius is slightly larger than the equilibrium bubble radius as discussed elsewhere (21,25). In Eq. (1), the terms $D_v C_v$ and $D_i C_i$ are the vacancy and interstitial fluxes impinging on the bubble and $D_v C_v^e$ is the self-diffusion coefficient. The bubble radius and the gas pressure in the bubble are computed using a hard sphere equation of state as described in Ref. 25.

For both models, the point defect concentrations are computed using the conventional rate theory and the temperature dependent sink strengths for extended defects discussed in Ref. 20. The sink strength of the sub-critical bubble population is insignificant when compared to the other sinks in the system, therefore it is not included when computing the point defect concentrations. The calculations assume that only the mono-defects and helium gas atoms are mobile and that the only defects which the bubbles emit are vacancies. The use of the principle of detailed balance and thermodynamic equilibrium (8,9) leads to the following form for the

vacancy emission rate from a cluster containing n vacancies and m helium atoms;

$$\alpha_V^n = F_S r(n-1, m) D_V C_V^e \exp \left\{ \frac{1}{kT} [G(n, m) - G(n-1, m)] \right\} \quad (2)$$

where the free energy in the equilibrium situation is:

$$G(n, m) = F_S r(n, m)^2 \gamma + mkT \ln(P) \quad (3)$$

where γ is the surface free energy, P is the pressure in the bubble and F_S is a geometric factor which can account for non-spherical clusters (20). For a sphere, $F_S = 4\pi$.

Nucleation by Stochastic Fluctuations

The method developed here is similar to that of Katz and Wiedersich (6), Clement and Wood (26) and Mansur and Wolfer (27). In general, a family of equations can be written describing the concentrations, C_k of discrete size classes containing $k, k+1, k+2, \dots$ up to $n_V^{\max}$ vacancies.

$$\frac{dC_1}{dt} = G_V + C_2(\beta_i^2 + 2\alpha_V^2) - C_1(\beta_V^1 + \beta_i^1) - D_V C_1 S_T^V \quad (4a)$$

$$\frac{dC_k}{dt} = \beta_V^{k-1} C_{k-1} + C_{k+1}(\beta_i^{k+1} + \alpha_V^{k+1}) - C_k(\beta_i^k + \beta_V^k) \quad (4b)$$

$$(k = 2 - n_v^{\max})$$

in which the $\beta_{i,v}^k$ terms are the interstitial and vacancy impingement rates on a cluster with k vacancies, the α_v^k has been given above and

$$\beta_{i,v}^k = F_s r(k,m) D_{i,v} C_{i,v} : \quad (5)$$

C_i of course equals C_v . The value of $n_v^{\max}$ can be arbitrarily large. A specific choice for its value will be discussed below. The term G_v in Eq. (4a) is the vacancy source term due to atomic displacements and vacancy emission from extended defects. The last term in Eq. (4a) represents the loss of vacancies to all the other sinks in the system. The sink strengths of the network dislocations S_p^v and the subgrain structure S_g^v are computed as described in Ref. 20,

$$S_T^v = S_g^v + S_p^v . \quad (6)$$

The temperature dependence of these sink strengths is consistent with experimental observation (20). It should be pointed out that the void nucleation rate is quite sensitive to the dislocation density (see Fig. 3b below). It is therefore important that calculations such as these include appropriate temperature dependent values of this and other microstructural parameters.

In order to simplify the equations used below, the terms which describe the shrinkage of a given cluster will be grouped together.

$$\gamma_k = \alpha_v^k + \beta_i^k \quad (7)$$

The nucleation regime can be defined as that region in vacancy cluster size space for which $\gamma_k > \beta_v^k$. Because of the radius dependence of the vacancy emission term and the existence of a supersaturation of vacancies under irradiation, a critical size is reached beyond which growth rather than shrinkage is the dominant process. The number of vacancies which correspond to this critical size will be designated n_v^* .

The steady state nucleation rate, J_{ss} , can be obtained from Eq. (4) in one of two ways. The first solution involves computing what is commonly termed the constrained equilibrium cluster size distribution which is obtained by imposing the requirement that there be no net flux from one cluster size class to the next, i.e.

$$J_k = \beta_v^k C_k - \gamma_{k+1} C_{k+1} = 0 \quad (8)$$

for all values of $k > 1$. This solution requires the calculation of pseudo-free energies of formation for clusters in each size class, resulting in an exponential cluster distribution for $n < n_v^*$. However, this method introduces the artificial result that the number of clusters begins to increase in size classes for which $n > n_v^*$. The values of the cluster concentrations in the constrained equilibrium distribution are elevated relative to the steady state distribution for $n < n_v^*$ also, this is accounted for in the theory by the introduction of the so-called Zeldovich factor. The use of this method to compute void nucleation rates is adequately discussed elsewhere (7,10).

The second method for obtaining the steady-state nucleation rate from Eq. (4) is to set all of the fluxes (Eq. 8) equal to the J_{ss} . This leads to a family of equations:

$$J_1 = \beta_V^1 C_1 - \gamma_2 C_2 = J_{ss} \quad (9a)$$

$$J_2 = \beta_V^2 C_2 - \gamma_3 C_3 = J_{ss} \quad (9b)$$

$$J_3 = \beta_V^3 C_3 - \gamma_4 C_4 = J_{ss} \quad (9c)$$

$$J_{N-1} = \beta_V^N C_{N-1} - \gamma_N C_N \quad (9d)$$

where to simplify the equations $N = n_V^{\max}$.

Let the ratio $\gamma_k/\beta_V^k = \rho_k$ for all $k > 2$ and let $\rho_1 = 1$. Then this system of equations can be solved by multiplying the equations for J_k by the product of all the ρ_k with $k < i$. Hence Eq. (9b) is multiplied by ρ_2 , Eq. (9c) is multiplied by both ρ_2 and ρ_3 and so on. If the resulting set of equations are summed, all the C_k are eliminated except C_1 and C_N , yielding

$$J_1 = J_{ss} = \frac{[\beta_V^1 C_1 - \gamma_N C_N \prod_{j=2}^N \rho_j]}{1 + \sum_{k=2}^{N-1} \prod_{j=2}^k \rho_j} \quad (10)$$

The term involving the products of the ρ_j can safely be eliminated by noting two facts. First, since the problem being considered is nucleation the concentration of the mono-defect, C_1 will in general be much greater than C_N . In fact the distribution is approximately exponential as the constrained distribution suggests. Secondly, it has already been pointed out that the ratio of the shrinkage to growth terms ρ_j is less than unity

for $n > n_v^*$. Hence for N sufficiently greater than n_v^* the product of the ρ_j contains many fractional terms. Therefore,

$$J_1 = J_{ss} = \beta_v^1 C_1 \left[1 + \sum_{k=2}^{N-1} \prod_{j=2}^k \rho_j \right]^{-1} . \quad (11)$$

The appropriate value of N can be determined numerically to ensure that J_{ss} has converged. The advantage of using this method to compute J_{ss} is the elimination of the Zeldovich factor and the approximations which must be made to compute it.

Each of the J_k in Eq. (9) can be defined in an analogous way to Eq. (10), i.e.

$$J_k = \beta_v^k C_k \left[1 + \sum_{i=k+1}^{N-1} \prod_{j=k+1}^i \rho_j \right]^{-1} . \quad (12)$$

The nucleation rate per cluster of a given size is then,

$$j_k = J_k / C_k = \beta^k \left[1 + \sum_{i=k+1}^{N-1} \prod_{j=k+1}^i \rho_j \right]^{-1} . \quad (13)$$

The characteristic time for this process would be just the reciprocal of this fractional nucleation rate:

$$\tau_k = j_k^{-1} . \quad (14)$$

Nucleation by Helium Accumulation

Voids can be formed as the result of a helium-stabilized bubble obtaining a critical number of gas atoms (18-21,25). This critical number

of gas atoms, m_{He}^* is a function of both the material and irradiation parameters,

$$m_{\text{He}}^* = f(\phi) F_v \left(\frac{\gamma}{kT} \right)^3 \left(\frac{\Omega}{\phi} \right)^2 \quad (15)$$

where $\phi = \ln S$, $f(\phi)$ is a real gas correction factor and F_v is a geometric factor to permit the consideration of non-spherical cavities. For a sphere $F_v = 4\pi/3$. The critical bubble size corresponding to m_{He}^* is in general not the same as the critical size associated with n_v^* discussed above. Their relationship is shown schematically in Figure 1. For a fixed value of the vacancy supersaturation, the family of curves shown in Figure 1 represent three different levels of helium. The curves labeled I and II contain a region in radius space for which the net growth rate is negative. This region corresponds to the void nucleation barrier and the stochastic nucleation theory deals with the probability that fluctuations will permit a bubble to grow the stable bubble radius r_b to the critical radius r_v^* . This latter radius is that corresponding to n_v^* . The curve labeled III is the curve for which $m = m_{\text{He}}^*$ and the growth rate is everywhere non-negative. The point at which curve III is tangent to the x axis is $r(m_{\text{He}}^*)$ and is also the minimum critical radius. Hence the minimum critical radius is a special case of the critical size calculated from the stochastic theory (18).

In order to compute a characteristic time for nucleation by helium accumulation it is necessary to assume a model for the partitioning of helium among the various microstructural features. Since the procedure used

here is similar to that which has been discussed previously (20) only a brief summary will be given. The model assumes that the sink strengths for helium are the same as those for vacancies except that the dislocation sink strength for helium is reduced by a factor f_p^{He} (28,29). This parameter has a nominal value of 0.5 and is varied in the analysis to determine its effect. Because the sink strength of the subcritical bubbles is small, it is neglected when computing the matrix helium concentration C_{He} but the subgrain structure (20) is included. The total system sink strength for helium is then:

$$S_T^{\text{He}} = S_g^v + f_p^{\text{He}} S_p^v \quad (15)$$

C_{He} can be computed at steady state as

$$C_{\text{He}} = \frac{G_{\text{He}}}{D_{\text{He}} S_T^{\text{He}}} \quad (17)$$

where G_{He} is the helium generation rate and D_{He} is the helium diffusivity.

The formation of bubbles on dislocations is accounted for by permitting a fraction of the helium trapped by dislocations to be "piped" to bubbles. This fraction, f_b^{He} is treated as a parameter in the analysis.

Based on the foregoing helium partitioning model, the arrival rate of helium atoms at a bubble with a radius $r(n,m)$ is

$$\beta_{\text{He}} = D_{\text{He}} C_{\text{He}} \left[F_S r(n,m) + \frac{f_b^{\text{He}} f_p^{\text{He}} S_p^v}{N_c} \right] \quad (18)$$

where N_c is the total number of bubbles among which the helium from the dislocations is partitioned. This bubble density is taken from Ref. 20 and reflects experimentally observed values. The temperature dependent expression for N_c is given in Table 1.

Equations 17 and 18 can be combined to eliminate the helium diffusivity.

$$\beta_{He} = \frac{G_{He}}{S_{He}} [F_S r(n,m) N_c + f_b^{He} f_{\rho}^{He} S_{\rho}^v] \quad (19)$$

The characteristic time for a bubble containing m helium atoms to reach the critical number is then:

$$\tau_{He} = \int_m^{m_{He}^*} \beta_{He}^{-1} dm \quad (20)$$

$$\tau_{He} \approx \frac{N_c S_T^{He}}{G_{He}} \sum_{i=m}^{m_{He}^*} [F_S r(n,i) N_c + f_b^{He} f_{\rho}^{He} S_{\rho}^v]^{-1} \quad (21)$$

RESULTS OF CALCULATIONS

The results compared in this section were computed as discussed above using the set of material and irradiation parameters listed in Table 1. The values of certain of these parameters listed in the table have been varied in the analysis and they will be discussed further in the text.

Representative values of n_v^* , r_v^* , m_{He}^* , and $r(m_{He}^*)$ which were computed at 400, 450 and 500°C using the base parameter set from Table 1 are listed in

Table 2. These parameter values are similar to those which have been used previously in rate theory simulations of void swelling (18-20).

Figure 2 provides a base case for comparison of the two alternate nucleation times as well as showing their temperature dependence. The void swelling incubation time should be approximately in the range of 10-50 dpa for the fast reactor conditions which are considered here. The values of the characteristic void nucleation times for both mechanisms are plotted in Fig. 2a and 2b as a function of m/m_{He}^* . The major differences between the two are the much greater temperature dependence and size dependence of the stochastic nucleation path. The relative magnitude of the two nucleation rates is shown in Fig. 2c where the ratio of the nucleation times has been plotted. In order for the nucleation times for the two processes to be comparable, the ratio of m/m_{He}^* must be about 0.80 for 400°C and greater than for 0.95 for 500°C. For lower gas contents the nucleation time due to helium accumulation is always much less than that for the stochastic process.

The parametric dependence of the ratio of the nucleation times at 450°C is shown in Figs. 3-4. The parameters which have been varied are the dislocation-interstitial bias, the network dislocation density, the self-diffusion coefficient, the surface energy and the two parameters in the helium partitioning model. The values of the parameters are indicated in the appropriate figure; in each case, all other parameters were maintained as listed in Table 1. The dependence seen in Figs. 3a-3c is a result of

the vacancy supersaturation varying in response to the parameter changes. Increases in the vacancy supersaturation increase nucleation by both mechanisms, but the nucleation rate by helium accumulation increases to a greater degree. Although the rate of helium absorption by a subcritical cluster is not a function of the supersaturation (Eq. 19), the critical number of gas atoms is reduced (Eq. 15). Similarly, while the critical number of gas atoms increases as the cube of the surface energy, higher surface energies also lead to increased vacancy emission (Eq. 2-3) and the net result of a higher surface energy is to decrease the stochastic nucleation rate relative to the helium mechanism (Fig. 3d).

The results are much less sensitive to the parameters in the helium partitioning model. Note that the vertical axis scaling has been changed in Figs. 4a and 4b in order to show the dependence. The fraction of dislocation-trapped helium which is directly "piped" to bubbles has only a modest effect on the ratio of the nucleation times (Fig. 4a) while varying the dislocation sink strength for helium has essentially no effect (Fig. 4b) as long as it is greater than zero.

DISCUSSION

The results presented above provide a comparison of two complementary nucleation paths which can lead to void formation in irradiated materials. The details of the results are certainly model dependent and parameter variations can selectively favor one process over the other. But the overall conclusions seem sound since the comparison is so striking. Because

the critical cluster sizes are relatively small for the temperatures discussed here, the characteristic nucleation times which have been computed should be somewhat less than the time at which measurable swelling is observed. However, even with moderate amounts of gas the stochastic nucleation theory predicts relatively long nucleation times at intermediate temperatures. At higher temperatures, this theory fails to predict finite nucleation rates with reasonable material parameters. This result is already evident in the values shown for 500°C in Fig. 2b and is in striking disagreement with the fact that void formation persists up to 700°C in austenitic stainless steels (30). Observed void swelling incubation times are much more consistent with the helium accumulation process as shown in Fig. 2a.

The role of fluctuations begins to be significant when a sufficiently large fraction of the critical number of gas atoms has been accumulated. However, at 400°C this fraction is already about 0.8 while at 500°C it is greater than 0.95. With these levels of helium, voids can nucleate due to fluctuations in a relatively short time as shown in Figs. 2b and 2c. This result is highlighted in Fig. 5 where the material parameters have been chosen to maximize the influence of the stochastic path; a low bias ($Z_i^n = 1.01$), a high network dislocation density ($\rho_n = 1.5 \times 10^{15} \text{ m}^{-2}$), a high self diffusion energy ($E_{SD} = 3.0 \text{ eV}$) and a low surface energy ($\gamma = 1.0 \text{ J/m}^2$). Even in this extreme case a relatively large amount of gas is required for stochastic fluctuations to make a significant contribution to the void nucleation rate. At 400°C 20% of the critical number of gas atoms is required and at 500°C over 80%. Hence, fluctuations can hasten the

final stages of void nucleation, but only if sufficient gas is available to assist the earlier stage.

This work highlights the importance of the role that transmuted helium plays in void formation. For temperatures of technological interest (i.e. $350 < T < 600^{\circ}\text{C}$), an examination of Figs. 2-4 indicates that fluctuations will contribute to the void nucleation rate only for cavities which are already near the critical size. At low temperatures the two processes contribute more equally to the total nucleation rate with the gas driven process dominant for smaller clusters. These conclusions support earlier work which explored the concept of the critical number of gas atoms and suggested that void formation was largely due to helium stabilized bubbles reaching the critical radius rather than due to stochastic fluctuations (18-23,25,31). The results are also consistent with the fact that higher cavity densities are observed in both neutron and ion irradiations in which the ratio of helium to displacement damage is increased relative to the fast reactor value (32,33). This further indicates that void formation may occur at much higher rates in a deuterium-tritium fusion reactor first wall than in fast reactor fuel cladding since the helium generation rate in the former will be about 30 times the latter.

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TABLE 1--Material and Irradiation Parameters

E_v^f	1.50 eV
E_v^{m*}	1.40 eV
D_v^0	$8 \times 10^{-4} \text{ m}^{-2} \text{ sec}^{-1}$
E_i^m	0.85 eV
D_i^0	$8 \times 10^{-5} \text{ m}^{-2} \text{ sec}^{-1}$
G_{dpa}	$0.25 \times 10^{-6} \text{ dpa/sec}^\dagger$
G_{He}	$3.5 \times 10^{-13} \text{ He/atom/sec}$
γ^*	$2.025-8.75 \times 10^{-4} T (\text{°C}) \text{ J/m}^2$
$\rho_n(T)^*$	$2.0 \times 10^{18} \exp [-0.016 T(\text{°C})] \text{ m}^{-2}$
$N_c(T)$	$2.53 \times 10^{25} \exp [-0.023 T(\text{°C})] \text{ m}^{-3}$
Z_i^{n*}	1.25
$f_b^{\text{He}*}$	0.5
$f_\rho^{\text{He}*}$	0.5

*Parameter varied in analysis.

† Equivalent to a 10^{-6} dpa/sec dose rate with a cascade efficiency of 0.25.

TABLE 2—Critical Cluster Parameters

T(°C)	m_{He}^*	$r(m_{\text{He}}^*)$ (nm)	$m = 0$		$m = 0.9$ m_{He}^*	
			n_v^*	r_v^*	n_v^*	r_v^*
400	33	0.574	121	0.692	95	0.638
450	98	0.890	481	1.10	353	0.989
500	576	1.82	4718	2.35	3387	2.10

FIGURE CAPTIONS

Figure 1. Schematic drawing comparing the cluster radii discussed in text.

Figure 2. Characteristic nucleation times for the helium accumulation process (2a), stochastic theory (2b) and their ratio (2c) at 400, 450 and 500°C.

Figure 3. Parametric variation in the ratio of the nucleation time by helium accumulation to that by the stochastic theory at 450°C. The parameters are the dislocation-interstitial bias (3a), the network dislocation density (3b), the activation energy for self-diffusion (3c) and the surface energy (3d).

Figure 4. Dependence of the nucleation time ratio on the parameters in the helium partitioning model; fraction of dislocation-trapped helium piped to bubbles (4a) and the fractional reduction in the dislocation sink strength (4b).

Figure 5. Temperature dependence of the nucleation time ratio with parameters chosen to minimize the influence of helium; $Z^n = 1.01$, $E_{SD} = 3.0$, $\rho_n = 1.5 \times 10^{15}$ and $\gamma = 1.0$.

Fig 1.

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Fig 1

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 Figure 2a

R-146
Stoller and Odette
Figure 2b.

R-146
 Stoller and Odette
 Fig. 2c

Stoller and Odette
R-146
Fig. 3a

Stoller and Odette
R-146
Fig. 3b

m/m_{He}^*

Stoller and Odette
R-46
Fig. 3c

Stoller and Odette
R-146
Fig. 3d

Stoller and Odette
 R-146
 Fig. 4a

Stoller and Odette
 R-146
 Fig 4.b

Stoller and Odette
R-146
Fig. 5

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