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Production bias: a proposed modification of the driving force for void swelling under cascade damage conditions

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A new concept of point defect production as the main driving force for void swelling under cascade damage conditions is proposed. This concept takes into account the recombination and formation of immobile clusters and loops of vacancies and interstitials th the cascade region. The lifetimes of the clusters and loops due to desolution are strong functions of the temperature, as well as their vacancy and interstitial nature. The resulting biased production of free point defects from the internal sources is shown to be a strong driving force for void swelling. The characteristics of void swelling due to production bias are described and compared with experimental results. We conclude that the production bias concept provides a good description of void swelling under cascade damage conditions.

Introduction

Recent studies [1–4] have shown that the number of freely migrating point defects that survive in typical cascades may be on the order of only a few per cent of those initially displaced. The observed swelling rate in metals and alloys irradiated with fast neutrons, on the other hand, can be up to 1–2%/dpa [5–7], depending on the irradiation temperature and dose. This swelling rate represents a 1–2% *net* rate of accumulation of vacancies over interstitials into voids alone. To produce this net accumulation rate the current accepted theory of void swelling requires many times more vacancies to be actually annihilated at the voids and dislocations.

Within the framework of the current accepted theory one is left with only two alternatives to resolve this dilemma – either the dislocation bias for interstitials is much larger than that derived from theoretical calculations [8], or the generation rate of freely migrating defects under cascade damage conditions is much larger than that deduced from various experiments [1–4] and computer simulation results (see sect. 2).

Comparison of the void swelling rates resulting from neutron and electron irradiations leads to further insight into this problem. The displacement damage produced by electrons in HVEM is predominantly in the form of freely migrating point defects. It is well known that the HVEM irradiation-produced swelling rates are comparable to those produced by neutron irradiation (~ 1% per dpa) [9–11]. Since there is no reason to expect any large difference between the dislocation bias between neutron and electron irradiations; it is difficult to reconcile, on the one hand, the large difference in the production rates of the freely migrating point defects and, on the other hand, the comparable vacancy accumulation rate in voids, in the two different cases. Unless, of course, the driving mechanisms for void swelling are different in the Frenkel pair and cascade-damage conditions. This possibility was suggested recently by Woo and Singh [12–13], who examined the effect of the intracascade clustering of both vacancies and interstitials on void swelling and proposed the concept of "production bias" as alternative driving force for void swelling under cascade damage conditions. Subsequently, Singh, Woo and Foreman [14] and Singh and Foreman [15] have shown that the swelling behaviour at very low doses can also be understood entirely in terms of production bias. In the present paper the main predictions of the production bias are compared with experimental observations.

2. Point defect clustering and production bias

The classical picture of a cascade with a vacancy-rich core surrounded by SIAs at the periphery has, in recent years, been confirmed by computer simulation experiments [16–18]. The MD simulations [17–19] have demonstrated that a large fraction of vacancies and SIAs produced in a collision cascade mutually recombine, while a substantial fraction of the surviving defects forms clusters (both vacancy and interstitial) and only small fractions of the initial displacements escape the cascade volume as freely migrating point defects. Evidence for both experimental and theoretical work supporting the clustering of both vacancies and interstitials have been reported [17–23].

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At temperatures higher than stage V, the lifetimes of vacancy loops due to thermal annealing becomes significantly reduced compared with that of vacancy clusters. If we consider the vacancy clusters from which the thermal emission of vacancies occurs, as internal sources, it is easy to see the inherent existence of a large asymmetry under cascade damage conditions and hence a bias between the production efficiencies of mobile vacancies and interstitials *available* for annihilation at voids. This asymmetry arises because of the likely difference between the fractions of vacancy and interstitial agglomeration. More importantly, while vacancy clusters may generate mobile vacancies by evaporation, interstitial clusters may not; this situation has also been pointed out by Simons [24]. We have termed this strongly temperature-dependent difference between the production (from internal plus external vacancies) of two kinds of available point defects the production bias [12,13].

3. Void swelling under cascade damage conditions

Due to intracascade recombination and agglomeration, the production rate of freely migrating defects is substantially reduced from the dpa rate K calculated by the NRT model [25]. Let α be the fraction of intracascade recombination, ϵ_i be the fraction of agglomeration of the surviving interstitials and ϵ_v, that for the vacancies. The effective generation rate of surviving point defects G is given by $(1-\alpha)K$ in our calculations.

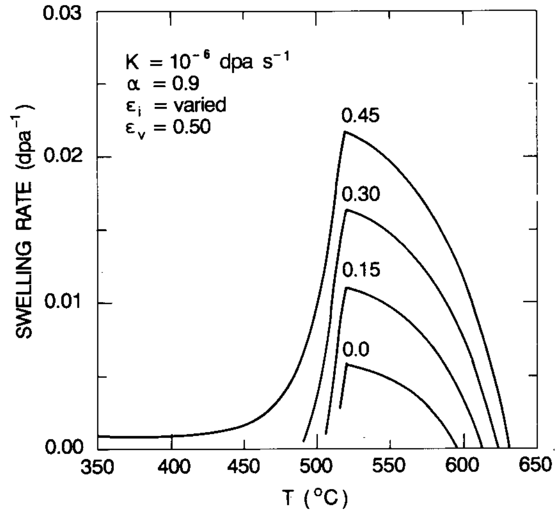

Fig. 1. The calculated temperature dependence of swelling rate for different values of ϵ_i (= 0.0–0.45) at $K = 10^{-6}$ dpa s^{-1}, representing the case of fast reactor irradiation.

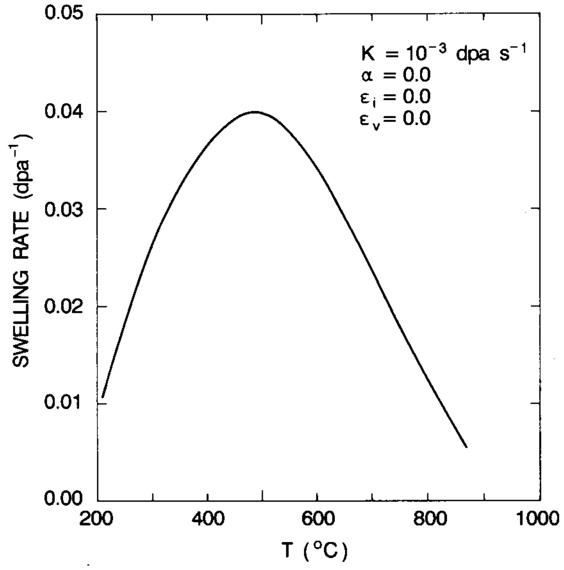

Fig. 2. The calculated temperature dependence of swelling rate for the case of low recoil energy irradiation where the damage is produced homogeneously and in the form of Frenkel pairs instead of multidisplacement cascades; this represents the case of 1 MeV electron irradiation, for example.

To illustrate the operation of the production bias, let us consider void swelling in a metal in which dislocation structure is made up of a network of density ρ_N, vacancy loops of density N_{vL} and interstitial loops of density N_{iL}. We also denote, for the vacancies, the total dislocation sink strength by k_d^2, the void sink strength by k_c^2, and the total sink strength by k_v^2. Let p be the dislocation bias then we may write down the total dislocation sink strength as $(1+p)k_d^2$, the corresponding total sink strength being k_i^2.

For the present steady-state point defect concentrations, the rate equations for the vacancies and interstitials can be written as

$$(1-\epsilon_i)G - \left[k_c^2 + (1+p)k_d^2\right]D_iC_i - \alpha_t C_iC_v = 0, \quad (1)$$

$$(1-\epsilon_v)G + K^e - \left(k_c^2 + k_d^2\right)D_vC_v - \alpha_t C_iC_v = 0, \quad (2)$$

where α_t is the coefficient of thermal recombination due to long-range migration, and K^e is the total vacancy emission strength for the various sinks, and has contributions from the voids, interstitial loops, the vacancy loops (clusters) and the network.

The solution of eqs. (1) and (2) is straightforward and is documented in ref. [13]. Using experimentally derived quantities for the microstructure (see table 1 of ref. [13]), the swelling rate as a function of temperature can be calculated for fast reactor (cascade damage) and HVEM (Frenkel pair production) irradiation conditions. The results are plotted in figs. 1 and 2. From fig. 1, the following characteristics of swelling under cascade damage conditions can be seen.

- The existence of two sharply separated temperature regimes: low sweling rate at lower temperatures and high swelling rate at the peak swelling temperature. At the peak temperature, the swelling rate could be almost a factor of 10 higher than that expected at lower temperatures. The magnitude of this factor is expected to be sensitive to material parameters and irradiation conditions (particularly recoil energies).

- The high swelling rate regimes are dominated by the production bias whereas the dislocation bias determines the low swelling rate regime.
- The transition from the low temperature (dislocation bias) regime to the high temperature (production bias) regime is generally very sharp. The steepness of the temperature dependence in this transition regime is governed by a relatively high activation energy, nearly equal to the activation energy for self-diffusion.
- The swelling rate at all temperatures increases with interstitial clustering.

Comparing with fig. 1, the following features of fig. 2 are noted:

- The peak swelling rates are comparable, despite a difference of a factor of ~ 20 in the production rate of freely migrating defects.
- There is no obvious and abrupt transition from a low-swelling to a high-swelling region as temperature increases. The smooth increase of swelling rate with temperature at lower temperatures in fig. 2 is due to the small activation energy ($E_m/2 \sim 0.7$ eV) involved in increasing the recombination, as compared to the large activation energy (2.7 eV) associated with the change in mechanisms in going from low to high temperature in fig. 1.

4. Discussion

In the following, the qualitative and semiquantitative swelling behaviour during neutron and electron irradiation calculated by the present theory are compared with observations from experiments.

As can be seen from fig. 1, the temperature range of interest for SS 316 is expected to be in the neighbourhood of ~ 400°C. To obtain an unobscured assessment of the temperature dependence near 400°C, it is important that variations of other factors, such as displacement rate gradient, specimen composition and microstructure be avoided. In this regard, fuel pin data from DFR fast reactor are suitable. In this case, an inlet temperature of ~ 270°C places, for many fuel pins, the region at ~ 400°C in the middle of the core where displacement rate gradients are relatively small. Fig. 3 demonstrates the swelling behaviour of typical 316 stainless cladding and the dramatic decline of the swelling rate in the temperature range around 400°C, even though this temperature range is associated with almost no significant variation in either displacement rate or neutron spectra [26–28]. The steep reduction in swelling becomes more pronounced at higher fluences as larger regions of the pin receive exposures beyond that of the transient swelling regime. This implies that it is the steady-state rate of swelling that is affected most strongly in this temperature range. This strong sensitivity to temperature appears to manifest itself below the range (~ 450–650°C) where the temperature-independent steady-state swelling rate of most austenitic stainless steels is ~ 1% per dpa and the variability of swelling is associated primarily with the influence of temperature, displacement rate and material variables on the duration of the transient regime of swelling [7,29–31].

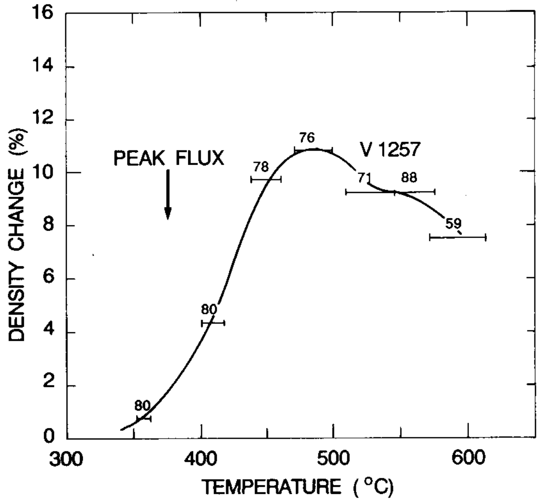

Fig. 3. Measured swelling in the fuel pin V1257 irradiated in the DFR fast reactor as a function of irradiation temperature. Note that the swelling declines precipitously between 350 and 450°C, even though there is only a little variation in displacement rate over this temperature range [25–27]. The numbers indicated on the curve refer to dose in terms of dpa (in the Half-Nelson units where 80 dpa (Half-Nelson) = 67 dpa (NRT) = 1.3×10^{27} n/m^2 ($E > 0.1$ MeV)).

The precipitous fall of the swelling rate around 400°C was also reported by Garner and coworkers, who showed that at ~ 400°C the steady-state swelling rate of various austenitic steels indeed falls to ~ 0.1% per dpa or less, depending on the alloy, its starting condition and the irradiation history [32–35].

Another recent experiment also demonstrates that the steady-state. regime indeed responds strongly when the temperature is changed from the 1% per dpa regime to a temperature below that regime [36]. In this experiment 316 stainless steel was irradiated at five stress levels at 550°C to 30 dpa whereupon most of the transient evolution was completed and swelling was accelerating rapidly. The temperature was then changed abruptly to 400°C in one stressed and one unstressed pin, and the irradiation continued. In both cases there was a dramatic reduction in the strain rate

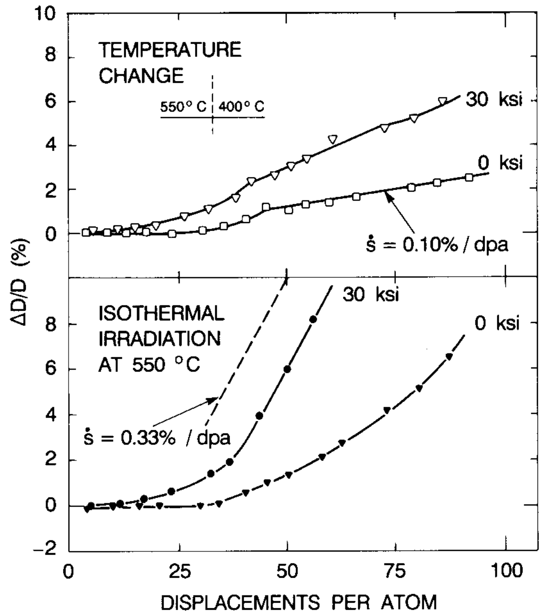

Fig. 4. Effect of temperature change (550 to 400°C) on creep and swelling in the 20% cold worked 316 stainless steel [35] as a function of displacement dose for a stressed and an unstressed samples [35]. Note a dramatic reduction in creep and swelling as the temperature is changed from 550 to 400°C.

thereafter with the swelling rate falling to 0.1%/dpa (see fig. 4). The behaviour observed in this experiment is consistent with the predictions based on the concept of a strongly temperature-dependent defect production term. One would expect, however, that if changes were made between two temperatures within the 1% per dpa regime (but after the transient was complete) that no change in the swelling rate would occur. This behaviour has indeed been observed by Yang and Garner [37].

Some indications of a rapid transition to a high swelling regime was also observed [38] in the swelling of pure copper in the ORR mixed spectrum reactor, showing a fivefold difference in swelling within 50°C.

It is worth pointing out that the void swelling calculated for neutron irradiated copper at very low doses (10^{-4}–10^{0} dpa), using the production bias as the only driving force for void growth, also agrees well with experimental observations [14,15]. In these calculations, the rate of vacancy escape from the cascade volume is calculated on the basis of cascade parameters determined from the MD simulations and is taken to be the effective vacancy production rate. It is relevant to point out here that the high swelling rate observed at low doses (e.g. ref. [6]) could not be explained in terms of the conventional rate theory using the dislocation bias as the only driving force for the void swelling [39].

5. Summary

We propose that the small production rate of freely migrating defects during fast neutron irradiation and the large steady-state swelling rates in the high swelling regime can be reconciled by a new void swelling mechanism based on the production bias as the main driving force. The physical origin and concept of the production bias is briefly reviewed. The characteristics of void swelling due to production bias were described and confirmed by comparison with experiments.

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