conf. 790327--34

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A HELIUM-INDUCED SHIFT IN THE TEMPERATURE DEPENDENCE OF SWELLING*

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In an annealed austenitic alloy undergoing bombardment with 4 MeV Ni ions to doses between 1 and 70 dpa at 840, 900, 950, 1025 and 1100 K, the introduction of simultaneously-implanted helium at a rate of 20 appm He/dpa moves the swelling versus temperature curve up the temperature scale by 40 to 70 K. Co-implantation of hydrogen (deuterium) at a rate of 50 appm D/dpa simultaneously with the helium causes little or no additional systematic effects. The major change in microstructure caused by the gases is an enhancement of cavity nucleation by factors of 2 to 5 at 840 to 950 K, increasing to factors of thousands at 1100 K. Concurrently there is a reduction in the size of cavities and in swelling at all temperatures below 1025 K, and an increase in cavity size and in swelling at 1100 K, where the cavities are gas stabilized. At 1025 K the increase in nucleation of cavities outpaces the reduction in size and causes increased swelling. The primary effects of the gases are decided at low doses below 1 dpa where cavity nucleation is completed and where the conditions governing cavity growth are established; at higher doses swelling is determined by cavity growth, which is dependent on dose only. The gases cause copious formation of cavities on grain boundaries, boding ill for mechanical properties.

1. INTRODUCTION

Gases are known to encourage the development of damage microstructure in crystalline solids during displacive irradiation. Helium [1–4] and hydrogen [3–5] are especially potent nucleators of voids and bubbles. In the first wall of a fusion reactor these two gases are expected to be generated at high rates from (n,α) and (n,p) reactions. The most recent estimates [6] of the generation rates in type 316 austenitic stainless steel are about 12 appm helium/dpa and about 45 appm hydrogen/dpa, while those in the nickel-base alloy PE16 are 20 and 77, respectively. Additional helium, together with deuterium and tritium, may be picked up from the plasma and the breeder blanket. This paper explores the effects of significant quantities of helium and hydrogen on swelling in an austenitic alloy using heavy ion bombardment and simultaneous implantation of the gases.

2. THE EXPERIMENT

The composition of the austenitic alloy, designated P7, is similar to that of type 316 stainless steel but with low levels of carbon and minor metallic elements such that it swells readily [7] but does not undergo radiation-induced phase instability which would complicate the study. Chemical analyses gave Fe–17 wt % Cr, 16.7 Ni, 2.5 Mo, 0.005 C with all other elements each less than 0.1%; the oxygen level was high, 0.03% (1062 appm). Annealed and polished disks of this alloy about 0.5 mm thick were bombarded in the Oak Ridge National Laboratory triple ion irradiation facility which was recently upgraded from its previous dual ion capability. The principle and practice of this improved facility are described briefly in two recent short papers [8,9], and full details and results of the present experiments will be made available in a laboratory report. In essence, the facility consists of two Van de Graaff particle accelerators, one of them delivering the displacement damage beam of 4 MeV ^{58}Ni ions and the other one simultaneously implanting alpha particles and deuterons from a source consisting of a mixture of helium and molecular deuterium. Deuterons are used as a substitute for hydrogen because the technique does not permit simultaneous implantation of helium and hydrogen, and because the deuterium is expected to function in the damage process in much the same way as hydrogen does.

Microstructures and swelling response as functions of bombardment temperature were assessed for three irradiation conditions, namely (a) nickel ion bombardments only, with no implanted gas, (b) dual ion irradiations with nickel and helium at He/dpa = 20 at the peak damage depth, and (c) triple ion irradiations with nickel, helium and deuterium at He/dpa = 20 and D/dpa = 50. The bombardment temperatures were 840, 900, 950, 1025 and 1100 K, with measured deviations less than ±5 K. The attempted nominal doses at the peak damage depths were 1, 10 and 70 dpa, estimated using the EDEP-1 code with an effective threshold energy of 40 eV and a correlation factor of 0.8 [10]. Actual doses on each disk depended on the Ni beam intensity profile which was measured continuously with an oscillating vane. The displacement damage rate in the peak

*Research sponsored by the Division of Materials Sciences, U.S. Department of Energy under contract W-7405-eng-26 with the Union Carbide Corporation.

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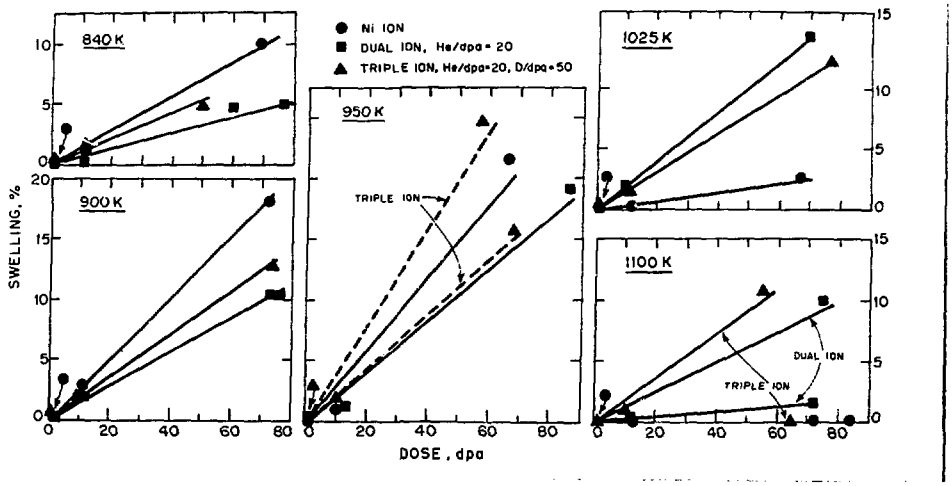

Fig. 1. Variation of swelling with dose and gas implantation at all test temperatures.

damage region was about 6×10^{-3} dpa s^{-1}. Transmission electron microscopy foils were prepared from the bombarded disks by electropolishing the bombarded surface to a depth of 0.6 μm, as indicated by Dektak profilemetry, then electrothinning from the back of the disk to electron transparency, thus exposing the peak displacement damage region for examination. Swelling is expressed [10] as $100\ V_c/(V_s - V_c)$, where $V_c = N\pi d^3/6$ and is the volume of cavities in a volume, V_s, of the swollen specimen, with N the cavity concentration and d the volume-averaged cavity diameter.

3. RESULTS AND DISCUSSION

3.1 Swelling

The dose dependence of swelling for each irradiation condition and temperature appears to follow linear relationships as shown in Fig. 1. However, there were some spurious results from duplicate specimens of the high dose triple ion irradiation at 950 K and from both of the high dose multiple ion irradiations at 1100 K. In these cases a single straight line can not describe the data so we have drawn several lines that define the observed limits of swelling. These deviants will be discussed in more detail when we describe the microstructures.

The temperature-dependence of the swelling rates from Fig. 1 are shown more clearly in Fig. 2. Note that for Ni ion bombardments without concurrent gas injection the swelling rate is a maximum at about 950 K and falls to zero at about 1050 K. Simultaneously-injected helium at a He/dpa ratio of 20 reduces the swelling rate at temperatures below 950 K, and increases the swelling rate at 1025 and 1100 K compared to bombardments with Ni alone. *Therefore, simultaneous implantation of helium at this high ratio translates the swelling curve up the temperature scale by 40 to 70 K.*

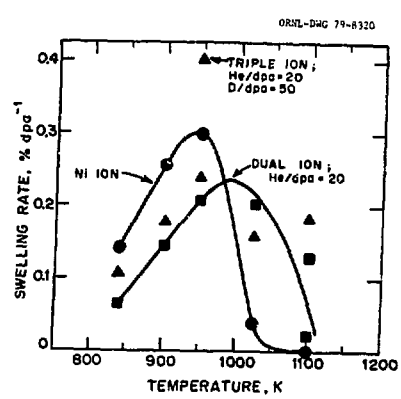

Fig. 2. Temperature dependence of swelling rates.

No attempt is made to draw a curve through the data points for the triple ion irradiations because of the aforementioned erratic swelling rates at 950 K. Although the triple ion bombardment data at the two lower temperatures appear to follow a sensible pattern with respect to the dual ion and Ni ion data, the pattern does not hold at higher temperatures. We consider that the scatter on the triple ion data *is too great for us to convincingly conclude that swelling under triple ion irradiation is systematically different from that for the dual ion irradiations.* In the same vein, it is not clear whether the apparent reduction in maximum swelling rate at the peak swelling temperature for the dual ion irradiations is real. If a single curve is drawn through the dual *and* triple ion datum points, the maximum swelling rate could be as high as that for the Ni ion case.

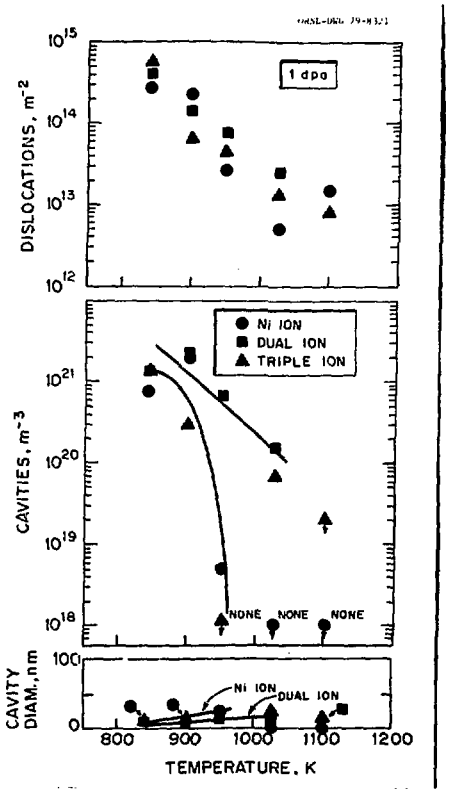

Fig. 3. Temperature dependence of microstructural data for a dose of 1 dpa.

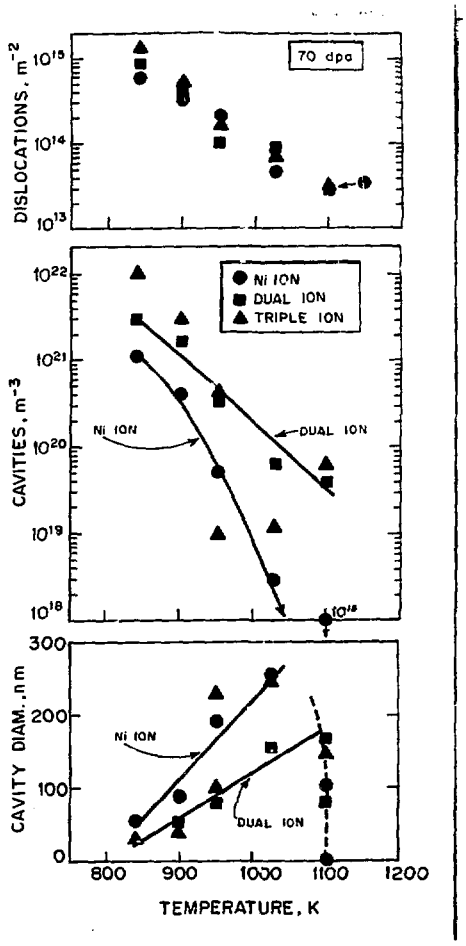

Fig. 4. Temperature dependence of microstructural data for a dose of 70 dpa.

3.2 Microstructural data

Quantitative microstructural data for 1 and 70 dpa are presented in Figs. 3 and 4. Data for 10 dpa are omitted because of space limitations, but they follow the same pattern as those in Figs. 3 and 4. There is a good deal of experimental scatter in the data but certain trends are clear and lead to some important conclusions.

First, *cavity concentrations decline with increasing irradiation temperature and are higher for dual ion irradiations than for Ni ion irradiations.* The fractional difference in concentration is a strongly increasing function of temperature being a factor of about 2 or 3 at 840 K and 900 K, increasing to 4 to 5 at 950 K and thereafter rising markedly to a factor of several thousands at 1100 K. This impressive difference at the higher temperatures is due the fact that *the cavity concentrations for the multiple ion irradiations do not decrease with temperature as sharply as those for the Ni ion irradiations.*

Second, *the concentrations of cavities are independent of dose between 1 and 70 dpa for all three types of irradiation at all temperatures.* The line drawn through the dual ion 70 dpa data in Fig. 4 can adequately represent the 1 and 10 dpa dual ion data, too, and the drooping curve for the 70 dpa Ni ion data is typical of those for the 1 dpa and 10 dpa data. The data for triple ion irradiations are highly erratic, sometimes falling on the dual ion line and sometimes on the Ni ion line, and occasionally in between. We conclude that *nucleation of cavities is essentially completed at doses below 1 dpa for all three types of irradiation at all temperatures. It follows that the extra cavity nucleation caused by simultaneous implantation of helium and hydrogen must have occurred at helium levels below 20 appm and hydrogen (deuterium) below 60 appm, and that subsequent increases in the levels of simultaneously implanted gases and in the dose do not encourage further nucleation.*

Cavity size increases linearly with irradiation temperature to 1025 K for 10 and 70 dpa doses, and to 950 K for 1 dpa, *then decreases sharply at 1100 K. The cavities are largest for the single ion irradiations and are almost 50% smaller for the dual ion bombardments, except at 1100 K, where the multiple ion cavities are the larger.* Cavities from the triple ion bombardments show variable behavior associated with their concentrations; when the concentration is lower than that for the dual ion case the cavity size is usually larger, and vice versa. For all three types of bombardment the cavity size increases with dose. The linear dependence on temperature over most of the temperature range is maintained at all doses. Within this temperature range there is a persistent roughly 50% reduction in cavity diameter at all doses for the multiple ion cavities. The slopes of the linear temperature dependencies for the 10 dpa and 1 dpa doses are about one-half and one-eighth, respectively, of those for the 70 dpa cases. At 1100 K the cavities also increase in size with dose, but in most cases at 1100 K the multiple ion cavities are significantly larger than the Ni ion cavities. *Over most of the temperature range, then, simultaneously-implanted helium at He/dpa = 20 increases the concentration of cavities and decreases their size.* On the contrary, *at 1100 K the simultaneously-implanted gases increase both the concentration and the size of the cavities.* Assuming that all of the helium is in the cavities we estimate that *for all temperatures below 1100 K the cavities are underpressurized; at 1100 K the cavities are fully pressurized. The size, or growth, of cavities is an increasing function of dose but is insensitive to increasing gas levels simultaneously implanted beyond a dose of 1 dpa. Moreover, the fractional difference in size, or growth rate, between single ion and multiple ion cavities is established at doses below 1 dpa and is thereafter independent of both dose and simultaneously implanted gas level.* Co-implantation of hydrogen (deuterium) gives widely varying behavior of cavity growth, seemingly being close to that for the dual beam irradiations at 840 and 900 K, and extremely erratic at 950 and 1025 K, i.e., in the vicinity of the peak swelling temperature. In only one sense is the triple ion data rational — the cavity sizes usually vary inversely with the cavity concentrations.

Dislocation densities decline progressively with increasing irradiation temperature. Their magnitude for a given dose, and their temperature dependence are seemingly invariant with gas level. The magnitude is dependent on dose, the levels at 1 dpa and 10 dpa being lower than those at 70 dpa by factors of two-thirds and one-half, respectively. Since cavity nucleation is completed below 1 dpa, these changes in dislocation densities with dose must be associated with cavity growth.

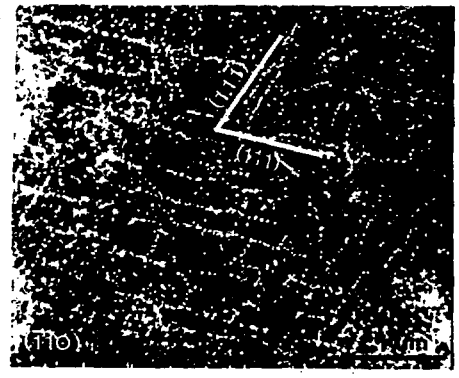

Fig. 5. Inhomogeneous cavity distribution at T = 900 K, dpa = 10, He/dpa = 20.

3.3 Specific microstructural features

Two particular aspects of the microstructures warrant especial mention. The first is that the spatial distribution of loops and cavities is highly heterogeneous at the 1 and 10 dpa levels for all irradiations. Details of these heterogeneities will be described in a later paper. Loop heterogeneity in the form of arrays is most obvious at 1 dpa and 840 K. Cavity heterogeneity is prominent at all temperatures, usually taking the form of short rows, sheets and large clouds. The short rows lie in discontinuous lines parallel to {111} (Fig. 5); some queues very similar to classical dislocation pile-ups are also seen: The observations indicate that loops and cavities are nucleated at preferred sites. We suggest that *nucleation occurs at clusters of residual impurities, possibly oxygen, that were associated with dislocations during prior thermomechanical treatment* and which resisted dispersal during the final anneal. Companion work on this material [11] shows that the heterogeneities can be erased by implanting helium prior to displacive bombardment.

The second aspect concerns grain boundaries. These display considerable evidence of migration during irradiation, and in the multiple ion specimens they become riddled with cavities of smaller size but much higher local density than those in the grain interiors (Fig. 6). *These grain boundary cavities* are most obvious at 1100 K but are resolvable at smaller sizes at temperatures down to at least 900 K. At the two higher temperatures it was easily seen that the grain boundary cavities increased in size with dose, but at a much slower rate than the larger cavities within the grains. *The presence of these grain boundary cavities is taken as an indication that they are gas stabilized,* and is also regarded as a warning that the high gas levels generated in first wall materials will not only influence swelling behavior but may seriously degrade the mechanical strength of grain boundaries. Indeed, *the*

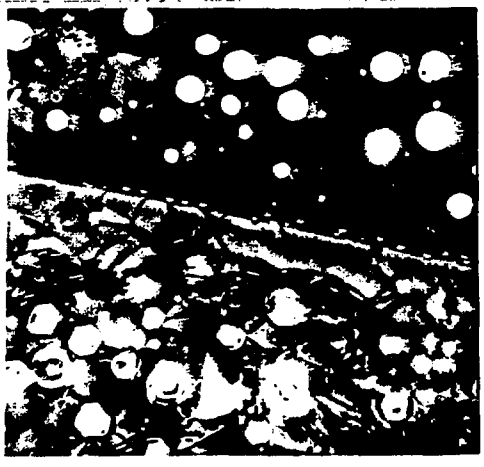

Fig. 6. Example of grain boundary cavities developed during simultaneous gas implantation. T = 1100 K, dpa = 70, He/dpa = 20, D/dpa = 50.

embrittling effects of helium may prove to be a much more serious problem than its effects on swelling.

3.4 Comments

Helium has a much stronger and more reliable role than deuterium in this work. A detailed analysis of its effects, and a thorough comparison with current swelling theory [12–15] and with recent experimental findings from other dual ion irradiations [16–20] will be made in later publications. *Clearly, the major feature that influences the swelling response and microstructural development in this alloy, and which is responsible for the upward temperature shift in swelling, is the enhanced concentrations of cavities caused by the gases.* Not only are these cavities developed at doses below 1 dpa, but their subsequent growth rate, and hence the swelling response, is also decided at such low doses. This emphasizes the importance of cavity nucleation processes and related events on the nature and degree of swelling, and draws attention to the need for more studies of damage structures at doses below the swelling incubation dose.

The erratic behavior and effects of hydrogen (deuterium) in some of the specimens in this study are puzzling. Nothing in our records or our observations of the bombardments indicate any unusual conditions for these specimens. That they should behave in such odd fashion even when helium is present is even more mystifying. We know that the deuterium rapidly escapes from the specimens at the elevated temperatures used in this work [9] but we do not know how long a residence period is required for it to influence the development of damage structure. Experiments are in hand to measure diffusion and trapping of deuterium in damaged specimens, and to see if the deuterium (and helium) affects the depth distribution of damage structure.

ACKNOWLEDGMENTS

This work could not have been done without the unstinting assistance of R. A. Buhl, M. Inman, C. G. McKamey and M. B. Lewis. We thank Connie Harrison for typing the manuscript.

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