CONF-791215-72

SIMULATION OF FIRST WALL DAMAGE: EFFECTS OF THE METHOD OF GAS IMPLANTATION*

N. H. PACKAN and K. FARRELL

Oak Ridge National Laboratory, Oak Ridge, Tennessee 37830

Cavity formation in an austenitic alloy of similar composition to type 316 stainless steel has been explored with regard to various methods of gas implantation. Irradiations were conducted at 900 K to doses of 1, 10, and 70 dpa with helium injection levels of 20 appm/dpa. Highest swelling (18%) was exhibited by the unimplanted reference material; a lesser amount by simultaneous helium injection (11%). Greatly reduced swelling due to profuse cavity nucleation was the result of the preinjection of 1400 appm He, either at room temperature (S = 1%) or at 900 K (4%). The dislocation density was not sensitive to helium injection technique. Simultaneous injection of 50 appm H/dpa, along with the He, may have caused a modest increase in the cavity and dislocation concentrations at higher doses. The observations are compared with a theory of void growth kinetics to estimate the relative influence of voids and dislocations as point defect sinks.

1. INTRODUCTION

The first wall of a fusion reactor will be exposed to an intense high-energy neutron flux which will generate both large numbers of atomic displacements and copious quantities of gaseous transmutation products (helium and hydrogen). The resulting high concentrations of vacancies and interstitials can, at certain temperatures, give rise to cavity formation. This process is promoted by the known facility of helium [1–4] and hydrogen [3–5] in stimulating the nucleation of bubbles. To explore such effects directly would require protracted neutron irradiations. An alternative increasingly used is to subject specimens to charged particle bombardment which generates in only a few hours damage levels that are equivalent to years of reactor exposure. The "transmutation products" must be separately added, either by prior injection using an accelerator or exposure to a radioisotope source [6], or preferably by gas injection simultaneous with the damage production. A recent estimate [7] of fusion reactor gas generation rates in type 316 austenitic stainless steel predicts about 12 appm helium/dpa and about 45 appm hydrogen/dpa, while those in the nickel-base alloy PE16 are about 20 and 47, respectively. In accelerator bombardments, the method of introduction of such quantities of gas might well be expected to influence the resulting damage microstructure. To investigate this, we have made a controlled comparison of several gas injection methods applied to a "high purity 316" austenitic alloy at a fixed irradiation temperature near 900 K and doses ranging from 1 to 70 dpa.

2. EXPERIMENTAL DETAILS

The composition of the austenitic alloy (Fe–17 wt % Cr, 16.7 Ni, 2.5 Mo) was similar to 316 stainless steel but with low (0.005 wt %) carbon and low residual elements (less than 0.1% each) to facilitate void formation and minimize phase instability. There was a relatively high oxygen content (1062 appm, equivalent to 0.03 wt %). The material was cold worked, with intermediate vacuum anneals at 1323 K, to a final 0.5 mm thickness out of which 3-mm disks were punched and then annealed for 15 min at 1323 K in argon. The disks were then mechanically polished through 0.1 μm diamond grit and electropolished at 233 K to remove any vestiges of surface mechanical deformation.

Ion bombardments were carried out using the ORNL dual accelerator irradiation facility which has recently acquired the capability [8,9] of injecting *both* helium and deuterium (equivalent to hydrogen) simultaneously with the damage-creating 4.0 MeV ^{58}Ni ion beam. Control of the irradiation parameters was similar to that described in previously published work [10]; in this case all bombardments were carried out at a temperature of 898 ± 5 K, which is below the peak swelling temperature of 950 K [11]. The vacuum at the target was typically better than 10 μPa. A sliding mask in front of the target permitted each row of disks in a 3 × 3 array to be bombarded to different doses. Nominally the attempted 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 [12]. The actual doses on each disk depended on the nickel beam intensity profile which was measured continuously with an oscillating vane. Typically the nickel beam current was 1 particle

---

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

CONF-790125--72

SIMULATION OF FIRST WALL DAMAGE: EFFECTS OF THE METHOD OF GAS IMPLANTATION*

N. H. PACKAN and K. FARRELL

Oak Ridge National Laboratory, Oak Ridge, Tennessee 37830

Cavity formation in an austenitic alloy of similar composition to type 316 stainless steel has been explored with regard to various methods of gas implantation. Irradiations were conducted at 900 K to doses of 1, 10, and 70 dpa with helium injection levels of 20 appm/dpa. Highest swelling (18%) was exhibited by the unimplanted reference material; a lesser amount by simultaneous helium injection (11%). Greatly reduced swelling due to profuse cavity nucleation was the result of the preinjection of 1400 appm He, either at room temperature (S = 1%) or at 900 K (4%). The dislocation density was not sensitive to helium injection technique. Simultaneous injection of 50 appm H/dpa, along with the He, may have caused a modest increase in the cavity and dislocation concentrations at higher doses. The observations are compared with a theory of void growth kinetics to estimate the relative influence of voids and dislocations as point defect sinks.

1. INTRODUCTION

The first wall of a fusion reactor will be exposed to an intense high-energy neutron flux which will generate both large numbers of atomic displacements and copious quantities of gaseous transmutation products (helium and hydrogen). The resulting high concentrations of vacancies and interstitials can, at certain temperatures, give rise to cavity formation. This process is promoted by the known facility of helium [1–4] and hydrogen [3–5] in stimulating the nucleation of bubbles. To explore such effects directly would require protracted neutron irradiations. An alternative increasingly used is to subject specimens to charged particle bombardment which generates in only a few hours damage levels that are equivalent to years of reactor exposure. The "transmutation products" must be separately added, either by prior injection using an accelerator or exposure to a radioisotope source [6], or preferably by gas injection simultaneous with the damage production. A recent estimate [7] of fusion reactor gas generation rates in type 316 austenitic stainless steel predicts about 12 appm helium/dpa and about 45 appm hydrogen/dpa, while those in the nickel-base alloy PE16 are about 20 and 77, respectively. In accelerator bombardments, the method of introduction of such quantities of gas might well be expected to influence the resulting damage microstructure. To investigate this, we have made a controlled comparison of several gas injection methods applied to a "high purity 316" austenitic alloy at a fixed irradiation temperature near 900 K and doses ranging from 1 to 70 dpa.

2. EXPERIMENTAL DETAILS

The composition of the austenitic alloy (Fe–17 wt % Cr, 16.7 Ni, 2.5 Mo) was similar to 316 stainless steel but with low (0.005 wt %) carbon and low residual elements (less than 0.1% each) to facilitate void formation and minimize phase instability. There was a relatively high oxygen content (1062 appm, equivalent to 0.03 wt %). The material was cold worked, with intermediate vacuum anneals at 1323 K, to a final 0.5 mm thickness out of which 3-mm disks were punched and then annealed for 15 min at 1323 K in argon. The disks were then mechanically polished through 0.1 μm diamond grit and electropolished at 233 K to remove any vestiges of surface mechanical deformation.

Ion bombardments were carried out using the ORNL dual accelerator irradiation facility which has recently acquired the capability [8,9] of injecting *both* helium and deuterium (equivalent to hydrogen) simultaneously with the damage-creating 4.0 MeV ^{58}Ni ion beam. Control of the irradiation parameters was similar to that described in previously published work [10]; in this case all bombardments were carried out at a temperature of 898 ± 5 K, which is below the peak swelling temperature of 950 K [11]. The vacuum at the target was typically better than 10 μPa. A sliding mask in front of the target permitted each row of disks in a 3 × 3 array to be bombarded to different doses. Nominally the attempted 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 [12]. The actual doses on each disk depende on the nickel beam intensity profile which was measured continuously with an oscillating vane. Typically the nickel beam current was 1 particl

---

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

μA, yielding a displacement rate of about 6.1×10^{-3} dpa·s^{-1} in peak damage region. The currents in the gas line were set by trial and error at about 110 μA of He^{+} and 140 μA of D_2^{+} so as to give 20 at. ppm He/dpa and 50 at. ppm D/dpa at the peak damage depth.

Following ion bombardment specimens were electrochemically thinned from the bombarded side to a depth of 0.6 μm, and then from the back side to perforation. They were then examined in a JEM 100-C electron microscope which could reveal cavities as small as 2 nm diam. Foil thicknesses were determined by stereo microscopy. Errors from these measurements, together with sectioning errors in obtaining a foil near the peak damage depth and problems with spatially-inhomogeneous defect distributions, contribute to the uncertainty in our values for the defect concentrations. Usually we considered a real difference in defect concentrations to be greater than a factor of 2 to 3. Swelling values were computed directly from the cavity sizes and concentrations per ASTM recommendations [12].

The alternative methods of gas injection which were explored in this experiment were: simultaneous injection of helium; preinjection of helium at room temperature (RT); preinjection at the subsequent damage production temperature of 900 K; and simultaneous injection of hydrogen (actually deuterium) along with helium. The preinjected specimens were either given a fixed 1400 appm He implantation regardless

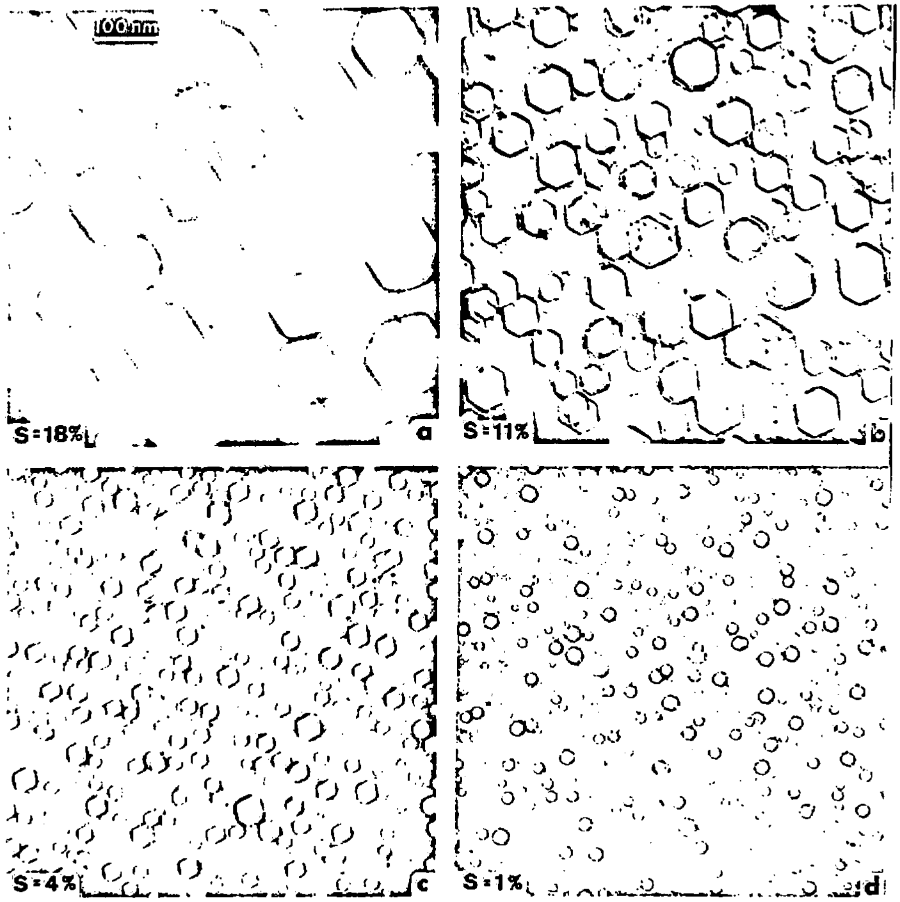

Fig. 1. Examples of void sizes and concentrations produced by a bombardment at 900 K to 70 dpa. (a) No added gas; (b) simultaneous injection of a cumulative 1400 at. ppm He; (c) preinjection of 1400 ppm He at 900 K; and (d) preinjection of 1400 ppm He at room temperature.

of their subsequent displacement-producing exposure, or were preimplanted at RT with 20 appm and 200 appm He and subsequently bombarded to 1 and 10 dpa, respectively. Specimens bombarded without any added gas were used as the reference condition.

3. RESULTS

Different methods of introducing helium can indeed profoundly affect the final damage morphology as is evident in Fig. 1. In each case (except 1a, no gas) the same amount of helium (1400 at. ppm) was introduced into a specimen that ultimately received about 70 dpa at 900 K. The largest swelling, 18%, was seen for the material with no added gas, Fig. 1a. Less swelling was exhibited by the specimens given simultaneous gas injection: the dual beam situation (Fig. 1b) yielded 11% while a triple ion bombardment (not shown) resulted in 13% swelling. The two preinjection techniques caused substantially lower swelling, apparently as a result of intensified cavity nucleation and restrained cavity growth. In Fig. 1c, preinjection at 900 K followed directly by nickel ion bombardment at the same temperature yielded a swelling of 4%, while preinjection at room temperature produced the most abundant nucleation and a swelling of only 1%.

The development of swelling at lower doses is shown in Fig. 2, wherein it appears that all but one of the various irradiation techniques reflect a linear swelling with dose, each with its own unique swelling rate. The rates for the no gas, simultaneous hydrogen plus helium, simultaneous helium, and RT preinjection cases are 0.26, 0.18, 0.15, and 0.016% per dpa. The elevated temperature preinjection data appear to share the slope of the simultaneous injections at low doses, but later drop down to approximately the swelling rate of RT preinjection. The RT preinjection experiment in which He/dpa = 20 was held constant, rather than a fixed 1400 ppm He, showed a very low swelling of 0.003% at 1 dpa and 2.9% at 11 dpa. Clearly the different methods of gas introducion create a disparity in swelling that increases with dose.

Figures 3–5 show data on the microstructural characteristics (mean cavity diameters, concentrations, and dislocation densities) resulting from the various bombardment methods. The reference condition evidently attained its high swelling by means of a high cavity growth rate and in spite of declining cavity numbers. Simultaneous injection of helium caused swelling roughly similar to that for the helium plus hydrogen case, but there is an interesting contrast in their cavity generation rates. The dual beam samples show roughly constant cavity numbers, whereas those of triple beam bombardment exhibit a steadily-rising cavity concentration through 70 dpa (from quite a low value at 1 dpa).

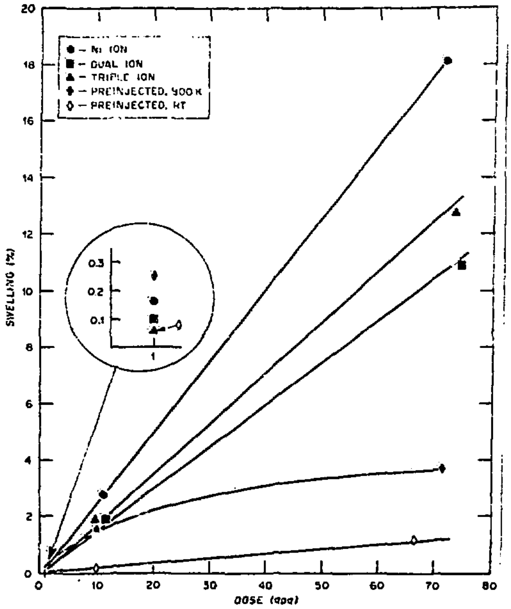

Fig. 2. Dependence of swelling upon dose for the gas injection methods studied.

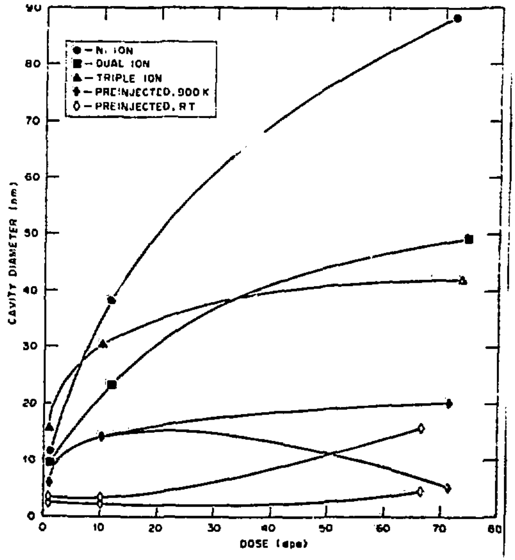

Fig. 3. Diameter of a cavity of mean volume as a function of dose. Certain preinjection conditions gave rise to bimodal size distributions, indicated here by pairs of data points.

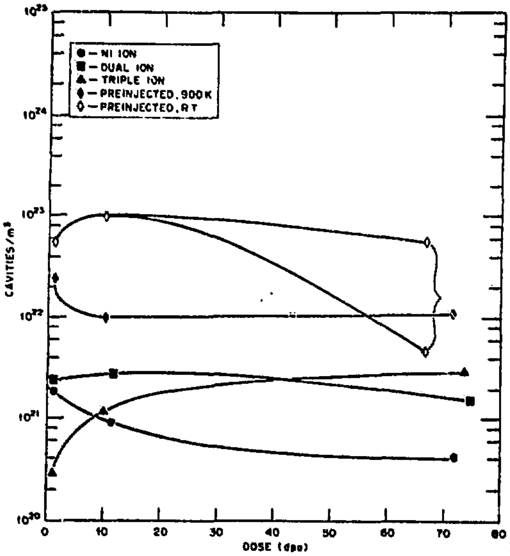

Fig. 4. Disparate behavior of the cavity concentration with dose for various methods of gas introduction.

Preinjection of 1400 ppm He brought about drastic swelling reduction through profuse initial nucleation of cavities which evidently competed with one another, allowing little cavity growth. The RT preinjection, in fact, gave rise to a sharply bimodal cavity size distribution. The smaller group (4 nm mean diam) grew negligibly over the dose range studies, perhaps because they were just below a critical size for void growth. Preinjection at 900 K ultimately generated a bimodal size distribution quantitatively similar to that of RT preinjection also at 70 dpa, but up to 10 dpa the cavities of high-temperature preinjection were more like those of simultaneous injection (or the no-gas case).

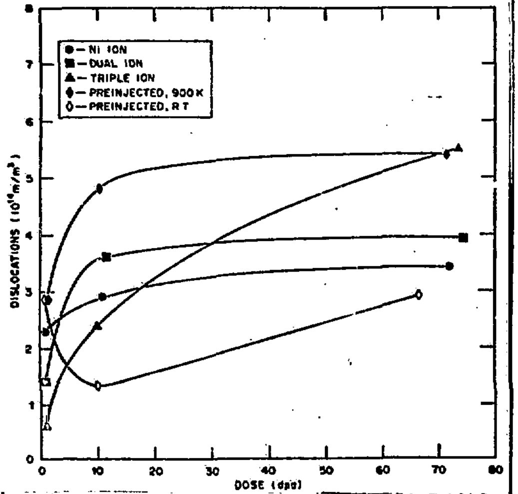

Fig. 5. Dislocation density is much less sensitive to gas implantation technique than is the void density.

The dislocation density (Fig. 5) was not especially sensitive to the method of gas injection, having variations of no more than a factor of 3. It may or may not be significant (above scatter) that while most specimens exhibited a plateau in dislocation density after ∿10 dpa, the material injected with helium plus hydrogen showed a persistent increase over the whole dose range, analogous to the increase it also showed in cavity density. The RT-preinjected material once more behaved in a contrary fashion.

4. DISCUSSION

The reference specimens bombarded with only nickel ions swelled significantly more than those subjected to any of the gas implantation techniques, a high swelling that resulted from the highest void growth rate. Such behavior can be expected from an inherently high-swelling material which fosters immediate void nucleation on preexisting sites and on a scale not so fine as to hinder subsequent void growth. Voids in this material were frequently observed in strings or other heterogeneous spatial distributions, most likely resulting from nucleation on similarly-distributed gases or other indigenous impurities. Simultaneous (dual and triple ion) gas injection resulted in reduced but still substantial swelling and lower cavity growth rates. The dual-ion irradiation also yielded cavity and dislocation concentrations comparable to the nickel-ion irradiation. These basically similar results despite the gradual addition of 1400 appm He indicate that again cavity nucleation must have been essentially set at the beginning, and that helium was added too slowly (even at our 20 appm He/dpa rate) to make a major change. Such a result would *not* be expected for a material in which cavity nucleation is difficult, and in fact Kenik [13] has observed a profound influence of simultaneously-injected He in the low swelling alloy LS1A. A similar variation in the effectiveness of simultaneously implanted helium depending on the relative ease of cavity nucleation without helium has been noted before by Brimhall and Simonen [14,15].

In the case of the triple-ion bombardments, both the initial (1 dpa) dislocation and cavity concentrations were notably low, yet both values increased continuously with dpa through the highest dose studied. This suggests that the simultaneous addition of hydrogen might play a modest but continuing role in sustaining cavity nucleation in this type of material. However, caution is advisable because such a trend was not found in our bombardments conducted at or above the peak swelling temperature [11].

The preinjection of 1400 appm He had a marked effect on the damage microstructures. Preinjection at room temperature produced 10^{23} cavities/m^3 and a swelling of only 1% at 70 dpa. The spatial distribution of these numerous cavities was quite uniform which implies that here the injected helium has assumed the dominant role in cavity nucleation. Evaluation of our observed microstructural parameters in a test parameter Q in the comprehensive model for void growth kinetics by Mansur [16] indicates that for 10 dpa and above, cavities are overwhelmingly the dominant sink for RT preinjection, whereas voids and dislocations are about equally important for the simultaneous injection and no-helium cases. The swelling vs dose behavior of 900 K-preinjected material was similar to that from dual-ion bombardment up to 10 dpa (though the cavity concentration was three times higher and the cavities only half as big). However, from 10 to 70 dpa this microstructure increasingly came to resemble that of RT preinjection. The initial dissimilarity from RT preinjection is presumably due to the fact that at the beginning of bombardment there were many more nuclei present that were greater than the critical size for growth; hence mean cavity diameter and swelling initially increased much more rapidly. Evidently the initial nucleation was nevertheless profuse enough to limit the cavities from growing as large as those from simultaneous injection.

That hot preinjection can give larger (and presumably fewer) nuclei than does RT preinjection was shown by Mazey and Nelson [17] who investigated *as-implanted* 316 stainless steel and observed defect clusters and helium bubbles for implantations made at 773 K and above. Comparing then specimens implanted with 100 appm He at RT and 873 K followed by bombardment with 46.5 MeV Ni ions to 40 dpa, they observed the same trends reported here: larger and fewer cavities and greater swelling for elevated temperature (ET) preinjection versus RT preinjection. A more recent comparison by McGruer et al. [18] of ET and RT-preinjection and simultaneous helium injection (80 appm/dpa) of 304 stainless steel is only partially consistent with these findings. Their results for RT preinjection and dual-ion bombardment (all to 5–15 dpa) are quantitatively similar to the 10 dpa values of this experiment. However, the hot preinjection (925, 975, and 1025 K) trials yielded cavities three times larger and seven times fewer than those of simultaneous bombardment, a relationship opposite to our result. Possibly the higher temperatures of their study may be partially responsible for the difference. One important point of agreement between the current investigation and all of the prior works [15,17, 18] is that the total dislocation content is very little affected by the method of helium introduction.

5. SUMMARY

Different modes of gas introduction strongly affect cavity formation in this free-swelling material. The disparities generally increase with dose as far as 70 dpa. The unimplanted reference material swells the most due to a high void growth rate. Simultaneous injection of helium (20 appm/dpa) yields a lower swelling and a nearly-constant concentration of cavities over the dose range studied. Preinjection of the same total amount of helium at room temperature gives profuse cavity nucleation and the lowest swelling. Preinjection of helium at the subsequent heavy ion bombardment temperature (900 K) results in low-dose swelling similar to simultaneous helium injection, changing over to resemble RT preinjection by 70 dpa. The total dislocation density is not significantly affected by the method of helium introduction. Simultaneous injection of 50 appm/dpa hydrogen may have a tendency to prolong cavity and dislocation nucleation for irradiations below the peak swelling temperature.

6. REFERENCES

[1] R. S. Nelson and D. J. Mazey, *Radiation Damage in Reactor Materials*, Vol. II, Vienna Symposium, IAEA (1969) 154–163.

[2] E. E. Bloom and J. O. Stiegler, *J. Nucl. Mater.* 36 (1970) 331.

[3] K. Farrell, A. Wolfenden, and R. T. King, *Radn. Effects* 8 (1971) 107.

[4] M. J. Makin, J. A. Hudson, D. J. Mazey, R. S. Nelson, G. P. Walters, and T. M. Williams, *Radiation Damage in Reactor Materials*, Proceedings of Conference, Scottsdale, Arizona, 1977 (Met. Soc. of AIME (1977) 645–665.

[5] J. T. Buswell, S. B. Fisher, J. E. Harbottle, and D.I.R. Norris, *Physical Metallurgy of Reactor Fuel Elements*, 1073 Berkeley Conf. (Metals Society, London, 1975) 170–174.

[6] N. H. Packan and W. A. Coghlan, *Nucl. Technol.* 40 (1978) 208.

[7] T. A. Gabriel, B. L. Bishop, and F. W. Wiffen, *Nucl. Technol.* 38 (1978) 209.

[8] K. Farrell, M. B. Lewis, and N. H. Packan, *Scripta Met.* 12 (1978) 1121.

[9] M. B. Lewis, *Proceedings of Conference on Use of Small Accelerators*, Denton, Texas, Nov. 1978, IEEE (to be published).

[10] N. H. Packan, K. Farrell, and J. O. Stiegler, *J. Nucl. Mater.* 78 (1978) 143.

[11] K. Farrell and N. H. Packan, this conference.

[12] ANSI/ASTM Std. E521-77, *Annual ASTM Standards*, Part 45 (American Society for Testing and Materials (1977) 984.

[13] E. A. Kenik, this conference.

[14] J. L. Brimhall and E. P. Simonen, *Nucl. Technol.* 29 (1976) 378.

[15] J. L. Brimhall and E. P. Simonen, *J. Nucl. Mater.* 68 (1977) 235.

[16] L. K. Mansur, *Nucl. Technol.* 40 (1978) 5.

[17] D. J. Mazey and R. S. Nelson, *Radiation Effects and Tritium Technology for Fusion Reactors*, USERDA CONF 750989, Vol. I (1975) 240–258.

[18] J. N. McGruer et al., *J. Nucl. Mater.* 74 (1978) 174.