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TEMPERATURE ASPECTS OF PULSED ION BOMBARDMENT IN AN AUSTENITIC ALLOY*

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The influence of pulsed ion irradiation to a damage level of 10 dpa at temperatures from 843 to 1023 K has been studied in a quaternary Fe-17Cr-17Ni-2.5Mo austenitic alloy. Pulsing periods of either 60 s on/60 s off or 1 s on/1 s off were used, together with continuous bombardment as a control. Helium (200 appm) was supplied either by simultaneous or room temperature preimplantation, or omitted. While most specimens experienced only 2-3 K temperature fluctuations with the beam pulses, certain specimens were deliberately allowed to undergo 15 to 50 K variations. Pulsed irradiation effects were found to range from little effect at 843 K, through a notable modification in the relationship between helium and cavity parameters at 938 K (near the peak swelling temperature), to a substantial reduction of swelling at 1023 K. The influence of the simultaneous temperature excursions compared with constant-temperature pulsed ion bombardment (mostly compared at 843 K) was small. These results are interpreted in terms of the effects of pulsing and helium on the stability of cavities at high temperatures through the critical size and number of gas atoms needed for a small cavity to begin bias-driven void growth.

1. INTRODUCTION

Partial simulation of the severe radiation environment of a fusion reactor first wall is possible through the use of heavy ion bombardment combined with helium injection. One aspect of many magnetic fusion reactor designs, the sequences of "burn" periods interspersed by off times, can be represented by interrupting (pulsing) the heavy ion beams at appropriately compressed periods, typically 1 to 60 s. Pulsed ion bombardment also has promise as a diagnostic tool for sorting out the kinetics of damage processes.

Many pulsing experiments to date have produced results that are not consistent from one experiment to another. A number of them employed pure nickel¹⁻⁴ and variously reported reduced,¹ increased,^{2,3} and little changed⁴ cavity mean sizes for pulsed compared to continuous bombardment. There were usually only minor changes to the overall swelling, however. Diverging results were also seen in "pure" ternary or quaternary austenitic alloys⁵⁻⁷ and

(partially) in two complex, low-swelling alloys.^{8,9} In most experiments temperature excursions of >10 K due to beam heating were apparently superimposed upon the pulsed damage production. In some cases,⁶⁻⁹ however, such temperature perturbations were limited to <2-3 K by using a lower beam intensity. The present work focuses on the changing influences of pulsing and helium over the temperature range for swelling on damage microstructure in a "pure" austenitic alloy analogous to type 316 stainless steel. The question of pulsing with and without concomitant temperature changes is also investigated, mainly at 843 K.

2. EXPERIMENTAL

The target material, designated P7 (Fe-17 wt % Cr-16.7 Ni-2.5 Mo), was a high-purity austenitic alloy similar to type 316 stainless steel but with low carbon (0.005 wt %) and low residual elements (less than 0.1% each). In previous continuous-irradiation experiments¹¹ the material swelled readily and remained single

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phase to high dose. Bombardments were carried out at the ORNL dual accelerator facility using 4 MeV Ni⁺⁺ and 0.2–0.4 MeV He⁺ ions. For those specimens that did receive helium, the amount was always 200 appm, implanted either at room temperature prior to bombardment (preimplantation) or simultaneously with the Ni ions at elevated temperature (coimplantation).

Bombardment temperatures were 843, 938, and 1023 K, spanning the swelling regime in this material.¹¹ Pulsing (interrupting both beams by means of synchronized shutters) was either "slow" (60 s on/60 s off) or "fast" (1 s on/1 s off) with steady beam as a control.

All specimens received a dose of nominally 10 dpa, at a common instantaneous dose rate of 2×10^{-3} dpa s⁻¹ (thus pulsed bombardments lasted twice the time of continuous bombardments). This threefold-reduced flux was employed so that beam on/off temperature fluctuations were normally less than 2–3 K. Exceptions to this rule were two cases in which a specimen was apparently not tightly clamped against the heat sink during slow pulse bombardments at 938 K, one using coimplantation and the other helium-free. These specimens exhibited ~40 K variations. Similar variations resulted from the deliberate incorporation of some loosely-mounted specimens in the helium-free and the helium preinjected 843 K bombardments.

3. RESULTS

Part of the results collected and analyzed here have been reported already: the 938 K data⁶ except for the temperature pulse points, the 1023 K data,⁷ and the 843 K data for coimplantation only.⁷ Illustrative electron micrographs of the respective damage structures are included in those papers.

As set forth in Table 1 and depicted as functions of temperature in Figure 1, the data first demonstrate how the choice of helium

TABLE 1
Values for cavity number density (N), mean void diameter (d), swelling (S), and dislocation density (L)

		Irradiation temperature (K)				
		843				
		d (nm)	N (10 ²¹ m ⁻³)	S (%)	L (10 ¹⁴ m ⁻²)	
No He implantation	no pulse	20	17–27	0.6–1.1	2.4–5.0	
	slow pulse	18.5	19	0.5–0.85	3.3	
	fast pulse	21.8	16–29	1.0–1.4	3.6	
Helium coimplanted	no pulse	*3.3 17	*68 25	*0.013 0.59	3.8	
	slow pulse	*4.2 19	*34 33	*0.014 1.2	4.3	
	fast pulse	*3.3 19	*24 30	*0.004 1.3	3.2	
Helium preimplanted	no pulse	*3.5 11	*83 20	*0.017 0.11	2.2	
	slow pulse	*4.7 12	*60 21	*0.035 0.18	3.2	
	fast pulse	*4.2 14	*72 26	*0.030 0.34	2.5	
		938				
		d (nm)	N (10 ²¹ m ⁻³)	S (%)	L (10 ¹⁴ m ⁻²)	
No He implantation	no pulse	55	2.5	1.8	1.2	
	slow pulse	50	4.0	2.8	1.8	
	fast pulse	65	1.1	2.4	1.5	
Helium coimplanted	no pulse	*4.2 35	*2 15	*0.001 3.7	1.9	
	slow pulse	*5.2 55	*1 4	*0.001 3.3	1.7	
	fast pulse	*5.2 44	*4 6	*0.003 2.6	1.5	
Helium preimplanted	no pulse	*4.4 19	*100 8–19	*0.03 0.25–1.0	0.4–1.5	
	slow pulse	*4.9 21	*30 2	*0.05 0.09	0.2	
	fast pulse	*4.9 13–16	*80 0.7–2	*0.05 0.02	0.7–0.2	
		1023				
		d (nm)	N (10 ²¹ m ⁻³)	S (%)	L (10 ¹⁴ m ⁻²)	
No He implantation	no pulse	84	0.18	0.45	0.21	
	slow pulse	0	0	0	0.15	
	fast pulse	0	0	0	0.18	
Helium coimplanted	no pulse	*9.8 39	*1.3 1.2	*0.007 0.48	0.44	
	slow pulse	15	6.1	0.10	0.13	
	fast pulse	20	2.0	0.074	0.23	
Helium preimplanted	no pulse	*6.2 18	*23 1.5	*0.030 0.054	0.22	
	slow pulse	5.6	39	0.041	0.09	
	fast pulse	6.3	50	0.064	0.11	

*Data of smaller cavities in bimodal distribution.

implantation method profoundly affects the damage features. With continuous irradiation helium refines the cavities by decreasing the sizes and increasing the number density, and generates a second component of small cavities (filled symbols in Figure 1) which are especially prominent with helium preimplantation.

The influence of pulsing is a bit more subtle than the effect of helium and is highly temperature sensitive. At 843 K, any influence of fast or slow pulsing on either the cavities

FIGURE 1

Variations in damage microstructure for P7 austenitic alloy bombarded to 10 dpa over the range 843–1023 K for several pulsing and helium implantation conditions.

or dislocations is small. At 938 K, near the peak swelling temperature, there is little variation in either the dislocation content or overall swelling with pulsing. However the refinement of the large cavities by helium coimplantation or preimplantation during continuous irradiation was not observed for pulsed irradiation; their statistics are similar to those of the helium-free case. Near the high temperature limit for swelling, 1023 K, either fast or slow pulsing suppresses all large cavities; by contrast, in the steady irradiations large cavities are observed with or without helium. However, the populations of small cavities are not greatly affected by pulsing at any temperature. The dislocation density is rather insensitive to pulsing except with preimplanted helium at the medium and high temperatures where it is somewhat depressed by pulsing.

The behavior of the specimens which experienced concurrent flux and temperature variations appears in Table 2 and is shown in Figure 1 as symbols with horizontal bars representing their observed temperature ranges. At 843 K the changes are not large but a 10–20% reduction in cavity sizes was generally realized. At 938 K, the no-helium and the coimplanted temperature-change specimens both had ~20% smaller cavity mean sizes but they diverged in opposite directions from their

isothermal counterparts for cavity concentrations and swelling. Interestingly, while all the 938 K isothermal coimplanted specimens exhibited both large and small cavities in approximately equal numbers, no trace of any small cavities could be found in the 921–944 temperature-change specimen.

4. DISCUSSION

In a recent review of helium interactions mechanisms, Mansur and Coghlan¹² explore the concept of a critical size r_C^C that a small cavity must attain to be able to grow further as a void by bias-driven vacancy acceptance. This critical size is strongly sensitive to gas: the more gas atoms in the cavity, the smaller is r_C^C . If the cavity contains more than a critical number of gas atoms n_g^* , there no longer is a critical radius because vacancy emission is then subordinate to vacancy acceptance for any size; the smallest critical radius, corresponding to n_g^* gas atoms in the cavity, is r_C^{C*} . They submit that the observation of a bimodal cavity size distribution in an irradiated material is evidence for the influence of the critical size: the largest cavities in the small cavity set should be $\approx 2r_C^{C*}$ in diameter. Promotion of any cavities from the subcritical to the void class is possible by random size fluctuations (boosting some of them over r_C^C) and/or by further gas accumulation.

Corroboration of these ideas can be found in the present data. At a high (1023 K) temperature, the expression for r_C^{C*} can be evaluated for our dose rate and observed dislocation density L and reasonable values for surface energy ($\gamma = 1.5 \text{ J}\cdot\text{m}^{-2}$) and the dislocation bias for interstitials ($[Z_V^d Z_V^C - Z_V^d Z_V^C] = 0.02$) to obtain the calculated result $r_C^{C*} = 8.7 \text{ nm}$ (or 17–18 nm diam). For our no-pulse, coimplanted specimens at 1023 K (Table 1), the small

TABLE 2

Data for pulse-irradiated specimens which also experienced synchronous temperature variations

843 K	Temp (°F) On Off	d (nm)	N (10^{20} m^{-2})	S (%)	L (10^{10} m^{-2})
No helium slow pulse	803 928	17	19	0.53	3.7
fast pulse	828 913	18	19	0.50	3.3
Preimplanted He slow pulse	849 917	4.3 10	105 20	0.037 0.13	3.7
fast pulse	828 917	4.4 11	98 24	0.039 0.19	3.2
938 K					
No helium slow pulse	953 913	40	10	4.0	1.9
Coimplanted He slow pulse	944 921	44	2.3	1.4	1.8

cavities had a mean diameter of 9.8 nm and a maximum size (not shown) of ~18 nm, consistent with the above-calculated r_C^* value.

What, then, is the influence of pulsing? For coimplanted helium at 1023 K only unimodal cavity size distributions were observed for fast or slow pulsing, with diameters of 15–20 nm (mean size) or 30–40 nm (maximum size). This indicates that pulsing may have (1) substantially raised the critical size for void growth, or (2) reduced the size of the small cavities hovering at and below r_C^* . The first result arises because r_C^* becomes infinite in the annealing period between pulses when the point defect concentrations have subsided. The second possibility arises because, as shown by Mansur and Coghlan,¹² the gas pressure in a cavity near r_C^* and n_g^* is only one-third of its equilibrium pressure at the same size and temperature. Thus during a beam-off interval the cavities near r_C^* will shrink toward a smaller size where the n_g^* contained gas atoms can support an equilibrium bubble. The method of introducing helium also seems to make a difference: with preimplanted helium, pulsing again suppressed large cavities but the sizes of the small cavities remained only about the same as for continuous irradiation.

At lower temperatures, analogous but lesser effects are seen. For 938 K, and coimplanted helium, the small cavity group had larger mean sizes (5.2 vs 4.2 nm) and maximum sizes (8–9 vs 6 nm) for either pulsing mode compared to no pulsing, and the concentrations of the large cavities were greatly reduced (though not eliminated) by pulsing. At 843 K, pulsing effects were minimal.

It seems plausible that pulsing could act upon both of the previously mentioned mechanisms by which subcritical cavities can surpass r_C^* , i.e., size fluctuations and further gas accumulation. The beam-off annealing intervals ought to sharply curtail upward size

fluctuations after the point defect concentrations have subsided — increasingly so the higher the temperature. And because our pulsing experiments entailed a halving of the average dose rate, the helium accumulation path may have been affected because the critical number of gas atoms per cavity n_g^* is increased for lower dose rates.¹² Thus both of the general effects of pulsing found in this experiment — the increasing importance at higher temperatures and the perturbations imposed upon helium effects — may be traceable at least in part to the influence of pulsed irradiation on cavity critical sizes.

Differences for the temperature-change specimens relative to their isothermal counterparts were generally small, which is consistent with the smaller magnitude of any pulsing effects at the lower bombardment temperatures. The 10–20% decrease in mean cavity sizes may only reflect the fact that these temperature variations were more downward than upward relative to the "normal" specimens; lower average temperatures yield smaller voids. For preimplanted specimens at 843 K the number of subcritical cavities increased while the respective large cavities decreased somewhat in both sizes and numbers, suggesting a possible further impediment in critical size attainment. Disparate behavior of the two ~920–950 K specimens, one helium-free and the other coimplanted, reiterates the sensitivity to helium in this range. The reason for the elimination of observable small cavities in the temperature-change, coimplanted specimens is under investigation. If small cavities do exist in it, their maximum sizes do not exceed 3-nm diam.

There are a few other theoretical treatments which can be related to these results. Simonen's analysis,¹³ neglecting any effects of gases, predicts that effects of pulsing on void annealing ought to be greatest near the upper

temperature limit for void formation, as indeed is observed here. Ghoniem and Kulcinski¹⁴ emphasized Inertial Confinement Fusion Reactor (ICFR) ultrafast pulsing in many of their analyses, but their predicted lowering of the high temperature limit for void swelling by up to ~50 K stands in good accord with these observations. Finally, Kmetyk et al.¹⁵ calculated that the special effect of pulses should enhance void growth for irradiations below the peak swelling temperature and decrease growth at higher temperatures. This was predicated on pulsing conditions pertinent to the LAMPF 800 MeV proton beam (essentially 0.5 ms on, 8.3 ms off) and the materials modeled: pure Al at 300–400 K and pure Mo at 1200–1600 K with small (10 K) for the Al and very large (~160 K) for Mo upward temperature excursions due to beam heating. This experiment's temperature range and especially pulsing profile differ so much from theirs that the lack of correspondence is not disturbing.

Both Taylor's experiment² and those of Brimhall et al.^{3,4} investigated void growth only, in pure Ni, by commencing pulsed bombardment of a preexisting void/dislocation microstructure; helium was not a parameter. Taylor observed pulsing to increase the cavity mean diameter by 50% and to double the swelling. Brimhall found that pulsing retarded the growth of only very small cavities (consistent with the critical size consideration here) and otherwise only manifested itself by increased coarsening of cavities that arose when high temperature bombardments succeeded lower temperature preconditioning irradiation. The helium-free irradiation of a Fe-15Cr-25Ni ternary alloy by Powell and Odette⁵ using 1 MeV electrons at 873 K (pulsing periods of 2, 20, or 60 s on/off) produced significantly reduced cavity diameters and swelling. Again, though our pulsing effects are not large, they are

different from the foregoing and it seems clear that the differences might be ascribed to helium.

Kaletta's irradiation¹⁰ of vanadium with helium ions (having consequent He/dpa ratios as high as 10^4) resulted in a marked increase in cavity sizes and swelling due to pulsing. This he attributed to enhanced mobility and subsequent coalescence of small helium-vacancy clusters, made possible by additional net vacancy condensation onto the clusters during beam-off periods. Such a mechanism would also be consistent with this experiment's observation of larger and fewer cavities at the peak swelling temperature (938 K) for pulsed as compared with continuous dual-beam irradiation. The same effect could alternatively ensue from a change in the effective helium diffusion coefficient D_{He}^{eff} for pulsed irradiation — D_{He}^{eff} should acquire different values depending on whether the beam is on or off.¹⁶

Several significant pulsing effects have been observed in a commercial, Ti-modified stainless steel.⁸ Pulsing (1) clearly restricted the size of interstitial loops at 1 dpa (thereby bearing out a prediction by Ghoniem,¹⁴ (2) delayed the replacement of early-to-form fine MC precipitates by the radiation-induced massive G phase, and (3) delayed the onset of void swelling. The swelling suppression was attributed both to effects on the critical size for void growth and to be a consequence of effect (2) above. That these effects were much more marked than those of the present experiment could indicate that phase stability and its effects on swelling are more sensitive to pulsing than are the processes leading to point defect clusters in simpler materials. There are more numerous and interdependent paths of microstructural development that pulsing could influence in such engineering materials; further exploration is needed.

5. SUMMARY

The effects of pulsed heavy ion bombardment to 10 dpa in a simple austenitic alloy depend upon (1) temperature and (2) the presence and method of introduction of helium. At higher temperatures pulsing greatly inhibits the development of large cavities. At lower temperatures the influences of pulsing are small, even with the addition of substantial beam-heating-induced temperature fluctuations. Pulsing can apparently influence the sensitivity of damage microstructure to helium. These effects may be caused by pulsing raising the critical size or gas content requirements for the growth of voids from small cavities or causing shrinkage during the off periods of cavities hovering near the minimum critical radius.

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