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EFFECT OF PULSED IRRADIATION ON VOID SWELLING
IN NICKEL

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EFFECT OF PULSED IRRADIATION ON VOID SWELLING IN NICKEL

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This study has compared the void microstructure in nickel induced by a pulsed ion bombardment to that induced by a steady-state irradiation. Pulse cycles of 10 seconds on and 10 seconds off produced no measurable difference in the void growth and swelling in the temperature range 775 to 975 K compared to continuous irradiation at the same instantaneous dose rate. Void annealing during the pulse annealing period was minimal due to the large void sizes which were obtained in these irradiations. Hence no measurable effect of pulsing on void growth was observed.

1. INTRODUCTION

This experimental study has compared the microstructural damage induced by pulsed irradiation to damage induced by steady state irradiation. The pulsed operation of proposed fusion reactors dictates that the radiation damage to the first wall materials will not build up in a continuous manner. Radiation damage accumulates overtime through the interaction of radiation produced defects which results in such microstructural features as void swelling. Periodic interruptions in the irradiation can lead to alteration of the microstructural development through annealing of the defect structure or changes in the defect nucleation behavior when the radiation begins again. A limited number of experiments have shown measurable effects of pulsing on void swelling.[1] However, the results have been contradictory and a consistent picture of pulsed radiation damage has yet to emerge.

In this initial study, the effect of a pulsed irradiation on the void swelling in nickel was evaluated utilizing high energy, heavy ion bombardment. More specifically, the void growth (or shrinkage) was determined as a function of dose for both pulsed and steady-state conditions. This void growth behavior was evaluated for three different temperatures. The experimental data were then compared with kinetic behavior expected from a model based on annealing of the point defects and microstructure during the off cycle. The results provide insight into the mechanisms of void growth in a pulsed irradiation environment.

2. EXPERIMENTAL PROCEDURE

The irradiation induced void microstructures were analyzed using transmission electron microscopy (TEM). Specimens suitable for TEM, i.e., 3 mm dia by 0.1 mm thick disks, were punched from high purity nickel foil obtained from the Johnson-Matthey Co. The TEM disks were mechanically polished, electrolytically

polished and given a vacuum anneal for 2 h at 1000 K prior to ion bombardment.

The specimens were irradiated at the PNL Tandem Accelerator Facility using 5 MeV Ni^{++} ions. Ion currents were selected to give a dose rate of 5×10^{-3} dpa/s at the region of observation (0.5 μm from the surface).[2] Control specimens were irradiated to 2 dpa and 10 dpa under steady state conditions. The pulsed irradiation specimens were first continuously irradiated to 2 dpa to nucleate an established void population. The beam was then periodically interrupted so that the irradiation was 10 seconds on and 10 seconds off. The pulsing was continued until a total dose of 10 dpa was accumulated so as to study effects of pulsing on void growth. The instantaneous dose rate remained constant for both continuous and pulsed experiments. Specimens were irradiated at temperatures of 775, 900 and 975 K. During pulsed irradiation of the nickel, beam heating to some specimens caused fluctuations in the test temperature. The maximum fluctuation observed extended from 865 K to 905 K. One specimen was also irradiated at 775 K to 10 dpa in the pulsed mode with no prior continuous irradiation.

A removal of approximately 0.5 μm of the ion bombarded surface was made prior to observation in the electron microscope. Void size was measured with a calibrated eyepiece for small voids or a Zeiss particle analyzer for large voids. Foil thickness was measured by stereo techniques.

3. RESULTS

The principal result was that a pulsed irradiation, after an initial steady state irradiation, produced approximately the same microstructure as the continuous irradiation. Typical microstructures of both pulsed and continuously irradiated specimens are compared in Figure 1 for the three irradiation temperatures. The results of the TEM analysis of the irradiated nickel in terms of void size, number

1. Continuous Irradiation to 10 dpa

2. Continuous Irradiation to 2 dpa
And Pulsed Irradiation to 10 dpa

Figure 1 : Void appearance in (1) continuously irradiated or (2) pulsed irradiated nickel at (a) 775, (b) 900, and (c) 975°K. In each micrograph the foil normal is near [110].

density and volume fraction are given in Table 1. Also shown are the low and high values measured from different areas of the specimen. Several regions were measured on each specimen, and data from duplicate specimens are shown where available. Table 1 also includes data from one test where pulsing was initiated with no prior steady state irradiation. In this case, there was also no significant influence of pulsing on the microstructure. There was, however, a greater variation in the void number density from region to region. In all the tests, the swelling did increase with dose.

It should be emphasized that due to inherent problems in TEM analysis and the nature of the microstructure in irradiated nickel, small effects can be missed. At the higher radiation temperatures, there was a large variation from region to region in the void number density, particularly in the pulsed specimens. The variation was due, in part, to the void size which was comparable to the thickness of the foil examined. In addition, a considerable variation in the void parameters in nickel specimens presumably irradiated under identical steady state conditions has also been reported by Packan et al.[2]. The void sizes reported here at 10 dpa are larger than those reported by Packan et al.[2] at 2 dpa. Our results are consistent with those of Packan et al. when this difference in dose level is accounted for. As a result of these wide variations in the measured values, a rather large perturbation in the defect state would have to occur in the pulsed microstructure before meaningful differences could be detected between pulsed and continuously irradiated specimens.

4. DISCUSSION

There have been several theoretical treatments which have analyzed radiation damage evolution during pulsed irradiation.[1,4-6] From these

analyses, there appears to be two basic mechanisms that can affect void growth rate during pulsing. One is the decrease in the void growth rate due to thermal annealing during the off cycle. The other is an increase in the growth rate due to vacancies released from the dissolution of vacancy loops, i.e., collapsed depleted zones, during the off period. The first mechanism would be predominant at high temperatures and the second mechanism at lower temperatures.

The feasibility of a particular mechanism can be evaluated by calculating the rate of change of a particular defect structure during the off cycle in a pulsed experiment. The calculated change in concentration of vacancies and sizes of vacancy loops and voids as a function of annealing time is shown in Figure 2. The normalized vacancy concentration is equal to $\exp(-D_v k_v 2t)$. The void and loop dissolution rate is estimated from $d(d_v)/dt = (2D_v C_v^e/d_v) \exp(-\gamma n/kT D_v)$.

where

$$D_v = 1.9 \times 10^{-5} \exp(-1.38/KT) \text{ m}^2/\text{sec}$$

$$C_v^e = \exp(1.5) \exp(-1.39/KT)$$

$$k_v 2 = 2 \cdot d_v \rho_v$$

$$\gamma = 6.8 \times 10^{-5} \text{ ev} \cdot \text{m}^{-2}$$

$$n = 1.09 \times 10^{-29} \text{ m}^{-3}$$

The loop (depleted zone) dissolution rate is estimated by assuming an initial size of 0.8 nm. The curves indicate complete vacancy but no void annealing during the 10 second off time for the void sizes observed in this study. Schiffgins et al. reached the same conclusion, i.e., void annealing during pulsing was only pronounced for the very small voids.[6] The annealing time for vacancy loops, Figure 2,

TABLE 1. Void Size (d_v), number density (ρ) and volume fraction (%) in Irradiated Nickel

Temp., K	Condition	d_v (nm)	ρ (m^{-3})	$\Delta V/V\%$
775	Steady 2 dpa	43 to 44	$4.3-4.8 \times 10^{20}$	1.8 to 2.1
775	Steady 2 dpa	43	3.8×10^{20}	1.6
775	Steady 10 dpa	45 to 64	$1.4-5.5 \times 10^{20}$	1.9 to 4.8
775	Steady 10 dpa	36 to 60	$2.7-6.9 \times 10^{20}$	1.5 to 3.0
775	Steady 2 dpa	46 to 63	$1.7-3.2 \times 10^{20}$	1.7 to 2.2
775	+ pulse to 10 dpa			
775	Pulse 10 dpa	45 to 56	$2.3-8 \times 10^{20}$	2.1 to 3.8
900	Steady 10 dpa	138 to 200	$8.9-9.1 \times 10^{18}$	1.3 to 4.2
900	Steady 2 dpa	178 to 210	$5-17 \times 10^{18}$	2.4 to 5.3
	+ pulse to 10 dpa			
975	Steady 10 dpa	170 to 260	$5-26 \times 10^{18}$	5.1 to 11
975	Steady 2 dpa	270	3×10^{18}	3.3
	+ pulse to 10 dpa

Figure 2 : Change in vacancy concentration and size of depleted zones and voids as a function of annealing time

is less than the pulse times so a contribution to void growth could be possible from this source. Ghoniem and Kulcinski have analyzed some data of Potter et al. on pulsed bombardment of nickel and shown that the increase in void size and swelling during pulsing observed by Potter et al. can be explained by absorption of vacancies which come from the dissolution of vacancy loops.[5] If we use the values of vacancy absorption and emission reported by Ghoniem and Kulcinski and apply them to the current study, the dissolution of vacancy loops would only cause a 50 nm dia void to increase to 52 nm for a void number density of $3 \times 10^{20} \text{ m}^{-3}$. Such a change in void size would be difficult to detect within the broad range of observed void sizes. Therefore, such a phenomena might be occurring but would not be detected.

Contrary to our experimental results and those of Potter et al.[5], Powell and Odette [4] reported a decrease in void size and swelling in an Fe-Cr-Ni alloy, electron bombarded in a HVEM under pulsed conditions. There was no pre-steady irradiation to a significant void concentration before pulsing in those experiments hence void nucleation would be an important factor. The model of Powell and Odette does show a decrease in the swelling rate during pulsing.

In our study and unlike Powell and Odette, no significant microstructural changes were noted

in the pulsed nickel specimen in which there was not pre-steady-state irradiation. Void nucleation is not well understood, so there could be differences in void nucleation behavior between an Fe-Cr-Ni alloy and pure nickel which may account for the contradictory results. Void nucleation is generally quite easy in pure nickel and it is possible that during the first 10 second pulse there was already a large void concentration developed in the nickel. So even with no deliberate pre-irradiation, we may still be observing void growth effects. The rather wide variation in results from the few pulsing studies reported does indicate a need for better understanding of pulsed effects on irradiation induced microstructures.

5. CONCLUSIONS

- Void growth and swelling in nickel were not measurably altered by a pulsed irradiation (10 sec on/10 sec off) as compared to a continuous irradiation in the temperature range 775 to 975 K.
- The results are consistent with models which show the pulse time too short for significant void annealing.
- Void growth due to dissolution of vacancy loops is possible but probably not detectable within the wide scatter of the data.

REFERENCES

- [1] Ghoniem, N. M., and Kulcinski, A critical assessment of the effects of pulsed irradiation on materials, UNFDM-311, (1979).
- [2] Brimhall, J. L., and Simonen, E. P., J. Nucl. Mat. **68**, (1977) 235.
- [3] Packan, N. H., Farrell, K., and Stiegler, J. O., J. Nucl. Mat. **78**, (1978) 143.
- [4] Powell, R. W., and Odette, G. R., J. Nucl. Mat. **85** and **86**, (1979) 694.
- [5] Ghoniem, N. M., and Kulcinski, G. L., Rad. Eff., **41** (1979) 81.
- [6] Schiffgens, J. O., Graves, N. J., and Doran, D. G., in: Proc. of Conf. on Radiation Effects and Tritium Technology, Gatlinburg 1975 ed. by J. S. Watson and F. W. Wiffen, (NTIS, Springfield 1976) 1-532.

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