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R. W. Powell
G. R. Odette (UCSB)

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THE EFFECT OF PULSED HVEM IRRADIATION ON MICROSTRUCTURE EVOLUTION IN A SIMPLE Fe-Ni-Cr ALLOY*

R. W. Powell

Hanford Engineering Development Laboratory, Richland, Washington, 99352, USA

G. R. Odette

University of California-Santa Barbara, California, 93106, USA

The effect of pulsed electron irradiation on microstructure evolution was studied in a simple Fe-Ni-Cr alloy and the results compared with a theoretical model. Pulse periods of 2.5 to 60 seconds (duty factor near 50%) at 600°C significantly reduced the maximum swelling rate compared to continuous irradiation. The void concentration was observed to increase and void size and swelling rates to decrease for the pulsed cases compared to the steady irradiation. Preliminary model calculations were used to guide the experiments and in the qualitative interpretation of the results. While there are several areas of agreement with experiment, the results indicate that further development of the models is required.

1. INTRODUCTION

Unlike fission reactors, fusion reactors are likely to operate in a pulsed mode [1] leading to cyclic variations in temperature, stress and radiation damage rates at the first wall. Such effects are not easily simulated in fission reactors, where most first wall materials studies will be conducted. Nonetheless, the extent and impact of these effects on materials behavior must be understood in order to project the large body of fission reactor data to fusion reactor conditions. The initial phase of this study was directed at understanding the effect of pulsed radiation damage rates on microstructure evolution.

Cyclic variation of the displacement rate can be categorized into three basic regions relative to the lifetimes of the point defects: (1) the pulse duration is short compared to the lifetimes of both vacancies and interstitials, (2) the pulse duration is short relative to the lifetime of vacancies but long relative to the lifetime of interstitials, and (3) the pulse duration is long relative to the lifetimes of both defect types. Previous experimental studies [2,3] have been performed in region (2) while theoretical treatments have addressed various aspects of all three regions. [4-6]

The current study is being conducted primarily in region (3), where tokamak fusion reactors would be expected to operate. [1] The effects on void formation of a range of pulse periods and duty factors is studied with electron irradiations and qualitatively compared with theoretical calculations.

2. THEORY

Analysis of the experimental results on microstructure evolution during pulsed irradiation is assisted by comparison with computer models. The interrelationship between dislocation loops, network dislocations and voids was treated previously for unpulsed irradiation conditions. [7] Both nucleation of defect aggregates and growth of the aggregates were treated with the model. A typical plot of the calculated fluence dependence of the various microstructural entities is shown in Figure 1 for an unpulsed irradiation.

In another study reported previously, the effect of pulsed irradiation on the nucleation of voids was considered. [6] Results of that study indicated that under some conditions there may be a significant reduction in the void nucleation rate for pulsed compared to the unpulsed conditions. The dominant mechanism for this

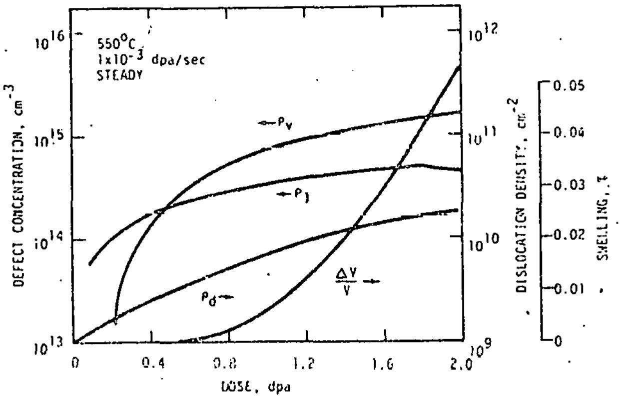

FIGURE 1. Typical Dose Dependence in Low Dose Region as Predicted by the Model.

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reduction was the annealing of pre-critical vacancy cluster embryos between pulses. Further, it was found that the results could be expressed as a quasi-analytical relationship between the throttling (reduction) of the nucleation rate and the pulse-on and pulse-off times normalized to the relaxation times for void nuclei build-up and decay. By using this relationship, it was possible to predict the throttling of the void nucleation rate for given pulsing parameters without going through the full computer calculations.

For the present study, the microstructure evolution model was modified to accept pulsed irradiation conditions, and the correlation for the nucleation rate throttling was incorporated into the void nucleation calculation routine. Additionally, the model treated void and dislocation loop growth (or shrinkage) rates during both beam-on and -off conditions.

Figure 2 illustrates the calculated microstructural evolution for a particular set of pulsed irradiation conditions. Comparison with the calculated evolution for similar unpulsed irradiation conditions shown in Figure 1 illustrates the significant effect pulsed irradiation is expected to have on the damage microstructure. The initial void nucleation rate is depressed many orders of magnitude by the pulsed radiation conditions chosen for Figure 2 but the final void concentration is less than one order of magnitude lower. The explanation for this is that the throttling of the nucleation rate declines as the microstructure evolves, resulting in a burst of nucleation at very nearly the same peak nucleation rate as for the unpulsed conditions. Identical calculations to those used for Figure 2, except that no nucleation rate throttling was applied, resulted in very little difference between the pulsed and unpulsed conditions. This demonstrates that the difference between Figures 1 and 2 is due to the nucleation rate throttling and not due to a change in the average displacement rate.

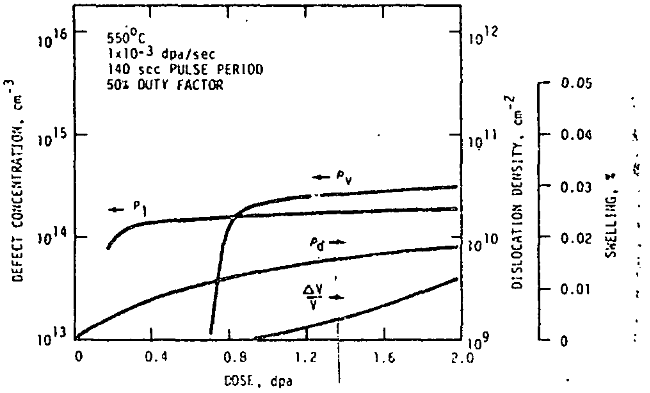

FIGURE 2. Typical Dose Dependence in the Low Dose Region for Pulsed Conditions. Other Parameters are identical to Figure 1.

The range of application of the combined model is determined by the simplifying assumptions made in its development. Point defect concentrations were assumed to react instantaneously to changes in the displacement rate. This is a valid approximation only if the pulse times are long compared to the time to achieve steady-state point defect concentrations. For pulse times significantly shorter than this, the vacancy concentration will oscillate about some average value. Under such conditions, the influence of pulsing on void nucleation rates is expected to be quite small since little embryo decay will occur during beam-off times.

Since the model is only approximate and may not yet contain all relevant mechanisms, it was used only to guide the experimental work of this study and to aid the qualitative interpretation of the experimental results. The model will be refined, however, and experiments will be used to test its validity, and as tools to calibrate model parameters.

3. EXPERIMENTAL PROCEDURES

A simple Fe-Ni-Cr alloy, designated E20, of nominal composition 15 wt.% Cr and 25 wt.% Ni, was used throughout this portion of the study. This simple alloy was chosen to eliminate as many variables as possible and still maintain a strong tie to technological materials of direct interest to the U. S. Fusion Materials Program.

Electron irradiations were performed at 1 MeV in a JEOL JEM-1000 using a double tilting goniometer heating stage. Pulsing of the electron beam was accomplished by deflection with two pairs of electrostatic deflector plates alternately charged and discharged with square wave voltage pulse generators. Rise and fall times of the voltage pulse were less than 1 msec and the positional stability of the electron beam was excellent.

An irradiation temperature of 600°C (including heating due to the electron beam) was used for all irradiations. Peak displacement rate for all pulsed and steady-state irradiations was 1.3×10^{-3} dpa/sec (40 barn cross section). Thus, assuming that the time during the beam-on condition was sufficient to achieve quasi-steady-state defect concentrations, there was no effective temperature shift due to differences in displacement rates [9] among any of the irradiations. A typical total dose of 4 dpa was employed which corresponds to 1 hour of steady irradiation or approximately 2 hours of pulsed irradiation.

A relatively low displacement rate was used in order to minimize the influence of temperature pulses on the results. The temperature rise due to the electron beam was less than 10°C as measured by the order-disorder transition in Fe_3Al [10].

Pulse periods of 2.5 seconds to 60 seconds with 40 to 60% duty factors (ratio of pulse-on time to pulse period) were employed in this investigation.

4. RESULTS

The dose dependence of swelling for unpulsed and pulsed conditions are shown in Figures 3 and 4. The swelling rates and void parameters for all the irradiation conditions studied in this experiment are given in Table 1.

These data indicate that: 1) the swelling rate decreases from unpulsed to pulsed conditions, 2) the swelling rate decreases with decreasing duty factor or increasing pulse period as shown in Figure 5, 3) the major effect of pulsing on swelling is due to a significant reduction in void size, 4) the void concentrations increase in going from unpulsed to pulsed conditions, and 5) pulsed irradiations appear to approach steady-state swelling more quickly than unpulsed irradiations.

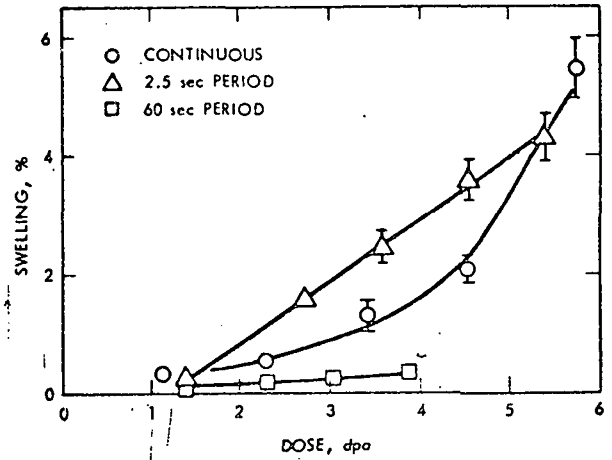

FIGURE 3. Fluence Dependence of Swelling for the Continuous Irradiation, the Shortest Pulse Period Irradiation and the Longest Pulse Period Irradiation.

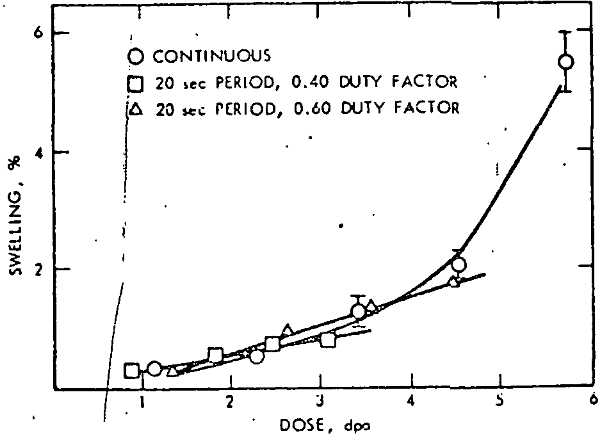

FIGURE 4. Fluence Dependence of Swelling for the Continuous Irradiation and Two Pulsed Irradiations With Equal Pulse Periods but Different Duty Factors.

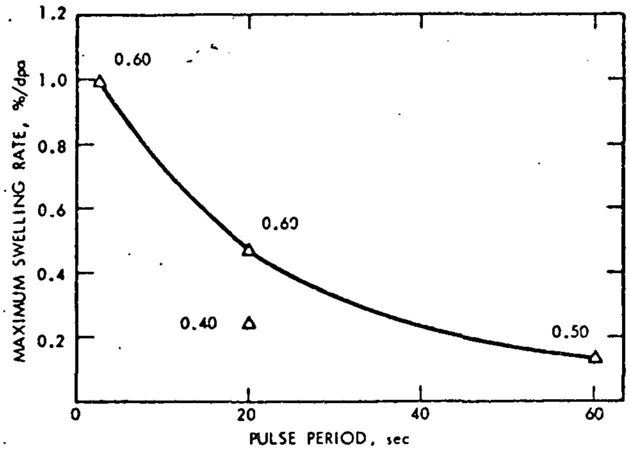

FIGURE 5. The Effect of Pulse Period on the Maximum Swelling Rate. Duty factors are as shown for each irradiation. For comparison, the maximum swelling rate for the continuous irradiation was 2.4%/dpa.

5. DISCUSSION

The major experimental result is that pulsed conditions in HVEM irradiations produced a significant decrease in swelling rates compared to unpulsed conditions. This is qualitatively consistent with theoretical considerations and the model predictions (see Figures 1 and 2). Further, the swelling rate decreased with increasing pulse period; the current model does not predict a decrease in this case.

The increased swelling rate at the lowest pulse period of 2.5 seconds compared to the swelling rates of the other pulsed irradiations can be partially rationalized if the vacancy concentration does not decay to thermal values between pulses as discussed in Section 2. Neglecting recombination, the time t for the vacancy concentration, initially at C_v, to reach thermal equilibrium C_v^{th} can be estimated as $t \simeq \frac{1}{D_v k_v^2} \ln \frac{C_v}{C_v^{th}}$, where D_v is the vacancy diffusion coefficient and k_v^2 the total vacancy sink strength. Assuming a vacancy migration energy of 1.5 eV, $C_v/C_v^{th} = 1000$ and a total dislocation density ranging from 1×10^9 to 1×10^{10} cm^{-2}, the time t ranges from 3 to .3 seconds. Hence, the experimental observations are qualitatively consistent with a theoretical minimum pulsing period below which microstructural evolution would not be influenced by time variations in vacancy concentrations. Note that this interpretation does not necessarily include all important transient effects. However, time averaged vacancy concentrations would be appropriate for use in rate theory models below this minimum pulse period.

The apparent increase in void concentrations in going from unpulsed to pulsed irradiations appears to contradict theoretical indications

TABLE 1

VOID STATISTICS FOR ELECTRON IRRADIATIONS

Pulse Period (sec)	Duty Factor	Maximum Void Concentration (cm^{-3})	Average Void Diameter at 3.5 dpa	Maximum Swelling Rate
Steady	1.0	1.1×10^{15}	29 nm	2.4%/dpa
2.5	0.60	2.9×10^{15}	26 nm	1.0%/dpa
20	0.60	4.0×10^{15}	21 nm	0.48%/dpa
20	0.40	3.7×10^{15}	17 nm	0.25%/dpa
60	0.50	3.0×10^{15}	12 nm	0.13%/dpa

that void nucleation rates can be reduced significantly by pulsing in some cases. This, however, illustrates the danger of simplistic interpretation of incomplete models. In the first place, the instantaneous nucleation rate for a particular set of conditions does not define the overall course of microstructural evolution as clearly illustrated in the model calculations in Section 2. There, it was shown that calculated final void densities differ by less than an order of magnitude for pulsed relative to unpulsed conditions in spite of a large initial reduction in nucleation rates in the former case. Further, these model calculations indicated that pulsing also influences the evolution of the dislocation structure, leading to a burst of void nucleation and an early onset of linear swelling which is also qualitatively consistent with observations. Secondly, the current model is obviously approximate in its present form since transient effects of defect concentrations are not treated. Hence, one possible interpretation of the data is that transient effects are significant for the experimental conditions in this study even if defect concentrations eventually approach equilibrium values between pulses. Therefore, the model will be extended to treat void nucleation between pulses when vacancy supersaturations remain large for some time after the interstitial transient has died away.

It should be emphasized that a meaningful quantitative comparison of models and experiment requires consideration of numerous combinations of mechanisms and model parameters. Further, the results of this study illustrate the need for a marriage of theory and experiment, and an iterative process of experiment and model development. Perhaps the most significant aspect of this work is the indication that pulsing introduces a new, independent, and highly controllable irradiation parameter which can be used to study damage mechanisms and to define damage model parameters.

6. CONCLUSIONS

This investigation has so far demonstrated the following major points concerning pulsed HVEM irradiation of a simple Fe-Ni-Cr alloy at 600°C.

1. Pulsed HVEM irradiation produced a significant decrease in swelling rate compared to continuous irradiation results.

2. The observed decrease in swelling rate was accompanied by an increase in void concentration and a decrease in void size.

3. Within the pulsing conditions employed, the swelling rate decreased with increasing pulse period or decreasing duty factor.

4. Analysis with a preliminary theoretical model of microstructure evolution indicated that the pulsing conditions employed should affect void densities, swelling rate, and the time of significant void nucleation. The discrepancy between model predictions of void density trends and experimental observations suggests that transients in the defect concentrations are important.

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