INVESTIGATION OF THE SYNERGISTIC INFLUENCE OF IRRADIATION TEMPERATURE AND ATOMIC DISPLACEMENT RATE ON THE MICROSTRUCTURAL EVOLUTION OF ION-IRRADIATED MODEL AUSTENITIC ALLOY Fe-15Cr-16Ni – T. Okita, T. Iwai and N. Sekimura (Tokyo University), F. A. Garner (Pacific Northwest National Laboratory)*

OBJECTIVE

The purpose of this effort is to determine the influence of dpa rate and irradiation temperature on void swelling of fcc alloys.

SUMMARY

A comprehensive experimental investigation of microstructural evolution has been conducted on Fe-15Cr-16Ni irradiated with 4.0 MeV nickel ions in the High Fluence Irradiation Facility of the University of Tokyo. Irradiations proceeded to dose levels ranging from ~0.2 to ~26 dpa at temperatures of 300, 400 and 500°C at displacement rates of 1 x 10^{-4}, 4 x 10^{-4} and 1 x 10^{-3} dpa/sec. This experiment is one of two companion experiments directed toward the study of the dependence of void swelling on displacement rate. The other experiment proceeded at seven different but lower dpa rates in FFTF-MOTA at ~400°C. In both experiments the swelling was found at every irradiation condition studied to monotonically increase with decreases in dpa rate.

The microstructural evolution under ion irradiation was found to be very sensitive to the displacement rate at all three temperatures. The earliest and most sensitive component of microstructure to both temperature and especially displacement rate was found to be the Frank loops. The second most sensitive component was found to be the void microstructure, which co-evolves with the loop and dislocation microstructure. These data support the prediction that void swelling will probably be higher in lower-flux fusion devices and PWRs at a given irradiation temperature when compared to irradiations conducted at higher dpa rates in fast reactors.

INTRODUCTION

It has recently come to the attention of the light water reactor community that void swelling is probably occurring in the austenitic components of pressure vessel internals, especially in pressurized water reactors (1). Similar concerns have been raised for fusion devices, especially those that will experience lower atomic displacement rates than found in fast reactors. Questions exist however, concerning the magnitude of swelling that might occur and what might be the effect of the major irradiation variables. These variables are the irradiation temperature, the rate of atomic displacements in dpa/sec and the generation rates of helium and hydrogen gas. A number of comprehensive irradiation experiments are in progress to quantify the answers to these questions and to provide microstructural data that will allow extrapolation to and prediction of swelling under conditions of lower dpa rate.

The experiment described in this paper is one of two companion experiments directed toward the study of the dependence of void swelling on displacement rate in particular, with additional information to be gained on the action of the other two variables. The other experiment proceeded at seven different but lower neutron-induced dpa rates in FFTF-

* Pacific Northwest National Laboratory (PNNL) is operated for the U.S. Department of Energy by Battelle Memorial Institute under contract DE-AC06-76RLO-1830.

MOTA at ~400°C (2). This range of dpa rates spanned the full set of dpa rates relevant to fusion devices, PWRs and fast reactors. In the neutron experiment the swelling was found at every irradiation condition studied to monotonically increase with decreases in dpa rate.

With some limitations the neutron experiment explored the effect of helium generation but it was conducted at only one irradiation temperature, 400°C, which lies just above or at the higher end of the PWR range of interest. The current experiment must of necessity proceed at dpa rates that or higher than that explored in fast rectors, but is capable of studying temperatures from the lower limit of fusion or PWR interest, 300°C and higher. The higher temperatures are necessary to explore the interaction of the two most important variables, that of dpa rate and temperature.

The on-going ion experiment is very large in scope, involving three alloys and simultaneous injection of helium and in some cases, helium and hydrogen. In the current paper we focus only on one alloy, the ternary Fe-15Cr-16Ni with very low levels of other solutes, irradiated without gas injection in the annealed condition.

Experimental Details

The model austenitic alloy, ternary Fe-15Cr-16Ni was prepared by arc melting from very pure Fe, Ni and Cr. The alloy was rolled to sheets of 0.2 mm thickness. Afterward, standard 3 mm microscopy disks were then punched and annealed for 30 minutes at 1050°C in a very high vacuum. The specimens used in the two companion experiments were identical in every way.

Irradiation proceeded with 4.0 MeV Ni^{3+} ions in the High Fluence Irradiation Facility operated by the University of Tokyo and located at Tokai-Mura, Japan. In this first series of irradiations no gas atoms were preinjected or simultaneously injected.

The irradiation conditions chosen are listed in Table 1. The damage vs. depth profile was calculated using the TRIM code (3) and the results are shown in Figure 1. After irradiation the specimens were electrochemically thinned to reach a depth of 600 to 700 nm, a depth chosen to avoid a strong influence of the “injected interstitial” effect (4) and also minimize the influence of the specimen surface. Analysis was conducted using a JEOL 200CX electron microscope operating at 200 KeV.

Results

In general, the radiation-induced microstructures were dense, especially at the lower temperatures, but rather simple, being comprised primarily of Frank interstitial loops, some unfaulted perfect loops, some network dislocations and faceted voids.

As shown in Figure 2, the swelling of Fe-15Cr-16Ni increases as the atomic displacement rate is lowered at all three irradiation temperatures studied. While swelling appears to increase with increasing temperature, it is significant to note that swelling occurs at 300°C, the lowest temperature relevant to PWR internals.

Figure 3, shows that there is a tendency for the cavity density to increase somewhat as the dpa rate decreases, although the cavity density also increases as the irradiation temperature falls. The Frank loop density also increases with decreasing temperature and decreasing dpa rate, as shown in Figure 4.

Table 1. Irradiation conditions studied to date

Temperature	Dose Rate	Dose
300°C 400°C 500°C	1x10^{-4} dpa/sec 4x10^{-4} dpa/sec 1x10^{-3} dpa/sec	0.17-0.21 dpa 1.6-1.9 dpa
300°C 400°C 500°C	4x10^{-4} dpa/sec	15.2-17.4 dpa
300°C	4x10^{-4} dpa/sec	26.2 dpa

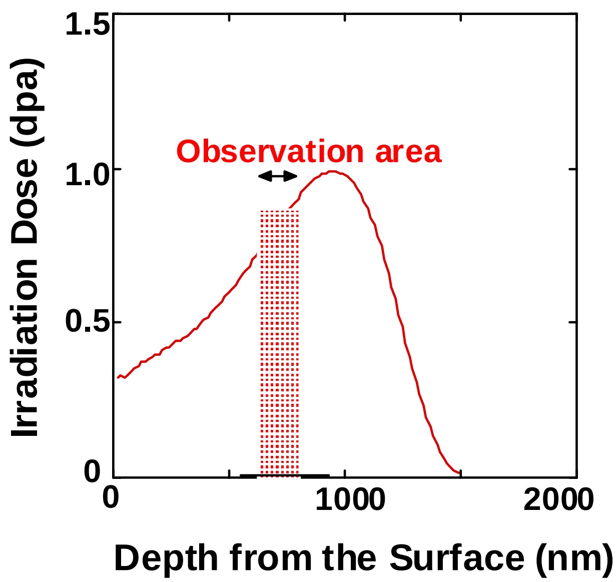

Figure 1. Depth distribution of ion-induced damage as calculated by the TRIM code, showing range of depths chosen for observation by microscopy.

<u>Discussion</u>

The most significant observation from this study is that swelling can occur at temperatures as low as 300°C, even in the absence of injected gases. This result appears to agree with the results of Porollo et al. (5) and Neustroev et al. (6). The second important observation is that void swelling is accelerated at lower dpa rates at every temperature studied, similar to the results of the companion irradiation study using fast neutrons.

When comparing the results of the ion and neutron studies (2) it becomes apparent that the level of swelling obtained at a given dpa level at the very high ion-induced dpa rates is much less than that obtained at lower neutron-induced dpa rates, confirming that dpa rate effects appear to be very consistent. It must also be recognized, however, that ion-induced swelling tends to be additionally retarded by the combined influence of the surface and the injected interstitial.

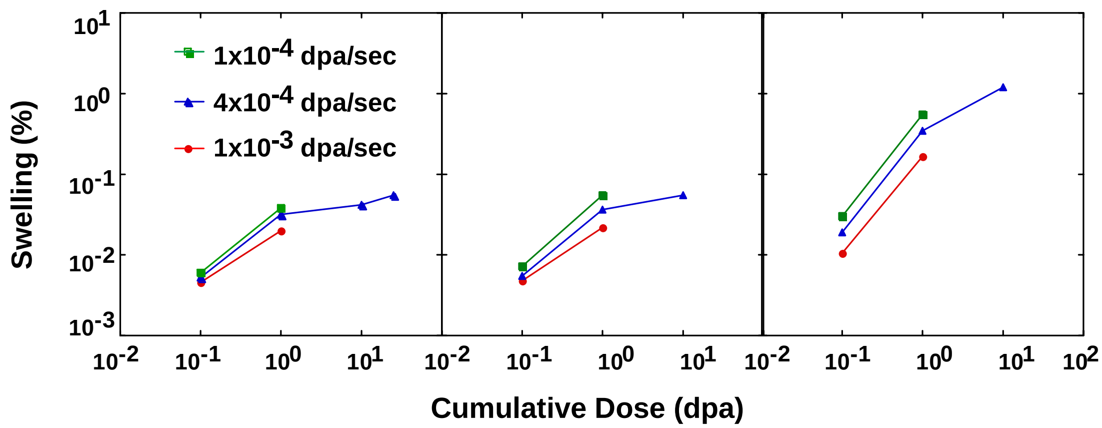

Figure 2. Dependence of swelling of annealed Fe-15Cr-16Ni on temperature, dpa and dpa rate. The irradiation temperatures are 300, 400 and 500°C, moving from left to right.

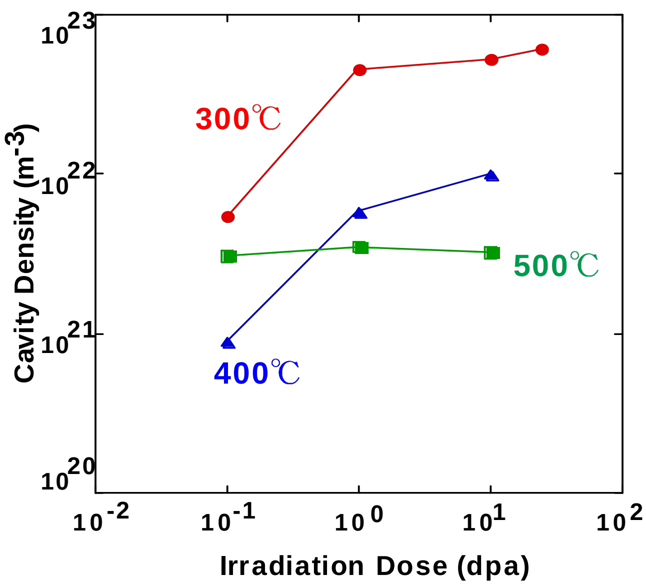

Figure 3. Dependence of cavity density of annealed Fe-15Cr-16Ni on temperature and dpa at $4x10^{-4}$ dpa/sec.

The swelling levels attained in the ion experiment have not yet reached large enough levels where it can be determined if changes in dpa rate are expressed primarily in the duration of the transient regime of swelling, as clearly observed in the neutron experiment. However, the ion experiments allow us the opportunity to test the conclusion of the neutron study

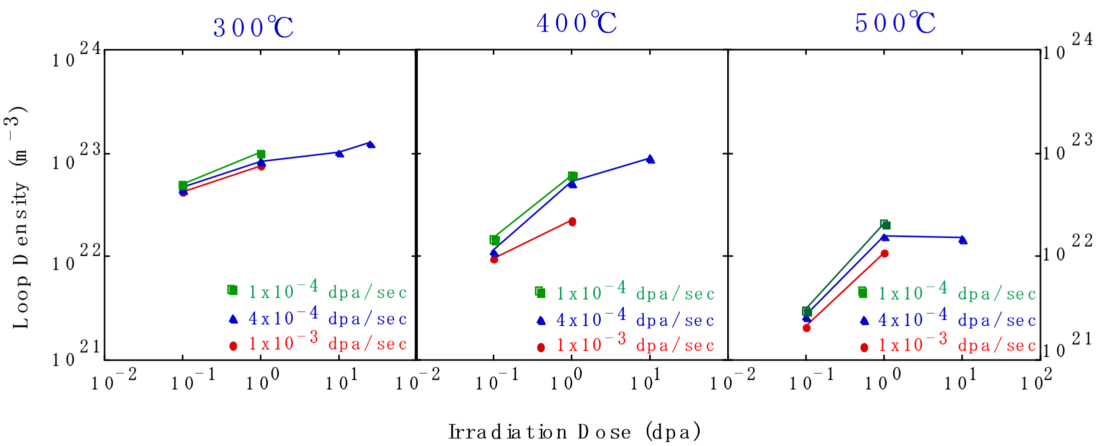

Figure 4. Dependence of loop density of annealed Fe-15Cr-16Ni on temperature, dpa and dpa rate.

concerning the origin of the transient shift. In the neutron study it appeared that the primary flux sensitivity was expressed in the evolution of the Frank loop population and its subsequent unfaulting. At all the neutron-induced dpa rates studied, however, most of the loops had already unfaulted and network formation was well in progress at even the lowest doses studied. In the ion experiment the specimens at lowest doses still retained most of the Frank loops.

As shown in Figure 5, however, it appears that the saturation loop population is temperature-dependent and also scales with the dpa rate to the ½ power, as predicted by the model of Watanabe et al. (7) and Muroga and co-workers (8). Figures 6 and 7 shows the evolution of loop evolution in simple austenitic Fe-Cr-Ni model alloys observed by Watanabe, indicating the strong role of temperature and dpa rate to determine the saturation density of Frank loops. Figure 8 shows the measured dependence of the saturation density as measured by Muroga in these same alloys.

A detailed model is now being constructed that incorporates the combined observation of the two companion studies. The model will describe how the microstructural evolution under both neutron and ion irradiation was found to be very sensitive to the displacement rate at all three temperatures. The earliest and most sensitive component of microstructure to both temperature and especially displacement rate was found to be the Frank loops, whose rate of unfaulting determines when the dislocation network starts to evolve. The second most sensitive component was found to be the void microstructure, which co-evolves with the loop and dislocation microstructure.

The data from these two companion experiments are very consistent and support the prediction that void swelling will probably be higher in lower-flux PWRs and fusion devices at a given irradiation temperature and dpa level when compared to irradiations conducted at higher dpa rates in fast reactors.

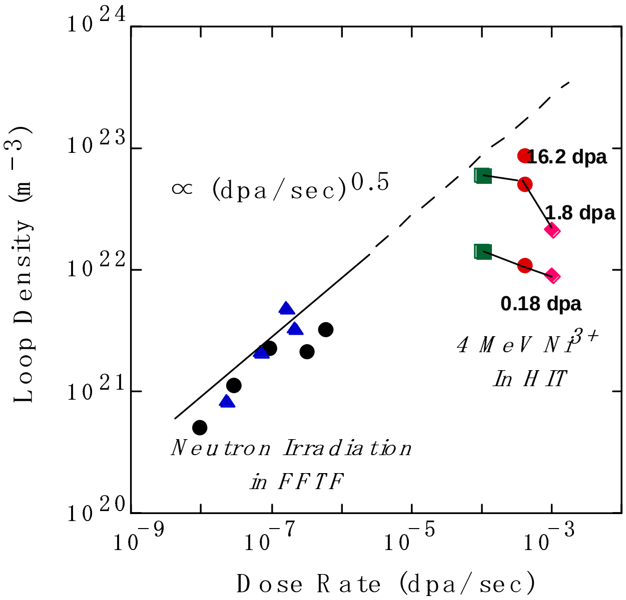

Figure 5. Dependence of loop density on dpa rate to the 1/2 power, as observed in this experiment and the companion neutron experiment in FFTF. Note that the ion-induced loop density moves toward the (dpa/sec)$^{1/2}$ trend line with increasing dpa, indicating that ion-induced loop saturation requires relatively high dpa levels.

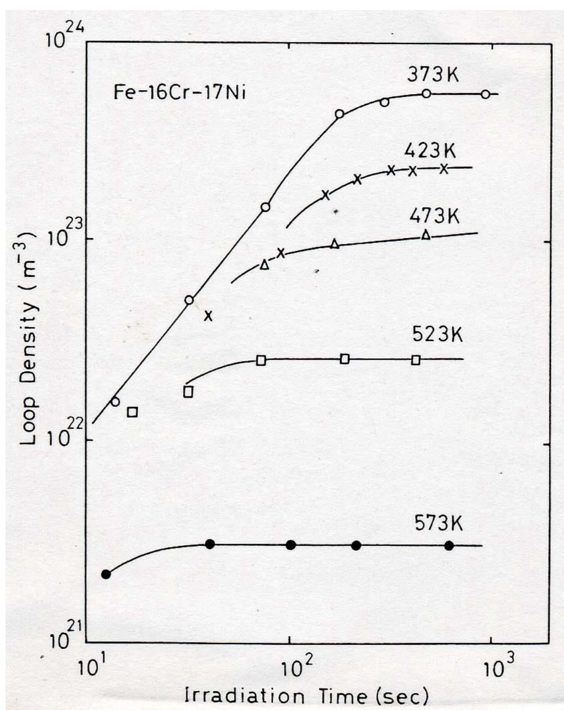

Figure 6. Development of Frank loop density vs. irradiation time and temperature, as observed during electron irradiation of simple ternary austenitic alloys by Watanabe and co-workers (7).

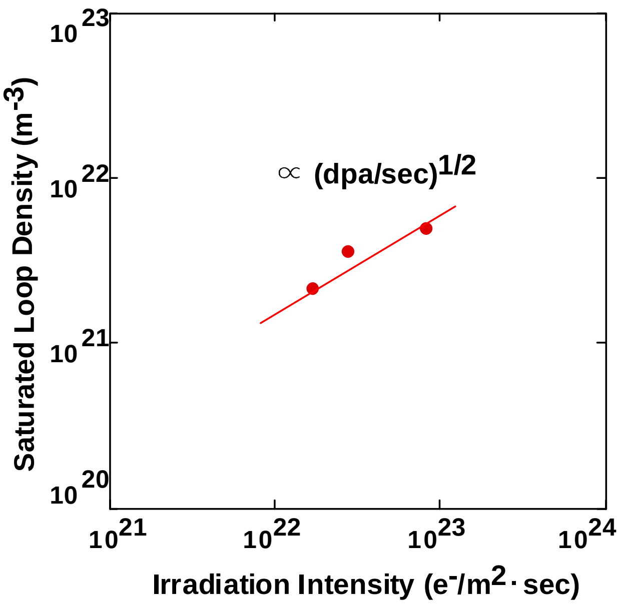

Figure 7. Flux dependence of Frank loop density vs. dpa rate, as observed by Watanabe and co-workers (7).

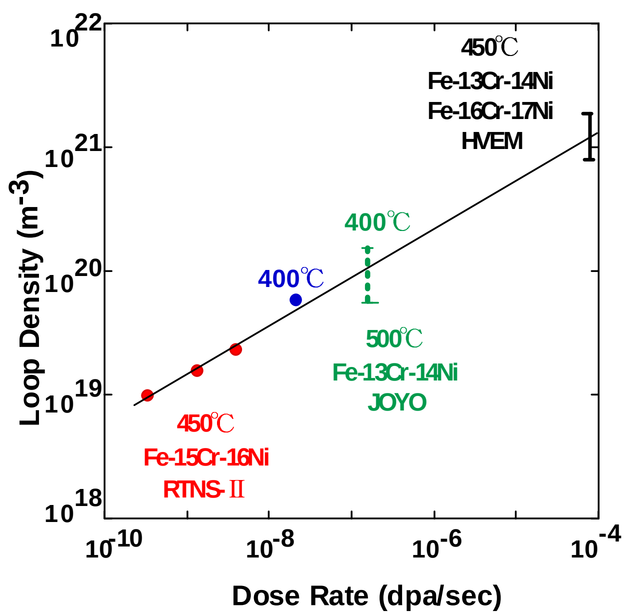

Figure 8. Dose rate dependence of saturated loop density in simple ternary austenitic alloys, as observed by Muroga and co-workers (8).

ACKNOWLEDGEMENTS

This work was supported by Monbusho, the Japanese Ministry of Education, Science and Culture under the FFTF-MOTA collaboration and the JUPITER program (Japan-USA Program for Irradiation Testing for Fusion Research), and the U.S. Department of Energy, Office of Fusion Energy, and Office of Basic Energy Sciences, under Contract DE-AC06-76RLO 1830. Pacific Northwest National Laboratory is operated for DOE by Battelle Memorial Institute.

REFERENCES

[1] F. A. Garner, L. R. Greenwood, and D. L. Harrod, "Potential High Fluence Response of Pressure Vessel Internals Constructed from Austenitic Stainless Steels", Proc. Sixth Intern. Symp. on Environmental Degradation of Materials in Nuclear Power Systems - Water Reactors, San Diego, CA. August 1-5, 1993, p. 783-790.

[2] T. Okita, N. Sekimura, F. A. Garner, L. R. Greenwood, W. G. Wolfer, and Y. Isobe, in this semi-annual report.

[3] J. P. Biersack and L. G. Haggmark, Nucl. Instr. And Meth. 174 (1980) 257.

[4] F. A. Garner, "Impact of the Injected Interstitial on the Correlation of Charged Particle and Neutron-Induced Radiation Damage", J. of Nucl. Mater. 117, (1983), p. 177-197.

[5] S. I. Porollo, Yu. V. Konobeev, A. M. Dvoriashin, V. M. Krigan, and F. A. Garner, “Determination of the Lower Temperature Limit of Void Swelling of Stainless Steels at PWR-relevant Displacement Rates”, Proc. of Tenth Inter. Conf. on Environmental Degradation of Materials in Nuclear Power Systems – Water Reactors, August 5-9, 2001, in press.

[6] V. S. Neustroev, V. N. Golovanov, Z. Ye. Ostrovsky, A. M. Pecherin, and V. K. Shamardin, “Radiation Effects in 18Cr-10Ni-Ti Steel Irradiated in Different Reactors under Conditions Close to VKU VVER Reactors Operating Conditions”, Proceedings of the Fourth International Ural Seminar on Radiation Damage Physics of Metals and Alloys, Feb. 25 – Mar. 3, 2001. Snezhinsk, Russia, in press.

[7] H. Watanabe et al., J. of Nucl. Mater. 155-157 (1988) 815.

[8] T. Muroga, H. Watanabe and N. Yoshida, “Correlation of Fast Neutron and Electron Irradiations Based on the Dislocation Loop Density”, J. of Nucl Mater. 174 (1990) 282-288.