10.2 A HIGH ENERGY SELF-ION IRRADIATION STUDY OF METALLIC REACTOR MATERIALS AT THE LLNL CENTER FOR ACCELERATOR MASS SPECTROMETRY (CAMS) —

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OBJECTIVE

The objective of the Lawrence Livermore National Laboratory (LLNL) Center for Mass Spectrometry (CAMS) experiment is to characterize the combined effects of charged particle ion versus neutron irradiations, including the effects of dpa and dpa rate, on an extensive matrix of materials for various reactor applications.

SUMMARY

The LLNL CAMS accelerator experiment was designed to irradiate a range of reactor materials (~80 samples per condition) with 70 MeV Fe^{+9} ions to a dose of 3 dpa at two dpa rates that differ by a factor of 10. The experiment also explores ion versus neutron irradiation effects by cross-comparing the CAMS results with previous neutron and charged particle irradiation (CPI) data for the same alloy. Following a prototype test on 10/8/15, the actual CAMS irradiation was successfully conducted beginning on 12/3/2015. Extensive PIE is planned for the FeCr alloys, NFA, TMS and other fusion reactor relevant alloys, including W. One objective is to explore the concept of using precipitation as a damage meter to evaluate the relative effects of neutron irradiations versus CPI; and to isolate the effects of dpa rate from that of the irradiating particle. Here some preliminary ion versus neutron results are reported.

PROGRESS AND STATUS

Introduction

The CAMS facility of LLNL houses a tandem accelerator that can be used for high-energy ion irradiations. The 70 MeV Fe^{+9} ions used in this study have a peak dpa range of ≈ 6.4 µm, as illustrated in Figure 1a, that is based on the SRIM code. In this case the total ion current produced a peak K&P dose of 3 dpa. However, the dpa varies widely over the ion range due to an increasing fraction of nuclear versus electronic ion energy loss rates. Thus a single irradiation can explore almost an entire dose (dpa)-response (hardening, loop formation, segregation, solute clustering and precipitation) behavior space. However, direct interpretation of such data is confounded by the simultaneous variation in the dose rate. Thus the initial experiment was conducted at two different dpa rates that vary by a factor of 10 as illustrated in Figure 1b.

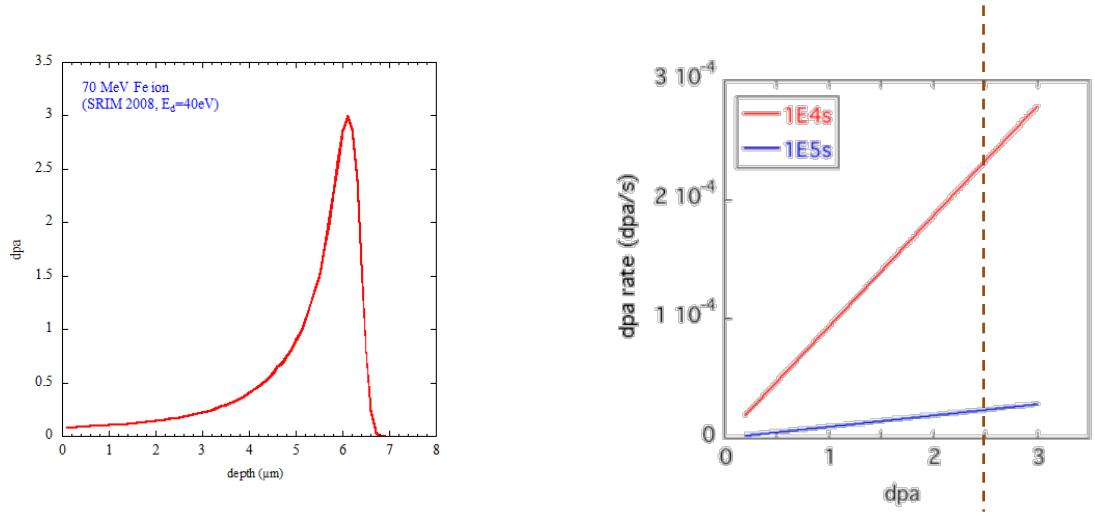

Figure 1. a) The dpa-depth profile in steel for 70MeV Fe ion calculated using SRIM 2008 with E_d = 40 eV and the KP model; b) two dose rate regimes that vary by a factor of 10.

Experimental Procedure

CAMS Irradiations

Ion beam irradiations are expensive (but trivial compared to neutrons), so maximizing the number of specimens is very important. Thus we developed and implemented a new target design to irradiate a large numbers of specimens while improving both the mechanical durability and heat transfer performance of the targets. The design is illustrated in Figures 2a and b. The holder contains 80 2x0.5x8 mm bars clamped by plates loaded by setscrews in a thermally conductive and highly rigid bundle of specimens in a pocket in the copper target assembly. The top surfaces of the specimens are polished together to finished amenable to FIB lift-outs for APT and TEM. The sides of the bars can also be polished without loss of irradiated material and used for measuring nano-hardness profiles. The specimens can also be used for EBSD as well as grazing incidence SAXS and XRD.

a b

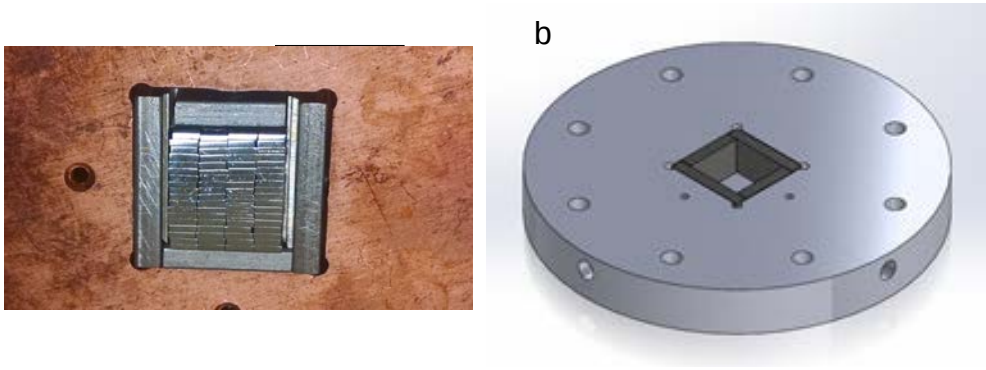

Figure 2. Sample holders used in CAMS experiment: a) Polished specimen array with ~80 4x0.5x8mm sample bars; b) a rendering of the bolt on copper target assembly developed by UCSB for use in the CAMS experiment

A prototype irradiation was successfully carried out on 10/8/15 to test the CAMS ion beam capabilities wand the new sample holder. Target irradiation conditions were achieved, at different dpa rates that varied by a factor of 10. The actual irradiations were carried out at 3 and 30 hours to reach a peak dose of 3 dpa on two identical targets starting on 12/3/15. LLNL scientists (S. Tumey, T. Brown) ran the irradiation with the help of a UCSB graduate student (N. Almirall), including calibrating the heaters, thermocouples, infared camera and ion beam distribution on target. The temperature of the sample was

held at 295±5°C provided by three cartridge heaters and controlled by an active air cooling system under a high vacuum of 10^{-7} torr. An infared camera was used to monitor the temperature and temperature fluctuations on the specimen surface over the entire experiment. Thermocouples were attached to the back of the target holder as well as in direct contact with the specimen bundle. There were no significant variations in temperature or vacuum over the entire irradiation.

The 70MeV ion beam using Fe^{+9} ions was aligned on target and tuned to obtain the desired gaussian beam envelope with a FWHM of 8 mm. The dpa profile was calculated using SRIM code for a peak K&P dose of 3 dpa There was a maximum of ≈ 5°C of beam heating, that was accounted for by the active cooling system. The total beam current and current spatial distrubtion was measured in front of the target by a multi-pin channel array faraday cup that will be translated to detailed dose and dose rate distributions on all individual samples. Based on these readings, relatively minor adjustments were carried out during the experiment.

Materials and Characterization Method

The alloy matrix consists of NFA/ODS, TMS, Fe-3 to 18Cr binary, RPV and maraging Fe-based alloys, tungsten and nickel alloys. CAMS alloys directly relevant to fusion materials, include: TMS Eur97, F82H IEA and M3, HT9 and NF616; NFA/ODS PM2, NFA-1, US and French MA957, MA956, PM2000; tungsten; and, model Fe 3-18%Cr binaries. FIB lift outs at selected depths were used for APT to characterize the high Ni alloy precipitates.

Results

A preliminary APT study of high Ni (3.5wt%) low alloy steels irradiated at the higher CPI dose rate found no precipitation at a depth of 1.3 µm, and a volume fractions (f_{ppt}) of f_{ppt} ≈ 0.51% and 1.3% of a Mn-Ni-Si intermetallic G-phase type precipitates at 3.3 and 5.5 µm, respectively. This alloy had previously been CPI under generally similar conditions in the DuET facility to 4.5 dpa at 290°C, that produced fppt ≈ 4.6% of precipitation. A similar alloy was irradiated in ATR-2 to ≈ 0.2 dpa producing f_{ppt} ≈ 2.6% precipitation. Correcting f_{ppt} to 2.1% to account for a lower Si content in this alloy yields an f_{ppt} ≈ 2.1%. Assigning crude and very preliminary dpa estimates for the CAMS irradiation, that will be refined in the near future, leads to the plot of f versus √dpa shown in Figure 3. These results suggest that the CPI dpa are ≈ 8.1 times less efficient in producing precipitation, primarily by radiation-enhanced diffusion, than fission reactor neutrons. This result is generally consistent with previous observations suggesting that an effective dpa (dpa_e) dose can be simply defined based on scaling dpa with relative dpa rates (dpa') as $dpa_e = dpa(dpa'_r/dpa')^p$ where dpa'_r is an arbitrary reference dpa rate. The p ≈ 0.23 observed in this preliminary, is consistent with previous estimates of p = 0.15 to 0.25.

Future Work

The PIE on FeCr alloys, NFA and other fusion alloys is planned in the near future. This microstructural characterization will be carried out primarily using APT, TEM and SAXS/XRD. Nanohardness as a function of depth, or dpa, profiles will be carried out at UC Berkeley.

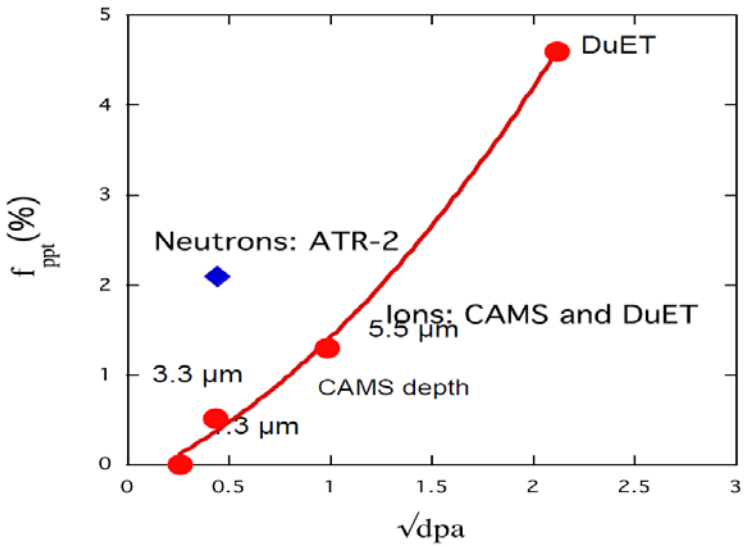

Figure 3. A comparison of the volume fraction (f_{ppt}) of Mn-Ni-Si precipitates formed under neutron (blue diamond) and CPI (red circles) irradiations of a 3.5%Ni low alloy steel as a function of √dpa.

Acknowledgments

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