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BEAM-BROADENING EFFECTS IN STEM/EDS MEASUREMENT OF RADIATION-INDUCED SEGREGATION IN HIGH-PURITY 304L STAINLESS STEEL*

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DISCLAIMER

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Radiation-induced segregation (RIS) is the spatial redistribution of elements at defect sinks such as grain boundaries and free surfaces during irradiation. This phenomenon has been studied in a wide variety of alloys and has been linked to irradiation-assisted stress corrosion cracking (IASCC) of nuclear reactor core components[1]. Therefore, accurate determination of the grain boundary composition is important in understanding its effects on environmental cracking. Radiation-induced segregation profiles are routinely measured by scanning-transmission electron microscopy using energy-dispersive X-ray spectroscopy (STEM-EDS) and Auger electron spectroscopy (AES). Because of the narrow width of the segregation profile (typically less than 10 -nm full width at half-maximum), the accuracy of grain boundary concentration measurements using STEM/EDS depends on the characteristics of the analyzing instrument, specifically, the excited volume in which x-rays are generated. This excited volume is determined by both electron beam diameter and the primary electron beam energy. Increasing the primary beam energy in STEM/EDS produces greater measured grain boundary segregation, as the reduced electron beam broadening a smaller excited volume. In this work, the effect of beam broadening is assessed on segregation measurements in a 304L stainless steel sample irradiated with 3.2 MeV protons at 400°C to doses of 3.0 and 0.1 dpa. The STEM/EDS measurements are also compared to measurements made using AES.

High-purity 304L stainless steel samples with the nominal composition of 20.9 at% Cr, 9.0 at% Ni, and 69.0 at% Fe as determined by electron microprobe analysis were used. Samples were irradiated with 3.2 MeV protons at a dose rate of approximately 7.0×10^{-6} dpa/s to 3.0 and 0.1 dpa. The sample temperature during irradiation was maintained at 400° ± 10°C. Further details of the sample preparation and irradiation are given in Ref 2.

Measurement of grain boundary composition by STEM/EDS was performed on a Philips EM400T/FEG and a Philips CM200/FEG at Oak Ridge National Laboratory. The EM400T has an incident beam size of 2 nm (full-width tenth maximum) and operates at 100 kV. The CM200 has an effective probe size of 2 nm (full-width tenth maximum) and operates at 200 kV. A double-tilt, liquid-nitrogen-cooled specimen holder was used to minimize contamination of the sample in the focused beam[3]. Grain boundaries were aligned parallel to the incident electron beam. Subtraction of "in-hole" spectra from the measured spectra was performed to correct for uncollimated radiation from the microscope illuminations system and radiation associated with residual radioactivity in the sample. Collected raw intensities were converted to atomic concentrations using *k*-factors[4] calculated from comparison of EDS-determined matrix intensities to the bulk alloy composition determined independently by electron microprobe analysis.

The same high-dose sample in the same geometry was examined in both instruments. The same grain boundaries were analyzed in the same region of the foil. Both grain boundary measurements and segregation profiles were taken on each instrument. The results of the grain boundary Cr and Ni measurements are illustrated in Figure 1. Clearly, instrument type and beam broadening shows a significant effect on the measured segregation. As expected, the CM200 operating at 200 kV measures more Cr and Ni segregation than the EM400T since less matrix material is excited by the higher energy incident beam. Results of AES analysis of samples from the same irradiation are also shown in Fig 1 in

order to compare the two techniques. The AES technique measures more segregation than either instrument using STEM/EDS. Results will also be compared with measurements taken on the same sample using a 300 kV dedicated STEM instrument and with measurements taken from the low dose sample on all instruments.

Segregation profiles were also taken for the high dose sample using both TEM instruments across the same grain boundaries. The Cr and Ni segregation profiles for one of these boundaries is shown in Fig. 2. The profile measured by the CM200 is deeper and more narrow than that measured by the EM400T. Again, as less volume is excited, more segregation is measured and the amount of detail in the profile is increased. These profiles will also be compared to those measured by a 300 kV dedicated STEM instrument.

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References

1. G.S. Was and P. Andresen, J.Metals 44 (4) (1992) 8.
2. D.L. Damcott, J.M. Cookson, R.D. Carter, Jr., J.R. Martin, M. Atzmon, and G.S. Was, Radiat. Eff. Def. Solids, 118 (1991) 383.
3. E.A. Kenik, Scripta Metall. 21 (1987) 811.
4. J.I. Goldstein, in Principles of Analytical Microscopy, eds. D.C. Joy, A.D. Romig and J.I. Goldstein (Plenum, New York, 1986).

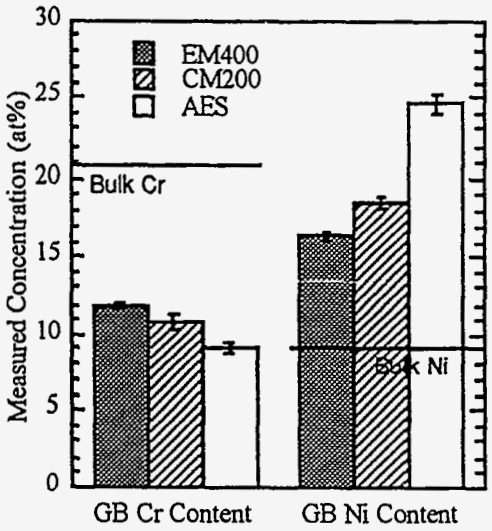

Fig. 1: A comparison of the GB Cr and Ni composition measurement for 304L SS irradiated to 3.0 dpa at 400°C with 3.2 MeV protons. Error bars represent the standard deviation of the mean.

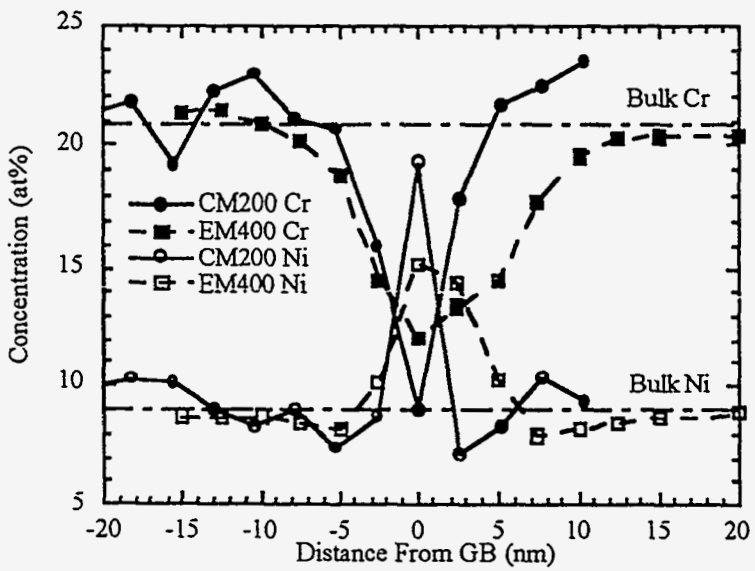

Fig. 2: A comparison of the GB Cr and Ni profiles for 304L SS irradiated to 3.0 dpa at 400°C with 3.2 MeV protons for the EM400T and CM200.