Measurement of Irradiation-induced Swelling in Stainless Steels with a New Transmission Electron Microscopy Method

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300 series stainless steels such as 316H are used in nuclear reactors where they experience neutron radiation damage. An important concern here is the volume increase or swelling and associated deterioration of mechanical properties. Traditionally, transmission electron microscopy (TEM) has been used to measure void size and number density to estimate the overall volume increase. However, this technique may not yield accurate results when the voids are small or cannot be effectively resolved in TEM. Here we have developed a method to measure swelling from the valence electron density of materials using plasmon energy loss of high energy electrons in TEM. The technique is potentially applicable to any metals regardless of composition or nature of phases. Furthermore, benefiting from the well-confined TEM probe, the technique is suitable to measure nanostructure density change.

The incident electrons in TEM lose energy to specimen atoms by displacing valence electrons and creating an oscillation of electrical field (plasmon). The square of plasmon peak energy E_p^2 in electron energy loss spectroscopy (EELS) is proportional to valence electron volume density [1]. A TEM sample is a thin membrane usually about 100 nm thick. It inevitably contains surface layer of either oxide, sample preparation damage, or other structures having different electron density from the intrinsic material. If ρ, V, and t represent valence electron density, volume, and sample thickness, respectively, and ρ_s, V_s, and t_s represent these quantities for the surface layer, plasmon energy is proportional to the average electron density:

$$E_p^2 = C \times \bar{\rho} = C \times \frac{\rho(V-V_s)+\rho_s V_s}{V} = C \times \frac{\rho(t-t_s)+\rho_s t_s}{t} = C\left[\rho - (\rho - \rho_s)\frac{t_s}{t}\right] \qquad (2)$$

By plotting and fitting $E_p^2 \sim 1/t$ linearly, the intersection to y-axis would be proportional to ρ.

TEM experiments were conducted at 200 kV using an FEI Titan S-Twin aberration corrected scanning transmission electron microscope (STEM) equipped with Gatan GIF (ER865). Figure 1(a) shows the annular dark field (ADF) image of 316H stainless steel irradiated with Fe^{2+} up to 100 dpa peak damage at 360 °C. Multiple EELS line scan spectra were collected. Figure 1(b) shows an EELS spectrum fitted in $f(x) = y0+A\times\exp\{-[(x-x_0)/width]^2\}+B\times x$. The fitting function is sought to match data trend near the top of plasmon peak without considering physical meaning. This yields a plasmon peak energy E_p= 25.95 eV.

Figure 2(a) shows a plot of E_p^2 of 202 EELS data points in the damaged region. From the difference of E_p^2 between damaged and undamaged regions, swelling is determined and is shown in Figure 2(b), along with SRIM calculated ion concentration [2]. SRIM calculations here used 3.7 MeV Fe^{2+} into 316H (Fe-17Cr-10Ni-2Mo-2Mn in weight percentage, density 7.86 g/cm^3); displacement energy values 40 eV for Fe, Cr, Ni, Mn, and 60 eV for Mo.

Swelling in Fe^{2+} irradiated 316H up to 100 dpa at 360 °C appears to correlate inversely with ion implantation depth profile. Swelling is large at surface and gradually decreases with depth. Near the ion implantation peak the material is denser than intrinsic state, suggesting suppression of void swelling by high concentration of implanted ions. On average swelling is 0.98 ± 0.06 % at depths of 0.16 - 1.85 μm in the damage layer.

References:

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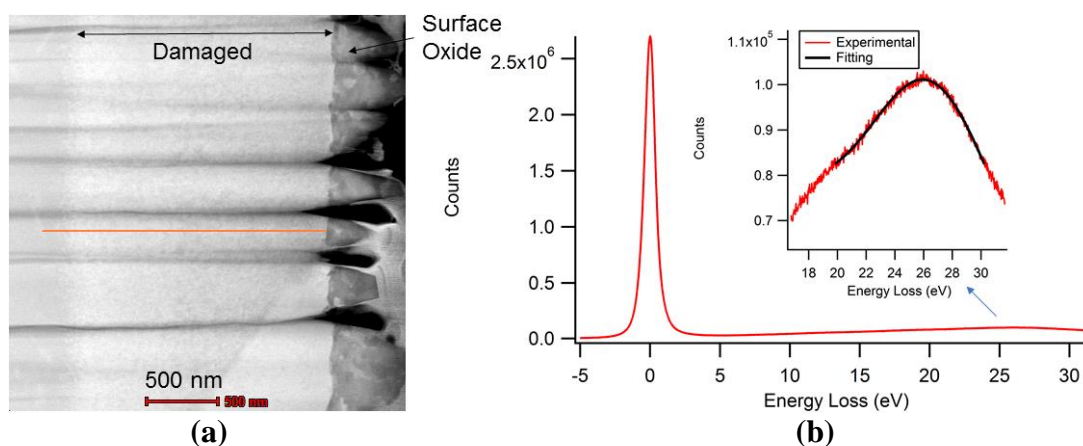

Figure 1. STEM and EELS measurement on Fe^{2+} irradiated 316H: (a) ADF image with beam semi-convergence angle 24.5 mrad and collection angle 25-55 mrad. (b) EELS spectrum acquired on the red line at 1.84 μm depth from oxide-metal interface in (a).
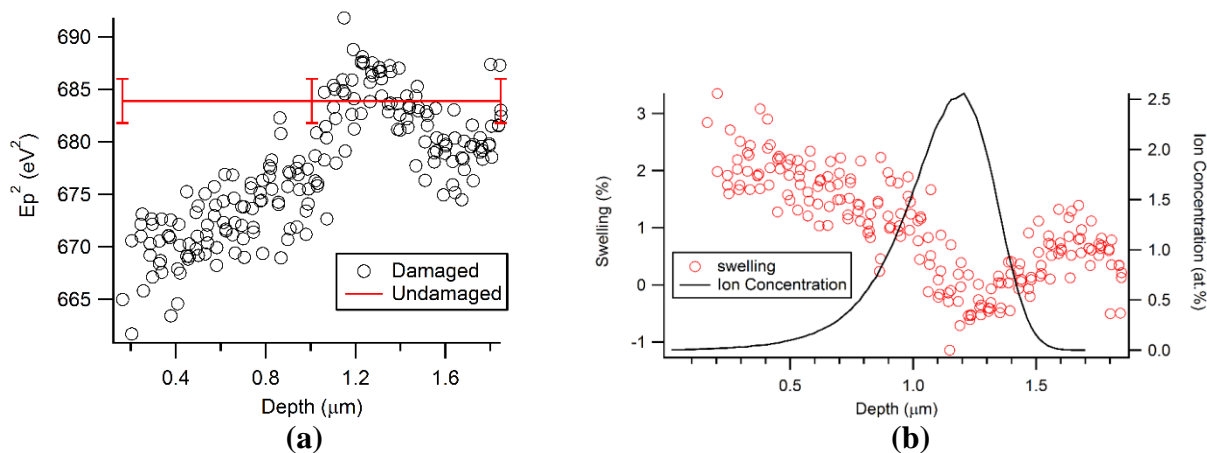

Figure 2. Swelling measurement of Fe^{2+} irradiated 316H up to 100 dpa at 360 °C: (a) plasmon peak energy square vs. depth in the damaged region after sample thickness correction, (b) swelling depth profile. Ion concentration was obtained from SRIM calculations [2].