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Inert Gas Measurement of Single Bubble in CeO_2

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Uranium dioxide (UO_2), an oxide with a fluorite crystal structure, is the main fuel used in commercial light water reactors. Inert fission gases such as Xe and Kr significantly impact the performance of UO_2 during reactor operation and in storage. These gases have a large yield of approximately 25% and have a low solubility in UO_2, resulting in the formation of large density of fission gas bubbles [1]. Such bubbles cause the fuel to swell, which promotes clad outward creep that shortens the cladding lifetime. Characterization of the inert fission gas content in bubbles can help us understand fuel swelling and fuel pin pressurization. This is performed for Xe bubbles in cerium dioxide, CeO_2, which is considered a surrogate for UO_2 due to similar crystal structure and properties.

Xe ion irradiation of CeO_2 was performed at room temperature using a 200 keV Danfysik ion implanter at the Los Alamos National Laboratory. The Xe ion energy was 400 keV and the total fluence was 1×10^{16} ion/cm^2. The Xe implanted CeO_2 was annealed in air at high temperature. A 200 kV FEI Titan scanning transmission electron microscope (STEM) with CEOS probe aberration corrector equipped with EDX and EELS was used to characterize Xe bubble microstructure and composition.

Figure 1 shows the STEM image and EELS elemental mapping of a Xe bubble. The bright signal of Xe M-edge and dark signal of Ce M-edge at the bubble region indicates that the bubble is filled with Xe. The atomic ratio of Xe to Ce in the yellow dashed square region is 3.75 and 9.58 at.% determined by EDX and EELS, respectively. The local foil thickness was measured through inelastic mean free path (IMFP) using EELS technique. The IMFP, λ, depends on the specimen density ρ, electron energy E_0 and collection semiangle (excitation semiangle α and collection semiangle β) according to following equations [2]:

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$$\lambda = \frac{200FE_0}{11\rho^{0.3}\ \ln\{(\alpha^2+\beta^2+2\theta_E^2+|\alpha^2-\beta^2|)/(\alpha^2+\beta^2+2\theta_C^2+|\alpha^2-\beta^2|)\times(\theta_C^2/\theta_E^2)\}} \qquad F = \frac{(1+E_0/1022)}{(1+E_0/511)^2},\ \ \theta_E = \frac{5.5\rho^{0.3}}{FE_0},\ \ \theta_C = 20\ \text{mrad}$$

Based on local TEM foil thickness, bubble size, and specimen density, the number of Ce atoms in the dashed square region was calculated to be around 1.0×10^6. The number of Xe atoms in the bubble is about 3.8×10^4 and 9.6×10^4 in terms of EELS and EDX results, respectively. The replacement of Ce by Xe in the bubble was calculated to be 0.12 and 0.31 according to EELS and EDX results, respectively. The red solid square regions R1 and R2 were selected for EELS spectrum analysis (Figure 2). Weak Xe $M_{4,5}$-edge at around 700 eV was only detected in R1 and strong Ce $M_{4,5}$-edge at about 900 eV was found in both R1 and R2. A method introduced by

Fortner and Buck [3] was applied for the EELS data analysis. The average branching ratio of Ce, M_4/M_5 at R1 and R2 is about 1.09 and 1.17, respectively, which correspond to an average valence state of +3.9 and +4, respectively. It indicates O vacancies were formed at bubble region. In short, chemical information and inert gas content in single bubble can be determined by STEM/EDS/EELS techniques. This research was supported as part of the Center for Materials Science of Nuclear Fuels, an Energy Frontier Research Center funded by the U.S. Department of Energy, Office of Science as well as Laboratory Directed Research and Development (LDRD) program at Idaho National Laboratory. The ion implantation was partially supported by the Center for Integrated Nanotechnologies (CINT), a DOE nanoscience user facility jointly operated by Los Alamos and Sandia National Laboratories.

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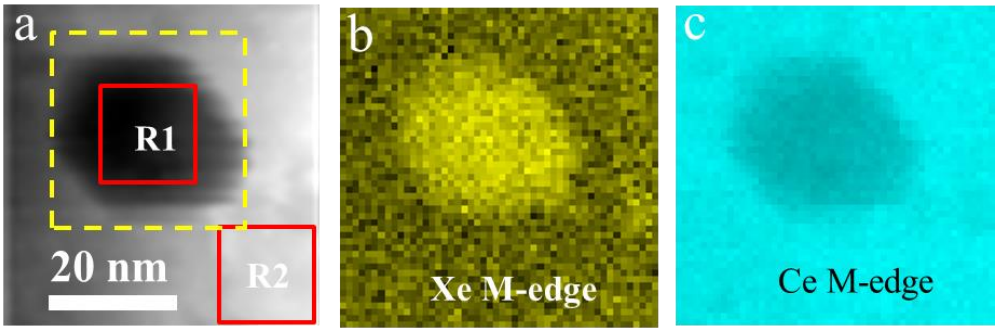

Figure 1. (a) STEM image of a single Xe bubble in CeO2 after post-irradiation annealing at 1200°C for 1 hour. (b) EELS Xe M-edge mapping. (c) EELS Ce M-edge mapping. The yellow dashed square was selected for composition analysis using EDX and EELS, and the red solid squares were selected for EELS spectrum analysis.

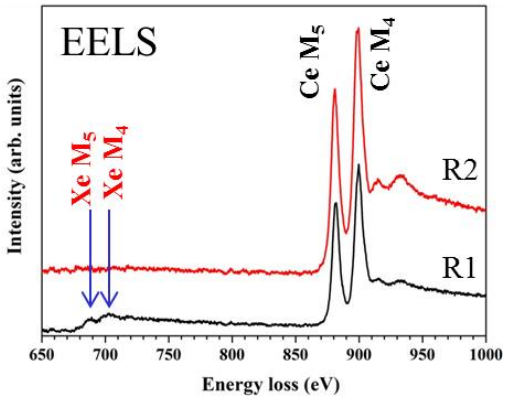

Figure 2. EELS spectra of R1 and R2 regions in Figure 1.