

Point defects production in ceria under high-energy electron irradiation by using *in-situ* cathodoluminescence

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Abstract

CeO₂ (ceria) is one of the remarkable materials as a surrogate for a nuclear fuel and transmutation target. We have investigated the production and charge state of point defects and F-centers in ceria by using *in-situ* cathodoluminescence (CL) spectroscopy under high-energy electron irradiation in a High Voltage Electron Microscope (HVEM).

Keywords: ceria, point defects, F-center, high voltage electron microscope, cathodoluminescence

1. Introduction

Understanding on the creation and charge state of point defects under irradiation is important for the safety evaluation of nuclear material. Ceria, a cubic fluorite-type of ceramics is a surrogate of PuO₂, and is induced oxygen point defects by elastic-inelastic collision under high-energy electron irradiation. These defects, charged by electron trapping (F-centers) emit visible light according to charge states (e.g. F⁰, F⁺, F²⁺) [1]. The production of F-centers was detected by *in-situ* cathodoluminescence (CL) spectroscopy during electron irradiation, which is interfaced with HVEM.

2. Experimental

A single crystal ceria in thickness of 1 mm, and sintered bodies of poly crystal in thickness of 150 μm were used. The HVEM in the Ultramicroscopy Research Center of Kyushu University was operated at energies ranging in 400 ~ 1250 keV and temperatures of 100 ~ 300 K. Electron beam with a flux of $\phi = 2.55 \sim 7.65 \times 10^{21} \text{ m}^{-2}\text{s}^{-1}$, measured by in-beam Faraday cup, was used with a beam diameter of 30 μm. The CL emission was collected in the HVEM by an optical fiber probe [2] and each spectrum was generally taken in 30 secs under averaged out over 5 recordings and repeated 5 times.

3. Results and Discussion

Figure 1 shows *in-situ* CL spectra of a single crystal ceria obtained under 20 keV and 600 keV electron irradiation by using Scanning Electron Microscope (SEM)-CL and HVEM-CL, respectively. Three broad bands were obtained with 600 keV electrons, centered at photon energies of 1.6 eV, 2.7 eV and 4.1 eV, assigned to be impurities, Ce³⁺ ions and F-centers, respectively. However, only one band (2.8 eV) was obtained for 20 keV, whose transferred energy is lower than the threshold displacement energy (E_d) of oxygen atom in ceria. Therefore, this band indicates Ce³⁺ ion and separates due to spin-orbit coupling. The F-center (4.1 eV) induced by 600 keV electrons is due to the displacement damage of oxygen sublattice. The electron energy, beam-flux and temperature dependences on CL spectra are discussed with a model taking cross-section of oxygen displacement and emission process of CL into accounts.

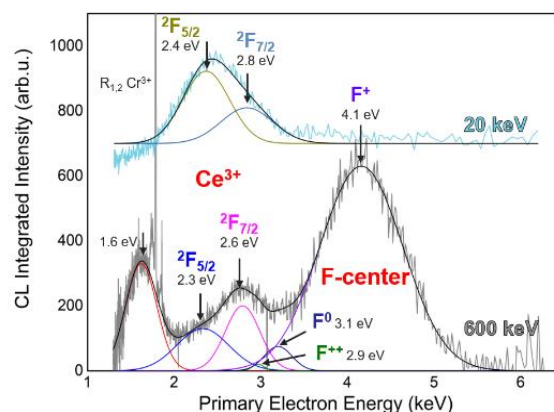


Figure 1. CL spectra are obtained at RT for 20 keV and 600 keV electrons in single crystal ceria. Gaussian fitting curves are also shown.

References

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