In-situ cathodoluminescence study of Gd_2O_3 doped CeO_2 in HVEM

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Abstract

CeO_2 (ceria) is a surrogate for PuO_2, and transmutation target and Gd_2O_3 doped ceria (GDC) can be used for burnable poison fuels. We investigated the production and charge state of F^+ centers in GDC by using *in-situ* cathodoluminescence (CL) spectroscopy under electron irradiation in a High Voltage Electron Microscope (HVEM).

Keywords: Gd_2O_3 doped CeO_2, Irradiation effects, High voltage electron microscopy, *In-situ* luminescence

1. Introduction

Understanding the production of point defect under irradiation condition is of importance to evaluate the safety of nuclear materials. We detected CL emission from F centers i.e. anion (oxygen) vacancies with trapping electrons, and from cerium ions in Gd_2O_3 doped ceria (GDC) varying with Gd atomic concentration (0 ~ 5 mol %) under high energy electron irradiation by using HVEM. To understand the kinetic behavior of point defects in GDC, we tried to interpret the in-beam charge states of F centers by CL spectroscopy and its thermal and concentration quenching effects.

2. Experimental

Single crystal ceria in thickness of ~1 mm and sintered poly crystal $Gd_xCe_{1-x}O_{2-\delta}$ (x = 0 ~ 0.05) in thickness of ~150 μm were used. A HVEM in Kyushu University was operated at energies ranging in 400 ~ 1250 keV and at temperatures of 100 ~ 300 K. Electron beam flux of φ = 2.55 ~ 7.65 × 10^{21} $m^{-2}s^{-1}$ was measured by in-beam Faraday cup with a beam diameter of 30 μm. An optical fiber probe in the HVEM [1] collected the CL emission and each spectrum was generally taken in 30 secs under the averaged out over 5 recordings and repeated 5 times.

3. Results and Discussion

Three broad CL bands appeared from the both ceria samples under irradiation. Two bands appeared at ~2.0 eV and ~2.9 eV by charge compensation and ionization of Ce^{3+} ions. The other appeared at 4.2 eV by F^+ center due to elastic collisions. The 4.2 eV band did not appear under 20 keV electrons irradiation whose transferred energy is lower than the threshold displacement energy of O ions. Fig. 1 shows the primary electron energy (E) dependence of CL integrated intensities (I_{CL}) of those three CL bands at 300 K. It shows maximum for ~600 keV electron irradiation. The displacement cross section for electron/hole trapping is increasing with E, whereas CL emission cross section is decreasing with E, which have apparent excitation cross section and radiative decay quenching effects by increasing of point defect concentration [2].

For the CL bands of GDC by high-energy electron irradiation, they have similar photon energies with ceria samples. However, I_{CL} of GDC decreases as increasing temperature and dopant concentration due to the quenching effects of CL by non-radiative decay.

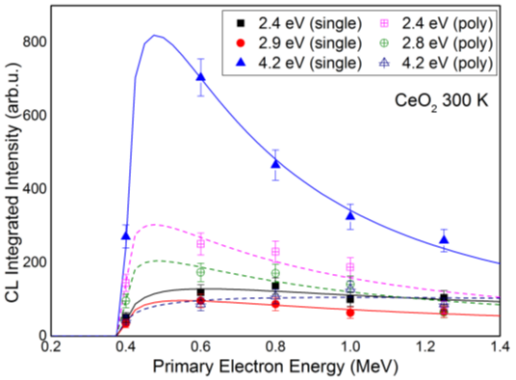

Figure 1. CL integrated intensities of CL bands at 300 K against primary electron energy for single crystal (full symbols) and poly crystal (empty symbols) CeO_{2-x}. Solid and dashed lines are least-square fit with equation of CL emission for each sample.

References

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