§37. Study on Mechanism of Luminescence Property Change in Irradiated Functional Materials

Yoshino, M., Nagasaki, T. (Nagoya Univ.),
Ogasawara, K. (Kwansei Gakuin Univ.),
Tanaka, T., Sakaue, H.A., Muroga, T.

In the development of fusion blanket systems, usage of various oxide, carbide, nitride, hydride functional materials have been proposed for electrical insulation, suppression of tritium fuel permeation, suppression of corrosion, radiation shielding, etc. In this study, cathodoluminescence (CL) measurement is performed on these functional materials to estimate crystallinity. The data about the changes in emission spectra with sample conditions varied by sintering temperature, ion beam irradiation damage, sample suppliers has been stored. In the previous study, it has been found that the intensity in 640-700 nm had a correlation with crystallinity in Er_2O_3 coatings [1]. On the other hand, the discussion on mechanism in CL of each materials is still begun to be studied, and will be proceeded with using examples from the earlier studies of each materials in the other research area. And, the analysis of electron energy level structure by the DV-Xα molecular orbital calculation method has also been implemented in this study, since each properties of the materials may depend on their electronic states. For promoting the discussion on the relationship between the results of electron energy level structure and emission mechanism, and the application of it for crystallinity estimation, SiO_2, whose emission mechanism and crystallinity may be investigated well, is selected to study.

SiO_2 films are synthesized by sol-gel method on SUS430 substrate and Si substrate with heating in air for 1 hour at 650 °C for studying the correlation between crystallinity and CL spectra. The CL spectra for these samples and commercial quartz plate are shown in Fig. 1. Emission peaks are observed around 280, 450, 650 nm in quartz plate as reported in some paper and around 650 nm also in SiO_2 on SUS430 as shown in Fig. 1(c) and (a). For investigating correlation between crystallinity and intensity ratio of these emission peaks, improvement of measurement sensitivity is needed to measure emission peaks around 280 and 450 nm. As shown in Fig. 1(b), in SiO_2 film on Si substrate, there are some emission peaks that is not observed in quartz plate. This implies the possible presence of composition or structure originate other than SiO_2, and will be discussed on the basis of the results of earlier studies and calculation results mentioned below.

As a first application of calculation to this study, electronic structures in SiO_2 are calculated by the DV-Xα method. The cluster models used in this calculation are shown as insets in Fig. 2. The models are constructed on the basis of the structure of SiO_2. In order to investigate the effect of the oxygen ion vacancy on the electronic structure, oxygen ion vacancy is introduced in the cluster in Fig. 2(b). Fig. 2 shows the electron energy level structure and the changes in it near the band gap with the introduction of oxygen ion vacancy. Here, the energy of highest occupied molecular orbital level in the cluster model without oxygen ion vacancy is set to 0 eV. The calculated band gap in this cluster is about 10 eV, in agreement with experimental results, about 9 eV. As shown in Fig. 2(b), when oxygen ion vacancy is introduced in SiO_2, new oxygen ion vacancy-related levels appear in the band gap.

The correlation between the crystal condition of SiO_2 and CL spectra will be continuously examined, and comparison with the change in the electron energy level structure obtained from theoretical calculation will also be studied. These experiments and the calculations about SiO_2 will be carried out to elucidate the mechanism of change in the luminescence properties with defect formation attributed to the irradiation damage envisioned in fusion reactor. Such studies will also be done for candidate blanket functional materials, Y_2O_3, ZrO_2, AlN.

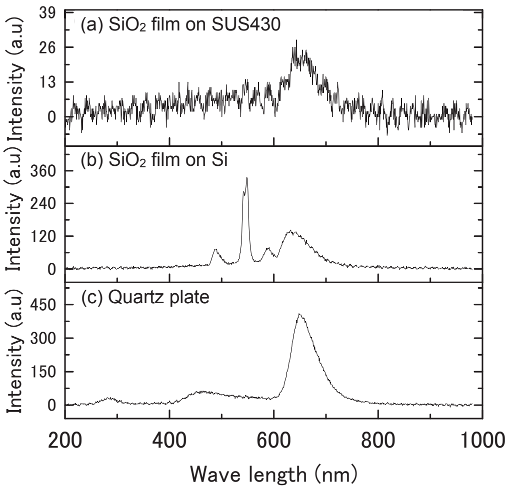

Fig. 1. CL spectra of SiO_2 samples and quartz plate.

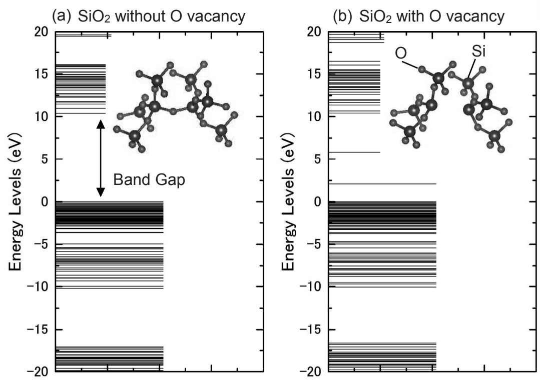

Fig. 2. Energy level structures near the band gap for SiO_2 (a) without oxygen ion vacancy (b) with oxygen ion vacancy.

[1] Tanaka, T., et al., Journal of Nuclear Materials 417 (2011) 794–797.