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Temperature quenching mechanism of cathodoluminescence in forsterite

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Cathodoluminescence (CL), the emission of visible light caused by electron irradiation, has been widely applied to geoscience by the reason of high detection sensitivity of emission center such as impurity elements and various structural defects. Recently, CL zoning of the forsterite in carbonaceous meteorites has been investigated to clarify the origin and thermal history of the chondrules. In this case, small amount of divalent Fe ions as quencher easily eliminate CL emissions caused by impurity and defect centers, so only near end forsterite can show CL emissions. Sample temperature is one of most important effects on CL in minerals, but has not been investigated to date for the forsterite. In this study, the effect on forsterite CL has been quantitatively analyzed by assuming the Mott-Seitz model for temperature quenching of luminescence.

Forsterite crystals (Fo: 99.1) in basalt from Mogok, Myanmar and micro-grains (Fo: 99.7-99.8) in chondrules from Allende and Kaba meteorites were prepared for CL spectral measurements. CL spectroscopy was made by a SEM-CL system, which is comprised of SEM (JEOL: JSM-5410) combined with a grating monochromator (OXFORD: Mono CL2) at accelerating voltage of 15 kV and beam current of 1.0 nA in a beam scan mode. The sample temperature was controlled by flowing liquid nitrogen and using an embedded heater in a cryostage. All spectra were corrected for total instrumental response, which was determined using a calibrated standard lamp.

At room temperature, the CL spectra show broad emission bands at around 400 nm in blue region and at around 650 nm in red region, and pronounced emissions increasing to IR region with small emissions at around 720 nm. These bands can be assigned to structural defect, divalent Mn and trivalent Cr impurities, respectively. Their emission intensities increase with a decrease in sample temperature. In general, luminescence efficiency decreases with rising temperature due to an increase in non-radiative transitions. This phenomenon is called temperature quenching.

CL spectral peaks in energy unit were deconvoluted by Gaussian curve fitting to determine the emission component for each emission center. The emission peak in blue region can be separated into two components centered at 3.15 and 2.99 eV, and the peaks caused by Mn and Cr impurity centers can be fixed at 1.91 and 1.74 eV, respectively. By assuming the Mott-Seitz model, activation energy in each temperature quenching process can be calculated by Arrhenius plots using integral intensity of each component. The straight-line relationships in the plots resulted in each activation energy as follows; blue emission at 3.15 eV: 0.08±0.04 eV, blue emission at 2.99 eV: 0.10±0.05 eV, red emission at 1.91 eV: 0.01±0.005 eV, red emission at 1.74 eV: 0.01±0.02 eV. The values of activation energies for blue emissions caused by structural defects correspond to the vibration energy of Si-O stretching mode in the lattice, and the values for red emissions caused by Mn and Cr impurity centers to Mg-O vibration energy. It suggests that the temperature quenching energy might be transferred as a phonon to the specific lattice vibration.

Keywords: Forsterite, Cathodoluminescence, Temperature quenching