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Detection of radiation-induced defect center in plagioclase using cathodoluminescence spectroscopy

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Cathodoluminescence (CL) techniques have been used as an effective method to visualize radiation halos in quartz. Recent scientific interests focus on the CL of quartz as a goedosimetry indicator to clarify the formation process of CL halos, whereas no method has yet been developed for a quantitative estimation of the radiation dose on quartz from disintegration of natural radionuclides. Feldspar is one of the most important rock-forming minerals in the Earth's crust, of which CL depends on the genetic conditions such as temperature, pressure, and radiation dose during various geologic processes. Geoscientific applications of the CL of feldspar minerals, therefore, have been extensively explored for the study of mineral genesis interpretation. No investigation of radiation effects on CL of feldspar has been performed to date, although the visible halos can be easily found in the feldspar directly attached to radioactive minerals. In this study, CL of plagioclase implanted by He^+ ion has been conducted to quantitatively estimate the radiation dose from disintegration of natural radionuclides possibly to be applied to geodosimetry.

Single crystals of albite (Or_1Ab_{99}) from Minas Gerais, Brazil, oligoclase ($Or_2Ab_{82}An_{16}$) from Inabu, Japan, andesine ($Or_1Ab_{53}An_{46}$) from Bekily, Madagascar; and anorthite (Ab_5An_{95}) from Yoichi, Japan were selected for CL and Raman measurements. He^+ ion implantation (dose density: 2.18 x 10^{-6} to 6.33 x 10^{-4} C/cm^2) on the samples was performed using a 3M-tandem ion accelerator at 4 MeV corresponding to the energy of alpha-particles from 238U. A scanning electron microscopy-cathodoluminescence (SEM-CL) was used to obtain CL spectra of these plagioclase. Operating conditions were set at 15 kV (accelerating voltage) and 1.0 nA (beam current).

CL spectra of unimplanted plagioclase consist of emission bands at 350, 420, 580 and 700?740 nm which are assigned to Ce^{3+} impurity, Ti^{4+} impurity or Al-O^--Al defect, Mn^{2+} impurity and Fe^{3+} impurity centers, respectively. Similar emission bands were recognized in CL spectra of the implanted plagioclase. The intensities of these CL emissions decrease with an increase in radiation dose of He^+ implantation. Raman spectroscopy on the plagioclase reveals that Raman peak at 505 cm^{-1} shows a decrease in the intensity and an increase in the full width at half maximum (FWHM) with increasing radiation dose of He^+ ion implantation. He^+ implantation destroys a linkage of T-O-T bond in the framework structure of the plagioclase, resulting in a reduction of CL emissions caused by impurity centers. CL spectra of implanted samples of albite and oligoclase exhibit characteristic red emissions at 700 nm, of which intensities increase with an increase in the radiation dose. It suggests that CL emissions at 700 nm are assigned to radiation-induced defect center formed by He^+ ion implantation. Deconvolution of CL spectra obtained from the implanted albite and oligoclase can successfully separate emission bands into six Gaussian components at 3.05, 2.82, 2.10, 1.86, 1.67, and 1.56 eV, where the component at 1.86 eV is undetectable in unimplanted samples. Integral intensity of the component at 1.86 eV linearly correlates with the radiation dose of He^+ ion implantation. The deconvoluted CL spectra of andesine and anorthite, however, have no component at 1.86 eV. These results suggest that the component at 1.86 eV might be assigned to O^{1-}/^{27}Al x 2^{23}Na center. The component intensity at 1.861 eV correlates with the radiation dose as a function of O^{1-}/^{27}Al x 2^{23}Na center, but does not depend on the concentration and distribution of the emission center, degree of Si-Al order and on the presence of microstructures or texture. CL spectral deconvolution, therefore, should be applied to quantitatively evaluate radiation dose of alpha particles from natural radionuclides on Na-rich feldspar as a geodosimetry indicator.

Keywords: cathodoluminescence, plagioclase, radiation-induced defect center, radiation halo, deconvolution