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Applicability of peroxy radical center in quartz to pre-Quaternary dating

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There are several lattice point defects in quartz that trap unpaired electrons excited by natural radiation. ESR (Electron Spin Resonance) spectrometers can detect such point defects as ESR signals. The ESR signal intensities increase with time under natural radiation. Many point defects in not only quartz but also other minerals have been applied mainly to Quaternary dating. A few researches on pre-Quaternary dating using ESR signals were reported, however at present they are very questionable. Thus, we examine the behaviors of ESR signals in quartz after gamma-rays irradiation at high-dose-level more than 1MGy and discuss their applicability to pre-Quaternary.

In this work, we used two natural quartz samples that were extracted using HF from the Cretaceous granitic rocks (Aio and Hofu granites) distributed in Yamaguchi, Japan. These granites have been dated as about 92Ma (Aio granite) and about 97Ma (Hofu granite), respectively, by Rb-Sr dating. In addition, we used quartz sand treated (Showa Chemical) as a standard quartz sample. ESR measurements were carried out at room temperature using an X-Band EPR spectrometer (Bruker E500 CW) with 100kHz modulation. As a result of ESR measurements, we detect from two natural quartz samples E` center derived from oxygen vacancies, peroxy radical center derived from oxygen interstitials and NBOHC derived from non-bridging oxygen atoms.On the other hand, we carried out gamma-irradiation from 100kGy to 1800kGy with a Co-60 source at Takasaki Research Establishment, Japan Atomic Energy Research Institute. By gamma-irradiation, the signal intensities of the E` centers and NBOHCs change irregularly, whereas the peroxy radical centers proportionally increase with radiation dose. Furthermore, the peroxy radical centers obtained from two natural quartz samples have much larger radiation sensitivities than that from the standard quartz sand. The peroxy radical center is easily created in silica glass by gamma-irradiation, however so far it has hardly appeared from quartz by gamma-irradiation at high-dose-level. A slight amount of radioactive elements exist inside the quartz samples. Therefore, alpha rays and recoils emitted out of these internal radioactive elements may have created amorphous local regions inside the quartz samples. The equivalent doses of two natural quartz samples are respectively estimated as about 224kGy (Aio granite) and about 171kGy (Hofu granite) by extrapolating the regression lines (curves) obtained from the intensities of the peroxy radical centers after gamma-irradiation using the least-square fitting method. The ESR ages of two natural quartz samples are calculated at about 58Ma (Aio granite) and about 60Ma (Hofu granite), respectively, by dividing the equivalent doses by the annual doses obtained from the concentrations of radioactive elements using Nambi and Aitken`s conversion table. These ESR ages are much younger than the Rb-Sr ages. In order to clarify the reason why the ESR ages are inconsistent with the Rb-Sr ages, we tried to analyze the concentrations of lead isotopes, 238U and 232Th in the whole rocks and quartz samples by ICP mass spectrometry. As a result, the ratios of lead isotopes in the whole rocks are almost consistent with those in the quartz samples within the range of errors. This result means that closed systems in both granites have held since their productions. On the other hand, the ratios of 206Pb/238U and 208Pb/232Th in the whole rocks are larger than those in the quartz samples. This means that the ratios of 206Pb/238U and 208Pb/232Th increase with geological time. Therefore, the ESR ages obtained in this work may show the ages of crystallization of quartz in both granites. Although the peroxy radical center is thermally very stable, we will examine in detail its closure temperature and the change of radiation sensitivity by alpha rays.