Article

Influence of Radioluminescence on Optically Stimulated Luminescence from Natural Quartz Grains

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The causes of the different optically stimulated luminescence (OSL) sensitivities for quartz aliquots from different origins were investigated in terms of radioluminescence (RL) during artificial irradiation. All RL spectra of red-thermoluminescence (RTL) quartz grains consist of two broad RL emission peaks, assignable to a violet region (V-RL, 400 nm) and a red one (R-RL, 630 nm). The OSL sensitivities were affected on the total amounts of V-RL intensities during irradiation of a fixed total dose (20 Gy) with different dose rates. Additionally, the bleaching effects of RL with shorter wavelength light than OSL-illuminating light (470 nm) were assured from another experiment with the combination of quartz slices and optical filter. Conclusively, it is suggested that the V-RL emissions appreciably affect the residual or naturally accumulated doses when OSL / SAR protocol is applied.

Key Words : optically stimulated luminescence, radioluminescence, dose rate effect, dosimetry, quartz

1. Introduction

An important application of the optically stimulated luminescence (OSL), which is the light emission during illumination of naturally or artificially radiation-irradiated materials, is the dating of ancient archeological samples as well as geological ones. Additionally, the OSL measurements have been utilized for retrospective dosimetry using quartz extracted from fired bricks and unfired mortars[1),2)]. In a pioneering work of OSL, Huntley and his workers have started the sediments dating using an argon ion laser[3)]. The recent OSL reader is able to illuminate the irradiated sample with blue light from light-emitting diodes (LEDs) or a blue laser light[4),5)]. On the OSL-dosimetry and dating, the OSL-response of the sample against some known doses could be calibrated in the laboratory, in which artificial doses are administered to higher dose rates in comparison with the natural ones. From these viewpoints, much attention must be paid on the question whether the measured OSL signals are dependent or independent on the dose rates. In OSL/SAR (single-aliquot regenerative-dose) protocol, the repeating cycle of heating, illuminating and irradiating procedures will give rise to systematic changes on luminescence sensitivity[6)]. According to Spooner[7)], the OSL-decaying (or shine-down) rates became 10 times faster at about 400 nm than that at 514.5 nm. In addition, the bleaching effects were found to be more effective at shorter wavelengths rather than longer ones[8].

Radioluminescence (RL) phenomenon, on the light emission during irradiation procedure, has been investigated using natural and artificial materials[9)-11)]. The different RL emission bands were reported for some quartz samples[10),11)]. If the RL emission could be observed during artificial irradiation in shorter wavelengths, the RL may be the cause of bleaching or diminishing effects of OSL in natural quartz itself just like during the OSL-stimulation.

In this viewpoint, the OSL sensitivities were studied from an aspect of influence of the dose rate changes during irradiation. Then, the RL spectra were measured using an RL spectrometric system. Subsequently, influence of 400 nm RL (V-RL) intensity on OSL intensity was examined by varying irradiation dose rates. Detected effects on OSL-bleaching phenomena regarding V-RL have been discussed.

2. Experimental

2・1 Sample preparation

Extraction of quartz grains (150 – 250 μm) from surface soil samples around JAEA (Tokai, Japan Atomic Energy Agency) was carried out by the following sequential procedures; acid-base treatments with 6 M HCl and 6 M NaOH solutions for a few hours, etching treatment with 46% HF for 20 min, and heavy liquid density separation (2.63 – 2.67 g cm^{-3}) using a sodium polytungstate solution. Further purification of quartz grains was done by hand selection under microscopic observation for the sake of elimination of feldspar grains. Single Japanese twin crystal from a museum collection was also employed in the present experiment after crushing and sieving to adjust the grains sizes ranging from 150 to 250 μm in diameter. Prior to irradiation, each sample was annealed at 450 ℃ for 5 min in an electric furnace to remove natural irradiation doses.

The quartz samples were then exposed to γ-ray of ^{137}Cs source (111 TBq, 365 mm distance). A fixed dose of 20 Gy was irradiated by a dose rate of approximate 1 Gy min^{-1} at room temperature in the Radioisotopes Center of Niigata University.

2・2 RL spectrometry

RL emission spectra of quartz samples were measured at room temperature using an on-line spectrometric system between 350 and 750 nm[12]. For RL-measurements, an X-ray irradiation system (Varian, VF-50J tube with W-target, 50 kV, 0.3 mA) surrounded by shielding lead blocks was set in front of quartz samples. RL observation was carried out twice, the first data being acquired for quartz sample and the second one without the sample as background. These raw data were reconstructed into two dimensional relationship draws consisting of RL wavelengths and intensities.

2・3 OSL/RL measurements

Both OSL and RL measurements were carried out by an automated TL/OSL-reader system, improved for the OSL/SAR as well as RTL-measurements[13),14)]. The OSL of quartz grains previously irradiated to 20 Gy was evaluated using the OSL/SAR protocol as described by Wintle et al.[15]. This protocol compensates for any sensitivity changes of the OSL signals by interpolating a test dose of 5 Gy for every regenerative dose[16].

The OSL-reader equipped sixteen blue light emitting diodes (blue-LEDs, Nichia Chemical Industries Ltd., NSPB-500S), which have an emission peak at 470 nm with 20 nm FWHM. The OSL signals were detected with a photo-

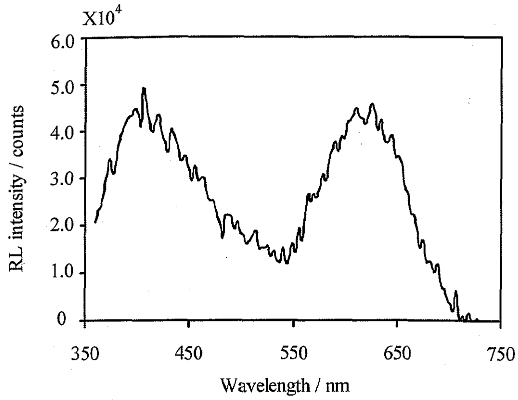

Fig. 1 RL emission spectrum from an aliquot of natural quartz at room temperature. A quartz grain aliquot was collected around JAEA and irradiated with X-ray at about 30 Gy.

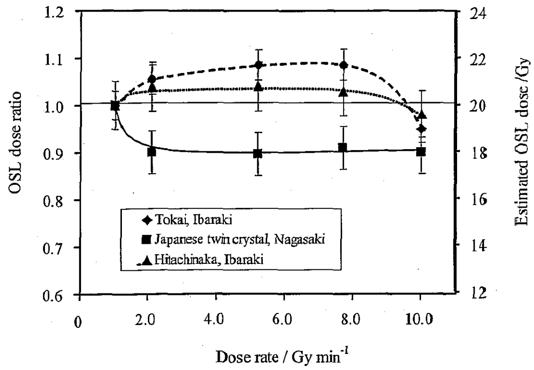

Fig. 2 Dose-rate dependences of estimated OSL-doses. OSL-doses are normalized to unity for OSL-doses evaluated at 1.0 Gy min^{-1}.

multiplier tube (PMT, Hamamatsu, R585S) by inserting a UG-11 (Schott) and an IRC-65L (Kenko) optical filters. In the OSL/SAR method, after 5 min preheating procedure at 220 ℃, OSL-decay curves were recorded every 0.2 s under blue light-illumination for 200 s.

For the RL-measurement, a small X-ray tube installed on the luminescence leader was conveniently utilized. The RL during X-ray irradiation was introduced to the PMT position in the TL/OSL-reader system through an optical bundle fiber. Combinations of the PMT and the filters analogous to the OSL measurements were employed to detect V-RL signals.

3. Results and Discussion

3·1 RL spectrometry

A typical RL emission spectrum of a natural quartz grain aliquot is shown in Fig. 1. Two broad RL emission bands are recognized at 400 nm (violet) and 630 nm (red), although the present quartz grains fraction shows the intrinsic RTL-property alone at higher temperature treatment beyond 250 ℃. There exists clear difference between the RL (two peaks) and the TL emission (single peak). From this result, V-RL emission centers were assumed to be unstable above room temperature, because intense violet TL (V-TL) was not observed or was bleached from the natural quartz grains.

3·2 Intensity variation of OSL and V-RL with irradiation dose rates

Variations of OSL intensities were practically examined for three kinds of natural quartz grains sampled at different places. Each aliquot has been pre-irradiated to 20 Gy. The OSL/SAR procedure was applied to confirm the fixed dose irradiation. Since the evaluated doses should agree to the fixed dose of 20 Gy, the dose-comparisons evaluated were carried out by normalizing to unity for the sample irradiated with the lowest dose rate of 1.0 Gy min^{-1}. Figure 2 shows dependences of OSL intensities on several dose rates. The experimental errors of doses are estimated with three or more aliquots. The figure confirmed that the estimated OSL-doses are meaningfully influenced on the irradiation dose rates beyond the experimental errors. Additionally, some different behaviors exist among samples ; samples A (Tokai) and C (Hitachinaka) showed overestimated result,

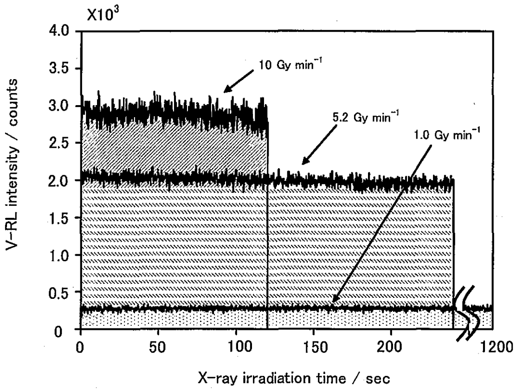

Fig. 3 Changes of V-RL intensities as a function of X-ray exposure period for three kinds of dose rates. A quartz grain aliquot was collected from surface soil around JAEA. Each shadow part in each dose rate is the integrated area when fixing dose of 20 Gy.

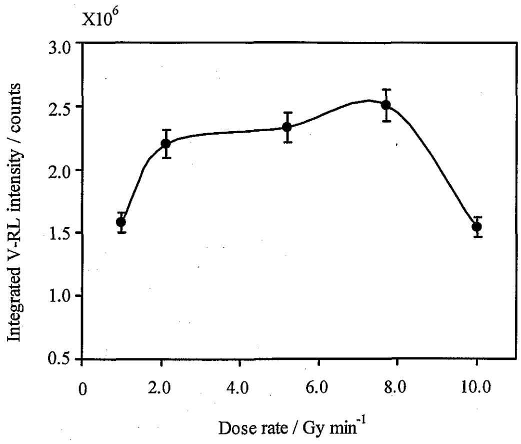

Fig. 4 Variation of integrated V-RL intensities by the dose rates of X-ray generator when the exposure dose was fixed to 20 Gy. The natural quartz grains were collected around JAEA.

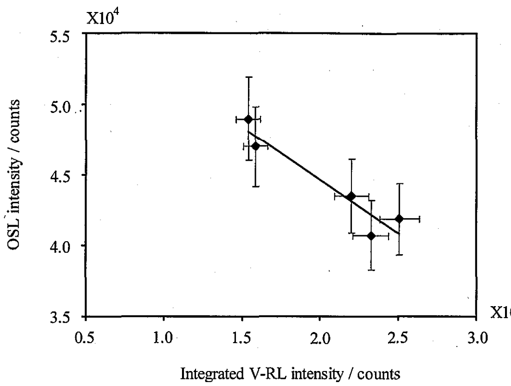

Fig. 5 Relationship between integrated V-RL intensity and OSL one when different dose rates were applied. A quartz grain aliquot was collected around JAEA. The total dose was fixed to be 20 Gy.

while sample B (Japanese twin crystal) underestimated one over 2 Gy min^{-1} dose rates. The former two samples show the reduction of OSL centers during artificial irradiation beyond dose rate of 2 Gy min^{-1}.

The broad V-RL emission spectrum, as seen in Fig. 1, belongs to slightly higher energetic photons close to the stimulating light (470 nm) for illumination. Spooner[7] had reported that illuminating photons with the shorter wavelengths cause the greater bleaching effects. According to Fig. 2, the apparent decreases of OSL signals from samples A and C (except for sample B from single Japanese twin crystal) might be attributed to self-bleaching effects from the V-RL during ionizing irradiation, resulted in higher dose estimation.

This reduction of OSL intensity accompanied with V-RL effects was studied further from V-RL intensity changes by varying dose rates. The behaviors of the V-RL signals as a function of irradiating duration are illustrated in Fig. 3. All of V-RL curves show respectively saturating tendency with irradiation, as well as V-RL counting rates increased with the increment of dose rates. The V-RL intensities are integrated during irradiation periods to accumulate the fixed dose of 20 Gy as seen shadow area in the figure. The resultant V-RL intensities are plotted as a function of dose rates and shown in Fig. 4. This figure exhibits that the total V-RL intensities were differently affected on the variations of irradiation dose rates.

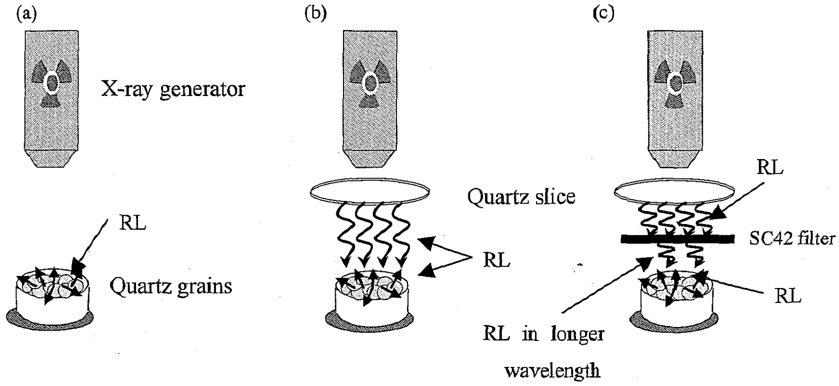

Fig. 6 Schematic X-ray irradiation arrangements for determining the influence of RL on OSL.
(a) a standard arrangement
(b) insertion of quartz slice
(c) insertion of quartz slice and SC42 filter
In (b) and (c) arrangements, quartz grains were covered with different slices of V-RL intensities during X-ray irradiation.

To clarify furthermore the bleaching effects of V-RL, the result of OSL intensities from the quartz grains irradiated with five different dose rates against the integrated V-RL intensities is shown in Fig. 5. The figure reveals that the OSL intensities possess a negative relation to the V-RL intensities. The same tendency was also observed for other natural quartz grain samples. This negative correlation of OSL intensities to V-RL intensities should support the certain presence of self-bleaching effects during artificial irradiation. This means that the RL in the violet region brings on the elimination (so-called self-bleaching effects) of OSL-source in quartz even during artificial irradiation.

3·3 Another evidence of self-bleaching effects with V-RL

The self-bleaching effects of OSL signals with V-RL intensities, as described in 3·2., were supported from the following experimental evidence. As seen in Fig. 6, two kinds of X-ray irradiation arrangements (b) and (c) were utilized by inserting each quartz slice, which brings on the different V-RL intensities derived from slice irradiation. In the arrangement (c), a gelatin optical filter was inserted between a quartz slice and a grain sample. Several quartz slices were employed to obtain different V-RL intensities. Three aliquots from the sample C were followed to the three kinds of irradiation arrangements and OSL-measurements with a fixed dose of 20 Gy. A type (a) arrangement was used to a standard OSL intensity, which was compared with the OSL-results from other types (b) and (c). The resultant OSL intensities are plotted against the integrated V-RL intensities which were separately measured for each slice. The results are summarized in Fig. 7. The remnant OSLs of an ordinate are the OSL intensity ratios from arrangements (b) and (c) against in (a), which are normalized to be unity in the case of the lowest V-RL intensity using a quartz slice with the dimmest V-RL intensities. Figure 7 indicates the ab-

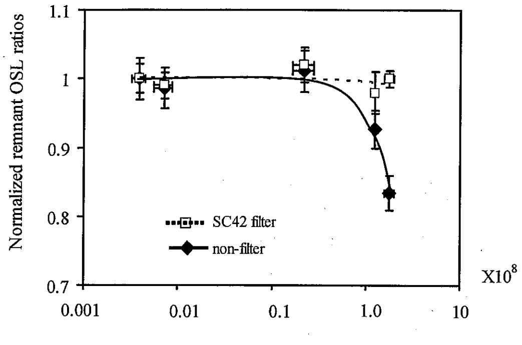

Fig. 7 Relationships between the normalized remnant OSL ratios and V-RL intensities of quartz slices. Each sample was irradiated to 20 Gy.
Remnant OSLs were normalized to unity from the lowest V-RL intensity quartz slice. A SC42 filter made from gelatin cuts enough the light shorter than 420 nm, but passes completely the light longer than 420 nm light as seen in the irradiation arrangement (c). The non-filter experiment was carried by the arrangement (b) in Fig. 6.

sence of bleaching effects with V-RL intensities lower than 2.2×10^7 counts for every case. However, the presence of bleaching effects is confirmed when V-RL intensities from quartz slice are beyond 2.2×10^7 counts with irradiation arrangement (b). In the case of the highest V-RL intensity, the OSL signals reduce by about 20% (arrangement b) without filter insertion. However, the OSL intensities with a sharp cut filter (Fuji SC42 filter) insertion (arrangement c) show almost no difference even in the case of high RL intensity region in comparison with the arrangement (b). This difference comes from whether SC42 filter was used or not. The former RL from the slice contains V-RL part, whereas the latter does not. These results reveal that the V-RL part could reduce the OSL of the quartz grain aliquot.

4. Conclusions

These experiments have allowed us to conclude that there exists apparent self-bleaching effect due to the RL on OSL-phenomena of natural quartz grains. The RL emission bands from natural RTL-quartz grains had two broad peaks, V-RL (400 nm) and R-RL (630 nm). Of two RL-emissions, V-RL one was found to be more effective for self-bleaching of quartz grains irradiated by different dose rates, together with covering the grain aliquot by quartz slices with different V-RL intensities coupled with an optical filter. Therefore, it should be noticed that such self-bleaching effects can influence the OSL-accumulation with different degrees dependent on the V-RL from the neighboring quartz grains. The reduction of OSL intensity would cause the RL not only from the slice and neighboring quartz grains, but also from the irradiated grains with higher efficiency. Therefore, much attention should be paid on the self-bleaching effects during artificial irradiation, particularly in accurate retrospective dosimetry and the OSL-dating.

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要　旨

天然石英からの光励起ルミネッセンスに対するラジオルミネッセンスの影響

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様々な起源の石英粒子からの光励起ルミネッセンス（OSL）感度が異なる原因について，照射中の発光であるラジオルミネッセンス（RL）の影響に注目した。RL 波長分光測定の結果から，全ての石英粒子からの RL 発光は，400 nm（V-RL）及び 630 nm（R-RL）に幅広いピークを有する 2 種であることがわかった。様々な線量率で一定線量（20 Gy）を照射した OSL の感度は，V-RL の全発光強度（20 Gy）に影響されることがわかった。更に，OSL の励起光である 470 nm よりも短い波長領域からの RL のブリーチング効果が，RL 強度の異なる石英薄片と光学フィルタを組み合わせた実験から確かめられた。これらの結果から，OSL/SAR 法により求めた積算線量には，V-RL 発光の影響があることがわかった。