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ScienceDirect
Radiation Measurements 41 (2006) 1015–1019
Radiation Measurements
www.elsevier.com/locate/radmeas

# Comparison of accumulated doses in quartz and feldspar extracts from atomic bomb-exposed roof tiles using several luminescence methods

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Received 22 August 2005; received in revised form 28 February 2006; accepted 1 June 2006

**Abstract**

Nine kinds of radiation-induced luminescence methods, including thermoluminescence (TL) and optically stimulated luminescence (OSL or IRSL), have been applied to the determination of accumulated doses on atomic bomb-explosion exposed roof tiles collected from Hiroshima and Nagasaki epicenter areas. RTL (red TL) measurements of the quartz fraction showed the highest accumulated doses after storage period of 59 years, which were consistent with the previous dose evaluations. However, in addition to the dose estimation with BTL (blue TL) and OSL from the same quartz fractions, all of the accumulated doses from feldspar extracts, including five kinds of TL and IRSL (infra-red stimulated luminescence), resulted in lower values than the RTL dose of quartz grains. The lower accumulated doses from the quartz and feldspar extracts might reflect unstable properties of luminescence centers and/or well-known anomalous fading effects, resulting in the underestimation of accumulated doses. Conclusively, the highest accumulated doses of RTL from atomic bomb-suffered roof tiles are considered to be an evidence of the most stable property of RTL centers in quartz.


*Keywords:* RTL (red thermoluminescence); Quartz grains; Atomic bomb-exposed roof tile; Retrospective dosimetry; OSL; IRSL; Feldspar fraction

## 1. Introduction

Nowadays, a variety of radiation-induced luminescence phenomena have been employed to the dosimetry tools such as thermoluminescence (TL) and optically stimulated luminescence (OSL) using either quartz or feldspar samples (Aitken, 1985, 1998; Bøtter-Jensen et al., 2003). Although quartz and feldspar commonly coexist in burnt archaeological and geological materials, there are a few reports on inter-comparisons of dose evaluations from different luminescence methods using mineral extracts from the same material. In burnt archaeological materials, earlier others have compared the naturally accumulated doses for pottery obtained by measurements of RTL (red TL), BTL (blue TL), and OSL from quartz extracts and IRSL (infra-red stimulated luminescence) from feldspar extracts (Hashimoto et al., 2003, 2005a). The highest naturally accumulated doses, which were obtained from RTL measurements of quartz extracts, led to the determination of luminescence ages that were concordant with the archeologically and stratigraphically predicted ages. Causes of different stability among luminescent sources have been considered to be attributed to relatively easy-to-bleach effects of OSL and BTL in quartz and anomalous fading effects in feldspar. It was concluded that the RTL dating was a favorable dating method when quartz grains were available in burnt archaeological materials from 3500 to 6000 years ago, corresponding to about the ranges of 7–24 Gy (Hashimoto et al., 2005a).

In addition to archeological dating, luminescence measurements of natural minerals are also useful for retrospective dosimetry. The RTL measurements of ceramics and quartz grains were successfully applied to two samples from the JCO (uranium fuel treatment company) critical accident site (Hashimoto et al., 2000) and some atomic bomb-exposed samples in Hiroshima (Hashimoto and Nomura, 2003). Evaluated doses were in concord with the calculated values.

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doi:10.1016/j.radmeas.2006.06.002

In the present paper, nine kinds of luminescence dosimetries were employed for two atomic bomb-exposed roof tile pieces collected from areas close to the epicenters in Hiroshima and Nagasaki. For quartz extracts, RTL, BTL, and OSL measurements were applied while VTL (violet TL), BTL, green TL (GTL), RTL, far-RTL, and IRSL ones were applied to feldspar grains extracted from the roof tiles using a SAR (single aliquot regenerative dose) protocol. Since various luminescent centers were assumed to be simultaneously created in the atomic bomb-exposed roof tiles, orders of the remaining absorbed doses from a variety of luminescence measurements should reflect the storage life (in other words, anti-fading property) of the related luminescent centers. From this viewpoint, the luminescence dose evaluations were carried out for two kinds of roof tile from Hiroshima and Nagasaki after a storage period of 59 years following the nuclear explosions, which is relatively short storage duration in comparison with archaeological materials.

## 2. Experimental

### 2.1. Preparation of measuring samples

Two pieces of roof tiles exposed to atomic bomb radiation have been excavated from the Hiroshima Peace Memorial Park and the Nagasaki Peace Park, where the atomic bombs were exploded on August 6 and 9, 1945, respectively. However, the accurate distances of both sampling locations from the epicenter are unknown. The surface of samples showed the distinctive bubbling pattern due to heat of the explosion.

Quartz and feldspar grains were extracted using a procedure as described in the preceding papers (Hashimoto et al., 2003, 2005a): the preparation was performed in dim red light to avoid optical-bleaching effects. After removing the outer layer (about 2 mm thickness) damaged by the heat, samples were crushed using an agate mortar and a pestle. Subsequently, ferromagnetic components were removed magnetically, followed by immersion in 6 M HCl for 1 day and in 6 M NaOH to remove metallic oxide contamination and organic materials, respectively. Heavy liquid separation using three solutions of sodium polytungstate (2.55, 2.63, and 2.67 $g\,cm^{-3}$) was used to separate the quartz (2.63–2.67 $g\,cm^{-3}$) and the potassium-rich feldspar (2.55–2.63 $g\,cm^{-3}$) fractions. The quartz fraction was further etched with 46% HF to remove the surface layer affected by $\alpha$-rays from surrounding materials. This procedure also removed any extant feldspar grains. Finally, all quartz and feldspar grains were sieved to obtain the 75–150 μm fraction.

### 2.2. Luminescence measurements

An automated TL and OSL-reader system with a small X-ray irradiator (Varian, VF-50J tube), developed for the SAR-protocol (Hashimoto et al., 2002, Nakagawa et al., 2003), was used for all luminescence measurements. The SAR sequence was slightly improved from the ordinal method by replacing the same TL-measurement procedure instead of cut-heat one for the test-dose run (Murray and Wintle, 2000). The dose rate by the X-ray irradiator was 5.5 $Gy\,min^{-1}$ when applying a 100 μm Al absorber, with a 50 kV tube potential and 0.1 mA tube current. Luminescence sensitivity changes during the repeated procedures were corrected by test dose irradiation of 5 Gy. Three aliquots consisting of 5 mg were measured in both cases of TL and OSL (and IRSL) measurements.

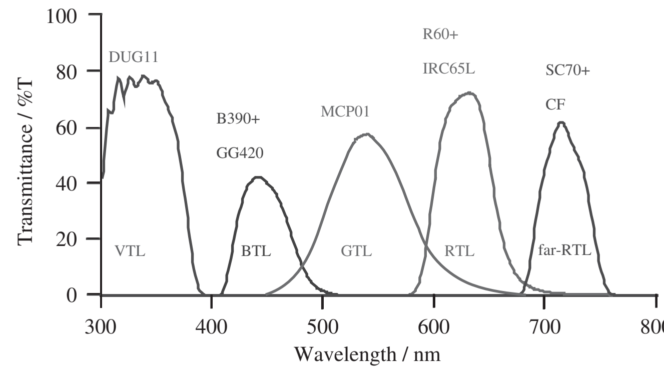


Fig. 1. Optical transmission properties of filter combinations for five kinds of TL measurements.

The color of the TL emission from quartz and feldspar was checked using color photographic images, involving TLCIs (TL color images), and an on-line spectrometric system combined with an image-intensifier photo-diode array (Hashimotto et al. 1989, 1997). On the basis of these results, the far-RTL, RTL, GTL, BTL, and VTL were measured with the detection windows of 700–750, 600–700, 500–600, 400–500, and 300–400 nm, respectively. The transmission characteristics of the used filter combinations are shown in Fig. 1.

OSL measurements were performed using a stimulation source with 16 blue diodes (blue-LED, Nichia, 470Δ20 nm; 12 $mW\,cm^{-2}$); the IRSL stimulation source was converted into 16 IR-LEDs (Hamamatu Photonics, 890Δ50 nm; 28 $mW\,cm^{-2}$). The OSL from quartz aliquot was recorded in every 0.1 s for 100 s while keeping the sample temperature at 125 °C.

The IRSL from feldspar was detected in the range of 350–600 nm wavelengths and the measurement conditions were similar to those for quartz (Hashimoto et al., 2002).

## 3. Results and discussion

### 3.1. TL, OSL, and IRSL

Fig. 2(a) shows TL-contour maps of quartz extracts from the Nagasaki-roof tile; they show a mixture of blue and red TL emissions, comprising peaks at 450–500 and 620–630 nm, respectively. A similar map was obtained from the quartz aliquot from Hiroshima-roof tile. It is to be noted that the RTL intensity is stronger than BTL intensity as also seen in quartz extracts from burnt archaeological materials or old roof tiles (Hashimoto et al., 2005a, b).

Besides the effects of impurities, the different TL properties of quartz extracts are expected to be greatly affected by their thermal history, involving cooling rate and atmospheric condition during the burning process of roof tiles (Hashimoto