Dose Dependence and Palaeodose Estimation for IRSL of Feldspar and Chinese Loess with Differenet Wave Ranges Using an Automated TL/OSL System with Four Optical Paths

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1. Introduction

Thermoluminescence (TL) and optically stimulated luminescence (OSL) emitted from minerals such as quartz and feldspar have been applied to archaeological and geological materials to date these materials (Kennedy and Knopff 1960; Huntley *et al.* 1985). The TL and OSL are caused by the electrons from the electron traps as they recombine with holes in the recombination center. When a mineral crystal has recombination centers of several types, luminescence of several wavelengths with different cross-sections might be emitted. Consequently, the dose dependence of a particular luminescence might differ from the other luminescence of a different wavelength. When applying TL or OSL to dating of archaeological and geological materials, linear dose dependence is preferred along with strong luminescence sensitivity.

Hashimoto *et al* (1986; 1997; 2003) measured TL and infra-red stimulated luminescence (IRSL) color image and TL contour maps. Baril (2004) observed IRSL of feldspars using charge-coupled device (CCD) cameras. In these cases, the samples were irradiated as much as several kGys before taking an image, which is far greater than the accumulated dose by geologic minerals used for luminescence dating.

The TL dating using quartz and feldspar extracted from two tephra were performed with five different optical filters (Nagatomo *et al.* 1999). For the quartz sample, they used three band-pass filters with respective wavelength passes of 415 nm, 500 nm, and 595 nm (± 20 nm, respectively) and estimated almost identical palaeodoses for three wavelengths. For the feldspar sample, the dose dependence of 630 ± 20 nm was the most suitable. They used a TL system with a single photomultiplier (PMT) and changed optical filters for each measurement. However, it is difficult to select the luminescence of optimum wave range to obtain the preferred dose dependence using a luminescence reader with a single PMT because too much sample and time are wasted for the complicated experimental work.

We designed and constructed a new TL/OSL system that has four light paths with four condensing lenses and four PMTs for choosing the suitable luminescence wave range for dating. Two feldspar specimens, albite and orthoclase, were tested to observe the difference of dose dependences for different wave ranges. The reader apparatus was also applied to a polymineral fine grain sample extracted from Chinese loess.

2. Automated TL/OSL System

We designed and constructed an original TL/OSL system (NRL-99-OSTL) in 1999 with two assemblies for heat control: one controlled from room temperature to 500°C and another kept at -150°C by flowing liquid nitrogen (Fig. 1). The TL/OSL system measures the TL and OSL up to 32 aliquots automatically. For OSL measurements, an excitation assemblage with 32-470 nm light emitting diodes (LEDs; Nichia Chemical Industries Co., Ltd., Japan) is installed. It is interchangeable with another assemblage with 32-890 nm LEDs (Hamamatsu photonics Co., Ltd., Japan). Luminescence from a sample is led to a PMT (R1140P; Hamamatsu Photonics

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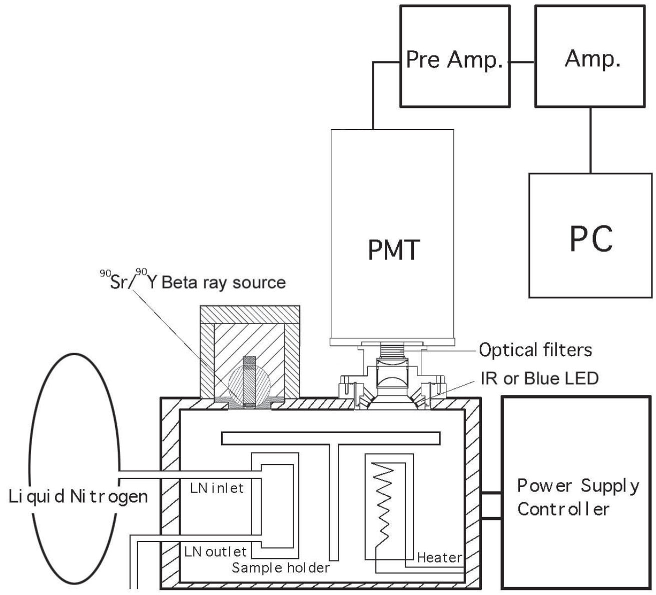

Figure 1 Schematic diagram of NRL-99-OSTL TL/OSL reader.

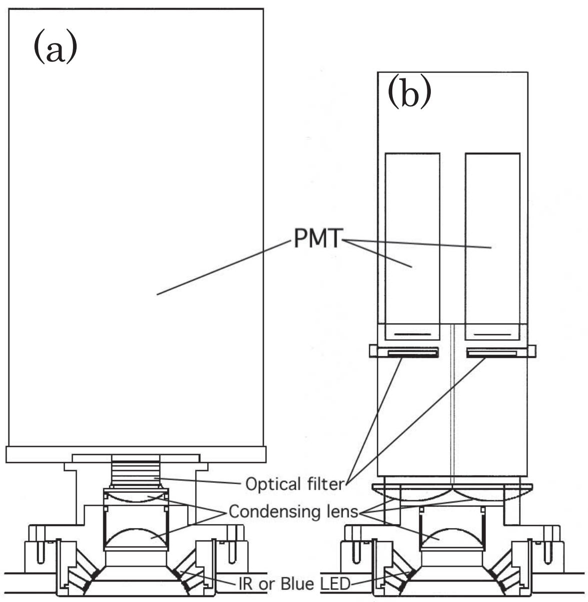

Figure 2 The luminescence detection assembly.
(a) is NRL-99-OSTL and (b) is NUE-05-OSLTL.

Co., Ltd., Japan) housed in a cooling unit (-20°C) through two condensing lenses and an optical filter set.

In this work, NRL-99-OSTL system was improved to detect four-difference wave range luminescence simultaneously and to perform the single-aliquot regenerative-dose (SAR) protocol measurements (Murray and Wintle 2000). The new NUE-05-OSLTL system is equipped with five condensing lenses and four optical filter units and four PMTs (H7360-01; Hamamatsu Photonics Co., Ltd., Japan) instead of two condensing lenses and a PMT of the NRL-99-OSTL system. Arrangements of these luminescence detection systems are shown in Fig. 2. The wave ranges selected by the four lens and optical filter assemblages are usually 300-390 nm, 390-590 nm, 590-650 nm, and 370-650 nm (Fig. 3). A detachable ^{90}Sr-^{90}Y beta source provided by Risø National Laboratory, Denmark is also installed for measurements by SAR protocol (Fig. 1).

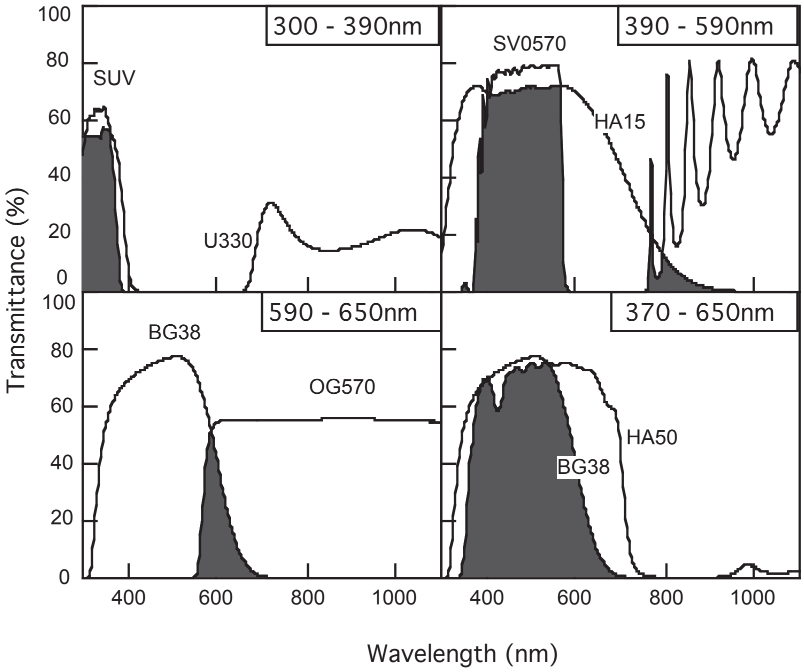

Figure 3 The four transmission wave ranges selected by the optical filters in NUE-05-OSLTL reader.

Comparing with the similar equipment developed by Daybreak Nuclear and Medical System, Inc. (Bortolot 2000), NUE-05-OSLTL provides four condensing lenses instead of a solid quartz light guide array for leading luminescent light to PMTs.

3. Measurements

3.1 Dose Dependence of Feldspars

Two feldspars, Brazilian albite and Arizonan orthoclase, were stimulated with the 890 nm LEDs to observe the dose dependences of IRSL for the four wave ranges. The specimens were gently crushed using a vise to obtain grains smaller than 250 μm; then they were treated using 20% hydrochloric acid (HCl) for 90 min to make the surface of the grains smooth. Finally, the grain size was adjusted from 75 to 150 μm using standard mesh cloth. Before irradiation, the samples were annealed at 350°C for 60 min in an electric furnace. These samples were exposed to 1, 5, 10, 25, 50, 100, 500, 1000, 2000, and 3000 Gy with a ^{60}Co gamma-ray at a dose rate of ca. 0.2 Gy min^{-1}.

The treated feldspar specimens were measured using multiple aliquot additive dose (MAAD) technique (Aitken 1998) with the four-wave ranges TL/OSL system (NUE-05-OSLTL). IRSL measurements were made at a sample temperature of 60°C for 100 s following a preheat treatment (160°C for 60 s) (Shitaoka and Nagatomo 2001). The stimulation intensity of the infrared LED was 86.9 mW cm^{-2} for IRSL measurement. The integrated IRSL signals from 40 to 80 s for each decay curve of four wave ranges (300-390 nm, 390-590 nm, 590-650 nm and 370-650 nm) were used for observing the dose dependence of IRSL intensity.

3.2 Palaeodose and IRSL Age Estimation of Chinese Loess

Chinese loess (ca. 14 ka; Nagatomo *et al.* 2009) collected from the Nihewan Basin in China was crushed in water, and polymineral fine grains (ca. 4-10 μm) (Zimmerman 1971) were separated from coarse grains; then treated with 10% hydrogen peroxide (H_2O_2) for ca. 16 hours. Finally, polymineral fine grains were treated with 20% HCl for 120 min.

MAAD protocol with NUE-05-OSLTL was also adapted to IRSL measurements of the polymineral fine grains to estimate the palaeodose (PD) of the Chinese loess. The measurements were made at a sample temperature of 60°C for 100 s following a preheat treatment (160°C for 60 s) (Shitaoka and Nagatomo 2001).

Five aliquots were used for each natural- and additive-dose measurements. Nonlinear dose dependence in the low-dose region was corrected using samples bleached under sun for 8 hr (Shitaoka and Nagatomo 2001).

The dose rate (annual dose) of the Chinese loess was evaluated from the concentrations of U, Th, and ^{40}K measured using a gamma-ray spectrometer with a highly pure Ge detector.

4. Results and discussion

4.1 Dose Dependence of Feldspars

The IRSL from albite and orthoclase are shown as a function of the irradiated dose from 1 to 3000 Gy in Fig. 4. Dose dependence curves were fitted to an exponential curve. IRSL from albite of four wave ranges shows similar dose dependence over 100 Gy. In the low-dose region, the dose dependences of 300-390 nm IRSL and 370-650 nm IRSL are almost linear, but those of 590-650 nm IRSL and 370-650 nm IRSL are exponential. The IRSL signals of 590-650 nm from orthoclase are several times more intense than those of the other wave ranges, but its saturation starts at a lower dose than those of the other wave ranges. In the low dose region (< 100 Gy), the dose dependence curves of 300-390 nm and 370-650 nm show near linearity compared to albite. Results suggest that the dose dependence of feldspar IRSL of a wavelength might differ from that of the other wavelength, especially in the low-dose region.

4.2 Palaeodose Estimation of Chinese Loess

IRSL of polymineral fine grains extracted from the Chinese loess was detected simultaneously through the four optical paths (300-390 nm, 390-590 nm, 590-650 nm and 370-650 nm) by MAAD protocol using the NUE-05-OSLTL. The dose responses of IRSL were linear up to the additive dose of 200 Gy. Therefore, the equivalent dose (ED) was evaluated from linear regression of the growth curve. The growth curves and the supralinearity correction (SPR) curves are presented in Fig. 5. The linear regressions of the SPR were made using the data for the given doses over 50 Gy. The PD, the sum of ED and SPR, are presented in Table 1 for the four wave ranges. The annual dose of the Chinese loess was 5.01 ± 0.37 mGy a^{-1}. Consequently, IRSL ages for each wave range were, respectively, 13 ± 2 ka, 10 ± 2 ka, 9 ± 2 ka, and 15 ± 4 ka (Table 1).

In the low-dose region of supralinearity correction curves, 300-390 nm and 590-650 nm IRSL show a supralinear response, 390-590 nm and 370-650 nm IRSL are sublinear. It is noteworthy that the supralinearity correction of 370-650 nm IRSL is remarkable compared with those for the other wave range IRSL because of the conspicuous sublinearity. Consequently, the estimated palaeodoses for both 390-590 nm and 590-650 nm IRSL are markedly smaller than those for 300-390 nm and 370-650 nm (Table 1). The PD and IRSL age show good agreement with data of the reference (Nagatomo,

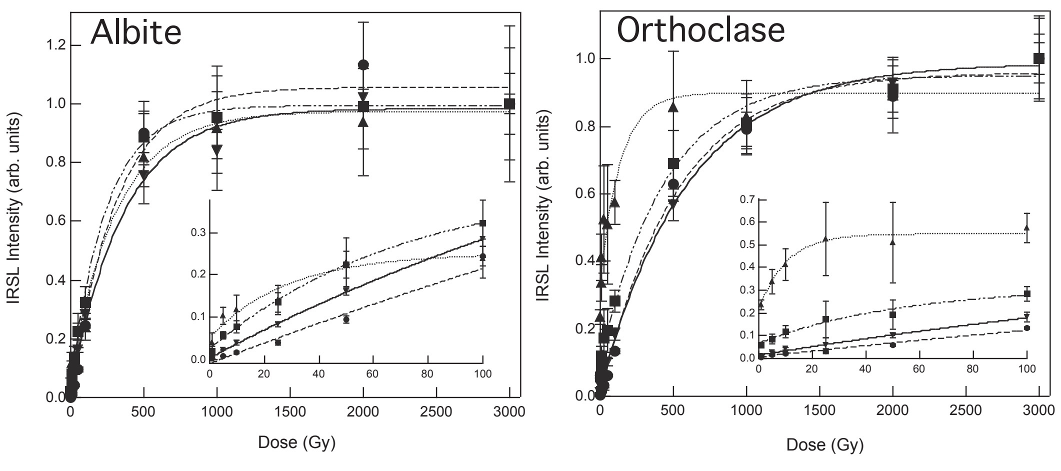

Figure 4 Dose dependences of IRSL from two feldspars, Albite and Orthoclase.
Circle is 300-390 nm, square is 390-590 nm, triangle is 590 - 650 nm and inverse triangle is 370 - 650 nm. The samples were exposed to 1, 5, 10, 25, 50, 100, 500, 1000, 2000, 3000 Gy with a ^{60}Co gamma-ray at a dose rate of ca. 0.2 Gy min^{-1}.

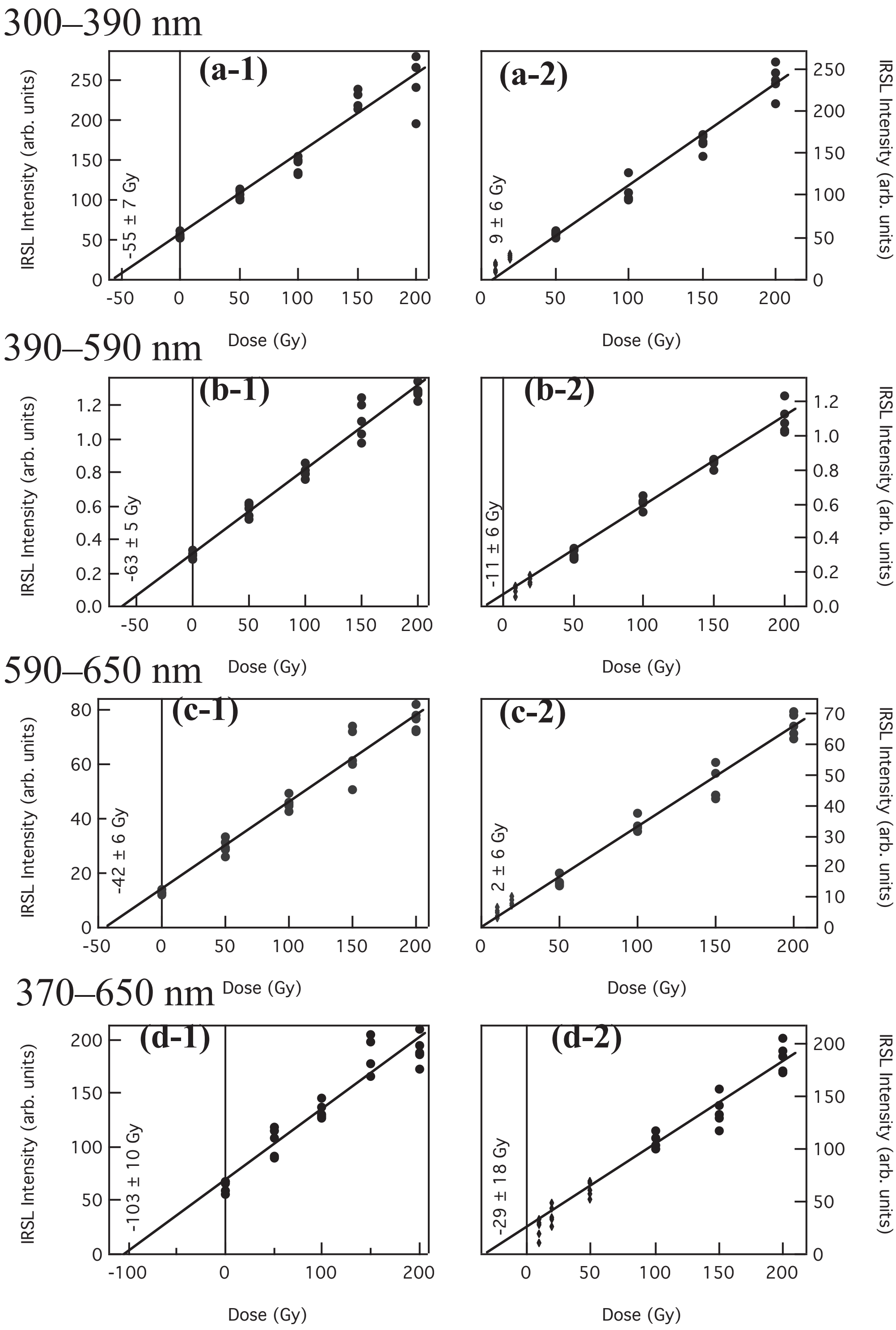

Figure 5 Growth curves of IRSL for four wave ranges.
(a-1), (b-1), (c-1), (d-1) are for the equivalent dose estimation and (a-2), (b-2), (c-2), (d-2) are for the non-linearity correction in the low dose region. In multiple aliquot additive dose protocol, palaeodose was obtained by the equivalent dose plus the non-linearity correction.

2009) except the low values by IRSL measurements using polymineral fine grains with 390-590 nm and 590-650 nm wave ranges. Although the short-term fading tests have not been performed for these samples, the low dose value for 390-590 nm wave range might be attributable to anomalous fading of feldspars in the samples. The low-dose value estimated for 590-650 nm might also be attributed to the fading of orange-red IRSL related to the influence of the yellow emission center (Stokes 2003).

Table 1 The estimated palaeodoses with four-wave ranges IRSL measurement, annual dose and IRSL ages of polymineral fine grains extracted from Chinese loess.

Detection wavelength (nm)	Palaeodose (Gy)	Annual dose (mGy a^{-1})	IRSL age (ka)
350–390	63 ± 10	5.01 ± 0.37	13 ± 2
390–590	52 ± 7		10 ± 2
590–650	44 ± 8		9 ± 2
370–650	74 ± 21		15 ± 4

The authors hope that 300-390 nm IRSL of polymineral fine grains will be checked carefully by other laboratories as an effective tool for use in OSL dating.

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波長４分割同時測定による長石および中国レスの線量依存性と蓄積線量評価

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要　旨：

熱ルミネッセンス（TL）および光ルミネッセンス（OSL）年代測定では、通常一本の光電子増倍管（PMT）と一本のライトガイドを用いて蓄積線量を評価する。しかし、年代測定に供する試料は様々であり、それらから年代測定に適した線量依存性を得られるように波長領域を選択することは難しい。今回、新しいルミネッセンス測定装置 NUE-05-OSLTL を作成した。この装置には、４本のライトガイドと４本の PMT を装着している。４本のライトガイドは、300〜390 nm、390〜590 nm、590〜650 nm そして370〜650 nm に波長を選択した。そして、1〜3000 Gy の γ 線を照射したアルバイトと正長石の赤外励起 OSL（IRSL）における線量依存性を観察した。アルバイトおよび正長石の370〜650 nm の IRSL は100 Gy 以下において、直線的な線量依存性が観察された。その一方で、590〜650 nm の IRSL は他の波長領域に比べて低い線量において飽和する線量依存性が観察された。

以上の測定結果を基に、約14 ka とされる中国レスから抽出した4〜10 μm の多鉱物微粒子を用いて、線量依存性と蓄積線量を評価した。その結果、50 Gy よりも低線量の300〜390 nm および590〜650 nm の波長領域において、IRSL がスープラリニアリティーな線量依存性を観察した。その一方で、390〜590 nm および370〜650 nm の IRSL では、サブリニアリティーな線量依存性を観察した。各波長領域から見積もった蓄積線量は、63±10 Gy、52±7 Gy、44±8 Gy および74±21 Gy となった。この中国レスの年間線量5.01±0.37 mGy ／年で年代に換算すると、13±2 ka、10±2 ka、9±2 ka および15±4 ka となった。このことから、この中国レスにおいては、300〜390 nm の波長領域を選択する必要があることが分かった。

キーワード： 波長四分割、IRSL、線量依存性、蓄積線量