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자연방사선과 인공방사선으로 조사된 석영의 선형변조 광 자극 냉광 빠른 성분의 연대측정과 관련된 광 자극 냉광 특성 연구

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요 약

자연방사선과 인공방사선에 의해 유도된 석영의 각각에 대한 선형변조 광 자극 냉광 빠른 성분들의 비교 연 구가 수행되었다. 우선적으로 측정된 선형변조 광 자극 냉광 신호를 수학적 방법으로 피크 분리하였으며 분리 된 성분들과 관련된 물리적 정보를 얻었다. 분리된 성분 중 빠른 성분들의 자연방사선과 인공방사선의 상대적 면적 비교를 통한 실험에서 석영 시료들은 세 그룹으로 구분되었다. 그룹 A는 상대적 면적 비가 1 값에 가깝게 주어졌으며 그룹 B 와 C 시료들은 1 보다 크거나 작은 값을 보였다. 재현 방사선을 통한 민감도 변화 실험 결과, 연속적인 재현 방사선에 대한 자연방사선과 인공방사선의 빠른 성분들의 상대적 면적 비도 앞의 결과와 같은 시료 그룹을 형성하였다. 결과로서 얻어진 자연방사선과 인공방사선의 빠른 성분들에 대한 민감도 변화는 선형 변조 광 자극 냉광 빠른 성분을 이용하여 얻은 자연축적선량 결정에 영향을 주었음을 보였다.

주요어: 자연/인공방사선, 선형변조 광 자극 냉광, 빠른 성분, 민감도 변화, 자연축적선량

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ABSTRACT: A comparative investigation for the fast component of quartz LM-OSL between a natural and an artificial dose was carried out. Using a mathematical peak deconvolution, the LM-OSL signal was separated and the related physical information was obtained. Three different luminescence groups were observed among the quartz samples in a comparison of the ratios of relative areas for fast components of a natural dose and an artificial dose. The ratios in group A were close to a value of unity, while those in group B and group C showed lower and higher values than unity, respectively. The relative ratios of sensitivity change for the fast component areas during each regenerative measurement were also classified for the above three groups. In addition, it was known that the sensitivity changes of each of the fast components from the natural and the artificial dose influenced the equivalent doses evaluated from the SAR method using the LM-OSL fast component.

Key words: natural and artificial radiation, LM-OSL, fast component, sensitivity change, equivalent dose

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1. INTRODUCTION

It is well known that optically stimulated luminescence (OSL) from quartz is composed of the sum of three or four components with different rates of charge loss and that each component corresponds to various physical characteristics. The components are described as fast, medium, and slow on the basis of the relative decay rates (Smith and Rhodes, 1994; Huntley *et al.*, 1996; Bailey *et al.*, 1997). The fast component is considered to originate from a trap corresponding to the 325° C TL peak (Smith *et al.*, 1986; Wintle and Murray, 1997). In optical dating, this has been of interest,

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| ~ . | _ | $D_{\rm e}$ value | Photoionization cross section $\sigma (\text{cm}^2)^1$ | | | | Relative area |
|--------|----------|----------------------------|--|------------------------|------------------------|------------------------|---------------|
| Sample | Туре | from CW-OSL (Gy 1 g SE) | Fast | Medium | Slow1 | Slow2 | ratio of fast |
| | | (0), 10 52) | | | | | component |
| CJU | Fluvial | 6 ± 0.1 | 2.21×10^{-17} | 5.84×10^{-18} | 6.21×10^{-20} | 4.10×10^{-21} | 0.16 |
| CSM2 | Fluvial | 24 ± 0.4 | 1.97×10^{-17} | 9.24×10^{-19} | 2.44×10^{-20} | 5.39×10^{-21} | 0.03 |
| WD2 | Fluvial | 49 ± 1.3 | 2.02×10^{-17} | 1.19×10^{-18} | 9.13×10^{-20} | 8.05×10^{-21} | 0.13 |
| KA4 | Paleosol | 94 ± 2.6 | 2.21×10^{-17} | 5.61×10^{-19} | 2.63×10^{-20} | 6.14×10^{-21} | 0.03 |
| SH2 | Paleosol | 125 ± 6 | 2.43×10^{-17} | 6.15×10^{-18} | 1.55×10^{-19} | 1.43×10^{-20} | 0.11 |
| CSM3 | Fluvial | 145 ± 4 | 2.69×10^{-17} | 5.63×10^{-18} | 9.21×10^{-20} | 1.27×10^{-20} | 0.05 |
| OS4 | Fluvial | 340 ± 9 | 2.15×10^{-17} | 2.67×10^{-18} | 9.21×10^{-20} | 5.87×10^{-21} | 0.08 |

Table 1. Photoionization cross sections obtained from peak deconvolution of natural LM-OSL.

1. The photoionization cross section is the mean value from three aliquots in each case.

2. The relative contributions of the fast component to the total LM-OSL signal are also indicated.

because of a fast bleaching characteristic, high signal-to-noise ratio, and sufficient stability to be used for quaternary dating.

The estimation of equivalent dose (D_e) is usually carried out by using a single aliquot regenerative dose (SAR) method with constant power stimulation to obtain a continuous-wave OSL (CW-OSL) signal (Murray and Wintle, 2000). The integration of the decay curve after subtraction of a background signal is applied for the first few seconds to select the fast component. However, such an isolation process cannot completely remove the medium and slow components. The existence of these components in the initial part of the CW-OSL signal may distort the D_e determination in the SAR method.

Peak-shaped linear modulation OSL (LM-OSL) is obtained by linearly increasing stimulation from zero. Peak deconvolution of the LM-OSL signal is a useful technique to extract only the fast component from other overlapping components according to each photoionization cross section (Bulur *et al.*, 2000). Especially, comparison of the D_e value from the LM-OSL fast component with that from CW-OSL by using the SAR method has been widely investigated (e.g., Choi *et al.*, 2003; Tsukamoto *et al.*, 2003; Li and Li, 2006). The physical characteristics and sensitivity changes of the LM-OSL fast component have also been examined (e.g., Jain

et al., 2003; Singarayer and Bailey, 2004; Moska and Murray, 2006). Despite these various studies, there have been no attempts to understand how the sensitivity of the fast component is affected by differences between natural and artificial irradiation.

In this study, we first calculated the physical information from each LM-OSL component by using peak deconvolution. Then, we preliminarily investigated the luminescence characteristics of the LM-OSL fast component between the natural dose and the artificial dose for several quartz samples with a wide range of equivalent dose. We also compared the D_e values obtained from the standard SAR method with those obtained only from the fast component separated from the LM-OSL.

2. SAMPLES AND EXPERIMENTAL DETAILS

Seven well-bleached samples from archaeological excavation sites in central Korea were used in this study. Samples of CJU, CSM2, WD2, CSM3, and OS4 were taken from fluvial deposits at the sites, and KA4 and SH2 were taken from paleosols (Table 1). Coarse grains of quartz ($125 \sim 250 \mu$ m) were extracted by routine chemical treatment with 10% HCl, 10% H₂O₂ and 1 hour of concentrated HF treatment. The purity of the resulting quartz against feldspar contamination was tested by in-

frared stimulated OSL measurement.

Quartz OSL measurements were performed by using an automated TL/OSL reader (Riso TL-DA-15) installed at the central laboratory of Kangwon National University and the Chungcheong Cultural Properties Research Institute, which was equipped with optical excitation units based on blue LED (470 ± 30 nm) arrays delivering about 50 mW/cm² passing through GG-420 filters. The luminescence was measured through a 7.5 mm thick U340 filter in front of a bialkali PM tube. At room temperature, all laboratory irradiations were carried out by using a calibrated ⁹⁰Sr/⁹⁰Y beta source, whose dose rate was 125 mGy/s at the sample position.

Before the LM-OSL measurement, D_e values using CW-OSL were previously determined, for future comparison, by using the conventional SAR method (Table 1). All CW-OSL measurements were made at 125°C, followed by 260°C preheating for 10 s to remove unstable signals. Samples received a total of 40 s stimulation and the CW-OSL signal of the initial 0.5 s stimulation was used for making the dose response curve.

3. RESULTS AND DISCUSSION

3.1 Deconvolution of LM-OSL curves

LM-OSL measurements were made to separate the several overlapping components. All natural quartz samples were preheated to 260° C for 10 s. LM-OSL signals were recorded at 125° C by linearly increasing the stimulation LED power from 0 to 50 mW/cm² over 3600 s. Three blank aliquots were measured under the same conditions, to subtract background signals and electrical noise.

LM-OSL curves were analyzed by assuming a linear combination of four first-order kinetics:

$$L(t) = \sum L_i(t) = \sum n_{0i} b_i \left(\frac{t}{T}\right) \exp\left(-\frac{b_i t^2}{2T}\right)$$
(1)

where n_0 is the initial trap population, b is the detrapping probability proportional to the photoionization cross section σ and T is the total measurement time (Bulur et al., 2000). The deconvolutions were performed by using commercial software SigmaPlot TM , which employs the Marquardt-Levenberg algorithm for non-linear fitting. The representative LM-OSL curve and each component from sample SH2 are shown in Fig. 1. Residuals were also calculated, to judge the reliability of the fitting results (Residual = 100*(I_{exp}-I_{decon})/I_{exp}). The resultant values of photoionization cross section for each component are summarized in Table 1. These are consistent with the result by Jain et al. (2003). The relative contributions of the fast component to the total LM-OSL signal are also indicated.

3.2 Relative area ratio for LM-OSL components between natural and artificial dose

We divided each quartz sample into two parts for comparison of the ratio of relative area between natural and artificial dose. One was directly used for natural LM-OSL measurement, and the other was bleached under sunlight for



Fig. 1. An example of LM-OSL peak deconvolution according to magnitude of photoionization cross section for sample SH2.

six hours before measurement. Then, the artificial beta dose was irradiated to each sun-bleached sample. LM-OSL measurements from the natural and the artificial dose were made under the same conditions as for the above measurements by using three aliquots in each case and then the CW-OSL responses to a 5 Gy test dose were measured to normalize the mass difference between aliquots and to correct any sensitivity change due to bleaching.

Based on the values of photoionization cross section (Table 1), LM-OSL curves were separated for four first-order components. Comparisons of the relative area ratios of the components between natural and artificial dose are shown in Fig. 2. Apart from the medium and slow components, three different luminescence groups for the fast component were observed among the quartz samples. The ratios in group A were close to a value of unity, while those in group B and group C showed lower and higher values than unity, respectively.

3.3 Comparison of sensitivity change due to bleaching for the fast component areas

Three representative samples were selected

from the different luminescence groups in Fig. 2: group A is CJU, group B is SH2, and group C is CSM3, because the amount of other quartz samples was not sufficient to make any further measurement. For the natural-dose part of each sample, seven regenerative LM-OSL measurements with the same beta dose irradiation were firstly performed to investigate the sensitivity change due to bleaching for the LM-OSL fast component area. All areas were normalized to the area of the first LM-OSL measurement. For the corresponding artificial-dose parts, the regenerative LM-OSL measurements and normalization were carried out in the same way. The relative ratio of sensitivity change due to bleaching for the fast component area was obtained by dividing the area change for the artificial dose by that for the natural dose.

Fig. 3 shows the relative ratios of sensitivity change due to bleaching, measured by LM-OSL fast component areas, for samples CJU, SH2, and CSM3, obtained by using the mean values obtained from three aliquots in each case. From the repeated LM-OSL measurements, the relative ratios of sensitivity change due to bleaching from the fast component areas for the natural dose and the artificial dose were also classified into three groups, showing a similar tendency to that



Fig. 2. Comparison of relative area ratios for four LM-OSL components between natural and artificial dose. Three different luminescence groups for the fast components were observed among seven quartz samples.



Fig. 3. Relative ratios of sensitivity change due to bleaching for LM-OSL fast component areas for samples CJU (group A), SH2 (group B), and CSM3 (group C).

of group A, group B, and group C in Fig. 2.

3.4 Comparison of De values obtained between CW-OSL and the LM-OSL fast component

In order to examine how the D_e value is affected by the sensitivity change of the fast component from natural and artificial irradiation, the SAR method using the LM-OSL fast component was applied to quartz samples CJU (group A), SH2 (group B), and CSM3 (group C), of which the D_e values were previously measured by the CW-OSL SAR method. LM-OSL measurements were made at 125°C, followed by 260°C preheating for 10 s to remove unstable signals. The sensitivity change of the fast component was corrected by using the CW-OSL intensity derived from the initial 0.5 s integration.

On comparing the D_e values obtained by CW-OSL with those found by using the fast component of LM-OSL, consistent result was obtained for the sample CJU (group A) only, which showed that the ratio of sensitivity change for the LM-OSL

CSM3

С

fast component was close to a value of unity. However, the ratio of sensitivity change for the LM-OSL fast component for the sample SH2 (group B) was less than unity, which made the regeneration curve for the fast component gentler than that for CW-OSL and consequently resulted in a larger equivalent dose when using the fast component (Fig. 4a). In contrast to the sample SH2, the ratio of sensitivity change for the LM-OSL fast component for the sample CSM3 (group C) was greater than unity, which caused a steeper regeneration curve for the fast component than for CW-OSL and consequently resulted in a smaller equivalent dose when using the fast component (Fig. 4b). For De determination, data was fitted by a single exponential saturating function expressed by $y = a^{(1-exp(-x/b))}$.

4. SUMMARY

With the use of several quartz samples, a comparative study on the fast component of quartz

 103 ± 9

| | • | * | 8 |
|--------|-------|---|--|
| Sample | Group | D _e from CW-OSL (Gy, 1σ SE) | <i>D</i> _e from LM-OSL fast comp. (Gy, 1σ SE) |
| CJU | А | 6 ± 0.4 | 7.5 ± 2.0 |
| SH2 | В | 125 ± 6 | 150 ± 12 |

 145 ± 4

Table 2. Equivalent doses resulting from CW-OSL and fast component of the LM-OSL using SAR method.



Fig. 4. Examples of determination of equivalent dose by using the SAR method applied to the LM-OSL fast component: sample SH2 (a), and sample CSM3 (b).

LM-OSL separated using peak deconvolution between a natural and an artificial dose was undertaken. From this study, we found that:

First, in the comparison of the relative area ratios for the fast component in the LM-OSL measurements for the natural and the artificial dose, three different luminescence groups were observed among the quartz samples. The ratios in group A were close to a value of unity, while those in group B and group C showed lower and higher values than unity, respectively.

Second, the relative ratios of sensitivity change due to bleaching for the fast component areas for the natural dose and the artificial dose were also classified into three groups, showing a similar tendency to that for the above three groups.

Third, on comparing the equivalent dose from CW-OSL with those obtained from the fast component of LM-OSL, consistent results were obtained for the sample of group A only.

However, further study with more quartz samples should not be excluded.

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