

## THERMOLUMINESCENCE PROPERTIES OF LiF:Mg,Cu,Na,Si PELLETS IN RADIATION DOSIMETRY

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**Abstract** — Sintered LiF:Mg,Cu,Na,Si thermoluminescence (TL) pellets have been developed for application in radiation dosimetry. LiF:Mg,Cu,Na,Si TL pellets were made from TL powders using a sintering process, that is, pressing and heat treatment. These pellets have a diameter of 4.5 mm, and a thickness of 0.8 mm are blue in colour and have a mass of 28 mg each. After 400 pellets had been produced they were irradiated with  $^{137}\text{Cs}$  gamma radiation and samples having a sensitivity within a  $\pm 5\%$  standard deviation were selected for experimental use. In the present study, the physical and dosimetric properties of LiF:Mg,Cu,Na,Si TL pellets were investigated for their emission spectrum, dose response, energy response and fading characteristics. Photon irradiation for the experiments was carried out using X ray beams and a  $^{137}\text{Cs}$  gamma source at the Korea Atomic Energy Research Institute (KAERI). The average energies and the dose were in the range of 20–662 keV and  $10^{-6}$ – $10^2$  Gy respectively. The glow curves were measured with a manual type thermoluminescence dosimetry reader (system 310, Teledyne) at a constant nitrogen flux and a linear heating rate. For a constant heating rate of  $5^\circ\text{C}\cdot\text{s}^{-1}$ , the main dosimetric peak of the glow curve appeared at  $234^\circ\text{C}$ , its activation energy was 2.34 eV and the frequency factor was  $1.00 \times 10^{23}$ . The TL emission spectrum appeared at the blue region centred at 410 nm. A linearity of photon dose response was maintained up to 100 Gy. The photon energy responses relative to the  $^{137}\text{Cs}$  response were within  $\pm 20\%$  in the overall photon energy region. No fading of the TL sensitivity of the pellets stored at room temperature was found over the course of a year. Therefore LiF:Mg,Cu,Na,Si TL pellets can be used for personal dosimetry, but more research is needed to improve the characteristics for repeated use.

### INTRODUCTION

ICRP recommend that a personal dose should be maintained as low as reasonably achievable (ALARA) to protect against the stochastic effect predicted by the linear non-threshold model in their report<sup>(1)</sup>. Precise dose evaluation is needed in the low-dose ranges of  $10^{-4}$ – $10^{-1}$  mGy to maintain this ALARA principle, and it can be achieved by using TL materials which are more sensitive than existing ones.

LiF:Mg,Ti TL material was first sold commercially as TLD-100<sup>(2)</sup> and it is still used for personal monitoring even though it has a low sensitivity. In 1978, Nakajima *et al.*<sup>(3)</sup> developed LiF:Mg,Cu,P TL material which has a sensitivity 20 to 40 times greater than the widely used TLD-100, and much research is now being done on this material in China (GR-200), Poland (MCP-N) and the USA (TLD-100H)<sup>(4)</sup>.

A new TL material, LiF:Mg,Cu,Na,Si phosphors, with a higher sensitivity than LiF:Mg,Cu,P and good fading characteristics, has been developed by Doh *et al.*<sup>(5)</sup> in Korea, and sintered LiF:Mg,Cu,Na,Si pellets have recently been developed at the Korea Atomic Energy Research Institute (KAERI)<sup>(6)</sup>. Because TL pellets have a totally different material structure and dosimetric characteristics from TL phosphors due to the high-temperature sintering process, research on the physical and dosimetric characteristics of these pellets is needed.

In the present work the physical and dosimetric

characteristics of LiF:Mg,Cu,Na,Si TL pellets were investigated. The glow curves were separated by a peak analysis program, and the activation energy and frequency factor calculated for the main peak. The three-dimensional emission spectrum was measured with the heating temperature and frequency. The energy response in the range of 20–662 keV, the dose response from  $10^{-6}$  to  $10^2$  Gy and the fading rate for a storage period of 1 year were also investigated for application to a personal dosimetry system

### MATERIALS AND METHODS

LiF:Mg,Cu,Na,Si TL pellets were made from TL phosphors by pressing at room temperature followed by a sintering process. These pellets have a diameter of 4.5 mm, are 0.8 mm thick with a blue colour and have a mass of 28 mg each. After 400 pellets had been produced they were irradiated with  $^{137}\text{Cs}$  gamma radiation and samples having a sensitivity within a  $\pm 5\%$  standard deviation were selected for experimental use.

The TL glow curves and intensities of the LiF:Mg,Cu,Na,Si TL pellets were measured using a commercial TLD reader (system 310 TLD reader: Teledyne Brown Engineering) controlled by a personal computer. Measurements were carried out with a linear heating rate of  $5^\circ\text{C}\cdot\text{s}^{-1}$  in a nitrogen flow of  $\approx 70$  kPa.

These pellets were irradiated with  $^{137}\text{Cs}$  gamma and X ray radiation on the  $10 \times 10 \times 2$  mm PMMA phantom at KAERI to investigate the physical structure, energy response, dose response, reusability and fading characteristics. The irradiating energies were in the range of 20 to 662 keV.

## RESULTS AND DISCUSSION

## Glow curve and TL sensitivity

Figure 1 shows the glow peak separated from the glow curve of LiF:Mg,Cu,Na,Si TL pellets using a TL analysis program. The heating rate was  $5^{\circ}\text{C}\cdot\text{s}^{-1}$  and the reading temperature was up to  $320^{\circ}\text{C}$ . The glow curve can be separated into five peaks at 104, 145, 189, 234 and  $268^{\circ}\text{C}$ . Of these peaks, the main peak which can be used for dose evaluation is the fourth peak (the  $234^{\circ}\text{C}$  peak), and the activation energy and the frequency factor of the main peak analysed by the TL analysis program are  $2.34\text{ eV}$  and  $1.00 \times 10^{23}\text{ s}^{-1}$  respectively.

## TL emission spectrum

Figure 2 shows a three-dimensional emission spectrum for TL pellets as a function of heating temperature and wavelength. Figure 2(a) shows the three-dimensional distribution of the TL emission, and Figure 2(b) shows the contour line distribution of the TL emission. The wavelength of the main peak is at  $410\text{ nm}$  with full width at half maximum (FWHM) of the  $92\text{ nm}$  monospectrum. The wavelengths of the low-temperature peaks have low intensities, but all of them are distributed to  $410\text{ nm}$ .

These characteristics are similar to those of the LiF:Mg,Ti pellets, which have a  $410\text{--}460\text{ nm}$  main peak emission spectrum with  $120\text{--}160\text{ nm}$  FWHM, and are slightly different from those of the LiF:Mg,Cu,P pellets which have a  $370\text{ nm}$  main peak emission, because the emission spectrum of LiF:Mg,Cu,P is dependent on the activator  $\text{P}^{(7)}$ . The TL spectrum distribution of LiF:Mg,Cu,Na,Si pellets corresponds very well to the spectral sensitivity of the photomultiplier tube of our TLD reader.

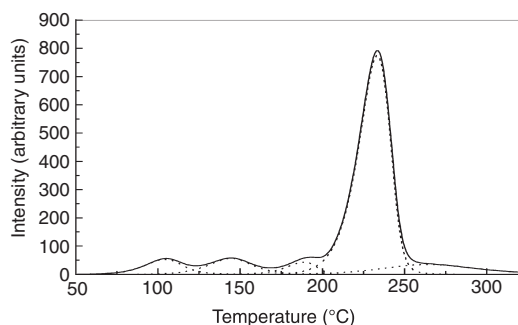


Figure 1. The typical glow curve for LiF:Mg,Cu,Na,Si TL pellets. Pellets were read out to  $320^{\circ}\text{C}$  using a linear heating rate of  $5^{\circ}\text{C}\cdot\text{s}^{-1}$ .

## Energy dependence

Figure 3 and Table 1 show the relative photon energy response,  $f(E)$ , normalised to the  $^{137}\text{Cs}$  radiation. A maximum  $f(E)$  of 1.20 was observed at  $48\text{ keV}$ , and this  $f(E)$  was flattened within  $\pm 20\%$  for the entire energy range. When a TL badge system is designed using LiF:Mg,Cu,Na,Si TL pellets it may not be necessary to use an energy compensation filter system to get the flattening energy response.

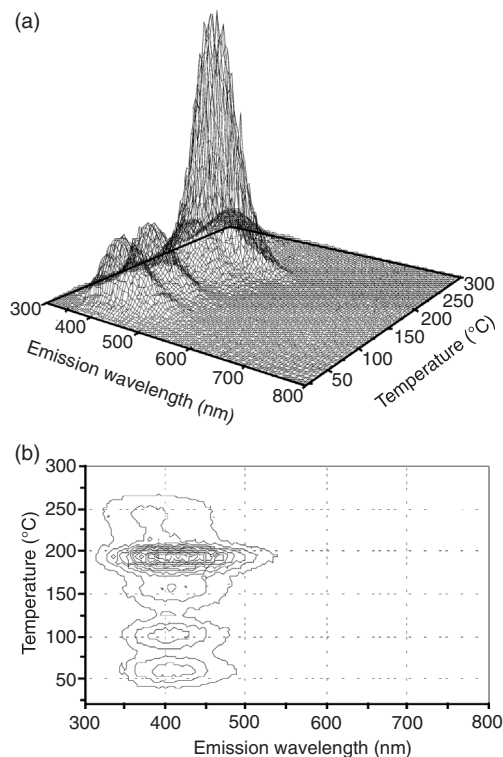


Figure 2. TL emission spectrum for LiF:Mg,Cu,Na,Si pellets: (a) isometric plot, (b) the corresponding contour plot.

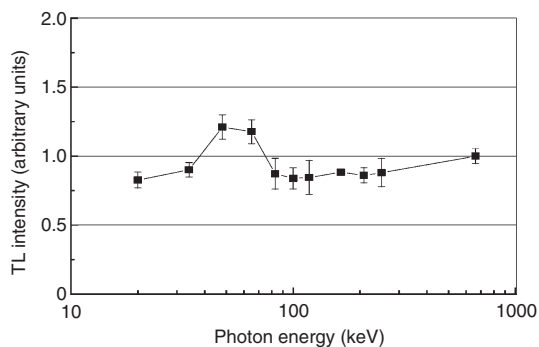


Figure 3. Photon energy response of LiF:Mg,Cu,Na,Si pellets.

**Dose dependence**

Figure 4 shows the dose response of LiF:Mg,Cu,Na,Si TL pellets over a range from  $10^{-6}$  to 100 Gy exposed to  $^{137}\text{Cs}$  gamma radiation with the height of the main glow peak. The TL response of the peak is linear from  $10^{-4}$  up to 100 Gy with no observed superlinearity or saturation.

**Reusability**

LiF:Mg,Cu,Na,Si TL pellets were irradiated with 10 mGy  $^{137}\text{Cs}$  radiation and the TL sensitivity was measured. These procedures were repeated eight times. The change in the TL sensitivity is shown in Figure 5. These pellets show a sensitivity change of about a 10% decrease within a reliability of 95% after a repeat of eight times. The PTB standard<sup>(8)</sup> says that the TL sensitivity changes for a personal dosimetry system after reusing 20 times should be less than  $\pm 5\%$ . To satisfy this standard for LiF:Mg,Cu,Na,Si TL pellets better sintering and reading conditions need to be found for continuous experiments.

**Table 1. Relative photon energy response f(E) for LiF:Mg,Cu,Na,Si pellets. The reference radiation was a  $^{137}\text{Cs}$  source.**

| Beam quality      | Mean energy (keV) | f(E)            |
|-------------------|-------------------|-----------------|
| M30               | 20                | $0.82 \pm 0.06$ |
| M60               | 34                | $0.90 \pm 0.05$ |
| N60               | 48                | $1.20 \pm 0.09$ |
| N80               | 65                | $1.18 \pm 0.09$ |
| N100              | 83                | $0.87 \pm 0.11$ |
| N120              | 100               | $0.84 \pm 0.08$ |
| N150              | 118               | $0.85 \pm 0.12$ |
| N200              | 164               | $0.88 \pm 0.01$ |
| N250              | 208               | $0.86 \pm 0.06$ |
| N300              | 250               | $0.88 \pm 0.10$ |
| $^{137}\text{Cs}$ | 662               | $1.00 \pm 0.05$ |

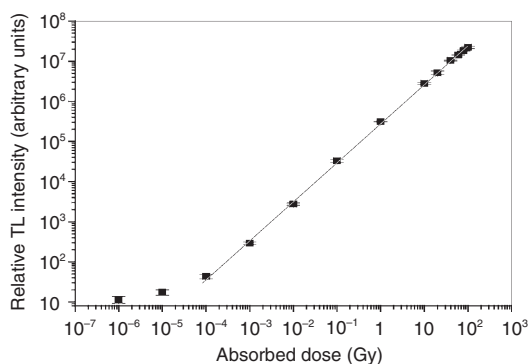


Figure 4. Dose response for LiF:Mg,Cu,Na,Si pellets.

**Fading**

The fading experiment was carried out over a period of 10 months after irradiation. The pellets were irradiated by 10 mGy  $^{137}\text{Cs}$  radiation. The fading characteristics are shown in Figure 6. The TL responses were normalised to the TL response of pellets obtained 2 h after irradiation. When pellets were stored at a room temperature there was no decrease in intensity after 10 months.

**CONCLUSIONS**

The dosimetric properties of LiF:Mg,Cu,Na,Si TL pellets made from TL powder using a sintering process, that is, pressing and heat treatment, have been investigated for glow curve, emission spectrum, dose response, energy response and reusability characteristics.

The glow curve can be separated into five peaks, and the activation energy and the frequency factor of the main peak were 2.34 eV and  $1.00 \times 10^{23} \text{ s}^{-1}$  respectively. The TL emission spectrum appeared at the blue region centred at 410 nm, and the linearity of the photon dose response was maintained up to 100 Gy. The photon energy responses relative to  $^{137}\text{Cs}$  response were within  $\pm 20\%$  in the overall photon energy region. No fading

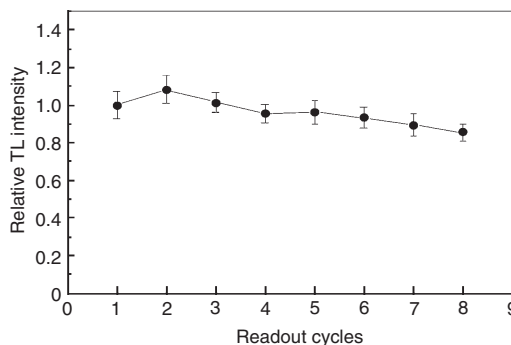


Figure 5. Reusability of LiF:Mg,Cu,Na,Si TL pellets.

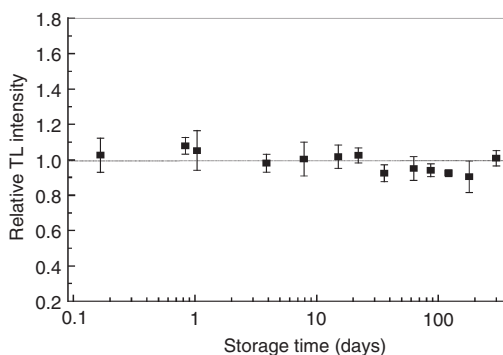


Figure 6. Fading characteristics of LiF:Mg,Cu,Na,Si TL pellets.

of TL sensitivity of the pellets stored at room temperature was found after a year.

From the above results, LiF:Mg,Cu,Na,Si TL pellets can be applied for personal dosimetry, but more research is needed to improve the characteristics of repeated use.

#### ACKNOWLEDGEMENT

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