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New LiF:Mg,Cu,Si TL Material (New KLT-300) with a Low Residual Signal and High Thermal Stability

Jang-Lyul Kim, Jung-Il Lee and Si-Young Chang
Korea Atomic Energy Research Institute, P.O Box 105 Yuseong, Daejeon 305-600,

1. Introduction

LiF-based TL materials have been widely used for a radiation dosimetry due to their various advantages. LiF:Mg,Cu,P is one of the most sensitive tissue equivalent TL materials, about 30 times higher than that of LiF:Mg,Ti (TLD-100)[1,2], but it has two main drawbacks: a thermal loss of the TL sensitivity in LiF:Mg,Cu,P annealed at temperature above 240 ^{o}C; a relatively high residual signal. Recently LiF:Mg,Cu,Na,Si[3] TL material was developed to overcome these drawbacks at the KAERI and there were some improvements in reducing the residual signal but they were not significant.

The newly developed LiF:Mg,Cu,Si TL material has a significantly lower residual signal and a better stability to a heat treatment. In this paper, the preparation method and some dosimetric properties (sensitivity and residual signal) of the LiF:Mg,Cu,Si TL material are presented.

2. Methods and Results

2.1 Sample preparation

A proper activation process is essential for the thermoluminescence properties of a powder and sintered pellets. Usually, two kinds of activation processes are performed for preparing LiF-based TL material. One is an activation by melting and the other is an activation by granulation. In this work, the granulation method was used for an activation. Granulation is a process of a growth of LiF micro-crystals from a size of about 5 μm to a size of about 100-300 μm. This growth can be performed at temperatures well below the LiF melting point in the presence of activators. The micro-crystals, grown in these conditions, show thermoluminescence properties. After that the sintering method was used to produce pellet-type LiF:Mg,Cu,Si TL detectors. The detailed sample preparation procedures are as follows: with LiF powder, 0.2 mol % of $MgSO_4 .7H_2O$, 0.05 mol % of $CuSO_4.5H_2O$, and 0.9 mol % of SiO_2 were mixed in distilled water and then dried at a temperature of 150 ^{o}C. For the activation process of the LiF crystal, the mixture of the LiF and 3 dopants as mentioned above were heated in an electric furnace at a temperature well below the melting point of LiF for 30 min in air. The material was quickly cooled to room temperature after an activation process and then pulverized. The pulverized crystals were rinsed with hydrochloric acid to remove any impurities between the micro-crystals, and then the hydrochloric acid in the material was removed by washing it with distilled water and then drying it. By a sieving with the reference sieve, the crystals of a grain size under 50- 150 μm were selected and abstracted. The abstracted crystals were fabricated to a pellet-type, with a size of 4.5 mm in diameter and 0.8 mm in thickness by a pressurizing with a mechanical press. The pressed pellet was sintered in an electric furnace at 835 ^{o}C under a nitrogen gas atmosphere and quickly cooled on a frozen aluminum plate.

2.2 The effect of dual-step annealing method

A main origin of the residual signal of the LiF-based TL material is the high-temperature peak which usually appears after the main dosimetric glow peak. In this work, a dual-step annealing method was introduced and applied to reduce the high-temperature peak as a final step of the sample preparation procedures. The dual-step annealing method is as follows: the sample is firstly annealed at 300 ^{o}C for 10 min and then quickly cooled, after that the sample is annealed at 260 ^{o}C for 10 min then quickly cooled again. Figure 1 shows the effect of the annealing treatments on the glow curve structure of LiF:Mg,Cu,Si. The glow curve for the sample annealed at 260 ^{o}C for 10 min has a relatively high high-temperature peak, but the high-temperature peak was significantly reduced in the case of the sample annealed at 300 ^{o}C for 10 min. In the case of the sample annealed by the dual-step annealing method, the high-temperature peak was also effectively reduced and the main-peak was significantly intensified. These results show that the dual-step annealing method is a very efficient method to reduce the high-temperature peak in the glow curve of the LiF:Mg,Cu,Si TL material.

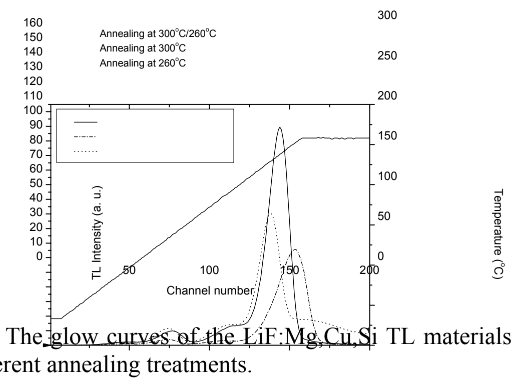

Figure 1. The glow curves of the LiF:Mg,Cu,Si TL materials with different annealing treatments.

2.3 Residual signal

The residual signal, which is defined as the percentage ratio of the second readout to the first readout with exactly the same reading programme, test for this new TL detector was carried out. The reading temperature profile was as follows. Prepared LiF:Mg,Cu,Si TL detector was preheated at 135 ^{o}C for 10 sec in the reader and then heated up to 260 ^{o}C with the heating rate of 10 ^{o}C /sec after an irradiation by a Cs-137 source with a dose of 10 mGy. After an annealing of the sample for a reuse, at 260 ^{o}C for 10 min in an electric furnace, the TL signal of the sample was measured. The Harshaw Model 4500 TLD Reader was used for this test. Table 1 shows the residual signal of the new LiF:Mg,Cu,Si TL detector and that for the other widely used TL detectors for a comparison. The residual signal of the new TL detector is 0.04 which is about 25 times less than GR-200A (LiF:Mg,Cu,P) and 15 times less than KLT-300 (LiF:Mg,Cu,Na,Si).

Table 1. The residual signals of the new LiF:Mg,Cu,Si and some LiF-based TL materials.

TL Materials	Maximum reading temperature (^{o}C)	Residual signal (%)
KLT-300 (LiF:Mg,Cu,Na,Si)	245	0.57
Chinese LiF:Mg,Cu,Na,Si	260	0.20
GR-200A (LiF:Mg,Cu,P)	240	0.92
New LiF:Mg,Cu,Si	260	0.04

2.4 Sensitivity

The sensitivity of this new TL detector was compared with TLD-100. To do this, five samples of both LiF:Mg,Cu,Si and TLD-100 were prepared and the TL glow curves were measured after an irradiation with a test dose. It is worth mentioning that the TLD-100 detectors were annealed under the conditions of 1 h at 400 ^{o}C and 2 h at 100 ^{o}C before an irradiation. The average values for both types of TL detectors were measured and the relative sensitivity was calculated. Figure 2 shows the typical glow curves of the new LiF:Mg,Cu,Si TL detector and TLD-100 after an irradiation with a dose of 10 mGy by a Cs-137 source. The heating rate of the sample was 10 ^{o}C/s and the maximum reading temperature was 260 ^{o}C. The relative sensitivity of this new LiF:Mg,Cu,Si TL detector is about 20 times higher than that of TLD-100.

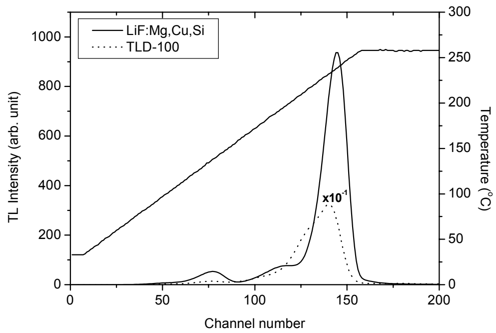

Figure 2. . The typical glow curves of the new LiF:Mg,Cu,Si TL detector and TLD-100 after irradiation with dose of 10 mGy by Cs-137 source.

3. Conclusion

A highly sensitive pellet-type LiF:Mg,Cu,Si TL detector with a extremely low residual signal and a high thermal stability has been newly developed. The preparation procedures of the new TL detector were composed of six steps: mixing of the raw chemicals, activation, pulverizing, pressing, sintering and an annealing. The activation was carried out by a granulation method.

A dual-step annealing method was introduced and this proved to be a very efficient method to reduce the high-temperature peak which is an origin of the residual signal. The residual signal was estimated to be about 0.04 % which is about 25 times less than GR-200A (LiF:Mg,Cu,P) and 15 times less than KLT-300 (LiF:Mg,Cu,Na,Si). The TL sensitivity of this new TL detector was about 20 times higher than that of TLD-100 by the light integration measurements.

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