Journal of the Korean Nuclear Society
Volume 36, Number 1, pp.47~52, February, 2004

Thermoluminescent Characteristics of Newly Developed LiF:Mg,Cu,Na,Si TL Detectors

J. I. Lee, J. L. Kim, and S. Y. Chang
Korea Atomic Energy Research Institute
150 Dukjindong, Yuseong-gu, Daejeon, 305-353, Korea
ex-jilee@kaeri.re.kr

(Received May 29, 2003)

Abstract

Recently, a new sintered pellet-type LiF:Mg,Cu,Na,Si TL detector which has a high sensitivity and good reusability, named KLT-300(KAERI LiF:Mg,Cu,Na,Si TL detector), was developed by the variation of the dopants concentrations and the parameters of the preparation procedure at KAERI (Korea Atomic Energy Research Institute). In this study, the thermoluminescent characteristics of the newly developed TL detectors were investigated. The sensitivity of the TL detector was compared with that of the TLD-100 by light integration. The dose linearity of the detector was tested from 10^{-6} Gy up to 30 Gy. The dose response was very linear up to 10 Gy and a sublinear response was observed at higher doses. The energy response of the detector was studied for photon energies from 20 keV to 662 keV. The result shows that a maximum response of 1.004 at 53 keV and a minimum response of 0.825 at 20 keV were observed. The reproducibility study for the TL detector was also carried out. The coefficients of variation for each detector separately did not exceed 0.016, and for all the 10 detectors collectively was 0.0054. Lower limit of detection for the detector was investigated at 70 nGy by the Harshaw 4500 TLD Reader and the residual signal of the TL detector was found to be 0.57 %.

Key Words : thermoluminescence, TLD, LiF

1. Introduction

During the last few years the LiF:Mg,Cu,Na,Si thermoluminescent(TL) material, doped with four different activators, has been studied for a practical pellet-type TL detector at Korea Atomic Energy Research Institute (KAERI)[1, 2] since it was introduced by Doh in Korea[3], and sintered LiF:Mg,Cu,Na,Si pellet has been developed[4]. In the report, the sintered pellet was made from the optimized powder-type LiF:Mg,Cu,Na,Si TL material. Those optimum concentration of dopants were Mg 0.6 mol%, Cu 0.8 mol%, Na 1.8 mol% and Si 1.8 mol%. The thermoluminescence properties of the sintered pellets were reported, but the sensitivity of the developed TL detector did not exceed 50% of the Chinese GR-200A and MCP-N, and had poor reusability, which was a

10% decrease of the readout values after a reuse of 8 times[5]. Therefore, there is a need to develop a new pellet-type LiF:Mg,Cu,Na,Si TL detector which has good reusability and high sensitivity.

Recently, a new sintered pellet-type LiF:Mg,Cu,Na,Si TL detector which has a high sensitivity and good reusability, named KLT-300(KAERI LiF:Mg,Cu,Na,Si TL detector), was developed by the variation of the dopants concentrations and the parameters of the preparation procedure at KAERI[6]. These newly optimized dopant concentrations of the LiF:Mg,Cu,Na,Si TL detector are Mg 0.2 mol%, Cu 0.05 mol%, Na and Si 0.9 mol%. These dopant concentrations are significantly lower than that of the previous report.

So, in this paper, some thermoluminescent characteristics of the newly developed LiF:Mg,Cu,Na,Si TL detector are reported. The parameters of the glow curve, TL sensitivity, dose response, energy response, reproducibility (reusability) and the detection threshold as well as the residual signal of the TL detector were investigated.

2. Materials and Methods

2.1. Sample Preparation

The sample preparation procedures were as follows. With the LiF powder, 0.2 mol% of $MgSO_4 \cdot 7H_2O$, 0.05 mol% of $CuSO_4 \cdot 5H_20$, and 0.45 mol% of $Na_2O \cdot 2SiO_2 \cdot 9H_2O$ were mixed in distilled water by a magnetic stirrer. The mixture of LiF and the dopants as above were dried and heated in an electric furnace at a temperature well below the melting point of LiF for 30 min under a controlled nitrogen atmosphere for the activation process of the LiF crystals. After the activation, the LiF was quickly quenched to room temperature and then pulverized. The pulverized crystals were rinsed with hydrochloric acid. The crystals of a grain size under 40~200㎛ were abstracted and fabricated to a pellet-type with a 4.5 mm diameter and 0.8 mm thickness by pressurizing with a mechanical press. To increase the mechanical stability of the pellet-type detector and to obtain the most preferable glow curve structure for a TLD, the pressed pellet was sintered in an electronic furnace at 830℃ under a controlled nitrogen atmosphere and cooled on a frozen aluminum plate. As a final step, the above cooled pellet was initially annealed in an electronic furnace at 250℃.

2.2. Glow Curve and Sensitivity

The glow curve measurement of the TL detector was performed using a manual-type commercial TLD reader (System 310, Teledyne Brown Engineering) in the range between room temperature and 300℃ with a heating rate of 10℃/sec in a N_2 atmosphere after a dose of 10 mGy with a 137-Cs source at KAERI.

The sensitivity of the new TL detector was compared with the TLD-100. Five samples of both LiF:Mg,Cu,Na,Si and TLD-100 were prepared and their thermoluminescent glow curves were measured. TLD-100 detectors were annealed under the conditions of 1 hour at 400℃ and 2 hours at 100℃ before irradiation. The mean value was calculated for the measured signal of the two kinds of TL detectors and the relative sensitivity was calculated.

2.3. Dose Response

To investigate the dose response of the TL detector, ten sample groups, which have ten samples per each group, were prepared and then the irradiations were performed at doses of 10^{-6},

10^{-5}, 10^{-4}, 10^{-3}, 10^{-2}, 10^{-1}, 1, 10, 20 and 30 Gy using a 137-Cs irradiator at KAERI with 2 mm PMMA (polymethyl methacrylate) as a buildup material.

In the TL measurement, to eliminate the low temperature peaks, the sample was preheated at 135℃ for 10 sec, and then the TL signals were recorded by increasing the temperature up to 245℃ with a heating rate of 10℃/sec using a Harshaw 4500 TLD Reader.

For the estimation of linearity, the normalized dose response function was found.

2.4. Energy Response

For the determination of the energy response of the new LiF:Mg,Cu,Na,Si TL detector, six sample groups, which have nine samples per each group, were prepared. The photon irradiations were performed at the photon energies of 20, 35, 53, 73, 118 and 662 keV using x-ray generator (20-118 keV) and a 137-Cs source (662 keV). The TL measurement was carried out using a Harshaw 4500 TLD Reader.

2.5. Reproducibility

The reproducibility (reusability) study for the newly developed LiF:Mg,Cu,Na,Si TL detector was carried out using the method which was presented in the IEC(International Electrotechnical Commission) 1066 International Standard "Thermoluminescence dosimetry systems for personal and environmental monitoring".

2.6. Detection Threshold

In the IEC 1066, the detection threshold of a TL detector is defined as the following formula.

$$\textit{Detection threshold} = t_n \cdot \sigma$$

where, t_n is the Student's t distribution for n^{-1} degrees of freedom (n = number of dosimeters used in the test) and σ is the standard deviation of the evaluated values for all the n dosimeters.

For determination of detection threshold of the new TL detector, ten unirradiated samples were prepared, and then the TL signals were measured with same temperature profile as mentioned in the dose response experiment.

2.6. 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. The prepared LiF:Mg,Cu,Na,Si TL detector was preheated at 135 ℃ for 10 sec in the reader and then heated to 245 ℃ with a heating rate of 10℃/sec after 10mGy 137-Cs γ-ray exposure and annealed at 245 ℃ for 10 sec in the reader. The Harshaw Model 4500 TLD Reader was used for this test.

3. Results and Discussion

3.1. Glow Curve and Sensitivity

Fig. 1 shows the typical glow curves of the new LiF:Mg,Cu,Na,Si TL detector and GR-200A(LiF:Mg,Cu,P) TL detector, obtained from the Solid Dosimetric Detector & Method Laboratory in China in June 2002, after a dose of 10 mGy with 137-Cs (-ray. The heating rate of the sample was 10 ℃/s and the reading temperature was up to 300 ℃.

The glow curve structure was very similar with that of the GR-200A but the temperature of peak 2 was slightly shifted to a higher temperature.

It was found that the relative sensitivity of this

new LiF:Mg,Cu,Na,Si TL detector is about 30 times higher than that of the TLD-100.

3.2. Dose Response

The results of the dose response experiment for the detector are represented in Fig. 2 as TL response versus absorbed dose. For clarity, the normalized dose response function is represented in Fig. 3. The dose response is linear from 10^{-4} to 10 Gy, but a sublinear response was observed at higher doses.

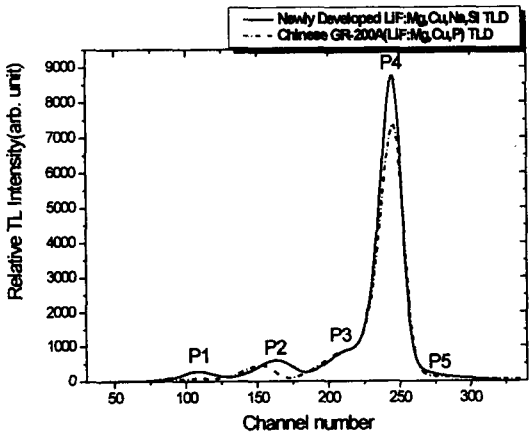

Fig. 1. Typical Glow Curves of Newly Developed LiF:Mg,Cu,Na,Si TL Detector and GR-200A

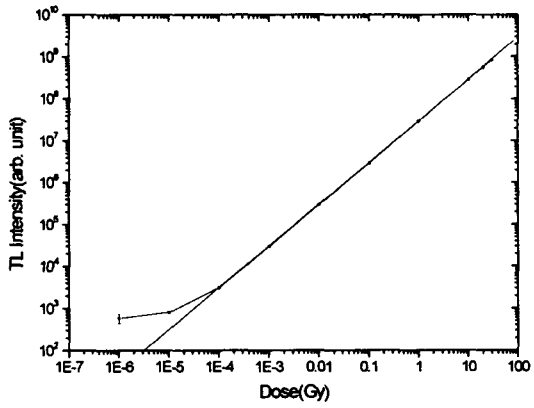

Fig. 2. Dose Response of LiF:Mg,Cu,Na,Si TL Detector as a Function of Absorbed Dose

3.3. Energy Response

Fig. 4 shows the relative energy response, normalized to the 662 keV photons (137-Cs) of the new LiF:Mg,Cu,Na,Si and MCP-N (LiF:Mg,Cu,P). The data for the MCP-N was obtained from the previous report [5]. The results for the LiF:Mg,Cu,Na,Si detector show that a maximum response of 1.004 at 53 keV and a minimum response of 0.825 at 20 keV were observed. In the energy range 73 keV and 118 keV, the response of the LiF:Mg,Cu,Na,Si is slightly higher than that of the MCP-N.

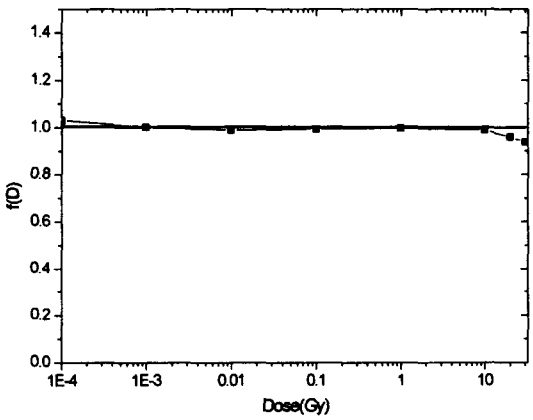

Fig. 3. Relative Dose Response Function of LiF:Mg,Cu,Na,Si TL Detector

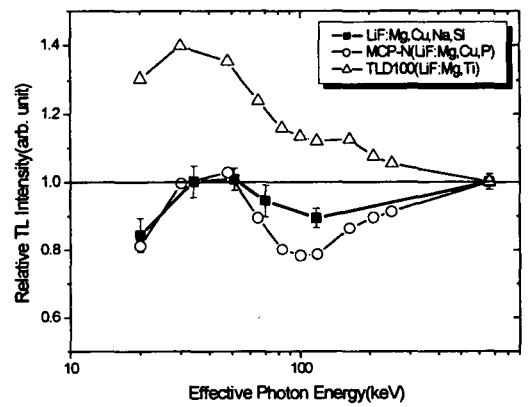

Fig. 4. Relative Energy Response of LiF:Mg,Cu, Na,Si TL Detector

3.4. Reproducibility

The coefficients of variation for each detector separately did not exceed 0.016, and for all the 10 detectors collectively was 0.0054. Fig. 5 shows the variation of the mean value of the TL signals of the LiF:Mg,Cu,Na,Si TL detectors with the number of reuse cycles.

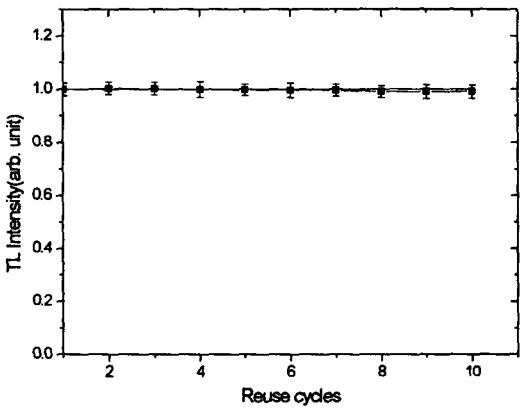

Fig. 5. Reproducibility of LiF:Mg,Cu,Na,Si TL Detector

3.5. Detection Threshold

Detection threshold of the detector was established as 70 nGy by the Harshaw 4500 TLD Reader.

3.6. Residual Signal

Fig. 6 shows the glow curves of the LiF:Mg,Cu,Na,Si TL detector for the residual signal test. In Fig. 6, (a) is for the first readout after a 10 mGy exposure and (b) is for the second readout. The total signals for the first and second readout were 1818836 and 10411, respectively. So the residual signal of the LiF:Mg,Cu,Na,Si TL detector was estimated to be 0.57 %.

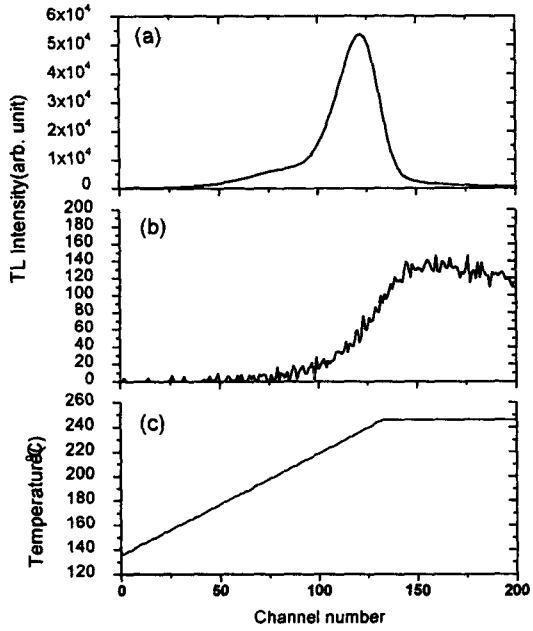

Fig. 6. The Glow Curves of the LiF:Mg,Cu,Na,Si TL Detector for the Residual Signal Test (a) the first readout after 10 mGy exposure; (b) the second readout; (c) the reading temperature profile

4. Conclusions

The thermoluminescent characteristics of the newly developed LiF:Mg,Cu,Na,Si TL detector were investigated for glow curve structure, sensitivity, dose response, energy response, reproducibility, detection threshold and the residual signal.

The glow curve structure and sensitivity of this new TL detector were similar with that of the GR-200A. The dose response is linear from 10^{-4} to 10 Gy, but a sublinear response was observed at higher doses. The energy response was slightly higher in the range of 73 to 118 keV compared to the MCP-N (LiF:Mg,Cu,P). This newly developed LiF:Mg,Cu,Na,Si TL detector has good reusability.

Acknowledgement

This study was the partial product of the national projects for long term nuclear energy development supported by the Ministry of Science and Technology.

References

1. Nam, Y.M., Kim, J.L., Chang, S.Y., Kim, G.D. The study of glow curves for LiF:Mg,Cu,Na,Si phosphor with different dopant concentrations, Ungyong Mulli. 11, 578-583 (1998).
2. Nam, Y.M., Kim, J.L., Chang, S.Y. Dependende of glow curve structure on the concentration of dopants in LiF:Mg,Cu,Na,Si phosphor, Radiat. Prot. Dosim. 84, 231-234 (1999).
3. Doh, S.H., Chu, M.C., Chung, W.H., Kim, H.J., Kim, D.S., Kang, Y.H. Preparation of LiF(Mg,Cu,Na,Si) phosphor and its thermoluminescent characteristics, Korean Appl. Phys., 2, 425-431 (1989).
4. Nam, Y.M., Kim, J.L., Chang, S.Y., Doh, S.H. Fabrication of LiF:Mg, Cu, Na, Si Pellets by Sintering Process, Saemulli, 40, 189-193 (2000).
5. Budzanowski, M., et al. Dosimetric properties of sintered LiF:Mg,Cu,Na,Si TL detectors, Radiat. Meas. 33, 537-540 (2001).
6. Lee, J.I., et al. Development of highly sensitive LiF:Mg,Cu,Na,Si TL detector. The First Asian and Oceanic Congress for Radiation Protection (AOCRP-1) (2002).