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LiF:Mg,Cu,Si - A high sensitivity TLD for personal and environmental monitoring of X and gamma rays

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1. Introduction

Ionizing radiations have found wide ranging applications in the field of industry, medicine agriculture and are an integral part of the nuclear facilities. X and gamma rays are by far the most encountered radiations. With increasing concerns of the effects of radiation, there have been continued efforts to develop highly sensitive dosimeters, both passive and active detectors, to measure lowest possible doses due to exposures to radiation. For X and gamma ray dosimetry, thermoluminescent dosimeters (TLDs) have been found to be the best suited and thereby most widely used for personnel and environmental monitoring. This has been because of their small size, possibility of placing at any site or on the body of a patient / person or affixing on the clothing, requirement of no electronics at the site of measurements (being passive dosimeters), repeated reusability by maintaining the same sensitivity, tissue equivalence and accuracy of dose measurement in a very wide range from a fraction of µGy to several tens of Gy. LiF:Mg,Ti, has been the most popular TLD but in the recent past more sensitive TLDs have become available. LiF:Mg,Cu,P has been one of the preferred replacement to LiF:Mg,Ti. However, in spite of the all round efforts of more than 25 years in improving the dosimetric properties of LiF:Mg,Cu,P, it is noted [1] that for the reuse, even an increase of 1 °C in the annealing temperature above 240 °C reduces the TL sensitivity significantly. Also, another recent study [2] demonstrated that the treatment needed for encapsulation of LiF:Mg,Cu,P in Teflon for making a commercial TLD badge reduces the sensitivity by more than a factor of 2. Also, Luo et al. 2006 [3] demonstrated a dependence of glow curve shape on heating rate and a decrease in its main dosimetric glow peak (peak-4) with increasing heating rate. This convinced us that fresh efforts are necessary to develop a high sensitivity LiF TL material to suit the needs of personnel and environmental dosimetry. This paper deals with the features of the recent development [4] of LiF:Mg,Cu,Si as a highly improved TL material.

2. Experimental

For the development of a new TLD, different preparation procedures and varying concentrations of Mg, Cu, Na and Si dopants were tried. An optimized method with dopant concentrations of 0.45 mol % of $MgSO_4.7H_2O$, 0.025 mol % of $CuSO_4.5H_2O$ and 0.9 mol % of SiO_2 was arrived at. The major features of the new method of preparation were 1- the avoidance of use of Na and P dopants in view of the evidence of enhanced sensitivity to thermal treatments with increasing Na content, 2- the use of method of melting for activation instead of granulation at temperatures much below the melting points and 3- the adoption of dual-step thermal treatment for stabilising the glow curve structure. Final dosimeters were in the shape of discs of thickness 0.8 mm and diameter 4.5 mm.

3. Results and Discussions

Replacement of dopant P by Si in LiF:Mg,Cu,P, has provided an improved LiF TLD material, LiF:Mg,Cu,Si. The glow curve structure of LiF:Mg,Cu,Si is similar to that of LiF:Mg,Cu,P (Fig.1) but the sensitivity was higher by about 10 %.

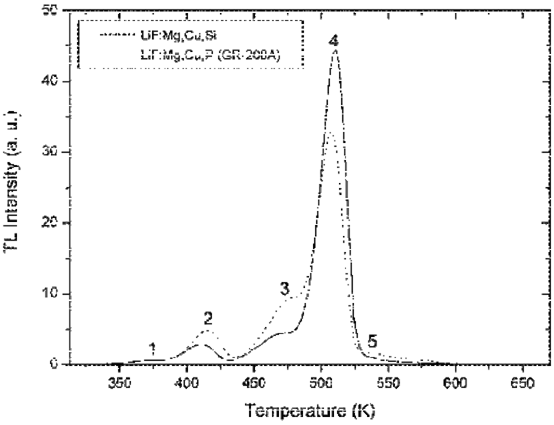

Fig.1 Comparison of TL glow curves of Korean LiF:Mg,Cu,Si with Chinese LiF:Mg,Cu,P (GR-200A).

The steps of heat treatments in the preparation procedure were found to be very critical for the temperature and the duration for the TL sensitivity and the glow curve structure. A final dual-step thermal treatment at 300 °C for 10 min followed by 260 °C for 10 min helped in stabilising the glow peak structure. Using a strict quality control, the TL sensitivity in different batches was reproduced. The dose response of the main dosimetric peak was linear from 1 µGy to 20 Gy of ^{137}Cs gamma rays. The fading of TLDs exposed to 10 mGy of ^{137}Cs gamma rays was less than 5 % for the post-irradiation storage period of 3 months (Table-1). Exposure to room light did not enhance the fading significantly. Also, there was no significant effect of pre-irradiation storage.

Table.1 Fading of TL signal on post-irradiation at room temperature.

Duration of Post-Irradiation Storage	Relative TL Response
10 min	0.970
1 h	1.000
24 h	1.001
3 days	0.997
10 days	0.960
30 days	0.973
2 months	0.968
3 months	0.957

The main features of LiF:Mg,Cu,Si material are the improved thermal stability permitting the TL readout at temperatures up to 300 ˚C, high sensitivity (55 times that of LiF:Mg,Ti, TLD-100), negligible (0.025%) residual TL signal and stable glow curve structure. No significant change (within 12%) in the TL was observed for 30 cycles of anneal (260 ˚C), exposure (6.2 mGy of ^{90}Sr-^{90}Y beta rays) and readout using the contact heating. LiF:Mg,Cu,Si has potential to withstand much faster heating rates. Table-2 shows the responses of different peaks relative to the values at a heating rate of 10˚Cs^{-1}. The integrated TL of the main peak, whether isolated or deconvoluted, did not reduce with increasing heating rate, on the contrary a small increase was observed (Table-2; columns 2 and 6).

Table-2 Relative response of total TL (from 50 to 300 ˚C), peak-2, peak-3, peak-4 and thermally isolated peak-4 (lower temperature peaks bleached by a thermal treatment of 160 ˚C for 10 min) at different heating rates.

Heating	Total	Individual Peaks			Isolated
Rate(˚Cs^{-1})	TL	Peak-2	Peak-3	Peak-4	Peak-4 .
1	0.94	0.84	0.46	0.97	0.94
2	0.95	0.86	0.62	0.97	0.95
5	0.99	1.00	0.87	1.05	1.00
10	1.00	1.00	1.00	1.00	1.00
20	1.03	1.09	1.28	1.00	0.99
30	1.02	1.08	1.32	0.95	1.02

The importance of the study of heating rates lies in ensuring the usefulness of this material for personnel dosimetry applications in which there is often a requirement of evaluating a very large number of dosimeters in the shortest possible time, obviously by using fast heating rates during TL readouts. Apart from the TL (integrated TL under a glow curve), the glow curve shape as a signature of exposure to ionising radiation is a very important aspect of quality assurance in these applications and changes, if any, due to reasons other than exposure to radiation need to be ensured. In routine use, a change in heating rate during the readout cannot be ruled out due to various reasons including varying contact with the heater.

TL emission spectrum of LiF:Mg,Cu,Si and LiF:Mg,Cu,P peaked (broad peak) at 384 nm and 368 nm, respectively. This shift in the peak with the replacement of P by Si clearly indicates the influence of these codopants on the emission spectrum. Neither Si nor P are the luminescent centres but are evidently associated with the luminescent centre, in this case Cu^{+}. It is inferred that the role of these codopants (Si or P) is to enhance the incorporation of Cu in the form of Cu^{+} as a luminescent centre in the complexes involving Mg, Cu and Si which would have otherwise gone as Cu^{++}, from the considerations of the ionic radii. Incorporation of Si is also inferred to alter the temperature of dissolution of precipitated phase consisting of the complexes responsible for TL and thereby to enhance the thermal stability.

4. Conclusions

The newly developed LiF:Mg,Cu,Si TLD exhibits much improved dosimetric characteristics over those reported so far and is expected to replace the other LiF based TLDs in personal and environmental dosimetry.

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