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Study of a new Lif:Mg,Cu,P formulation with enhanced thermal stability and a lower residual TL signal

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Abstract

This paper presents results obtained for a new LiF:Mg,Cu,P (HMCP) preparation with modified Mg and Cu concentrations. The shape of the HMCP glow curve shows minimal differences for annealing in the range from 523 to 543 K for 10 min. The thermoluminescence (TL) readout value remained stable when annealed in the range from 513 to 543 K for 10 min. The new formula allows heating of the material to higher temperatures than that originally employed for the well-known GR-200A dosemeter, practically without losses in sensitivity. The TL sensitivity is approximately half of that for the GR-200A, and still 29-fold greater than that for the TLD-100 dosemeter, and the residual signal is approximately five-fold lower than for the GR-200A. These results indicate that the new TL material shows enhanced thermal stability and a lower residual TL signal at a small TL sensitivity cost. The heat treatment temperatures are related to concentrations of Mg and Cu in LiF:Mg,Cu,P.

Keywords: Thermoluminescence; TL; LiF:Mg,Cu,P; Sensitivity; GR-200A

1. Introduction

One of the most promising thermoluminescent (TL) materials is LiF:Mg,Cu,P. Its main advantages are high sensitivity, good tissue equivalence, almost flat energy response, low fading rate, linear dose response and good stability at ambient temperatures, and shorter annealing procedures (oven annealing at 513 K for 10 min) (Oster et al., 1993). It has become a popular TL detector material in routine applications for personal, environmental and clinical dosimetry. The drawbacks of standard LiF:Mg,Cu,P are the thermally induced loss of TL sensitivity for material annealed at temperatures above 513 K and the relatively high residual signal (Delgado, 1996).

To eliminate the residual signal, pre-irradiation annealing at 513 K for 10 min has been widely accepted. In personnel monitoring services, it is important to omit the high-temperature annealing process to achieve economic management of large numbers of TL detectors. There are three efficient methods for reducing the residual signal without pre-irradiation annealing. The first is an increase in the readout maximum temperature (Oster et al., 1993). In our experience (Tang et al., 1999), an initial decrease in TL sensitivity of approximately 20% for the first several readouts was found as the readout was increased to 543 K in our TL system. Above 533 K the sensitivity shows some tendency to decrease (Delgado, 1996). The second is an improvement in the preparation procedure. The residual signal of LiF:Mg,Cu,P can be decreased by increasing the Cu concentration and the sintering temperature (Tang et al., 1999). It is also possible to prepare a material (LiF:Mg,Cu,P powder) with a residual signal of only 0.25–0.5% following a readout at 513 K for 10 s (Horowitz and Horowitz, 1992). However, the influence of heat treatments on TL response has not been investigated. Third, the separation of the dosimetry signals from the residual signal by computerised analysis of glow curves. A 1-μGy test dose could be well and reproducibly measured at 200 μGy (Delgado et al., 1995) but it offered poor results after measurement at 500 μGy, after which the residual doses were so high that the program was unable to produce reliable results.

The trapping sites all appear to be related to Mg, and heat treatments of LiF:Mg,Cu,P at temperatures near 523 K or above

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lead to dissolution of the Mg precipitate phases, which in turn alters the glow curve structure and leads to a reduction in TL sensitivity (McKeever, 1991). A change in valence of copper from Cu^{+} to Cu^{2+} occurs upon annealing above 513 K (Chen and Stoebe, 1998). The presence of Cu^{+} ions may be essential for the high TL sensitivity of LiF:Mg,Cu,P and a change in the valence of copper to Cu^{2+} reduces the TL sensitivity. The shape of the glow curve for LiF:Mg,Cu,Si, which is doped with much higher concentrations of Mg and Cu than in standard LiF:Mg,Cu,P (GR-200A), shows minimal differences and its sensitivity remains stable when annealed in the range from 533 to 573 K for 10 min (Tang, 2003). However, the TL sensitivity of LiF:Mg,Cu,P (GR-200A) decreases when annealed at temperatures above 513 K. It seems that the annealing temperature is related to the concentrations and properties of Mg and Cu in the LiF lattice. So far, it is uncertain whether it is possible to increase the annealing temperature for LiF:Mg,Cu,P and retain a stable TL sensitivity by modifying the concentrations of Mg and Cu. To investigate this possibility, a new LiF:Mg,Cu,P material was prepared by modification of the concentrations of Mg and Cu to allow heat treatments above 513 K. The aim was to reduce the residual signal while retaining a stable TL response in re-use cycles without the need for an initialisation procedure.

2. Experimental

A new LiF:Mg,Cu,P formulation doped with much more Mg and Cu than in the GR-200A dosemeter and a conventional concentration of P (abbreviated as HMCP) was developed in the Solid Dosimetric Detector and Method Laboratory (DML), and the standard LiF:Mg,Cu,P round GR-200A dosemeters manufactured by DML in 2000 were also employed. Both the HMCP and GR-200A dosemeters are 4.5 mm in diameter and 0.8 mm thick. The new production process is the same as previously reported for the GR-200A (Shen et al., 2002; Zha et al., 1993). The other TL material used in this study was LiF:Mg,Ti (TLD-100) kindly provided by Bicron Co., with dimensions of $3 \times 3 \times 0.89\,mm^3$. The new HMCP and the GR-200A dosemeters were compared, examining the influence of the annealing temperature on TL glow curves, the re-usability characteristics at different annealing and readout temperatures, and the residual signal and sensitivity. All samples are irradiated with the same dose of 1-mGy ^{60}Co gamma radiation. For annealing, an oven with air circulation and temperature control within ± 1 K was used. The TL readout equipment used was a type RGD-3 manual reader manufactured by DML. The light intensity and planchet temperature were sent to a PC via an RS232C interface and the data were stored as identifiable and recoverable files. Two readout programmes were applied: (i) for plotting glow curves, a linear heating rate of $6\,K\,s^{-1}$ from 323 to 623 K and (ii) for quantitative measurements, a heating rate of $15\,K\,s^{-1}$ with pre-heating at 413 K for 8 s and readout between 413 and 513–543 K and hold at 513–543 K for 12 s.

3. Results and discussion

3.1. TL glow curves

Fig. 1 shows the glow curves for the new LiF:Mg,Cu,P (HMCP) and the standard LiF:Mg,Cu,P (GR-200A) chip. Pre-irradiation annealing programs of 533 K for 10 min for HMCP and 513 K for 10 min for GR-200A were used. Readout programme (i) for plotting glow curves was used. The shape of the glow curve for HMCP is similar to that for GR-200A. The glow curve for HMCP consists of several overlapping glow peaks, namely low-temperature peaks (nos. 2 and 3), the main dosimetric peak (no. 4) and high-temperature peaks. The shape of the glow curve—i.e. the positions of the TL peaks—is independent of the concentrations of Mg and Cu and this result supports McKeever's conclusion that the number and position of the various TL peaks are dictated by the presence of Mg impurities (McKeever, 1991). The height of the high-temperature peaks for HMCP is lower than that for GR-200A, and the low-temperature peaks for HMCP are higher than those for GR-200A. The height of the main peak for HMCP is approximately three-fold lower than that for GR-200A. It was reported that the height of the low-temperature peaks increases and the main peak and high-temperature peaks are reduced with an increase in Cu concentration (Tang et al., 1999), and that for Mg concentrations above 0.2%, the high-temperature peaks increase and the main peak is reduced with an increase in Mg concentration (Shoushan, 1988). It seems that the increase in the low-temperature peaks is related to the high Cu concentration in HMCP, while the decreases in the main peak and the high-temperature peaks are related to the high Mg and Cu concentrations.

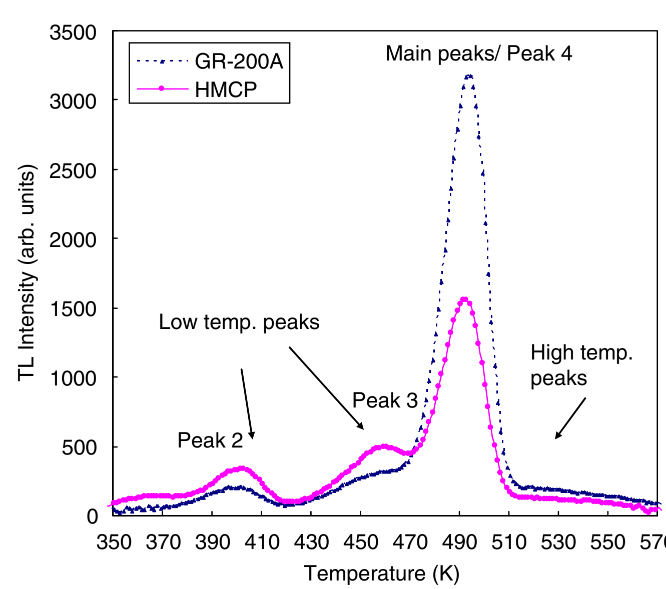

Fig. 1. Glow curves for the new LiF:Mg,Cu,P material and the standard LiF:Mg,Cu,P chip (GR-200A).