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Improvement in the Preparation Method of LiF: Mg, Cu, P Thermoluminescent Phosphor

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Abstract: This study describes the preparation method of a new LiF:Mg, Cu, P thermoluminescent (TL) phosphor which can be heated up to 553 K without any change in their reestablished dosimetric characteristics. The main tests performed were detection threshold, sensitivity and TL response as a function of dose. Results showed that this material, submitted to a thermal annealing at 553 K for 10 min followed by 373 K for 2 h, is approximately 20 times more sensitive than the commercially available TLD-100.

Key words: Thermoluminescence, LiF:Mg, Cu, P, gamma radiation

INTRODUCTION

Thermoluminescence dosimetry (TLD) is a highly confident method to be used in medical physics especially in radiodiagnosis and radiotherapy[1,2]. For this reason, it is necessary to count with adequate TLDs which could be useful in this field. At present time, existing TLDs can be classified into two groups: those having good tissue equivalence but poor sensitivity such as LiF:Mg, Ti (TLD-100) and those with high sensitivity and poor tissue equivalence such as $CaSO_4$:Dy. Even though both two types of TL phosphors have demonstrated being useful for a number of applications, the search of a new phosphor having similar properties to those of LiF:Mg, Ti but with higher sensitivity continues. Among the different TL phosphors produced in Mexico, there is special interest in LiF:Mg, Cu, P due to its dosimetric properties[3]. In this study, the preparation method and the dosimetric characteristics of a new LiF:Mg, Cu, P phosphor, which can be heated up to 553 K, are presented.

MATERIALS AND METHODS

Irradiation of the samples was performed using the following sources: ^{137}Cs gamma Calibrator J.L. Shepherd and Associates, model 28–6B (44.4 GBq) at a dose rate of 2.87 mGy/h; ^{60}Co gamma irradiator, Vickers model VickRad 2000 series (380.31 GBq) at a dose rate of 26.28 Gy/h; ^{60}Co gamma irradiator, Nordion model Gammacell 220 series (10.55 TBq) at a dose rate of 173.82 Gy/h. Readings were made using a Harshaw TL Analyser model 4000. In order to avoid spurious contributions on TL signal, all readings were made in a nitrogen atmosphere.

For preparing the TL material, 12 g of commercial LiF powder analytical grade were placed in a platinum crucible to be dried and dopants were added in aqueous solutions ($MgCl_2$, $CuCl_2$ and $(NH_4)_2HPO_4$), at the following concentrations 0.2 M%, 0.05 M% and 3 M%, respectively.

The powder obtained was dried and submitted to a thermal treatment at 673 K for 15 min, followed by heating at 1423 K during 30 min, finally obtaining the LiF:Mg, Cu, P TL phosphor. All thermal treatments were performed in a nitrogen atmosphere. Material thus obtained was crushed and sieved choosing the polycrystalline powder with size in the range of 70 to 210 µm. This powder was used to make pellets of a mixture (2:3) of LiF:Mg, Cu, P and PTFE with the following dimensions: 5 mm diameter, 0.6 mm thickness and an average mass of 20±3mg. These pellets were submitted to a special thermal treatment for sintering[3].

To study the effect of the pre-irradiation thermal treatment on the TL response of LiF:Mg, Cu, P, four samples of the TL powder were placed in silver crucibles labeling them as A, B, C and D which and submitted to four different thermal treatments.

The four samples were heated during 10 min: sample A was heated at 513±2 K, sample B at 553±2 K, sample C at 573±2 K and sample D at 673±2 K. All the four samples

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were left cooling down to room temperature and then irradiated with ^{60}Co gamma radiation at an absorbed dose of 100 mGy. TL readings were made 1 h after irradiation. The influence of the thermal annealing on the low temperature peaks were evaluated submitting two batch of LiF:Mg, Cu, P+PTFE to two different thermal treatments: (A) 553±2 K for 10 min; (B) 553±2 K for 10 min followed by 373±2 K during 2 h. Both samples were left cooling down to room temperature and then irradiated with ^{60}Co gamma radiation at an absorbed dose of 100 mGy.

Performance tests

Batch homogeneity: To test the batch homogeneity, 568 LiF:Mg, Cu, P+PTFE pellets were submitted to a thermal treatment at 553±2 K for 10 min followed by 373±2 K during 2 h. This thermal treatment was considered as the standard thermal annealing. Later, samples protected from the light, were exposed to environmental radiation up to 45 days. The samples of LiF:Mg, Cu, P+PTFE were grouped in plastic holders. According to the results of this test, 489 pellets (86%) were selected at 95% confidence level rejecting only 79 pellets (14%).

Sensitivity: Sensitivity was tested irradiating one batch of LiF:Mg, Cu, P+PTFE pellets jointly with a similar batch of LiF:Mg, Ti (TLD-100) and commercial LiF:Mg, Cu, P (GR200A) with ^{60}Co gamma radiation at an absorbed dose of 102 mGy. Previous to irradiation all TLDs were submitted to their respective established thermal annealing[3-7]. All readings were performed 1 h after irradiation in a nitrogen atmosphere.

Detection threshold: To determine the detection threshold one batch of 21 LiF:Mg, Cu, P+PTFE pellets was submitted to its corresponding thermal treatment annealing, followed by cooling to room temperature and reading the background signal. For comparison, a batch of 16 TLD-100 chips was submitted to the same process.

TL response vs dose: Thermoluminescence response as a function of absorbed dose was determined irradiating a batch of 24 LiF:Mg, Cu, P+PTFE pellets with ^{137}Cs and ^{60}Co gamma radiation at different absorbed doses in the range from $5x10^{-5}$ to 10^3 Gy. Readings were made 24 h after irradiation in a nitrogen atmosphere.

Repeatability: Repeatability of the TL response was evaluated irradiating a set of LiF:Mg, Cu, P+PTFE pellets jointly with a set of GR200A at an absorbed dose of 101.7±1.32 mGy of ^{60}Co gamma radiation, reading them and submitting to the standard thermal treatment annealing. This procedure was repeated for 10 cycles of irradiation readout annealing.

Fading: Fading of TL response at room temperature was tested using a batch of 21 LiF:Mg, Cu, P+PTFE pellets. Samples were submitted to the standard thermal treatment annealing and then irradiated with ^{60}Co gamma radiation at an absorbed dose of 100 mGy and stored at room temperature protected from the light. Readings were taken at different times after irradiation along a time interval of two months.

RESULTS

Figure 1 shows the glow curve of LiF:Mg, Cu, P powder submitted to different annealing thermal treatments and then irradiated with ^{60}Co gamma radiation at an absorbed dose of 100 mGy. It is clearly appreciated in Fig. 1 that the sample submitted to the thermal treatment to 553±2 K for 10 min, exhibits a TL glow curve optimal for dosimetric applications as well as the highest TL intensity (Table 1).

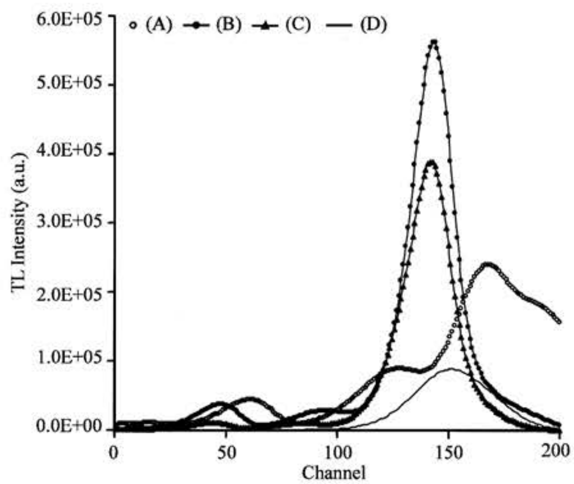

Fig. 1: Glow curves of LiF:Mg, Cu, P (powder) submitted to different thermal annealing procedures: (A) 513 K; (B) 553 K; (C) 573 K; (D) 673 K

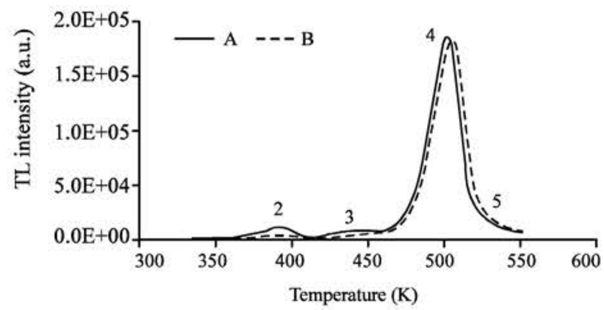

Fig. 2: Glow curves of LiF:Mg, Cu, P +PTFE submitted to different thermal annealing procedures: (A) 553 K; (B) 553 K followed by 373 K

Glow curves of LiF:Mg,Cu,P+PTFE exhibited 5 peaks, low temperature peak (peak 1) disappears few hours alter irradiation, meanwhile, the other peaks 2, 3, 4 and 5, appears at 391, 448, 505, 524 K. The influence of the thermal annealing on the low temperature peaks is shown in Fig. 2. It may be appreciated that, when the material is submitted to a thermal treatment consisting of heating at 553 K for 10 min followed by an additional heating at 373±2 K for 2 h; the intensity of the first peak is reduced by 64%; the second peak intensity diminishes 34.6% while the dosimetric peak is only reduced by 2% (Fig. 2). It must be noted that after this thermal treatment is applied the material did not exhibit any appreciable fading.

Sensitivity results are summarized in Table 2. These results showed that LiF:Mg, Cu, P+PTFE is 23.1 times more sensitive than TLD-100. Measured detection threshold of LiF:Mg, Cu, P was as low as 26.25 μGy; compared to TLD-100 measured detection threshold equal to 240 μGy.

The TL response as a function of ^{137}Cs and ^{60}Co gamma absorbed dose in the range of $5x10^{-5}$ to 10^3 Gy is shown in Fig. 3. In Fig. 4a it may be appreciated that the TL response of LiF:Mg, Cu, P+PTFE is linear in the range

Table 1: TL intensity of LiF:Mg, Cu, P (powder) submitted to different thermal annealing procedures

Thermal annealing (K)	TL intensity (nC)	Sensitivity (nC/mg·mGy)	Relative sensitivity*
513	7051.66	3.97	16.91
553	9479.78	5.42	23.11
573	7003.52	3.87	16.52
673	2416.00	1.36	5.82

* Assuming TLD-100 sensitivity is equal to unity

Table 2: Sensitivity of LiF:Mg, Cu, P and GR200A

	LiF:Mg, Cu, P+PTFE		GR200A	
Batch	Sensitivity (nC/mg·mGy)	Relative Sensitivity*	Sensitivity (nC/mg·mGy)	Relative sensitivity*
1	3.4117	21.9402	3.6759	23.6392
2	3.7684	24.2341	3.3874	21.7839
		$\bar{x}$ = 23.0872		$\bar{x}$ =22.7116

* Assuming TLD-100 sensitivity is equal to unity

Table 3: Repeatability of the TL response of LiF:Mg, Cu, P and GR200A

	LiF:Mg, Cu, P+PTFE			GR200A	
Cycle	Dose (mGy)	TL intensity(nC)	Sensitivity (nC/mg·mGy)	TL intensity (nC)	Sensitivity (nC/mgmGy)
1	103.02	1700.89	2.33	6242.88	2.17
2	101.41	1672.01	2.33	6412.24	2.27
3	99.79	1711.98	2.42	6151.65	2.21
4	102.95	1710.63	2.35	5905.11	2.06
5	103.33	1717.25	2.35	5899.76	2.05
6	99.48	1653.38	2.35	5906.48	2.13
7	102.79	1654.23	2.27	5725.65	2.00
8	101.41	1679.45	2.34	5811.34	2.06
9	100.56	1645.98	2.31	5805.85	2.07
10	102.56	1627.92	2.24	5768.84	2.02
$\bar{x}$	101.73	1677.37	2.33	5962.98	2.10
SD%	1.32	1.79	1.97	3.63	4.02

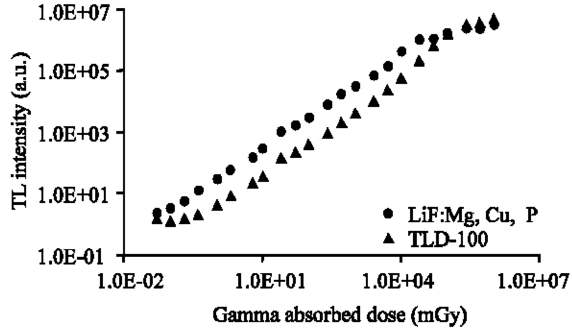

Fig. 3: TL response of LiF:Mg, Cu, P +PTFE and TLD-100 as a function of absorbed dose

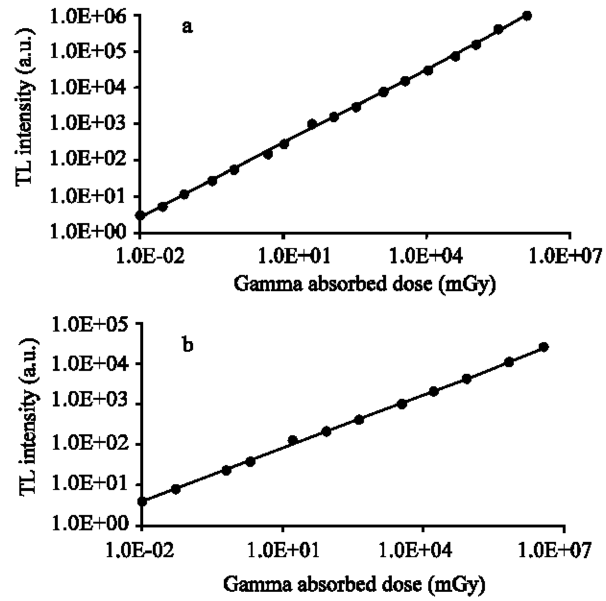

Fig. 4: Linearity range of the TL response; (a) LiF:Mg, Cu, P +PTFE, (b) TLD-100

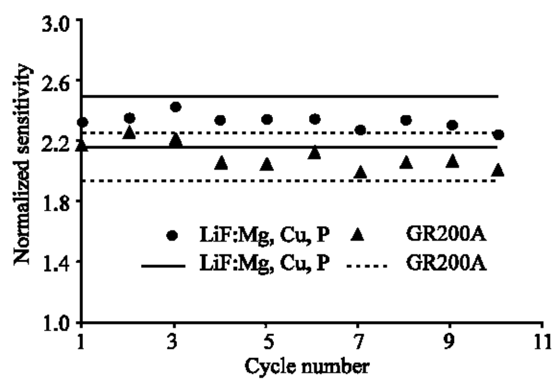

Fig. 5: Repeatability of the TL response of LiF:Mg, Cu, P and GR200A (±7.5% SD)

of 10^{-4} to 25 Gy. Compared to TLD-100 TL response which is only linear from 10^{-3} to 5 Gy (Fig. 4b).

Supralinearity effect was not observed in LiF:Mg, Cu, P+PTFE, i.e. saturation was observed immediately after the linear region; while, TLD-100 exhibits supralinearity either before and after the linear region, reaching saturation at 250 Gy.

Table 3 summarizes the repeatability results. Table 3 shows the dosimeter identification, the absorbed dose given for each cycle and the TL response normalized with respect to the mass. Average values and standard deviations are shown. Results show (Fig. 5) that repeatability of LiF:Mg, Cu, P+PTFE is in accordance with the requirements of international standards (±7.5%SD)[8].

No fading was observed after two months in LiF:Mg, Cu, P+PTFE when annealed at 553±2 K for 10 min followed by 373±2 K for 2 h.

DISCUSSION

The present new preparation method of LiF:Mg, Cu, P involves the use of analytical grade LiF instead of suprapure LiF, but following the procedure previously reported[4] consisting of heating the TL material at 513 K for 10 min. However, this new method allows to heat the material up to 553 K for 10 min without any changes in its TL characteristics.

LiF:Mg, Cu, P submitted to a thermal annealing treatment, consisting in heating the material at 553 K for 10 min, resulted to be 23 times more sensitive than TLD-100 and presented a threshold detection limit 10 times lower than TLD-100. When the thermal annealing at 553 K for 10 min is followed by 373 K for 2 h, it is observed that peaks 2 and 3 reduce their intensities by 64 and 35%, respectively while the dosimetric peak (peak 4) diminishes by only 2 %. For this reason the fading is found to be negligible along a two month time interval.

In order to be reused, LiF:Mg, Cu, P prepared by the present method must be submitted to a thermal annealing treatment consisting in heating the material at 553 K for 10 min followed by 373 K for 2 h. This thermal treatment erases completely all remaining TL signal, resulting in a background signal similar to the one obtained for the unexposed material.

From the results of these tests it can be concluded that LiF:Mg, Cu, P+PTFE prepared by the new improved method fulfils the requirements to be considered as a highly confident dosimetric system to be used in low dose gamma applications of medical physics.

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