Optik 156 (2018) 416–423

Contents lists available at ScienceDirect

Optik

journal homepage: www.elsevier.de/ijleo

Full length article

Investigation Of the low-dose-thermoluminescence characteristics of LiF:Mg,P

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ARTICLE INFO

Article history:
Received 7 September 2017
Accepted 6 November 2017

Keywords:
Thermoluminescence
Dose response
Annealing
Sensitivity
Fading
Lithium fluoride

ABSTRACT

Thermoluminescence (TL) characteristics of LiF:Mg,P with varying concentrations (0.4%, 1% and 2%) of phosphorous are investigated and compared with those of the widely used TLD-100. The TL glow curves of the three LiF:Mg,P samples exhibited two apparent peaks around 120 °C and 239 °C. The TL response of each of the two peaks vary linearly with increasing dose (between 0.4 mGy and 12.0 mGy) for each of the three samples. The thermolumiscence intensities from samples with 0.4%, 1.0% and 2.0% of phosphorous concentration were respectively at least 26, 18 and 21 times that of TLD-100. TL response from a repeat use of a single aliquot of the samples with 0.4%, 1.0% and 2.0% of phosphorous concentration did not vary by more than 6%, 8% and 10% respectively. There were 3%, 10% and 12% fading of TL response over three-week storage period at room temperature for samples with 0.4%, 1.0% and 2.0% of phosphorous concentration. Pre-irradiation annealing up to 600 °C did not affect the glow curves structures of all the samples. However, while the sensitivity of samples B and C did not change with the annealing, the intensity of sample A first decreased with temperature up to 500 °C before it significantly rose to a value above that of the unannealed sample. TL intensity of the sample annealed at 500 °C decreased with increasing annealing duration up till one hour beyond which it remains constant. The linearity of the dose-response, negligible fading of response and unchanged sensitivities with pre-irradiation annealing for samples B and C qualify them as suitable TL dosemeters.

1. Introduction

Several thermoluminescent (TL) dosimeters are commercially available for wide range of applications: personnel, environment, medical dosimetry, nuclear accidents, other biological sciences and radiation protection [1]. Lithium fluoride doped with magnesium and titanium, known commercially as TLD-100, is still the most commonly used for personnel dosimetry. It is widely used because of several properties such as tissue equivalence, relatively low fading, and the possibility to manufacture the material with acceptable reproducibility [2–5]. However, it is not entirely suitable for very low dose measurements because of its relatively low sensitivity, poor detection threshold and disagreement in several reports about the fading [6–8].

Another material with nearly tissue equivalence is lithium fluoride doped with magnesium, copper and phosphorus (LiF:Mg, Cu, P) [9,10]. It has several important advantages over TLD-100 such as higher sensitivity, repeated use has been shown to cause low fading, good detection threshold, ability to detect considerably lower radiation levels and lower photon

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https://doi.org/10.1016/j.ijleo.2017.11.039

energy dependence than TLD-100 [11]. Furthermore, LiF:Mg,Cu,P shows no supralinearity up to saturation above 1 kGy, a particular plus for accident dosimetry [12]; the discovery of LiF:Mg,Cu,P as tissue equivalent and highly sensitive TLD material fascinated the radiation dosimetry community the world over [13]. However, LiF:Mg,Cu,P has its own associated problems and this has led to the large number of publications on the numerous attempts to overcome the problems, but with a limited success. Even dual – step annealing tried for LiF:Mg,Cu,P [13–15] did not work satisfactorily.

In an attempt to investigate the reasons for the problems associated with LiF:Mg,Cu,P, Mckeever [16] found that Cu did not play any role in the TL emission process but that phosphorus is the main activator responsible for TL emission in the phosphor. Some other studies [16–19] have also shown that while P is responsible for high TL sensitivity of LiF:Mg,Cu,P, Cu did not play any role in the formation of traps and luminescent centres. The loss of TL sensitivity of LiF:Mg,Cu,P when annealed at temperature above 240 °C has also been attributed to the presence of Cu [12,20].

The aim of this study, therefore, is to investigate the thermoluminescence characteristics of LiF:Mg,P (i.e LiF:Mg,Cu,P without Cu). The performance of the phosphor as a dosemeter will be compared to those of LiF:Mg,Cu,P and TLD-100.

2. Materials and methods

2.1. Samples preparation and characterization

Samples of LiF:Mg,P were prepared from commercially available lithium fluoride, magnesium chloride ($MgCl_2$) solution (0.2 M) and phosphoric acid solution (1.25 M). 13.5 cm^3 $MgCl_2$ and 11.2 cm^3 H_3PO_4 solutions were added to 35 g of lithium fluoride. The mixture was heated at 800 °C for 1 h to form LiF: Mg (0.2%), P (2%). The Mg concentration was kept constant at 0.2% while P was varied to achieve two other phosphorous concentrations (0.4% and 1%). Hereafter the samples with 0.4% P, 1% P and 2% P concentrations are referred to as sample A, sample B, and sample C respectively. The concentration of phosphorous was varied because it is reported as the one responsible for TL emission in LiF:Mg,Cu,P. The LiF:Mg,P samples in this study were used in powder form with grain sizes between 90 μm and 105 μm.

The samples' compositions were determined using Rutherford Backscattering spectrometry using 2 μC of 2.2 MeV $^4He^+$ beam from ion beam analysis facility at the Centre for Energy Research and Development, (O.A.U) Ile-Ife, Nigeria. This facility is centered on a NEC 5SDH 1.7 MV Pelletron Accelerator, equipped with a RF charge exchange ion source. The ion source is equipped to provide proton and helium ions. A quantitative analysis of the RBS spectra was made using the computer code SIMNRA [21]. The analysis comprises of simulating each spectrum in comparison with the experimental one.

2.2. Thermoluminescence measurements

For each of samples A–C, thermoluminescence measurements were made by weighing about 15 mg of the sample into a powder disc of 8 mm in diameter by 1 mm in thickness. The grains of the sample were properly spread and fixed on the disc so as to ensure good thermal contact between the disc and the sample. The measurements were made using TLD Cube reader (manufactured by Radpro International, Wermelskirchen, Germany) at a heating rate of 2 K/s. Glow curves for dose-response study were acquired from aliquots of each sample irradiated to doses ranging from 0.4 mGy–12 mGy. The irradiation was done at room temperature using a ^{90}Sr-^{90}Y beta-ray irradiator with a dose rate of 0.04 mGy per revolution. All the TL measurements were made in a nitrogen atmosphere, hence thermal contact between sample and heating plate was further improved.

Effects of annealing on the glow curve structure and TL sensitivity of each of the samples were investigated by subjecting the samples to pre-irradiation annealing at 300 °C, 400 °C and 500 °C and 600 °C each for a duration of 30 min. Another set of measurements were carried out at an annealing temperature that produced maximum sensitivity but varying durations of 20 min, 30 min, 40 min, 60 min and 90 min. After each of the annealing, the sample was irradiated to a dose of 2 mGy follow by TL readout at a heating rate of 2 K/s

The intrinsic TL sensitivity of a thermoluminescent material is expressed as the TL yield per unit of mass per unit of ionizing radiation exposure. In this study the TL sensitivities of the three samples relative to that of TLD-100 were determined. For these experiments, two sets of each of samples A–C and a similar batch of LiF:Mg,Ti (TLD-100) were irradiated to doses of 3.6 mGy and 180 mGy. TL readouts were performed 1hr after irradiation in a nitrogen atmosphere using heating rate of 2 K/s. The relative sensitivity was determined as the ratio of the response of each of samples A, B and C to that of TLD-100.

Reusability is an another important TL property any dosimetric material should possess. To test the reusability of samples A–C, eight cycles of repeated annealing, irradiation and readouts were performed for the same aliquot of each sample. For this test, each irradiation was to a dose of 2 mGy and annealing was at 400 °C.

Fading is a loss of stored thermoluminescence signal after irradiation, which is not desirable for a good dosemeter. Fading depends on several parameters such as storage temperature, time, radiation type and annealing procedure. To determine the thermal fading features of LiF:Mg,P, several aliquots of each sample were annealed and irradiated to a fixed dose of 2 mGy. After irradiation the aliquots were stored in dark conditions (light-tight box) at room temperature. The aliquots were grouped into six sets of three each. They were readout at six different periods (immediately, 1 h, 1 day, 1 week, 2 weeks and 3 weeks) after irradiation. The average of the TL responses of the three aliquots in each set represents the TL signal for that period after irradiation.