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Dosimetric properties of in-house prepared MgB_4O_7:Dy

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ABSTRACT

The dosimetric characteristics of prepared MgB_4O_7 doped with Dy have been studied using the thermoluminescence (TL) technique. The TL was observed at the most intense temperature of 187±2 °C that is applied on varied Dy concentration. The response recorded was linear with gamma doses in the range from 1 Gy up to 2 kGy and a good linear index coefficient at all applied doses but notin dose range from 100 to 500 mGy. The MgB_4O_7:Dy dosimeters have shown a TL-sensitivity of 2.2 times higher than that of TLD-100. Furthermore, the study indicated that the dosimeters subject of study, are highly affected by fading.

KEYWORDS

Thermoluminescence;
Magnesium borate;
Dosimetry;
TL Linearity;
TL Fading.

INTRODUCTION

Individuals occupational and medical exposure of to ionizing radiations is a common feature of person day life. The detrimental effects of excessive exposure are well recognized. However, the accurate measurements of dose exposure in some situations as in medicine and occupational regions are significant. In the first case, the minimum exposures consistent with good diagnostic and treatment should be achieved, and in the later case, the monitoring of persons with not exceeding the recommended permissible doses. In the last few decades, the thermoluminescence detectors could measure the properties stated above with higher accuracy.

Thermoluminescence (TL) is the phenomenon of light emission during warming a previously irradiated substance with uniform heating rate. Most of substances either organic and inorganic show the property of luminescence, but principal materials used in various application of luminescence, involves inorganic solid insulating materials such as alkali and alkaline earth halides, Quartz (SiO_2), Phosphates, Borates, and Sulphates etc. Luminescence solids are usually referred to as Phosphors.

While lithium fluoride has been an extremely popular since it gives consistent response to radiation, compared with many other substances such as: oxides (e.g. Al_2O_3:C; MgO), fluorides (e.g. LiF:Mg,Cu,P; CaF_2:Mn), sulphates (e.g. $CaSO_4$:Dy) and borates (e.g. $Li_2B_4O_7$:Mn; MgB_4O_7:Tm) exhibit a different response for the different types of radiation. The different in response of the TL dosimeters gives advantage to be used in many fields as clinical, retrospective, space and personal dosimetry.

Due to borates significant properties: being equivalent to human tissue with an effective atomic number of 8.4 and their considerable response for all types of radiation[1], the borate dosimeters shall br condidered for further study. In addition, the borates are relatively stable chemical compounds and respond to radiation without serious problems to attempts to dope them with TL sensitisers such as the rare earths.

Schulman et al. 1967[2], concluded the first study on the TL of lithium borate compounds, afterwards many studies on various alkali and alkaline earth with detailed TL properties were done[3-7]. In spite of the fact that the magnesium borate (MgB_4O_7) compound were first prepared by Prokic[8,9], there are very few related issues were published. Driscoll et al.[10] found the sensitivity of their prepared magnesium borate (MgB_4O_7) was up to 12 times of Harshaw LiF chips after measuring the TL immediately after irradiation. They also found also a reduction in TL response with 90($\pm$2) % after 100 hours. At the same year, Barbina et al.[11] prepared magnesium borate with decrease in TL response for about 65% from the original value after 15 days and with less sensitivity 4 times that of LiF chips. In 1999, Furetta et al.[12] activated the MgB_4O_7 dosimeters with Dy and Na, this change led to increase in the sensitivity to be 25 times and the fading was improved to be only 35 % after storage at room temperature for 17 days.

All the previous work led to a conclusion that MgB_4O_7 is a significant dosimeter that can be motivated to prepare a homemade MgB_4O_7. Therefore, we shall attempt practically to improve the dosimetric characteristics of that substance.

EXPERIMENTAL PROCEDURE

Samples of magnesium borate (MgB_4O_7) with dysprosium (Dy) as a doped activator were prepared. The doping material Dy_2O_3 of 99.90% purity ranging from 0.01 to 0.4 wt% was added. Glass system composed 75 B_2O_3. 20 MgO. 5Na_2O. x Dy_2O_3 mol. %, ($0 \le x \le 0.4$ gm) was arranged. The materials used were of chemically pure grade in the form of H_3BO_3, MgO, Na_2Co_3 and Dy_2O_3.

The MgB_4O_7:Dy specimens were prepared by melt quenching technique using platinum 2% rhodium crucibles in an electric furnace. The batch was pre-heated at 500–600°C for almost an hour to evaporate the carbonates. The temperature of melting was 1200°C for one hour after ensuring the last traces of batches were disappeared. To avoid the presence of bubbles the glasses have been continuously stirred during the glass preparation. After that, the melt is poured onto stainless steel mold, then annealed at around 400 °C to remove thermal strains. Slabs of the desirable samples were prepared by grinding, then polishing with parafûn oil and adding a minimum amount of water. The thickness of the MgB_4O_7:Dy slabs was about 3 mm; whereas both the width and the length was not exceed than 6mm (the dimension of the TL-tray). The polishing process was completed with stannic oxide. The homogeneity of the glasses was examined using two crossed polarizers.

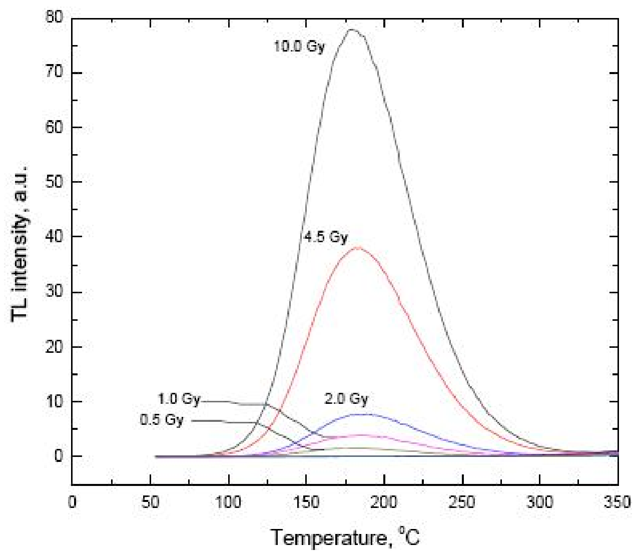
Two gamma sources are used in this study for both low and high doses irradiation. High gamma doses irradiation ranging from 500 Gy up to 7 kGy was performed using a ^{60}Co point gamma source (irradiation cell) installed at the National Center for Radiation Research and Technology (NCRRT) of the Egyptian Atomic Energy Authority (EAEA). The source activity is 50 KCi, with a dose rate of about 4.0 kGy/h at sample position. Low gamma doses irradiation ranging from 100 mGy up to 20Gy was performed using ^{60}Co Gammatron therapy unit (manufactured by Siemens, Germany) at National Inistitute of standards (NIS). The dose rate is 2.97 Gy/h at one meter from the center of the, calibrated ^{60}Co source, calibrated by the secondary standard dosimetry system of NIS (NPL system). The Harshaw 4500 TLD Reader was equipped with two photomultiplier tubes that can read independently; the reader operates on WinREMS software, which runs under Windows on a separate computer. Glow curves were performed over the experimentally studied range of temperature (100 - 400 °C). All TL measurements were taken after 24 hours with rate of 2°C/s. The prepared dosimeters were reused by annealing them at 400°C for one hour.

RESULTS AND DISCUSSION

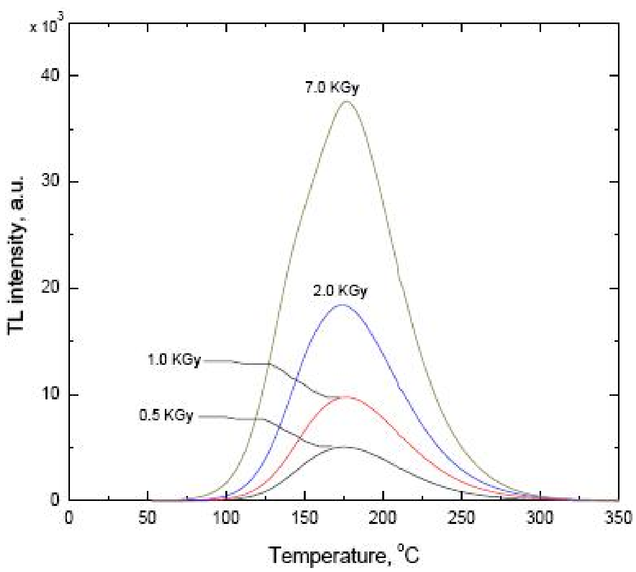
Glow curve structure

By changing the concentrations of the doping Dy from 0.01 up to 0.4 Wt %, it was found that the highest

TL was achieved with doping Dy of 0.3 wt%. The shape of the glow curves for different dose values recorded from 100 mGy to 7 kGy are represented in Figures. (1a and b). The glow curve structure of the home made MgB_4O_7:Dy phosphor is very simple and consisting of a single well defined peak which maximum value is achieved at around 187°C, which seems to be constant in position within ±2°C for all doses delivered. Non-existence of high temperature peaks means that there is no non-radiative recombinations of the luminescence centers and trapped electrons (retrapping) through the irradiation process.



(a)



(b)

Figure 1 : Typical glow curves of MgB_4O_7:Dy obtained with a linear heating rate of 2°C/s for different dose, (a) of low values from 0.1 to 10 Gy, and (b) of high values up to 7 kGy

Dose response

The TL response as a function of the exposure to gamma radiation was measured from 100 mGy up to 7 kGy as represented in (Figure 2). Two gamma sources were used to study this wide range of doses. It was found that the dose response curve can be divided into three ranges. The first range represents the sub-linear portion from 100 mGy to 200 mGy, the second range represents the linear region from 0.2 Gy up to 2 kGy (fitted region) and the last range specified the supra-linear range from 2 kGy up to 7 kGy. Each of these ranges can be represented by the power equation:

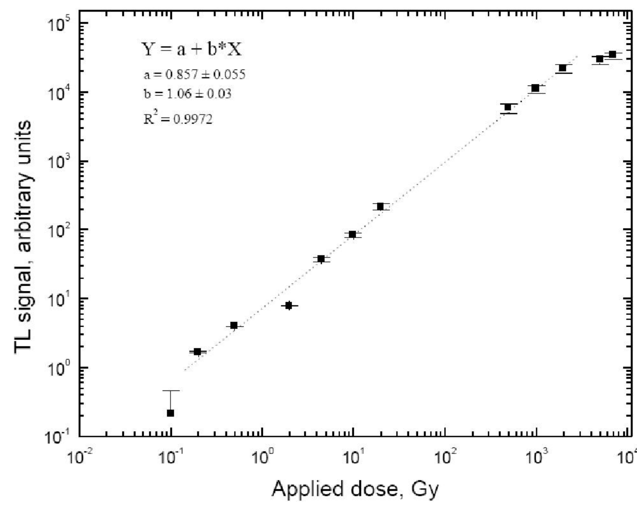


Figure 2 : The TL-dose dependence.

$$H_{peak} = a * D^b \text{ (Gy)} \quad (1)$$

where H_{peak} is the height of the main peak, D is the dose in Gy and a, b are the slop and the order of the power equation. As Figure 2 is represented on log-log scale which means that the linear part that has the order of the power equation (b) is equal to unity. Fitting that portion, it yields a value of b=1.02 with a regression coefficient R^2 = 0.9972. The results displayed a linear dose response ranging from 0.2Gy up to 2 kGy wider than that obtained by previous researchers. kGy For example, Barbina et al.[11] and Furetta et al.[13] had obtained a linear response up to only 1Gy and 40 Gy, respectively.

A good check of the linear property of the material can be done using the so-called linearity index or dose function f(D) (*14*).

$$f(D) = \frac{H_{peak}/D}{H_{n(peak)}/D_n} \quad (2)$$

where, $H_{n(peak)}$ is the height of the applied dose of the normalized D_n. Figure 3 shows the plot of f(D) versus dose. There is a high divergence from the normalized line at the lowest values (100 mGy) which can be attributed to the high uncertainty at that range. Also, a slight supralinearity effect seems to start from 2kGy up to the highest values of the delivered doses. The starting and ending points of the linear part represent the minimum and maximum detectable dose respectively. The minimum detectable dose depends essentially on the grain size of the detector, where it increases as the grain size decreases. However, the low detectable dose depends mainly on the heat treatments and evaluation procedures to eliminate the residual dose (zero dose or unirradiated readout) and decrease the standard deviation of this value in order to minimize the low detectable dose.

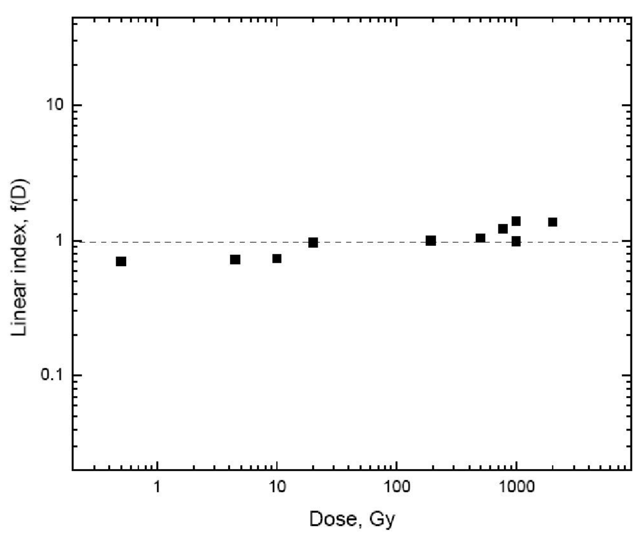


Figure 3 : The linearity index, f(D), plotted against the dose

TL sensitivity

The intrinsic TL sensitivity of a thermoluminescent material which is expressed as the TL yield per unit of mass divided into units of dose of ionising radiation. It provided a correction is applied to take into account the spectral TL emission to match the peak response of the TLD reader. In this study the TL sensitivity is expressed as the area under the glow curve area per unit of mass of dosimeter and per unit of dose of gamma rays ($TLmg^{-1}Gy^{-1}$) and compared relatively to the TL sensitivity of LiF:Mg,Ti (TLD-100). The investigated MgB_4O_7:Dy TL dosimeters show a TL sensitivity equal to 2.2 times higher than that of (TLD-100) as measured with the 4500 TL reader, taking into account the dependency on the spectral response of the TLD reader.

The current study revealed a sensitivity of 2.2 times higher than that of TLD-100; Furetta et al.[12] with the same dooping (1.9 times of TLD-100), while, the sensitivity was increased by adding Tm with Dy to 7 times[8] and 6 times after adding Na with Dy[13].

As the sensitivity depends on the readout and annealing regimes, it is very critical to know the impacts of that system for both the investigated material and the standard reference one (such as TLD-100) in order to compare between different data. The annealing system depends on annealing temperature, time of annealing and cooling mechanism. The readout system depends on the temperature range, the readout heating rate, the pre-irradiation readout (zero dose). Abd El-Hafez and Maghraby[15] concluded that TLD-100 is affected dramatically with the pre-irradiation readout, while that is not the case in other material such as CaF_2:Tm (TLD-300) as found by Abd El- Hafez et al.,[16]. The same behavior of the investigated dosimeter (MgB_4O_7:Dy) and CaF_2:Tm (TLD-300) are noticed, i.e. there is no high temperature peaks in both detectors. In other words; there is no non-radiative recombination of the luminescence center and the trapped electrons in the irradiation process.

Fading

As mentioned in the introduction section, all types of the previous prepared magnesium borate TL materials showed high fading of the main peak ranging from 35 - 65% through two weeks, at room temperature[11,12]. This shortcoming has been overcome by the development of MgB_4O_7:Dy with a simple glow curve, i.e., a single peak at about 187°C, and consequently fading was found to be drastically reduced. In order to determine the fading characteristics, MgB_4O_7:Dy samples were annealed and irradiated to a dose of 1.0 Gy of ^{60}Co. The investigated samples were stored in dark conditions at room temperature between 25-30°C. In order to minimize the effect of a possible reader drift, the samples were exposed at different periods of time prior of readout, from 1 day to 1 month. Readout was performed for all the irradiated samples at the same time.

Figure 4 illustrates the TL output as a percentage value referred to that at zero storage time versus the storage time in days. As per the figure, it is remarkable that the MgB_4O_7:Dy - TLD samples demonstrated a decrease of the TL response during the elapsed period of time in an exponential function.

$$Y=Y_0+A*\exp(-x/t) \quad (3)$$

where (Y_o) is represented the corresponding background TL i.e. the residual signal after annealing the samples two times at least; (A) is the measured highest TL intensity immediately after the irradiation process (t = 0 time). Whereas, (Y) is represented the residual TL signal after storage time (t). It was found that main peak was faded about 39% from its original value in about two weeks and decreased with 54% in one month at the ambient temperature. The fitted curve in Figure 4 has a regression coefficient R^2 equal to 0.95987. The results are in good compliance with the lowest fading of the previously published results although they are not satisfying. Further efforts should be exerted to minimize that unacceptable value of the fading effect.

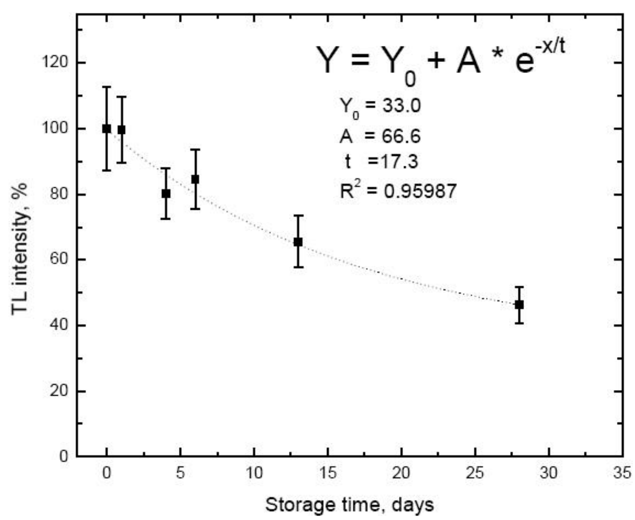


Figure 4 : Fading characteristic of MgB_4O_7:Dy dosimeters at the ambient temperature up to 30 days.

Reproducibility

The reproducibility of the dose measurements was assessed by reiterating the readouts of irradiated MgB_4O_7:Dy dosimeters, subject of study, irradiated with a dose of 1 Gy. Figure 5 shows the TL intensity against number of repetition for 7 repeated cycles. The illustrated TL values represented the average of readings of three dosimeters that read out at the same conditions and with standard deviation (σ), taking into account that this property was achieved with the same dosimeters. The results show that the dosimeters can measure gamma doses with less than 11.5 % as variation based on standard deviation for 7 sequential measurements. The other researchers who used the same doping material Dy did not mention the effects of reproducibility. Our results are in compliance with a value of 10 % that was obtained by Dogan et al., 2009 for Ce as a doping material.

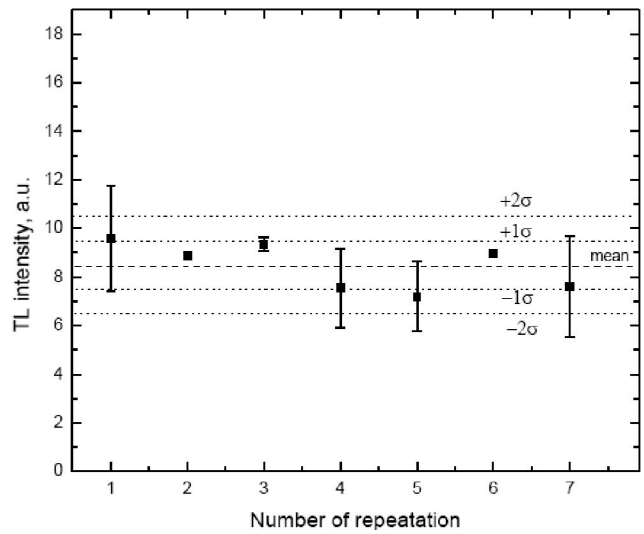


Figure 5 : The reproducibility of the radiation response of MgB_4O_7:Dy dosimeters for seven successive cycles

CONCLUSIONS

Using the thermoluminescence technique, the dosimetric characteristics of in-house prepared MgB_4O_7 doped with Dy were investigated and compared with results obtained by previous researchers studied the dosimetric properties of MgB_4O_7:Dy. The reason for this shortage is most probable attributed to their high fading in very short time. However, the current study revealed a higher sensitivity than that prepared with similar doped dosimeters by Barbina[11] and Furetta et al.[13]. In addition, the range of the photon response is 0.2 Gy-2 kGy is bigger than that obtained by the previous researchers.

The prepared MgB_4O_7:Dy ships displayed a good dosimetric properties, however further study is needed to add a different doped materials like Na and Tm with Dy to find some way to minimize the percentage of fading in order to increase the application of MgB_4O_7:Dy in different fields especially in higher dose dosimetry.

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