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Thermoluminescence properties of Li₂B₄O₇:Cu material

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Abstract. The copper-doped lithium tetraborate $Li_2B_4O_7$:Cu (LBCu) is one of the famous tissue equivalent materials for the thermoluminescent dosimetry, very useful for applications in medical and personal dosimetry measurements. This material in powder and pellet forms was prepared by sintering method in the Laboratory of Applied Spectroscopy and Gemology-IMS-VAST. In this study the dosimetric characteristics of LBCu powder were examined: homogeneity of batch, reproducibility, dose response and evaluation kinetic parameters.

Keywords: Lithium Tetraborate, Thermoluminescent, Dosimetry

1. Introduction

The copper-doped lithium tetraborate $Li_2B_4O_7$:Cu (LBCu) is one of the well-known materials for thermoluminescence dosimetry with effective atomic number $Z_{eff}=7.4$ close to that of soft biological tissue. This material has high sensitivity, linear dose dependence, a wide operation dose range, and a weak dependence of the ionizing radiation energy. LBCu is very promising material for applications particularly in medical and personal dosimetry.

The first thermoluminescent material based on lithium tetraborate activated by manganese is commercialized by Harshaw under the name TLD-800. This material presented a very poor sensitivity, mainly caused by the light emission in the 600 nm region of the spectra, far from the response region of most of the photomultipliers used in commercial reader. The use of copper as activator instead of manganese overcomes the drawback of poor sensitivity, shifting the emission spectrum to about 370 nm [1, 2].

LBCu materials in powder and pellet forms have been prepared by sintering and melting method in the Laboratory of Applied Spectroscopy and Gemology-IMS-VAST. The LBCu samples were irradiated with ⁶⁰Co gamma source or X-ray tube copper anticathode at 25 kV, 5 mA. Thermally stimulated luminescence (TSL) measurements were carried out on the Harshaw Model TLD 3500 Reader.

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2. Results and discussion

2.1. Preparation and optimization

The LBCu powders [2, 4], and pellets [3, 5] were prepared by sintering method. Ethanol solution of CuCl₂ was mixed with stoichiometric amount of commercial $Li_2B_4O_7$. The dispersion was homogenized by stirring the mixture for 30 min, using a magnetic stirrer. Afterwards alcohol was allowed to evaporate at ambient temperature. Drying was completed in a laboratory furnace at 100 °C for 10-12 hours.

The pellets (4.5 mm in diameter and 0.95 mm in thickness) were then prepared by cold pressing the as-prepared LBCu powders.

Powder and pellets were kept in a platinum crucible and sintered at $870 \,^{\circ}\text{C}$ for 30 min in air atmosphere, followed by a natural cooling to room temperature. Powder was then ground and sieved to obtain grain size in the range of 75–200 μm , and the obtained powder was then heated again at 400 $^{\circ}\text{C}$ for 1h.



Fig 1. TSL glow curve of LBCu sample as a function of Cu concentration.

Fig 2. Maximum peak intensity and response of LBCu sample versus sintering temperature.

The sintering temperature and Cu concentration are important parameters which influence TSL properties of the material. Figure 1 and 2 show the TSL-response as a function of Cu concentration and sintering temperatures. It can be seen that the TSL-response increases gradually with increasing copper concentration and sintering temperature within the range from 0.1 to 0.15 mol% and from 870 to 880 °C respectively. Above these temperatures the samples undergo a complete melting and the TSL signal can not be collected again. At the temperature 890 °C, samples were melted. Therefore, the optimal temperature of sintering process is $870 \,^{\circ}$ C for copper concentration of 0.15 mol%.

2.2. Homogeneity

Evaluation of the homogeneity was carried out with 10 samples taken from a given batch and irradiated at the same radiation dose. The material exhibiting standard deviation about 2 % could be accepted for the dosimetric application.

Every batch of more than 3 g in weigh has to be homogeneity tested. The resultant TSL response was between 2.0-3.0 % variations based on standard deviation of 10 sequential measurements. According to the evaluating of the Radiotherapy Laboratory in Henry Mondor Hospital, our dosimeters show a standard deviation of 2.25 %.

2.3 Reproducibility

The TSL sensitivity of sample T4 is almost unchanged after 10 times of the TSL reading out. The fluctuation of the TSL intensity was estimated about 3.6 %. It indicates that LBCu is the reusable material in the dosimetric application.

2.4 TSL dose response

The dose response was checked by Henry Mondor hospital. The results of the measurements show a linear dose response in the study range of dose until 30 Gy (show in fig.3).



Fig 3. TSL response of the main glow peak versus gamma ray dose.

2.5 Evaluation of the kinetic parameters

Some selected TSL glow curves of sample after irradiation of different dose levels are shown in fig.4. The glow curve shows 4 peaks in the range from 50 to 450 °C. The temperature of every peak in the glow curve of LBCu does not shift with increasing dose levels. This means that all of the glow peaks in the investigated region ought to be of the first- order of TSL kinetics.

The variable heating rate method consists of monitoring a glow peak temperature with different heating rates. In accordance with the method, the temperature T_m depends on the heating rate β this dependence can be defined by the following equation:

$$ln(T_m^2/\beta) = E/kT_m + ln(E/ks)$$

where $s(s^{-1})$ is the frequency factor, E (eV) is the trap depth, $T_m(K)$ is the maximum temperature of the glow-peak and k is Boltzmann's constant. If T_m is measured for a number of different heating rates, E can be found from the slope of the straight line obtained from the plot of the logarithmic term $ln(T_m^2\beta)$ against $1/T_m$. The value of s can also be obtained from the intercept of the slope. Figure





Fig 4. The TSL glow curve of LBCu with different Xray irradiation times (at a heating rate of $2 \,^{\circ}\text{Cs}^{-1}$).

Fig 5. Glow curves of LBCu measured at various heating rates.

Table1. Activation energy E and frequency factor s are obtained using the variable heating rate method

Peak	Energy (eV)	Frequency factor (s^{-1})
1	0.43	$1.3 x 10^4$
2	0.98	7.3×10^8
3	1.17	4.1×10^9
4	1.46	$1.7 x 10^{10}$

5 shows TSL glow curves of LBCu irradiated x-ray at various heating rates and obtained results are presented in table 1.

3. Conclusion

The Lithium tetraborate doped with copper in powder and pellet form were prepared by sintered method. For the preparation of this dosimeter the optimum sintering temperature was $870 \,^{\circ}$ C and the optimum concentration of Cu was 0.15 mol%. The homogeneity from batch to batch was accepted to dosimetric application. The reusability of the dosimeters has been investigated for nearly 10 repeated readouts using the readout anneal, with no change on the TSL response.

Represented dosimetric characteristics make sintered solid lithium tetraborate dosimeters very promising and suggest potential use in different TSL dosimetry applications, particularly in medical dosimetry, and also for individual monitoring.

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