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RESEARCH ARTICLE

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Investigation on relation between TL and OSL of $Li_2B_4O_7$:Cu, In

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Abstract Polycrystal $Li_2B_4O_7$ (LBO) doped with Cu and In was prepared and then sintered at different temperatures. X-ray diffraction (XRD) was applied to get the parameters of the LBO structure, thermoluminescence (TL) and optically stimulated luminescence (OSL) were measured, and a second-order exponential decay model was fitted to the OSL decay curves. The results indicate that the original number of OSL traps that have captured electrons is linearly related with the sum of TL decay during the OSL process. Mean decay constant of OSL is related to the sintered temperature. The possible reason is that the sintered temperature affects the crystal sizes of the polycrystal, and consequently affects the stimulating light's intensity and the photoionization cross-section of the electrons, which have been captured by the traps.

Keywords $Li_2B_4O_7$, thermoluminescence (TL), optically stimulate luminescence (OSL)

1 Introduction

$Li_2B_4O_7$:Cu phosphors with high TL sensitivity, and the wide dose range of linear TL response [1], are most attractive because the effective atomic number Z_{eff} of 7.3, which is very close to that of the biological tissue [2], makes them an excellent tissue equivalent. Although $Li_2B_4O_7$:Cu phosphors had been investigated before 1980, commercially available phosphors are inconvenient for routine dosimetric use [3]. The direct sunlight causes considerable bleaching [3] and the OSL of the radiated phosphors can be observed. This means that this kind of phosphor can be possibly developed as an OSL dosimetric material.

The detailed mechanism of OSL and the relation between OSL defects and TL defects are not very clear in many materials. The relation between OSL defects and TL defects in $PbWO_4$:Y crystal has been studied through dose response [4], and the result shows that OSL defects are different from TL defects in $PbWO_4$:Y crystal. Because of the possible application in dosimetry and the considerable TL fading caused by light [5], $Li_2B_4O_7$:Cu, In phosphors can be used to study the relation between OSL and TL.

2 Materials and methods

2.1 Preparation of LBO:Cu, In

Copper(0.1 wt%) was added into lithium borate and the mixture was mixed thoroughly to form a homogenous powder, and then indium(0.1wt% or 0.5 wt%) was added and mixed again. The mixture was sintered for 0.5h at different temperatures (600°C, 650°C, 700°C, 750°C, 800°C, 850°C, 880°C).

2.2 TL and OSL

The measuring procedure of TL and OSL can be depicted as follows:

① The sample was irradiated in a dose of 0.1 Gy by a ^{90}Sr radiation source.

② Measuring the TL curve (TL1) with a heating rate of 2°C /s.

③ The sample was irradiated in a dose of 0.1 Gy by the ^{90}Sr radiation source again.

④ Measuring the OSL curve of the sample excited by a blue light with power 50 mw/cm^2 and central wavelength of 470nm for 40 seconds.

⑤ Measuring the TL curve (TL2) with a heating rate of 2°C/s.

The radiation and the measurement of TL and OSL were all carried out on a RisфTL/OSL DA-15B/C apparatus, with a U340 color filter in front of a photomultiplier tube. The TL curves are shown in Fig. 1.

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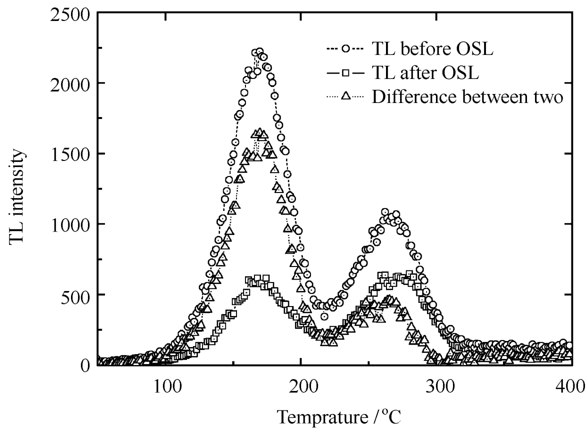

Fig. 1 TL curve of $Li_2B_4O_7$:Cu (0.1%), In (0.5%)

In Fig.1, the "···○···" denotes TL1, the TL curve before OSL. The "—□—" denotes TL2, the TL curve after OSL. The "···△···" denotes the TL1 minus TL2, the difference between TL1 and TL2.

The TL curves of $Li_2B_4O_7$:Cu (0.1%), In (0.5%) with different sintering temperatures are shown in Fig. 2. The result shows that the sintering temperature affects the TL peaks at about 180°C remarkably (including sensitivity and peak temperature), but less to the TL peaks at about 260°C.

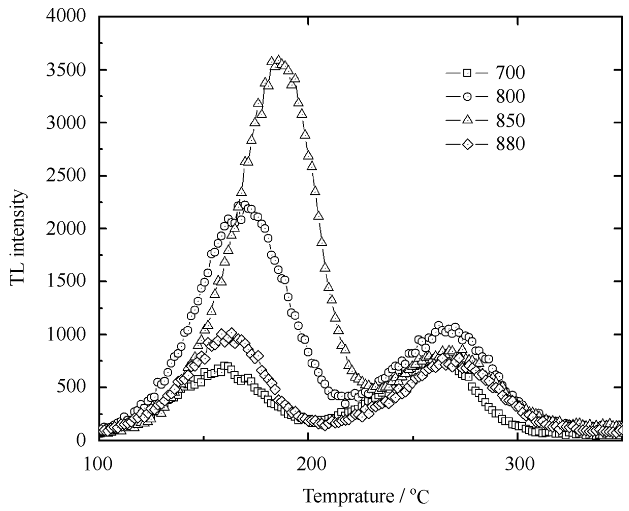

Fig. 2 TL of $Li_2B_4O_7$:Cu (0.1%), In (0.5%) sintered at different temperatures

The phosphor sintered at 850°C is the most sensitive among $Li_2B_4O_7$:Cu (0.1%), In (0.5%) phosphors, but the one sintered at 800°C is the most sensitive among $Li_2B_4O_7$: Cu (0.1%), In (0.1%) phosphors.

The OSL curves of $Li_2B_4O_7$:Cu (0.1%), In (0.5%) sintered at different temperatures are shown in Fig. 3. It can be seen from Fig. 3 that the OCL curves change greatly with the variation of sintering temperature. The OSL curves of $Li_2B_4O_7$:Cu (0.1%), In (0.1%) are similar to the ones of $Li_2B_4O_7$:Cu (0.1%), In (0.5%).

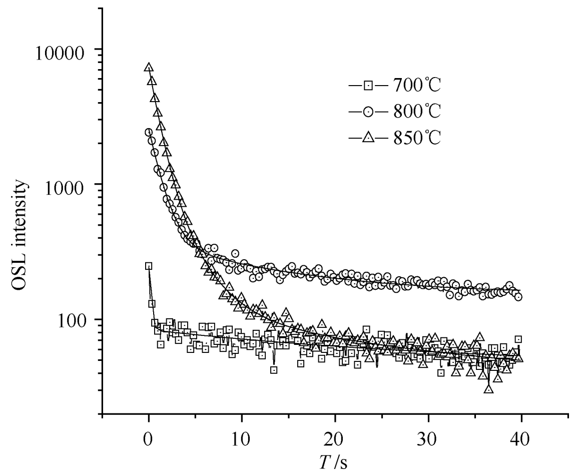

Fig. 3 OSL of $Li_2B_4O_7$:Cu (0.1%), In (0.5%)

2.3 XRD

The XRD curves of all phosphor powders were measured with a BD90 XRD apparatus, and they are all similar to the XRD spectrum of $Li_2B_4O_7$:Cu (0.1%), In (0.1%) sintered at 800°C as shown in Fig. 4a. It indicates that the crystal lattices of the phosphors do not change much with the variation of sintering temperature; all the crystal lattices of the phosphors are the space group of 4 mm, and the indexes of primary crystallographic plane are marked in Fig. 4a.

The details of maximum XRD peaks (the (112) crystal plane) of some phosphors are shown in Fig. 4b; the curves were fitted with Gaussian peak, and the half widths were marked in Fig. 4b. The curves of $Li_2B_4O_7$:Cu (0.1%), In (0.1%) phosphors are all similar to the ones of $Li_2B_4O_7$:Cu (0.1%), In (0.5%) phosphors, which were sintered at different temperatures, and the temperatures were marked on Fig. 4b too. The relation between the crystal size and the half width can be expressed with the expression [6]:

$$d = 0.89\lambda / w\cos\theta \tag{1}$$

The calculated crystal sizes of phosphors are 15.7 nm (650°C), 208nm (750°C) and 36.8nm (850°C). The actual sizes may be larger than the calculated ones because of the instrumental width increment. The result shows that the higher the sintered temperatures, the narrower the XRD peak, and the larger the crystal size.

3 Results and discussion

3.1 OSL curve fitting and the decay constant

Based on the OSL theory [7], the OSL intensity can be expressed as follows:

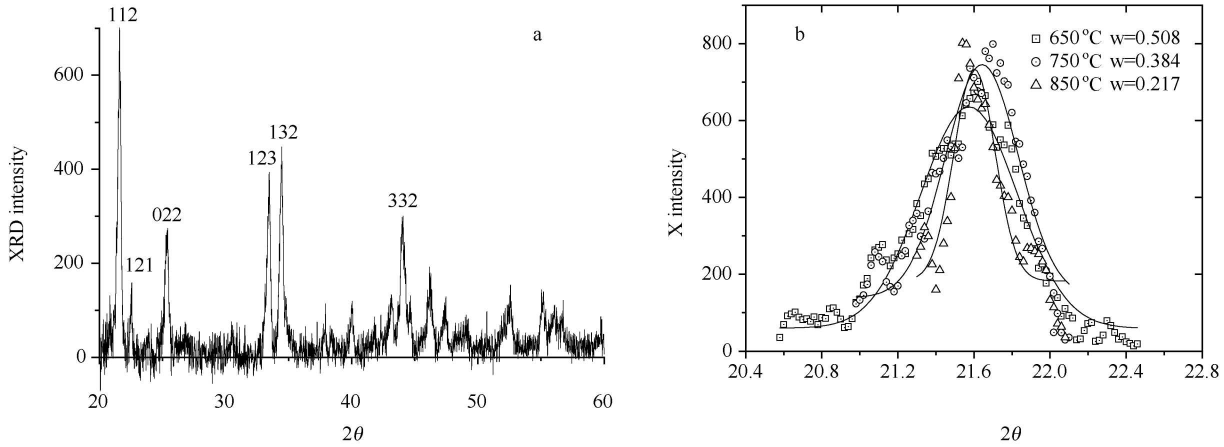

Fig. 4 XRD of LBO:Cu, In phosphors

a. XRD of $Li_2B_4O_7$:Cu(0.1%),In(0.1%)(800°C) b. Peak of (1,1,2) in XRD of $Li_2B_4O_7$:Cu(0.1%), In(0.5%)

$$I_{OSL} = \sum_{i=1}^{p} I_{i0} \exp(-t/\tau_i) = \sum_{i=1}^{p} \frac{n_{i0}}{\tau_i} \exp(-t/\tau_i) \qquad (2)$$

where p is the category number of optically active traps, I_{i0} is the initial CW-OSL intensity at $t = 0$ and τ_i is the decay constant, I_{i0} is directly proportional to the stimulation intensity Φ and the photoionization cross-section σ, the decay constant is proportional to $1/(\Phi\sigma)$, n_{i0} is the initial concentration of electrons trapped in traps. The OSL curves can be fitted with two exponential functions, the representative fitted curves are shown in Fig. 3, and the parameters are tabled out in Table 1 and Table 2.

The result of the tables shows that the fast decay constant has a trend to grow with the increase of sintering temperature, but the slow decay constant shows a contrary tendency.

3.2 The relation between TL1-TL2 and n_0

Based on the TL theory [8], when the irradiated phosphors are heated, the electrons trapped in traps are excited to the conduction band, where they combine with holes and emit light. The TL peak temperature is determined by the excited energy and frequency value of the traps.

TL intensity or TL peak area is directly proportional to the concentration of initial electrons before being heated. Some of the electrons trapped in TL defects are excited to conduction band by light during OSL measurement, and TL intensity decreases after OSL measurement. Therefore the area of TL1-TL2 in Fig. 1 is directly proportional to the decrease of the electrons trapped in TL defects.

The area of TL1-TL2 is proportional to the parameter n_0 ($n_0 = n_{10} + n_{20}$, the initial concentration of electrons trapped in OSL traps). The good linear relation between TL1-TL2 and n_0 is shown in Fig. 5.

In Fig. 5, the "—△—" denotes $Li_2B_4O_7$:Cu (0.1%), In (0.1%) phosphors, the "—○—" denotes $Li_2B_4O_7$:Cu (0.1%), In (0.5%) phosphors, and the dashed denotes the fitted line of the points. All the points of phosphors sintered at different

Table 1 OSL curve parameters of $Li_2B_4O_7$:Cu (0.1%), In (0.1%) sintered at different temperatures

T/℃	I_{10}	τ_1/s	n_{10}	I_{20}	τ_2/s	n_{20}
600	30.86	0.51	15.71	98.73	30.80	3041.08
650	32.46	0.44	14.24	75.06	38.92	2921.28
700	135.43	0.77	104.83	157.69	23.22	3662.13
750	214.86	0.19	40.05	197.52	17.48	3451.75
800	2833.26	1.08	3063.86	270.91	13.87	3757.67
850	1334.84	1.38	1844.96	215.26	11.92	2566.02

Table 2 OSL curve parameters of $Li_2B_4O_7$:Cu (0.1%), In (0.5%) sintered at different temperatures

T/℃	I_{10}	τ_1/s	n_{10}	I_{20}	τ_2/s	n_{20}
600	41.36	0.16	6.71	6.68	61.69	412.25
650	41.48	0.09	37.46	133.26	25.75	3431.02
700	159.77	0.20	32.59	56.49	54.81	3096.19
750	475.65	0.24	112.83	67.14	28.89	1939.91
800	2178.86	1.27	2759.59	216.39	15.19	3285.86
850	6557.81	1.12	7313.01	652.95	5.01	3268.59

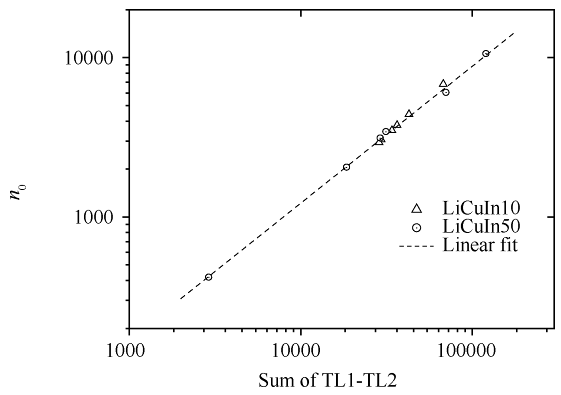

Fig. 5 The relation between the area of TL1-TL2 and n_0

temperatures are on the line. The parameters of $Li_2B_4O_7$: Cu (0.1%), In(0.5%) phosphors have a larger range of numerical value than that of $Li_2B_4O_7$:Cu (0.1%), In (0.1%) phosphors.

Because the decrease of electrons trapped in TL defects is directly proportional to the area of TL1-TL2, the n_0 (the initial concentration of electrons trapped in OSL traps) is generally proportional to the decrease of electrons trapped in TL defects. It means that OSL traps are correlative with TL traps in LBO:Cu, In phosphors. The relation between TL traps and OSL traps in LBO:Cu, In phosphors is different from that in $PbWO_4$:Y crystal [4].

3.3 The relation between OSL and sintering temperature

The result in Sect. 2.3 shows that the crystal size is related to sintering temperature: the higher the sintering temperature (below melting point), the bigger the crystal size will be. When the sintering temperature is low (for example T=650°C), the crystal size is small and there are more borderlines or defects in the polycrystal. Some small cavities may even exist in phosphors because the phosphors are not so compact. When light is transmitted in phosphors, most of it would be absorbed or scattered by the borderlines or defects. Therefore, the intensity of the light casting on the electrons seized by TL or OSL defects is weak, and the OSL decay constant is long. When the sintering temperature is high (for example T=850°C), the crystal size is larger and there are less borderlines or defects in the polycrystal. Moreover, fewer small cavities exist in phosphors too, because the phosphors are relatively compact. When light is transmitted in phosphors, little would be absorbed or scattered by borderlines or defects. Therefore the intensity of the light casting on the electrons seized by TL or OSL defects is stronger, and the OSL decay constant is shorter. This may be the reason why the OSL slow decay constants in Table 1 and Table 2 tend to decrease with the sintering temperature. The OSL fast decay constants may have some relation with the surface characteristics of the phosphors, and the reason why the OSL fast decay constants in the Tables tend to increase with the sintering temperature needs to be further investigated.

4 Summary

The OSL defects are closely related with the TL defects in polycrystal phosphors of $Li_2B_4O_7$:Cu, In. The sintered temperature affects the crystal sizes of the polycrystal, the stimulating light's intensity and the photoionization cross-section of the electrons captured by OSL or TL defects, and consequently affects the OSL decay constants.

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