JNS 1 (2012) 199-203

Journal of
NANOSTRUCTURES
JNS
NANOSTRUCTURES
JNS.kashanu.ac.ir

Thermoluminescece properties of BeO:Mg nanoparticles produced by sol-gel method

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Article history:
Received 3/12/2011
Accepted 22/2/2012
Published online 1/3/2012

Keywords:
Nanostructure
BeO
Sol-gel
Thermoluminescence

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Abstract

Beryllium oxide (BeO) nanopowder was synthesized using sol-gel method and the effects of changes of chemical reagents were investigated. Because of tissue equivalence and other outstanding thermoluminescence (TL) properties of the bulk material in X and γ dosimetry, the applicability of this nanostructure in TL dosimetry was studied following irradiation to the ^{137}Cs source. The TL glow curve of this nanoparticle shows 3 component glow peaks at 391, 429 and 481 K. Computerized glow curve deconvolution program was used for obtaining the number of constituent glow peaks and trapping parameters. Activation energies of three components were obtained to be 1.4, 1.39 and 1.39 eV with kinetic orders of 2.03, 2.12 and 2.09 respectively. TL dose response of this phosphor is also investigated.

1. Introduction

Beryllium oxide (BeO) because of its various properties like heat conductivity, thermal stability, electrical resistivity, hardness, a wide energy gap, high transparency over wide spectra range, high radiation resistance and so onhas attracted considerable attentions since about 1950 [1, 2]. It is important that the effective atomic number of BeO is Z = 7.13, which is close to biological tissue (Z = 7.42). This property encourages one to test this composite in nano size as a biological dosimeter [3]. In contrast to most of Be compounds which are toxic, BeO is a nontoxic material which makes it a good candidate for personal radiation protection dosimeter [4]. Recently the thermoluminescence properties of

different nanoparticles have been studied [5-9]. In a bulk form, Beryllium Oxide (BeO) shows intense TL and especially good OSL peaks [10]. In dosimetry of X- and gamma rays, some of the advantages of beryllium oxide over other TL materials are their resistance to mechanical shocks, low fading, the absence of low temperature peaks and moderate energy dependence. The low neutron sensitivity makes the application of BeO promising in mixed gamma-neutron radiation fields. Considering the above mentioned outstanding properties of the bulk BeO, it is important to investigate the properties of this material in nano scale. The current paper discusses the thermoluminescence behaviour of the nanopowders of BeO produced by sol-gel method.

2 Experimental

2.1 Materials and characterization

All the chemical reagents such as $BeSO_4$, ethylene glycol, citric acid, $MgSO_4$ and DI water were purchased from Merck Chemicals. The structural characterization of nanoparticles was supported by X-ray diffraction (XRD) with Rigaku D-maxc III diffractometer using CuK_α radiations. SEM images were obtained using a scanning electron microscope model Philips XL-30 ESEM. All irradiations were made with ^{137}Cs gamma source. The TL response was recorded by a Harshaw model 4500 computer based TL reader. A heating rate of 1 °C/Sec was used for heating the samples and recording the glow curves.

2.2. Synthesis of BeO nanoparticles

This procedure in brief uses acid (citric acid usually) to the solution of $BeSO_4$ in ethylene glycol to construct polymer,then the polymer is built up by increasing the temperature up to about 150-200 degrees centigrade resulting $BeSO_4$ solution to become trapped in the holes through the polymer. By further heating the sample, the $BeSO_4$ molecules break to BeO and make up crystals in nanoholes where they are jailed [11]. The nanocrystals exposed to gamma ray were readout by the TL reader system. Knowing that any changes in the synthesis procedure leads to some changes in the material's properties and sensitivity, all different synthesis conditions were tested as far as possible. Results show that the produced nanoparticles via sol-gel method show higher thermoluminescent sensitivity for high doses than that of bulk material [12]. The production procedure of the BeO:Mg nanopowder is as following.

2 mmol of $BeSO_4$ (0.354 g) was dissolved in 10 ml ethyleneglycol 90% on the stirrer. Then, the heater was turned on and citric acid and Be salt with molar ratio of 15:1 was added to the solvent (for 2 mmol Be, we need 30 mmol acid, 6.304 g). Finally, 0.1 mol of $MgSO_4$ was dissolved in 10 ml DI water and 2 ml of the resulted solution (equal to 0.2% mol of Mg to Be) was added to the above mentioned solution. The heater temperature was increased slowly from 80 to 240 degrees centigrade through about 2 hours. In this time interval the gel (polymer) was formed and then burnt. As the gel formed, the magnet was pulled out of the solution. Now the burnt polymer should be removed from the produced powder and a white soft powder is achieved. The powder is heated up to 670–700 °C by a rate of 11 °C/min and annealed at this temperature for 1 min to mostly half an hour then cooled down slowly. Once before receiving this pre-irradiation annealing procedure is required before irradiating the sample to gamma ray. All irradiations were made with ^{137}Cs source.

3. Results and discussion

3.1. SEM and XRD results

Fig. 1 shows the XRD pattern for BeO:Mg nanoparticles. This spectrum shows that the sample is BeO with hexagonal structure and corresponds with JCPDS card No 78-1562.

By the use of Debye-Scherer equation, crystallite sizes of calcined samples were estimated to be approximately 25 nm, which is in good agreement with the SEM images [13]. The SEM photograph of nanoparticles are shown in Fig. 2.

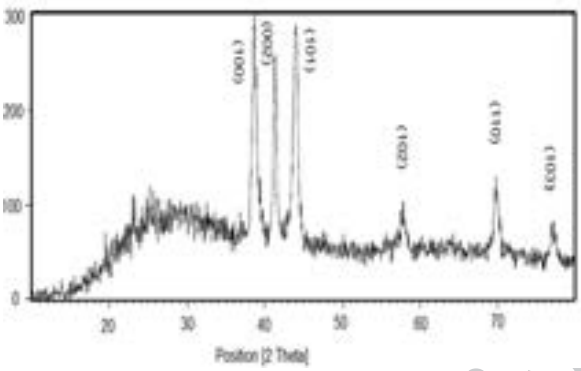

Fig. 1. XRD pattern of the synthesized nanoparticles.

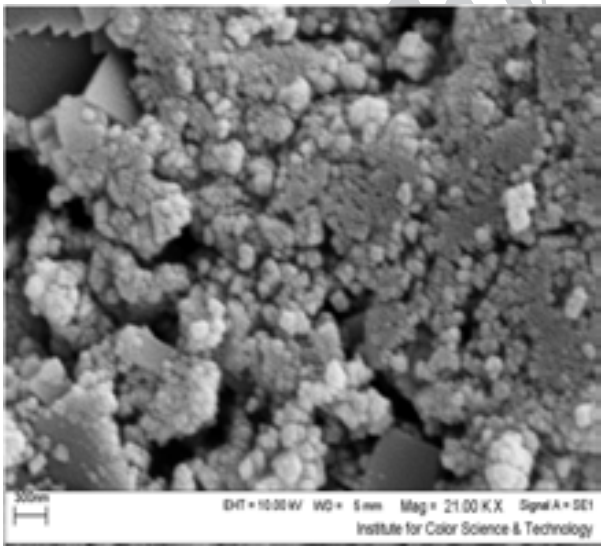

Fig. 2. SEM image of produced nanoparticles.

3.2. Thermoluminescence glow curve

Fig. 3 shows TL glow curve of BeO:Mg nanopowders exposed to γ-rays from ^{137}Cs source (open circles). TL glow curve of the bulk and nanostructure BeO:Mg has not reported yet. As is evident in Fig. 3, the glow curve contains three overlapping peaks around 490, 505 and 521 K.

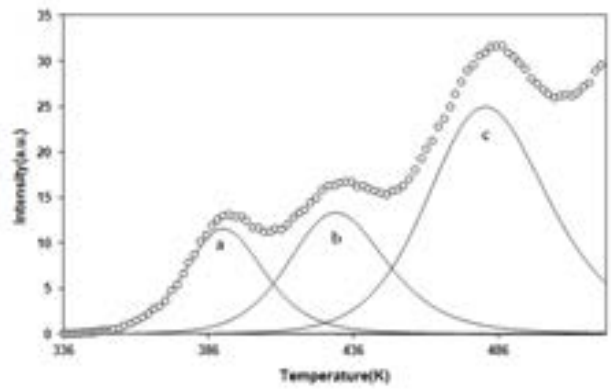

Fig. 3. Complex TL glow curve of thesynthesized nanoparticles which contains 3 overlapping glow peaks at 391, 429 and 481 K. Computerized glow curve deconvolution procedure was used for deconvolution procedure and obtaining the kinetic parameters.

Computerized glow curve deconvolution technique was used for obtaining the trapping parameters of component glow peaks based on three overlapping peaks. The computer program has produced in our laboratory using Levenberg-Marquart algorithm based onnon-linearleast square method. The function for thermoluminescence general order kinetics model was employedin our program for curvefitting is [14]:

$$I(T) = I_m b^{\frac{b}{b-1}} \exp\left(\frac{E(T-T_m)}{kTT_m}\right)\times$$

$$\left\{\frac{T^2}{T_m^{\,2}}(b-1)(1-\frac{2kT}{E})\exp(\frac{E(T-T_m)}{kTT_m})+1+(b-1)\frac{2kT_m}{E}\right\}^{\frac{-b}{b-1}} \quad (1)$$

In which I_m is the TL intensity, T_m the maximum temperature, T the absolute temperature, E the

activation energy, k the Boltzmann constant and b the kinetic order. The free parameters of each TL glow peak in computerized glow curve deconvolution procedure are I_m, T_m, E and b. The used glow curve deconvolution function(in terms of I_m, T_m, E and b) has advantage over the deconvolutionfunction in terms of n_o (initial concentration of electrons in trapping states), s (the pre exponential factor), E and b as I_m and T_m can easily be estimated from the experimental glow curve asthe initial values for curve fitting procedure. Also the used function for general order of kinetics is more useful than the deconvolution functions for limiting cases of first and second- orders of kinetics since intermediate cases in which $1 < b < 2$ can be dealt with and it smoothly goes to first and second orders when $b \rightarrow 1$ and $b \rightarrow 2$ respectively. For testing the goodness of fit, the figure of merit (FOM) has been used [15]. FOM values lower than 2.5% show a good fitness to experimental glow curves. The activation energy, kinetic-order and maximum temperature of three mentioned peaks can be observed in Table 1.

Table 1. Kinetic parameters of three component glow Peaks of the synthesized nanoparticles.

Peak list	E(eV)	I_m	T_m	b
a	1.4	11.541	391	2.03
b	1.39	13.282	429	2.12
c	1.39	24.951	481	2.09

Fig. 4 shows the dose response of the synthesized nanoparticles. As in evident, the response is linear to absorbed dosesfrom up to about 1 kGy. Also the exposed phosphors were stored in dark before reading to reduction of fading effects.

4. Conclusion

Because of some known exceptional properties of the bulk BeO in TL radiation dosimetry, the synthesis and thermoluminescence characteristics of the magnesium doped BeO nanoparticles were investigated for the first time. The structural characterization of nanoparticles was supported by X-ray diffraction and SEM images.

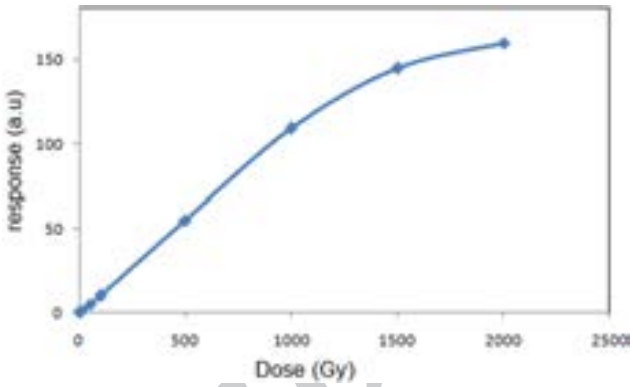

Fig. 4. Dose response of the BeO nanoparticles.

The TL glow curve of this phosphor contains three overlapping glow peaks around 490, 505 and 521 K. The linearity of dose response is of vital importance in TL dosimetry. A linear dose response up to about 1000 Gy was observed for this phosphor which makes it proper for high dose dosimetry. Also considering the fading of the low lying glow peaks, the third high temperature glow peak is recommended for using in TL gamma dosimetry.

Acknowledgements

Authors are grateful to the research council of the University of Kashan for financial support of this work.

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خواص ترمولومینسانس نانو ذرات BeO:Mg تهیه شده به وسیله ی روش سل ژل

چکیده:

نانوپودر بریلیوم اکسید با استفاده از روش سل ژل تهیه شده و اثرات تغییر معرفهای شیمیایی مورد توجه قرار گرفته اند. به دلیل اهمیت تعادل و دیگر خواص برجسته ترمولومینسانس مواد توده ای در تعیین مقدار مواد جذب شده X و γ کاربرد این نانو ساختار در تعیین مقدار مواد جذب شده ترمولومینسانس با استفاده از تابش به وسیله منبع ^{137}Cs دنبال شده است. انحنای تابش ترمولومینسانس این نانو ذرات سه پیک تابشی جزء سازنده در ۳۹۱، ۴۲۹ و ۴۸۱ کلوین را نشان می دهد. انرژی های فعال سازی سه جزء سازنده ۱.۴، ۱.۳۹ و ۱.۳۹ الکترون ولت با مرتبه سینتیکی ۲.۰۳، ۲.۱۲ و ۲.۰۹ بدست آمده اند..