EFFECT OF COBALT-60 RADIATION ON THERMOLUMINESCENCE PROPERTIES OF COPPER DOPED AND TERBIUM DOPED SODIUM MAGNESIUM BORATE

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I specially dedicate this work

To my dear parents

Hj. Ab Hamid Bin Ibrahim

Hjh. Che Salamah Bt Omar

Whose love, kindness, encouragement and prayer have brought me this far

To my siblings

For their endless laughs and tears

To my supervisor and ex-supervisor

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ABSTRACT

This thesis presents the investigation of thermoluminescence (TL) properties of copper (Cu) doped and terbium (Tb) doped sodium magnesium borate ($NaMgBO_3$) as a thermoluminescence dosimeter (TLD) material. Undoped $NaMgBO_3$ and two doped samples with various concentrations of dopant Cu (0.1- 0.5 mol%) and Tb (0.02- 0.1 mol%) were successfully prepared by combustion synthesis method. The effect of two different dopants and their concentrations was studied for TL properties subjected to Cobalt-60 gamma irradiation. The structure of the undoped sample was studied using X-ray diffraction and showed the presence of polycrystal structure with average size of 32 nm. The doped samples exhibit simple glow curve with single peak that occurs around 175 ^{o}C and 280 ^{o}C for Tb doped and Cu doped sample respectively. The undoped sample exhibits double peak glow curve with peaks located around 175 ^{o}C and 300 ^{o}C. Sample doped with 0.07 mol% Tb shows the highest TL response with activation energy of 1.00 eV. It also exhibits the highest TL response sensitivity which is 222 times compared to that of the undoped sample. Furthermore, it also gives the lowest fading effect with 5% signal loss over 28 days. Tb doped sample also shows good reproducibility with slightly different TL response reading within the range of (1.3 – 1.4) × 10^8 nC/g over 6 times readout. All samples show good linearity with linear correlation coefficient of 0.99518 for undoped sample, 0.99713 for Cu doped sample and 0.99518 for Tb doped sample. In conclusion, $NaMgBO_3$: 0.07 mol% Tb exhibits the best TL properties among the other samples and has the potential to become a new TLD material for radiotherapy and radiation processing.

ABSTRAK

Tesis ini mengemukakan kajian sifat luminesens terma (TL) sodium magnesium borat ($NaMgBO_3$) yang berdop kuprum dan berdop terbium sebagai bahan dosimeter luminesens terma (TLD). Sampel $NaMgBO_3$ tidak berdop dan dua sampel berdop dengan pelbagai kepekatan dopan Cu (0.1- 0.5 mol%) dan Tb (0.02- 0.1 mol%) telah berjaya disediakan dengan kaedah sintesis pembakaran. Kesan terhadap dua dopan berbeza dan kepekatanya dikaji bagi sifat TL dengan mendedahkan pada sinaran gama Cobalt-60. Struktur bahan dikaji menggunakan pembelauan sinar-X dan keputusannya menunjukkan kehadiran struktur polihablur dengan saiz purata 32 nm. Sampel berdop mempamerkan lengkung bara mudah satu puncak masing-masing berlaku sekitar 175 ^{o}C dan 280 ^{o}C bagi sampel berdop Tb dan berdop Cu. Sampel tidak berdop menunjukkan lengkung bara dua puncak dengan puncak terletak sekitar 175 ^{o}C dan 300 ^{o}C. Sampel berdop 0.07 mol% Tb menunjukkan sambutan TL yang paling tinggi dengan tenaga keaktifan 1.00 eV. Sampel ini juga mempamerkan kepekaan sambutan TL yang paling tinggi iaitu 222 kali berbanding sampel tidak berdop. Tambahan, sampel ini juga memberi kesan kepudaran terma yang paling rendah dengan 5% kehilangan isyarat selama 28 hari. Sampel berdop Tb juga menunjukkan sifat kebolehgunaan semula yang cukup baik dengan hanya sedikit perubahan pada bacaan sambutan TL dalam julat $(1.3 – 1.4) \times 10^{8}$ nC/g yang berlaku dalam 6 kali bacaan. Semua sampel menunjukkan sifat linear yang bagus dengan pekali korelasi linear 0.99518 untuk sampel tidak berdop, 0.99713 untuk sampel berdop Cu dan 0.99518 untuk sampel berdop Tb. Kesimpulannya, sampel $NaMgBO_3$: 0.07 mol% Tb mempunyai sifat TL yang terbaik antara sampel yang lain dan berpotensi untuk dijadikan bahan TLD yang baharu untuk radioterapi dan pemprosesan radiasi.

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LIST OF SYMBOLS

T	-	Temperature
m	-	Mass
R^2	-	Regression coefficient
M	-	Molar weight
θ	-	Diffraction angle
k	-	Boltzman's constant
τ	-	Half width at low temperature side of the peak
δ	-	Half width at high temperature side of the peak
ω	-	Total half width
E	-	Activation energy
s	-	Frequency factors
Z_{eff}	-	Effective atomic number
hv	-	Absorb energy
E_g	-	Trapping energy
p	-	Probability trapping electron

b	-	Kinetic order
T_M	-	Temperature at peak glow curve
I_M	-	TL response at peak glow curve
μ_g	-	Geometrical factor
CV	-	Coefficient of variation
σ	-	Standard deviation
D	-	Dose response
F	-	TL response
S	-	Sensitivity

LIST OF ABBREVIATIONS

TLD	-	Thermoluminescence dosimeter
TL	-	Thermoluminescence
OSL	-	Optically stimulated luminescence
CPT	-	Co-precipitation technique
CS	-	Combustion synthesis
SHS	-	Self-propagating high temperature synthesis
SSC	-	Solid state combustion
IR	-	Infra red
RE	-	Rare Earth
GCD	-	Glow curve computerized deconvolution
XRD	-	X-ray diffraction

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CHAPTER 1

INTRODUCTION

1.1 Overview

Radiation is the emission of energy in the electromagnetic (EM) wave form or high-energy particles moving through space or medium that cause ionization. There are many sources of radiations including nuclear, light from sun, lamps and even our gadget emits EM wave. Now days, radiation have been used in many field to improve quality of human daily life such as in medical field, agriculture and others. Unfortunately, too much exposure to ionizing radiation are harmful to living cells and human such as it induce DNA mutation and the most extreme effect is it can cause cancer and lethal effect. This case was experienced by Marie Curie where she died because of over-exposure to radium while she was discovering it. The other researcher who died caused by over exposure to the ionizing radiation was Thomas Edison who discovered fluoroscope. Hence, ionizing radiation can be highly hazardous where stringent prevention principles applied, and protective measures have to be taken to ensure safe work practice (Ali, 2011). Over the years researchers have been working to find the device for radiation detection purposes.

1.2 Research Background

The amount of radiation received by the body or radiation dose can be monitored by devices known as radiation dosimeter. It is an, instrument or system that measures or evaluates, either directly or indirectly, the quantities exposure, kerma absorbed dose or equivalent dose, or their time derivatives (rate), or related quantities of ionizing radiation (Podgorsak, 2005). There are four types of device that had been used as radiation dosimeters which are ionization chamber dosimetry system, film dosimetry, luminescence dosimetry and semiconductor dosimetry.

The amount radiatio received can be analyzed by exciting the trapped electron to higher energy level by heat so that it will return back to it ground level and emttig light that is known thermoluminescence (TL) phenomena and the material is called themoluminescence material. On the other hands, if the electrons have been exciting by light, this phenomenon is known as optically stimulated luminescence (OSL). This study will explore a new material for radiation dosimeter based on TL phenomena only.

To date the most common radiation dosimeter make used of of lithium fluoride doped with magnesium and titanium (LiF:Mg, Ti) with commercial name TLD-100. It has a high sensitivity to low dose and it is reusable compared to silver halide salt use in film badges and used once. These radiation dosimeters are used in medical, radiation worker personnel, archaeological dating and environmental monitoring applications. The other materials that can be considered as radiation dosimeters are calcium fluorite, calcium sulphate, lithium borate and quartz. However, these materials have disadvantages in terms of pre annealing procedure and reproducibility. These types of material need complicated annealing procedure and are not reproducible (Jiang *et al*., 2009). Nanomaterial are capable to enhance the device efficiency due to increasing the surface area of the particle. Nanoscale material exhibits unique chemical and physical properties, substantially differing from the bulk counterpart (Elssfah *et al*., 2007) especially in luminescence field.

Nano-pigment is inorganic or organic material, insoluble, chemically and physically inert into the substrate or binder with a particle size less than 100 nm. The particle size in nano scale means it has a high surface area which ensures higher surface coverage, higher number reflectance point and improves scattering (Ahmed *et al*., 2011). So, to enhance the efficiency of TLD response, nanomaterial is the most suitable material to replace glass material. Therefore, borate compounds composed of nanoparticles have been widely studied due to their features as glass former and also reported of being very advantageous materials for radiation dosimeter application (Santiago *et al.*, 1998 and Haghiri *et al*., 2013). The borate compound is being used as radiation dosimeter due to its high sensitivity, low cost and easy preparation. (Jiang et al., 2009) Borate compound presents an effective atomic number which is very close to human tissue ($Z_{eff} = 7.42$). Hence, borate is an ideal material to be considered to use as TLD material especially for medical and environment applications.

1.3 Problem statement

Several borate based TLD materials in glass form were successfully prepared using melt-quenching technique (Kar *et al*., 2015, Anjiah *et al*., 2014, Elkholy *et al*., 2010, Aboud *et al.*, 2014 and El-Adawy *et al*., 2010). Bahri et al. (2014) was studied calcium borate glass subjected to 6 MV and 10 MV X-ray irradiations. They found the sensitivity of germanium doped sample subjected to 6 MV is 1.28% higher compared to sample subjected to 10 MV. Same result was also shownh by Aboud et al. (2014) on the study of TL properrties of Cu-doped lithium potassium borate subjected to 6 MV photon. The Cu doped sample had 0.32 higher sensitivity compare to undoped one and both sample exhibited a good properties of TL. Based on above mentioned articles, the borate host provide good TL properties. However, Kar *et al*. (2015) was comparing TLD properies of lithium tetraborate (LTB) in varius forms as single crystal, polycrystalline and glass when subjected to 3-10keV X-rays. They found, Cu doped LTB single crystal shows the highest TL count. Several researcher come out with various method of synthesizing TLD material based on borate compound in crystaline form and in nano size such as solution combustion (Neharika et al., 2015, Bajaj et al., 2013, Nagpure *et al*., 2012, Bahl *et al*., 2013 and Guifang *et al*., 2010) and solid state method (Palan *et al*., 2016, Li *et al*., 2008 and Li *et al*., 2007). Materials prepared by both methods exhibit good TL and high TL sensitivity. However, by comparing the synthesizing methods, solution combustion method is much easier and quick than solid state where solid state method need the sample in thermal carbon reducing atmosphere at high temperature.

Based on TL properties studies by Kar et al. (2015) using lithium tetraborate glass they found that Cu doped sample show high TL response compare to Mn doped sample, the similar results also published by Faramawy et al., (2000). The studies show Cu doped in lithium borate glass have ability to enhance the material sensitivity. Bahl *et al.* (2013) synthesised magnesium borate doped with dysprosium and sodium *(MgB_4O_7:Dy,Na)* as a TLD material using combustion method. The sample was subjected to gamma (0.662 MeV) and proton (150 MeV). They succeed to get nano size 50 nm of *MgB_4O_7:Dy,Na* and the glow curve exhibits two peaks. The material also gives a simple glow curve structure, linear response over wide range of exposure, low fading, and good reusability. This material was found to be sensitive for high energy photon and low energy gamma radiation. Based on good result exhibited by (MgB_4O_7:Dy,Na), by Bahl et al (2015), Khan *et al.* (2015) work on sodium magnesium borate and used rare earth as a dopant *($NaMgBO_3$:Dy and Sm)* . The result found that the TL response for nano particle is far much higher, and have low fading affect which is about 5% from both material (MgB_4O_7:Dy,Na) and ($NaMgBO_3$:Dy and Sm). Based on these finding, a new nano size TLD material based on $NaMgBO_3$ can be prepared by combustion method. It is believed that nano particle be able to enhance the efficiency of TLD due to increasing surface area of the material. The material will be doped with Cu and also Tb where Cu dopant is abled to enhance the TL properies, whereas rare earth ion Tb replace the Sm or Dy. . This study aims to find the effect of dopat to the TL response of the material by choosing dopat from different group which is rare-earth and transition metal. Further reasons to explain the choosing dopant is explain in section 2.4. So, this study conduct to explain further the properties of TL from this material such as TL response, kinetic parameter, linearity, sensitivity, fading effect and reproducibility by comparing two different dopant that from two different group.

1.4 Objective

In order to gain the knowledge of the new TLD material, the study will focus on the following objectives.

I. To determine the crystalline structure and size of $NaMgBO_3$:Cu and $NaMgBO_3$:Tb phosphors using combustion method.
II. To determine basic TL properties such as glow curve, kinetic parameter, dose response, sensitivity, reproducibility and fading effect of $NaMgBO_3$:Cu and $NaMgBO_3$:Tb by subjected to Cobalt-60 gamma irradiation.

1.5 Scope of study

This study may provide a basis of synthesizing nano-size TL dosimeters material by combustion method. The preparation method makes use of urea and ammonium nitrate as a fuel and being heated at low temperature bellow 600 ℃ in 5 minutes. Three sample were prepared mainly undoped $NaMgBO_3$, Cu doped $NaMgBO_3$ where Cu in the range of 0.1-0.5 mol% and Tb doped $NaMgBO_3$ where Tb in the range of 0.02-0.00 mol%. XRD analysis for undoped sample was carried to determine the sample structure and size of particle. Sodium magnesium borate doped with copper and doped with terbium phosphors were exposed to Cobalt-60 gamma irradiation within dose ranges 10-80 Gy for TL properties analysis. The general characteristics of thermoluminescence dosimeter were read out using TLD reader model 3500 Harshaw at temperature setting 0-300 ℃. In TL glow characteristic measurement and analyst of kinetic parameter using peak-shape method, for undoped, Cu doped sample and Tb doped sample, the phosphors were exposed to 50 Gy gamma irradiation. The same 50 Gy dose exposure was used in fading characteristic which was read in a month and reproducibility was read in 6 cycles. TL-dose relationship using dose in the range 10-80 Gy gamma irradiation and the slope was analyst for sensitivity.

1.6 Significance of study

This study may explore a new nanosize TL material based on $NaMgBO_3$:Cu and $NaMgBO_3$:Tb phosphor as a new material for thermoluminescence dosimeter especially in radiation processing such as plant growth inhabitance and insects sterillisation and also radiotheraphy (Bos, 2001). It hopes that the study will provide some information on the studied nanomaterial in order to establish $NaMgBO_3$:Cu and $NaMgBO_3$:Tb phosphor as a potential radiation dosimeter material and enhance the efficiency of TLD.

1.7 Thesis Outline

Chapter 1 is a brief introduction to the study that consists of problem statements, objectives of the study, scope, significance, and finally thesis outline. The previous related literature is reviewed in chapter 2, which covers thermoluminescence theory, combustion method and advantages of borate compound. Chapter 3 explains the experimental methods used in the present work starting from the material identification, sample preparation and thermoluminescence measurement. Chapter 4 presents and discusses the results obtained from $NaMgBO_3$:Cu and $NaMgBO_3$:Tb phosphor. Last but not least, Chapter 5 provides the summary of the results from this study and gives some recommendations for future study.

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