THERMOLUMINESCENCE PROPERTIES OF DYSPROSIUM-DOPED CALCIUM BORATE GLASS FOR DOSE MEASUREMENT SUBJECTED TO IONIZING RADIATION

HAMIZA BINTI AHMAD TAJUDDIN

UNIVERSITI TEKNOLOGI MALAYSIA

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HAMIZA AHMAD TAJUDDIN

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I specially dedicate this work

To my lovely parents and parents in law
Halijah Binti Haji Zabidi
Muhammad Amin Chen Bin Abdullah
Suhana Binti Sudin
Mohd Shafie Bin Sukaimi

Whose love, kindness, patient and prayers have brought me this far

To my beloved husband
Mohd Hairi bin Shafie
For his love, understanding and support through my endeavor

To my lovely daughters
Arissa Amani and Damia Darwisyah
They fill my life with bundle of joy

To my sibling and friends

For their endless laugh and tears

Thank you so much!!!

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ABSTRACT

Thermoluminescene (TL) dosimeters are widely used in radiation therapy to verify the radiation dose received by cancer patients. This research was carried out to study the TL properties of Dysprosium (Dy) doped borate glass as a TL dosimeter subjected to photon and electron irradiations. The glass samples with various Dy concentrations were prepared by melt-quenching technique. X-Ray diffraction analysis indicates that the glasses are amorphous. The performance of the glass samples were compared to the TLD-100 in terms of TL response, linearity, sensitivity, dose response, fading and reproducibility. The addition of Dy_2O_3 in borate glass enhanced TL sensitivity and improved the TL intensity of the glass. It was found that calcium borate with Dy concentration of 0.30 mol% was the optimum concentration to produce the highest TL response. The TL intensity increased with Dy concentration up to 0.40 mol% before it begin to gradually decrease. This phenomenon is due to the concentration quenching. The TL properties were studied over the useful radiotherapeutic dose in the range of 0.5 Gy to 4 Gy when subjected to 6 and 10 MV photon and 6 and 12 MeV electron irradiation. The sensitivity of Dy doped calcium borate was 5.8 times more sensitive than that of undoped sample and 6.8 times less sensitive compared to TLD-100. A correlation coefficient of 0.98 was obtained for reproducibility and the minimum fading for this glass was 10% for 60 days observation. By using XCOM software, the Z_{eff} of the glass was found to be 12.14 and 13.60 for undoped and Dy doped glass, respectively.

ABSTRAK

Dosimeter luminesens terma (TL) digunakan secara meluas dalam terapi sinaran untuk mengenal pasti jumlah dos sinaran yang diterima oleh pesakit kanser. Kajian ini dijalankan untuk mengkaji sifat TL kaca borat yang didopkan dengan Dysprosium (Dy) sebagai dosimeter TL terhadap penyinaran foton dan elektron. Sampel kaca dengan pelbagai kepekatan Dy disediakan dengan teknik pelindapan lebur. Analisis pembelauan sinar-X menunjukkan kaca tersebut adalah amorfus. Prestasi sampel kaca telah di bandingkan dengan TLD-100 dari segi sambutan luminesens, kelinearan, kepekaan, sambutan dos, kepudaran dan kebolehulangan. Penambahan Dy_2O_3 ke dalam kaca borat meningkatkan kepekaan dan keamatan luminesens terma kaca tersebut. Kajian mendapati kalsium borat dengan kepekatan Dy sebanyak 0.30 mol% adalah kepekatan optimum yang menghasilkan sambutan luminesens terma tertinggi. Keamatan luminesens terma meningkat dengan kepekatan Dy sehingga 0.40 mol% sebelum ia mula berkurang secara beransur-ansur. Fenomena ini disebabkan oleh berlakunya lindapan kepekatan. Ciri luminesens terma telah dikaji pada julat berguna dos radioterapi 0.5 Gy hingga 4 Gy terhadap penyinaran foton 6 MV and 10 MV dan penyinaran elektron 6 MeV dan 12 MeV. Kaca borat yang didopkan dengan Dy mempunyai kepekaan 5.8 kali ganda berbanding dengan kaca yang tidak didopkan dan 6.8 kali ganda kurang kepekaan berbanding TLD-100. Pekali korelasi sebesar 0.98 dihasilkan untuk kebolehulangan dan kepudaran minimum bagi sampel ini ialah 10% untuk tempoh 60 hari. Dengan menggunakan perisian XCOM, Z_{eff} bagi sampel kaca ialah 12.14 bagi kaca tidak didopkan Dy dan 13.60 bagi kaca yang didopkan Dy.

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(where x = 0, 0.05, 0.1, 0.2, 0.3 and 0.4 mol %

LIST OF SYMBOLS

Å	-	Angstrom
B_2O_3	-	Pure borate
°C	-	degree calcius
C	-	Coulomb
CaO	-	Calcium Oxide
$CaCO_3$	-	Calcium Carbonate
d	-	Thickness
D	-	Absorbed dose
Dy	-	Dysprosium
Dy_2O_3	-	Dysprosium dioxide
E	-	Energy difference
ε	-	Energy
F (D)	-	Dose response
Gy	-	Gray
k	-	Boltzmann’s constant
LiF	-	Lithium fluoride
LiO	-	Lithium Oxide
m	-	Mass
M	-	TL signal
MgO	-	Magnesium Oxide
N_2	-	Nitrogen
n	-	Refractive index
n	-	number if trapped electron
P	-	Probability of escaping per unit time
T	-	Temperature
t	-	Time
τ_c	-	Characteristic time

w_i	-	Fraction of that element
Z	-	Atomic number of the atom
Z_{eff},	-	The effective atomic number
λ	-	Wavelength
σ	-	The standard deviation

LIST OF ABBREVIATIONS

EDXS	Energy dispersive X-ray spectroscopy
ECC	Element correction coefficient
ICRP	International Commission on Radiological Protection
LINAC	Linear accelerator
MU	Monitor Unit
PMT	Photomultiplier
RCF	Reader calibration factor
SSD	Source to surface distance
TLD	Thermoluminescence dosimeters
TL	Thermoluminescence
TTP	Time temperature profile
XRD	X-ray diffraction
WinREMS	Window Radiation Evaluation and Management system

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CHAPTER 1

INTRODUCTION

1.1 Research Background

Marie Curie died because of over expose to her discovery which is radium. Thomas Edison discovers about the fluoroscope but he stopped his work in this area after his assistant died due to an X-ray overdose. Two cases above had proved that radiation can give bed effect if wrongly handled it. After many years later, there are some improvements in understanding of the dangers of radiation, and yet human often fail to handle it safely. It is easy to become pleased about the dangers of radiation as it is invisible and odourless. As a result, radiation practitioner may expose patients as well as workers to higher levels of radiation than necessary. The hazard of excess radiation exposure is not insignificant and leading to a variety of health issues.

Recently, research groups have reported a number of radiation effects on the applications of glass as such the development of the optically stimulated luminescence methodology for use with doped glass material with its possible use as dosimeter material, measurement *in-vivo* of the absorbed dose in patients exposed to radiation therapy and diagnosis (Aznar *et al.*, 2002), and the use of borate glass as TLD material (Espinosa *et al.*, 2006).

Many authors have studied polycrystalline glass of alkaline and earth borate compounds. Recently, there are many studies on thermoluminescence dosimetry focusing on improving the TLD characteristic performance. Furthermore, the increasing use of ultraviolet, X-ray and gamma radiations in industrial and medical applications has motivated researchers on the search for new host materials with high potential of TL properties (J. Li *et al*, 2005). Why choose borate instead of other glass material? It is because borate glass compounds have been widely studied due to their features as glass formers and also reported of being very advantageous materials for radiation dosimetry application (Satiago *et al* 2001)

There are numbers of advantages of doped borate glass as radiation dosimeter. For instance, unlike conventional TLDs, the borate glass has very low hygroscopic nature. This is an important point for TL dosimeters because the water can cause an adverse effect on the TL efficiency of the material associated with non-radioactive relaxations during thermal stimulation. (McKeever, 1985).

Commercially available TLD materials such as lithium fluoride, calcium fluorite, calcium sulphate, lithium borate and quartz are used as dosimeters in medical, personnel, archaeological and environmental applications. However, they have many disadvantages such as long annealing time and not reproducible (Li Juan *et al*, 2008). In recent study, TLD materials have been extended to nano material and optical fibre. Fibre Optics for example is impervious to water and for some instance it becomes possible to locate the fibre dosimeter within a particular tissue.

The wide variety of TLD materials and their different physical forms allow the determination of different radiation qualities at dose levels from µGy to kGy. Most applications and major part of the literature are based on lithium fluoride doped with magnesium and titanium (LiF:Mg, Ti). However other materials such as $CaSO_4$: Dy is widely used and recently TL material LiF:Mg, Cu, P also show great potential as radiation dosimeter. In most materials, the linear interval is limited by super linearity and decay of the TL intensity at large doses.

The useful range was determined by linear dose dependence. High sensitivity such as high TL signal per unit absorbed dose is also one of the requirements for TLD material. High sensitivity is an important requirement if the TLD materials are used as personal and environmental radiation monitoring. Low dependence of TLD response on the energy of the incident radiation and long fading, i.e the ability to store dosimetric information for a long time are also required as TLD materials. The luminescence spectrum should match the maximum spectral sensitivity of the photomultiplier and the TL disimetric material should be mechanically strong, chemically inert and radiation resistant. Generally, these requirements should be fulfilled in order to produce high quality TL dosimeter.

Rare earth elements are interesting dopants donating material in host glass. In many cases, such as in optical absorption or light emission, there exists a direct relation to the energies in the ground and excited states of the electron system. In recent years, studies on rare earth doped glasses have gained much interest of researchers for the reason that the particular 4f electronic configuration of rare earth in varied glass matrixes leads to emissions from ultraviolet to infrared with many potential uses including in thermoluminescence dosimeter (Ferhi *et al.*, 2009).

The present studies intend to investigate new material based on borate (B_2O_3) glasses and mixed with other material such as calcium. The presence of dopants in calcium borate glass can greatly enhance the TL sensitivity to ionizing radiations by providing an increase number of traps. Furthermore, addition of dopant can enhance the chemical durability and stability of the glass host.

1.2 Problem Statement

Although the fast development of active real-time electronic dosimeters, passive dosimeters which integrate absorbed dose over a period of time are still in demand and are frequently obligatory in radiation protection. Thermoluminescence dosimeters are now gaining popularity in individual dosimetric services, replacing dosimetric films, which were commonly used in the past century (Olko *et al*, 2006). The property of thermoluminescence materials to ionizing radiation depends on the relationship between the absorbed dose and the intensity of light emitted. In personal dosimetry, thermoluminescence detectors are commonly used due to their major advantages such as high sensitivity, low doses as low as 1 μGy to be measured, and linear dose response up to at least 1 Gy, good energy response, reproducibility, and resistance to high humidity and magnetic fields.

Despite all advantages stated above, there are also some disadvantages of any thermoluminescence detectors such as signal erased during readout, easy to lose reading, no instant readout, readout and calibration time consuming and not recommended for beam calibration. TLD phosphors that most frequently used in medical applications are LiF:Mg,Ti and LiF:Mg,Cu,P due to their tissue equivalence characteristics. However, these well-established materials have several notable drawbacks, including being hygroscopic and not able to store permanently dose information since the reading of the detectors erases the dosimetric information. TLDs only provide integrated dose information sometime after the patient has been irradiated. Therefore, the radiotherapist cannot adjust the exposure in real-time to ensure that the proper dose is delivered to the desired region. Additional limitations of TLDs include their poor dose reproducibility, limited dynamic range and in certain cases nonlinear responses (Soltani *et al*., 1992).

Even though many new long lasting thermoluminescence materials are developed, the thermoluminescence phenomenon by calcium borate remains to be explored. Thus, in this study, TL properties of Dy-doped calcium borate will be carried out.

1.3 Research Objectives

1.3.1 To prepare and determine the optical properties of undoped and Dy-doped calcium borate glass system.

1.3.2 To investigate the thermoluminescence properties of undoped and Dy-doped calcium borate glass subjected to photon and electron irradiations.

1.4 Scope of study

In this study, an important characteristic of dosimeters such as fading, TL glow curve, linearity, reproducibility and sensitivity characteristic of undoped and Dy-doped calcium borate glass subjected to photon and electron irradiation will be explored. This material might have potential as TLD material and can be used for variety application such as environmental or personal dosimetry.

The irradiation on the glass sample has been conducted at dose ranging from 0.5 – 4.00 Gy subjected to photon and electron irradiation. The ionizing sources were delivered by using Primus MLC 3339 linear accelerator machine (LINAC). The energy used in this study was 6 and 10 MV photon beams and 6 and 12 MeV electron beams.

The study of fading on calcium borate glass has been performed by using 10 MV photon beams for 1, 2, 3 and 4 Gy. Readings of TL response were obtained until 60 days after 24 hours irradiation. For reproducibility characteristic, the glass samples were exposed to 6 MV photon with 4 Gy absorbed dose.

In this study, the effective atomic number, Z_{eff} for glass sample has to be determined by using XCOM method. Besides that, XRD analysis was carried out to give confirmation on amorphous nature of sample. Since this glass sample was a new material prepared, an investigation of sample composition is very important. Therefore, Energy Dispersive X-ray Spectroscopy (EDXS) was used in this study.

The current chapter provide introduction to the problems associated with TL and a review of the existing literature regarding the subject. Chapter 2 explained previous study and theories regarding the TL models, radiation interactions, principle of TLD and their important characteristics as a radiation dosimeter and brief introduction to the technique related in this study. Chapter 3 describes the methodology and equipment used. In Chapter 4, a range of thermoluminescence studies and the results obtained are presented and discussed in detail. Chapter 5 summarizes the findings of this investigation, and provides an outlook for future study in this area.

1.5 Significant of the research

This work intends to study TLD material with chemically and physically stable and also has high TL response and good sensitivity. Calcium borate glass can easily be prepared at low melting temperature and low cost compared to other TL materials such as lithium fluoride, lithium borate, calcium fluoride and calcium sulphate. The doping ion, Dy in borate host glass is expected to enhance the TL intensity. Such study will gain knowledge for further research in the development of new TL materials for medical and environmental applications.

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