ELECTRON PARAMAGNETIC RESONANCE DOSIMETER MATERIAL PROPERTIES OF POTASSIUM HYDROGEN TARTRATE AND POTASSIUM TARTRATE HEMIHYDRATE

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ABSTRACT

The electron paramagnetic resonance (EPR) of potassium hydrogen tartrate (PB) and potassium tartrate hemihydrate (PT) as a EPR radiation sensitive material was investigated. The aim of this study is to find a new bone-equivalent dosimeter material in clinical use. PB and PT irradiated with X-ray were studied. The energy dependence, dose-response, and radical stability of PB were studied. The simulation data was verified with the experimental data in order to study the capability of the spectra generated by the EasySpin. Both samples were exposed to X-ray in the range of absorbed dose, 1 - 9 Gy. The data measurement for EPR spectra was conducted 24 hours after the irradiation process. The energy dependence of PB sample is equivalent to the energy dependence of bone from 1 keV to 2 keV and above 100 keV to 1 MeV while the energy dependence of PT is bone-equivalent from 1 keV to 2 MeV and from 5 keV to 1 MeV. The features of the spectrum for PT sample that irradiated with X-ray is the same as that irradiated with gamma ray in the previous study. For PT samples, 3 peaks were observed and second peak was used throughout the study. The g-factor of PT samples was 2.00316 ± 0.00024. There were 2 peaks obtained from the spectrum of PB sample. First peak was used throughout the study. The g-factor obtained in this study for PB sample was 2.00942 ± 0.00130. The effect of the modulation amplitude and microwave power was studied within the range of 0.2 - 2 mT and 0.1 - 5 mW, respectively. In order to obtain the optimum EPR spectra, the selected modulation amplitude and microwave power were 0.7 mT and 0.6 mW, respectively. Linear dose-response was yielded for PT sample irradiated with X-ray between the dose range of 1 - 9 Gy while linear dose-response is obtained between the dose range of 2 - 9 Gy for X-ray irradiated PB sample. The normalised signal intensity of PT sample that irradiated with X-ray was higher than that irradiated with gamma ray. The signal of PB sample increased in the first 10 days after irradiation and maintained at a stable value for at least 50 days after irradiation. The EPR spectra generated by the simulation is the same as the experimental result. Nevertheless, the simulation had showed that 0.8 mT is the optimum modulation amplitude. This value is different with the value of experimental optimum modulation amplitude because the factor of signal-to-noise ratio and the simulation does not include the parameter of microwave power, as mentioned in previous study. In conclusion, PB sample is not that suitable for medical usage for low dose measurement that lower than 1 Gy. The higher absorbed dose range and potential voltage, or changing the radiation source to gamma rays, and the effect of temperature with relative humidity during the irradiation process is worth to be studied in the future.

ABSTRAK

Resonan paramagnet elektron (EPR) untuk kalium hidrogen tartrat (PB) dan kalium tartrat hemihidrat (PT) sebagai bahan peka sinaran EPR dikaji. Kajian ini adalah bertujuan untuk mencari bahan dosimeter baharu bagi tulang untuk tujuan perubatan. PB dan PT yang disinar dengan sinar-X dikaji. Kebergantungan tenaga, sambutan dos dan kestabilan radikal PB dikaji. Data simulasi disahkan dengan data eksperimen untuk megkaji keupayaan spektrum yang dihasilkan oleh EasySpin. Kedua-dua sampel disinar dengan sinar-X antara 1 - 9 Gy. Proses pengambilan data dijalankan 24 jam selepas proses penyinaran. Kebergantungan tenaga PB adalah sama dengan kebergantungan tenaga tulang dari tenaga 1 keV hingga tenaga 2 keV dan dari tenaga 100 keV hingga tenaga 1 MeV manakala kebergantungan tenaga PT adalah sama dengan kebergantungan tenaga tulang dari tenaga 1 keV hingga tenaga 2 keV dan dari tenaga 5 keV hingga tenaga 1 MeV. Ciri-ciri spektrum sampel PT disinar dengan sinar-X adalah sama dengan yang disinar dengan sinar gama dalam kajian sebelum ini. Sampel PT mempunyai tiga puncak dan faktor-g bagi puncak kedua yang digunakan dalam eksperimen ini ialah 2.00316 ± 0.00024. 2 puncak didapati dari spektrum sampel PB. Puncak pertama telah digunakan dalam kajian ini. Faktor-g untuk sampel PB ialah 2.00942 ± 0.00130. Kesan amplitud modulasi dan kuasa gelombang mikro dikaji dalam julat 0.2 – 2 mT dan 0.1 – 5 mW masing-masing. Amplitud modulasi yang dipilih ialah 0.7 mT manakala kuasa gelombang mikro yang dipilih ialah 0.6 mW untuk mendapat spektrum optimum. Sambutan dos linear didapati bagi sampel PT yang disinarkan dengan sinar-X antara dos 1 - 9 Gy manakala sambutan dos linear didapati bagi sampel PB yang disinarkan dengan sinar-X antara 2 - 9 Gy. Keamatan sinaran dinormalisasikan bagi sampel PT yang disinarkan oleh sinar-X adalah lebih tinggi daripada yang disinarkan oleh sinar gama. Isyarat sampel PB meningkat hingga hari kesepuluh selepas penyinaran dan menjadi stabil sekurang-kurangnya 50 hari selepas penyinaran. Spektrum EPR yang didapati melalui simulasi adalah sama dengan EPR spektrum yang didapati daripada eksperimen. Walau bagaimanapun, simulasi telah menunjukkan bahawa 0.8 mT ialah amplitud modulasi yang optimum. Nilai ini berbeza dengan nilai amplitud modulasi optimum eksperimen kerana faktor nisbah isyarat kepada bunyi dan parameter kuasa gelombang mikro tidak termasuk dalam simulasi, seperti kajian sebelum ini. Kesimpulannya, sampel PB tidak sesuai untuk kegunaan perubatan semasa pengukuran dos yang rendah daripada 1 Gy. Penggunaan julat dos terserap dan voltan yang lebih tinggi, atau menukar sumber radiasi kepada sinar gama, serta kesan suhu dan kelembapan relatif semasa proses penyinaran bernilai untuk dikaji pada masa akan datang.

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LIST OF ABBREVIATIONS

WHO	-	World Health Organization
MRI	-	Magnetic Resonance Imaging
CT	-	Computer Tomography
IGRT	-	Image-Guided Radiotherapy
IMRT	-	Intensity-Modulated Radiotherapy
SBRT	-	Stereotactic Body Radiation Therapy
EPR	-	Electron Paramagnetic Resonance
ESR	-	Electron Spin Resonance
UPM	-	Universiti Putra Malaysia
PB	-	Potassium Hydrogen Tartrate
PT	-	Potassium Tartrate Hemihydrate
UTM	-	Universiti Teknologi Malaysia
R.H.	-	Relative Humidity
Q-Dip	-	Quality-Dip
SHF	-	Super High Frequency
OSL	-	Optically Simulated Luminescence
S/N	-	Signal-to-Noise Ratio

LIST OF SYMBOLS

$\hbar$	-	Reduced Planck's constant
S	-	Spin Angular Momentum
m_l	-	Magnetic Quantum Number
μ_β	-	Bohr Magneton
$\bar{\mu}$	-	Magnetic Moment
B_0	-	Magnetic Field
ms	-	Electron Spin
g_e	-	Spectroscopic g-factor of Free Electron
L	-	Orbital angular momentum
h	-	Planck's constant
f	-	Frequency
g	-	g-factor
Z_C	-	Electrons Number of Carbon
Z_H	-	Electrons Number of Hydrogen
Z_O	-	Electron Numbers of Oxygen
Z_K	-	Electrons Number of Potassium
frac	-	Fraction of Electrons in The Molecule
Z_{eff}	-	Effective Atomic Number
μ_{en}	-	Linear Attenuation Coefficient
ρ	-	Density
T	-	Temperature
P	-	Pressure
k_{TP}	-	Correction Factor of Temperature and Pressure
ΔH_{pp}	-	Peak-to-Peak Linewidth

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CHAPTER 1

INTRODUCTION

1.1 General Introduction

In Chapter 1, the importance and objectives of this dissertation were stated. Cancer has become a major cause of death in the world nowadays. Different treatments conducted in order to cure cancer. One of the methods is radiation therapy. However, for different kinds of methods in radiation therapy, its radiation dose is a very important parameter. Alanine, the Electron Paramagnetic Resonance (EPR) dosimeter material has quite low sensitivity for clinical use. Hence, the search for a new bone-equivalent dosimeter material is necessary. In this study, the main aim is to search for a new EPR dosimeter material for bone. The objectives of this study are to investigate the EPR dosimeter material properties of potassium hydrogen tartrate (PB), and potassium tartrate hemihydrate (PT). The simulation was also conducted and compared with experimental result to understand its capability. However, in this study, only the material properties in the physics aspect are focused on.

1.2 Background of Study

Cancer is the major cause of death and becomes a very serious setback for every country in the world in order to increase life expectancy in 21st century (Bray et al., 2018). From the investigation was done by the World Health Organization (WHO), there are almost 50% of the new cases of cancer worldwide happen in Asia. Despite that, global patterns also show that more than half of cancer cases that lead to death in the world are reckoned to occur in Asia too (International Agency for Research on Cancer, 2018). Hence, many researchers have conducted experiments and researches in order to improve our understanding and technology in the field of cancer treatment. Nowadays, there are different possible methods that can be carried out for cancer treatment, such as surgery, chemotherapy, hormone therapy, bone marrow transplant, immunotherapy, and radiation therapy. Different fields of researchers, from biomedical, chemical, or physics fields, are putting effort by applying all their knowledge and experience during the process of conducting their new research to improve the methods of cancer treatment.

Radiation therapy is one of the important methods of cancer treatment. Radiation therapy can be considered as a vital role in order to cure cancer because 50 - 60% of patients require radiation therapy to treat cancer (Rosenblatt et al., 2015). Radiation energy can be used to kill cancer cells. An electrically charged particle, known as an ion, is formed by ionizing radiation. When the ion passes through the cancer cells, the ion and photon will deposit energy in the cancer cells. Therefore, the cancer cells are killed (Baskar et al., 2012). The radiation therapy technique for cancer treatment is still undergoing quick development nowadays. The radiation dose is a very important parameter during the procedure of radiation therapy. A suitable dosimeter that can help to measure the radiation dose during radiation therapy is required. If the oncologists are more familiar with the value of the radiation dose during radiation therapy, the process during cancer treatment can be optimised too (Ciesielski et al., 2003). Hence, the optimum goal when using radiation therapy for cancer treatment is to minimise the radiation dose to normal function cells and maximize its exposure to the cancer cells. Therefore, the value of radiation dose, especially during the exposure of low-dose radiation in clinical use, should be obtained precisely to avoid life-threatening incidents to occur during the process of cancer treatment for a patient.

Alanine is widely used as dosimeter material nowadays in clinical application. EPR/alanine system is used currently in the clinical application because it can fulfill a lot of important properties in clinical use, such as properties of independent of the sensitivity for energy range during the process of radiotherapy process and water-equivalent composition (Baffa and Kinoshita, 2014).

1.3 Problem Statement

Since radiation dose is an essential information that is required to be handled precisely during the process of cancer treatment, many types of research were conducted in order to search or synthesize a new material for dose measurement and dosimetry. EPR dosimetry has become a very important scientific tool for dosimetry application because it has been selected to quantify the concentration of radicals in metrology and routine now (Regulla, 1999). Alanine is one of the EPR dosimeter materials that has been used 60 years ago (Bradshaw et al., 1962). The dose of alanine-EPR dosimetry usually can be used within the range of 1 Gy to 100 kGy. Alanine can be used during measuring high radiation dose, which is typically more than 1 kGy. However, it is suitable to be served as an EPR dosimeter material during making measurements of low doses. This is because the signal to noise ratio obtained is low and the time taken for making the measurement is long (Nor et al., 2016). The tolerance levels of reference and treatment condition for 10 Gy, which is commonly the absorbed dose in clinical application, were found to be 5% and 6% (Baffa and Kinoshita, 2014). 5% of dose errors can result in up to huge probability changes in the tumour control during the radiotherapy process (Helt-Hansen et al., 2019). This limitation has become the main obstacle that required to be overcome in order to make alanine for clinical application.

Hence, many types of research have been conducted to improve the sensitivity of alanine such as the application of the "spiking" method (Geso et al., 2017). Despite improving the sensitivity of alanine for clinical use during low dose measurement, many researchers are also attempting to find new EPR dosimeter materials with higher sensitivity to replace alanine. However, many of the materials encounter setbacks, such as facing the problems of lower sensitivity than alanine and hence are not suitable to be used as an EPR dosimeter material. A new research field in searching for a material that is able to fulfill criteria and has higher sensitivity than alanine is currently explored by researchers from the worldwide country. The higher sensitivity of new materials can increase the signal during the process of dose estimations will enable it to be used in low photon energy medical applications, such as cancer treatment.

Besides that, there are experiments showing that the properties and conditions of human bones will be affected after the process of radiation therapy. The blood flow in the bone is reduced after radiation therapy. Bone strength and bone density are affected too (Hopewell, 2003). Furthermore, after the radiation therapy treatment for breast cancer, there were rib fractures found in the bone of patients. The probability of occurrence of rib fracture increases with a larger dose per fraction delivered to the patient (Overgaard, 1988; Aoki et al., 2015). Hence, since the bones will be damaged after radiation therapy and all of these damages are radiation dose-related, a new EPR dosimeter material irradiated with X-Ray may help us to reduce these damages caused to patients by obtaining the data of radiation dose deposited in the bones of patients. The new dosimeter material should be bone-equivalent energy dependence to detect the radiation dose deposited in the human bones. Besides that, high sensitivity and detectable in low dose range is preferred in order to increase the efficiency of the new dosimeter material in clinical use.

In this dissertation, a new material, potassium hydrogen tartrate, as known as potassium bitartrate (PB), is studied. This compound (PB, chemical formula: $C_4H_5O_6K$) was usually formed during the fermentation of wine. The organic properties of this material make this material serves to be a potential new EPR dosimeter material. Besides that, potassium tartrate hemihydrate (PT) that irradiated with X-ray together with PB was investigated in this study too. From the previous experiment of potassium tartrate hemihydrate that had irradiated with the gamma ray, the results show that the sensitivity of PT is 30 % higher than the sensitivity of alanine within the range of 1 – 9 Gy (Nor et al., 2016). This makes PT is possible to serve as a new potential EPR dosimeter material in order to measure the absorbed dose in the human bone. However, PT has some drawbacks that limit it to be used in medical applications. During the first two weeks after the exposure to gamma rays, the EPR signal is deemed to be unstable (Nor et al., 2016). After understanding the information and possibilities to serve as a new EPR dosimeter material, PB that had never been studied as an EPR dosimeter material before was investigated in this study.

1.4 Objectives of Study

The main aim of this dissertation was to search for a new bone-equivalent dosimeter material. The materials focused on this study were PB and PT. This study investigated the properties of PB and PT. Its ability to serve as a new EPR dosimeter material was investigated. There are some objectives required to be achieved in this research that are listed below:

1. To evaluate the dose-response and radical stability of PB.
2. To compare the energy dependence of the normalised mass-energy absorption coefficient for PB, PT, and bone.
3. To study the capability of the generated EPR spectra obtained by using simulation.

The first objective listed above could be achieved by studying the repercussion of microwave power, modulation amplitude, and dose absorbed by the irradiated samples. The parameters, such as modulation amplitude and microwave power, were adjusted in order to obtain optimum EPR spectra. Therefore, by manipulating the parameters mentioned above, the optimum results for radical stability and dose-response could be obtained in this experiment.

Secondly, the energy dependence of the normalized mass-energy absorption coefficient for PB was investigated and compared with the energy dependence of PT, alanine, bone and soft tissue. Bone-equivalent dosimeter material was preferred in this study. If the energy-dependence of the material found to be bone-equivalent, the amount of ionizing radiation that deposited inside the bone can be obtained during the process of radiation therapy (Greene et al., 2018).

Besides that, simulation by using Matlab with the EasySpin was also conducted in this dissertation. The samples that simulated by using Matlab were PB and PT. The PT sample and PB sample were tested by simulation in order to calculate the parameter based on theoretical meaning. The results of EPR spectra and optimum modulation amplitude obtained through simulation by using Matlab with the EasySpin were compared with the experimental result to understand the capability of the EPR spectra obtained by using EasySpin.

1.5 Scopes

In this experiment, the EPR dosimeter material properties of PB sample and PT sample were focused. The energy dependence of both samples was investigated and compared with the energy dependence of bones within the range of 1 keV to 1 MeV. The EPR dosimetry of the low dose range (1 - 9 Gy) was investigated. Hence, the PB and PT sample were irradiated with 150 kV, 6 mA X-rays with an absorbed dose range from 1 - 9 Gy. The relative humidity, temperature and the pressure during the irradiation process was 68.1%, 22.9°C and 1002.2 mbar, respectively. The absorbed dose that lower than 1 Gy was not investigated. It is because no signal was obtained in that range in the previous study, that PT was irradiated with gamma ray (Nor et al., 2016). The microwave frequency during EPR data measurement was lying within the X-band frequency, which was approximately 9.2 GHz. Despite that, in this experiment, the radical stability of the PB sample was investigated up to 50 days after irradiation. The range of modulation amplitude and microwave power investigated in this experiment were 0.2 mT – 2.0 mT and 0.1 – 5.0 mW, respectively. A bone-equivalent dosimeter material is more preferred than tissue-equivalent dosimeter material. In this experiment, the physical aspect of EPR dosimeter material for PT and PB samples are focused only.

1.6 Significance of Study

The problem of cancer has become an obstacle in the effort to extend the life expectancy of a human. One of the methods of cancer treatments involving radiation therapy, in which the radiation dose is the critical information required to be known during the process of treatment. 7% of dose error are clinically detectable nowadays. However, 5% of dose error can cause 10% to 20% of changes in tumour control probability and result in 20% to 30% to complication probabilities of normal cell (Helt-Hansen et al., 2019). Some bones may be destroyed during the process of radiotherapy. Hence, a new dosimeter material for bone can help to speed up the development of radiotherapy technique by leading the researchers to a further understanding of the dose absorbed by the bone of the patient and reduce the probability of dose-error in clinical use. For example, the technique of In vivo alanine/EPR dosimetry result in uncertainties with 6.6% for 0.5 Gy to 3.2% for 2 Gy in clinical use (Ciesielski et al., 2003). Therefore, the probability of recovering from cancer can be increased and the life expectancy of human will increase if a better EPR dosimeter material for low dose measurement in clinical use is found.

Furthermore, the nuclear plant leaking accidents, such as Fukushima Daiichi Accident in 2011 and Chernobyl Disaster in 1986 may occur more frequently due to increasing number of nuclear power plant in the world (Lu et al., 2020). A suitable dosimeter material for bone may become a potential tool to be used during the medical treatment by understanding the radiation dose deposited in the human bone in these disastrous accidents. Therefore, a new EPR dosimeter material for bone with higher sensitivity and detectable at low dose is essential in clinical use. The bone-equivalent dosimeter material can help to save lives too during the exposure to radiation due to the act of war or terrorism in future too (Swartz et al., 2007).

1.7 Outline of Dissertation

This study included five chapters. In Chapter 1, the background of the study and the significance of the study were highlighted and the gaps of research were mentioned in the part of the problem statement. The main aim and objectives associated with scopes of this research were stated in order to be achieved in this dissertation.

Chapter 2 had presented the brief history, theory, and function for the electron paramagnetic resonance (EPR). A piece of important information, g-factor that must be studied in this experiment is explained too for a better understanding of this experiment. The conditions to become a good dosimeter material were stated with examples given from the study by using potassium tartrate hemihydrate conducted before. More previously tested materials were discussed in this chapter. The material properties of potassium hydrogen tartrate are studied too.

Chapter 3 outlined the research methodology in detail. The procedures for four important stages in order to achieve the aim and objectives stated above were listed and explained thoroughly. The four important stages outlined in Chapter 3 including the preplanning stage, the irradiation stage, the data taking stage, and the simulation stage.

In Chapter 4, the results obtained from Chapter 3 were recorded down. The EPR spectrums obtained were shown and discussed in this chapter. The effects caused by different modulation and microwave power were mentioned too. The dose-response, radical stability, and the energy dependence of the normalized mass energy absorption coefficient for potassium hydrogen tartrate were discussed. Besides that, the EPR spectrums obtained through simulation by using Matlab with EasySpin were compared with the EPR spectrums obtained from the experiment to understand the capability of the EasySpin App.

Chapter 5 was the conclusion part that concludes this study. The conclusion part included the brief information and results discussed in Chapter 4. The efforts and the outcomes in order to achieve the aim and objectives for this dissertation were concluded too in this chapter. Some suggestions were provided in order to improve the quality of the study and the future research direction in order to search for new dosimeter material could be determined too.

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