ENHANCEMENT OF DOSE RESPONSE AND NUCLEAR MAGNETIC RESONANCE IMAGING OF PAGAT POLYMER GEL DOSIMETER BY SILVER AND PLATINUM NANOPARTICLES

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By

NAJMEH DEYHIMIHAGHIGHI

Thesis submitted to School of Graduate Studies, Universiti Putra Malaysia, in Fulfillment of the Requirements for Degree of Master of Science

May 2014

With great respect, I would like to dedicate my dissertation work to my family. A special feeling of gratitude to my loving parents, Mohammad Deyhimihaghighi and Marzieh Ameli, whose words of encouragement and push for tenacity ring in my ears, for all their love, sacrifices and faith. My sister, Maryam, has never left my side and is very special person for me.

I also dedicate this dissertation to the memory of my grandmothers for their blessfull prayers.

Abstract of thesis presented to the senate of Universiti Putra Malaysia in fulfillment of the requirement of the degree of Master of Science

ENHANCEMENT OF DOSE RESPONSE AND NUCLEAR MAGNETIC RESONANCE IMAGING OF PAGAT POLYMER GEL DOSIMETER BY SILVER AND PLATINUM NANOPARTICLES

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May 2014

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Radiotherapy is a treatment technique used to inactivate cancerous cells using ionizing radiation, typically of high-energy photons or electrons beam, which are delivered to the tumor volume without destroying the healthy surrounding tissues. The absorbed dose distributions in tissue volume can be studied in three dimensions (3D) by using a soft tissue equivalent polymer gel dosimeter, which centers on polymerization of monomers induced by free radicals as a result of radiolysis of water by ionizing radiation. In the presence of metal nanoparticles, the dose sensitivity of a polymer gel may be improved, from which its 3D dose distribution of nuclear magnetic resonance imaging (MRI) can be used in radiotherapy treatment plans.

In the present study, the normoxic polyacrylamide gelatin and tetrakis hydroxy methyl phosphoniun chloride (PAGAT) (4.5% N, N′-methylen-bis-acrylamide (bis), 4.5% acrylamid (AA), 5% gelatine, 5 mM tetrakis (hydroxymethyl) phosphonium chloride (THPC), 0.01 mM hydroquinone (HQ) and 86% deionized water) polymer gel dosimeters were synthesized with and without the presence of silver (Ag) and platinum (Pt) nanoparticles. The Ag nanoparticles with average particle sizes of 20 nm and particle concentration of 3.14×10^{-2} mg/l and Pt nanoparticles with average particle sizes of 10 nm and particle concentration of 1×10^{-2} mg/l were synthesized by laser ablation method from their respective metals in distilled water. The concentration of metal nanoparticles were varied from 3.14×10^{-2} to 9.42×10^{-2} mg/l for Ag nanoparticles and 0.5×10^{-2} to 3×10^{-2} mg/l for Pt nanoparticles to form two types of PAGAT polymer gel dosimeters before irradiating with 6 to 25 Gy of 1.25-MeV ^{60}Co gamma rays. The predominant gamma rays interaction with matter is by Compton scattering effect as the photoelectric absorption effect is diminished.

MRI evaluated the polymerization of the dosimeters and the gray scale of the MRI film was determined using an optical densitometer. The results of optical densities demonstrate that the amount of polymerization increased with an increase in absorbed dose, while the increase of depth inside the dosimeters has a reverse effect. Moreover, it was found that there was a significant increase in the optical density-

dose responds by 27.10% for dosimeters adding with 1×10^{-2} mg/l Pt nanoparticles and by 11.82% for dosimeters with 6.28×10^{-2} mg/l Ag nanoparticles.

Abstrak tesis yang dikemukakan kepada senate Universiti Putra Malaysia sebagai memenuhi keperluan untuk ijazah Master Sains

PENINGKATAN SAMBUTAN DOS DAN PENGIMEJAN RESONANS MAGNET NUKLEAR DOSIMETER GEL POLIMER PAGAT OLEH NANOPARTIKEL PERAK DAN PLATINUM

Oleh

NAJMEH DEYHIMIHAGHIGHI

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Radioterapi adalah satu teknik rawatan yang digunakan untuk melumpuhkan aktiviti sel-sel kanser menggunakan sinaran mengion, biasanya daripada alur foton atau elektron bertenaga tinggi yang ditujukan kepada isipadu tumor tanpa memusnahkan tisu-tisu sekeliling yang sihat. Pengagihan dos diserap dalam isipadu tisu boleh dikaji dalam tiga dimensi (3D) dengan menggunakan dosimeter gel polimer bersifat setara dengan tisu lembut, yang tertumpu kepada pempolimeran monomer oleh radikal bebas akibat daripada radiolisis air oleh sinaran mengion. Kehadiran nanopartikel logam menyebabkan sensitiviti dos gel polimer boleh ditingkatkan, yang pengedaran dos 3D daripada resonans magnetik nuklear pengimejan (MRI) boleh digunakan dalam pelan rawatan radioterapi.

Dalam kajian ini, poliacrilamida gelatin normoxic dan tetrakis hidrolesil metil fosfonium klorida (PAGAT) (4.5% N, N′ bis-metilin-acrilamida (bis), 4.5% acrilamida (AA), 5% gelatin, 5 mM tetrakis (hidroksimetil) fosfonium klorida (THPC), hydroquinone (HQ) 0.01 mM dan 86% deionized air) polimer gel dosimeters telah disintesis dengan dan tanpa kehadiran nanopartikel argentum (Ag) dan platinum (Pt). Nanopartikel Ag dengan saiz zarah purata 20 nm dan ketumpatan zarah 3.14×10^{-2} mg/l dan nanopartikel Pt dengan saiz zarah purata 10 nm dan ketumpatan zarah 1×10^{-2} mg/l telah disintesis oleh kaedah laser ablasi daripada logam masing-masing di dalam air suling. Amaun nanopartikel digunakan adalah berbeza dari 3.14×10^{-2} hingga 9.42×10^{-2} mg/l bagi nanopartikel Ag dan dari 5 hingga 3×10^{-2} mg/l bagi nanopartikel Pt untuk membentuk dua jenis polimer gel dosimeter PAGAT sebelum penyinaran dengan dos dari 6 hingga 25 Gy oleh sinar gama ^{60}Co 1.25-MeV. Saling tindakan utama sinar gama dengan bahan ialah melalui saling tindakan serakan Compton dimana penyerapan fotoelektrik tidak memberi kesan.

Pempolimeran dosimeter diukur dengan MRI dan skala kelabu filem MRI ditentukan dengan menggunakan sebuah densitometer optik. Keputusan ketumpatan optik menunjukkan bahawa amaun pempolimeran meningkat dengan peningkatan dalam dos terserap, manakala pertambahan kedalaman di dalam dosimeter yang mempunyai kesan sebaliknya. Selain itu, didapati bahawa terdapat peningkatan ketara dalam tindakbalas ketumpatan optik-dos sebanyak 27.10% untuk dosimeter diisi dengan nanopartikel Pt 1×10^{-2} mg/l dan sebanyak 11.82% untuk dosimeter dengan nanopartikel Ag 6.28×10^{-2} mg/l.

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Najmeh Deyhimihaghighi

2014

I certify that a Thesis Examination Committee has met on 26 May 2014 to conduct the final examination of Najmeh Deyhimihaghighi on her thesis entitled "Enhancement of Dose Response and Nuclear Magnetic Resonance Imaging of Pagat Polymer Gel Dosimeter by Silver and Platinum Nanoparticles" in accordance with the Universities and University Colleges Act 1971 and the Constitution of the Universiti Putra Malaysia [P.U.(A) 106] 15 March 1998. The Committee recommends that the student be awarded the Master of Science.

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LIST OF ABBREVIATIONS

AgNPs	Silver Nano Particles
AuNPs	Gold Nano Particles
a.u	Arbitrary unit
(AA)	Acrylamide
AAS	Atomic Absorption Spectroscopy
Bis	N, N′-methylene-bis-acrylamide
^{10}B	Boron-10
BNCT	Boron Neutron Capture Therapy
^{60}Co	Cobalt -60
^{137}CS	Cesium-137
CT	Computed Tomography
CTRx	Computerized Tomography scanner
^{12}C	Carbon-12
CPE	Charged Particle Equilibrium
DAA	Diacetone Acrylamide
DEF	Dose Effect Factor
FOV	Field Of View
FT raman	Fourier transform Raman spectroscopy
FID	Free-induction decay
Fe^{2+}	Ferrous ion
Fe^{3+}	Ferric ion
Gy	Gray
Hz	Hertz
HQ	Hydroquinone
^{131}I	Iodine-131
ICRU	International Commission on Radiological Units
^{192}Ir	Iridium-192
IMRT	Intensity Modulated Radiotherapy
LET	Linear Energy Transfer
LINAC	Linear Accelerator
MRI	Magnetic Resonance Imaging
^{60}Ni	Nicle-6-0
Nd: YAG	Neodymium-doped Yttrium Aluminum Garnet
NMR	Nuclear Magnetic Resonance
NPS	Nano Particles
NVF	N-vinyl formamide
PtNPs	Platinum Nano Particles
PVC	Polyvinyle Chloride
PVA	Polyvinyle Alcohol
OD	Optical Density
PDD	Percentage depth dose
PH	Power of Hydrogen
QA	Quality Assurance
RF	Radio Frequency
R_1	Relaxation Rate for T_1
R_2	Relaxation rate for T_2
SV	Sivert

SNR	**Signal to Noise Ratio**
SSD	**Source Sample Distance**
SRS/SR	**Stereotactic Radiosurgery**
SDS	**Sodium Dodecyl Sulfate**
SPR	**Surface Plasmon Resonance**
$AgNO_3$	**Silver nitrate**
SE	**Spin Echo**
THPC	**Tetrakis (Hydroxymethyl) Phosphonium Chloride**
T	**Tesla**
T1	**Longitudinal relaxation times**
T2	**Transverse relaxation times**
TEM	**Transmission Electron Microscope**
TR	**Repetition Time**
TE	**Echo Time**
TLD	**Thermoluminescence Dosimeter**
UV	**Ultra Violet**
UV-vis	**UV visible**
VP	**1-vinyle-2-pyrrolidinone**
XO	**Xylenol**
2D	**2 Dimension**
3D	**3 Dimension**

CHAPTER I

INTRODUCTION

1.1 Background of Study

1.1.1 Radiotherapy

Radiotherapy is a form of cancer treatment using high energy x-rays or gamma rays and electrons beam that are delivered to stop cancer cells from growing. The radiation beams affect the cells' ability to multiply. It can affect both cancer cells and normal tissues alike. The principal goal in radiotherapy is to deliver a determined dose to the tumor, although decrease the amount of dose that reaches normal tissues, which results in eliminating the diseased cells and improving the quality of life (Venning, 2005). There is an important stage of planning before irradiating cancerous cells. An x-ray machine and computerized tomography (CT) scanner are used to visualize the anatomical structure of the patient. These two methods give us a good overview of exact position of tumor in the body. In the radiotherapy treatment planning, parameters like tumor volume, distance of source to tumor volume, absorbed dose, and surrounding tissues are determined. With all this information, the planning computer program patterns the best position of treatment to deliver high doses to tumor and lowest amount to healthy tissue. For tumors that sit in deep, it is needed to manage several beams from different angles to obtain better result. Normally, one method to reduce the harmful effect of dose is to deliver the dose at several treatment times which is called "fractional treatment", to give time for healthy tissues to recover and destroy cancerous cells. However, a rapid advancement in the field of dynamic treatment techniques such as stereotactic radiosurgery and intensity-modulated radiotherapy (IMRT) for treatment of cancer require 3D dose distribution (Maryanski, et al., 1992). Current dosimeters like thermoluminecent dosimeters (TLD) and radiographic film are able to achieve 1-D and 2-D doses, thus limiting the spatial resolution doses in 3D (Venning, 2005). In these limitations, polymer gel is a useful tool to measure dose in 3D and plays an important role. The amount of produced polymer inside the gel, allows a dose distribution in 3D form that can be visualized by MRI scan, and be used in radiotherapy treatment planning.

1.1.2 Polymer Gel Dosimeters

By knowing the energy of ionizing radiation and composition of materials, the absorbed dose may be calculated, which is basic information of dosimeters. A device that indicates quantifiable and reproducible changes in physical or chemical properties is defined as dosimeter, which is related to dose delivered to material. The technique for measuring the radiation dose can be divided into the absolute method, which involves direct measurement of radiation dose such as calorimeters, ionizing chambers. And secondary method such as radio-chromatic dosimeter film, TLD, Fricke (ferrous sulfate) and polymer gel, which involves indirect measurements. Polymer gel along with MRI as mentioned before are the first 3D dose distribution dosimeter.

1.1.3 Nanoparticles in Treatment and Imaging

Applying high Z material as contrast agent in low energy x-ray is one of the raised concerns about risk in radiology procedure. As the bone is composed of high Z element, calcium, there is the same concern for it through radiologic process. High doses were reached to bone marrow when using kilo-voltage range x-rays. Increase in dose could be seen in situations where high Z materials like iodine are applied as contrast agent. Adams et al., (1977) showed the effect of ionization radiation combine with high Z element to chromosome aberration. It was observed that the presence of high Z material caused increase absorption of x-ray and thus increases the breakage of chromosome. The unpleasant effect in diagnosistic radiology can be used in radiotherapy desirably. It was proposed to load the tumor with high Z materials that cause an increase in the amount of dose delivered to the tumor, then this will allow maximum dose to be received by tumor and much less harm effect to healthy tissues.

Nanotechnology is a field involving chemistry, physics, biology and medicine, has predominant potential for early detection, diagnose accurately, and personalized treatment of cancer (Cai and Chen, 2007). Nanoparticle especially noble nanoparticles like silver (Ag), gold (Au), and platinum (Pt) nanoparticles are versatile agent with biomedical applications in the field of sensitive diagnostic experiment, radiotherapy enhancement, drug and gen delivery. They were proved to be non-toxic in gen and drug delivery applications. Characterization of nanoparticles and comparing them to their bulk material counterparts make them a significant choice to get a better result in different fields. In the field of radiotherapy, metal nanoparticles are studied because of their potential application in the enhancement of radiation dose regarding the treatment or improving the planning stage in determining the exact dose. Metal nanoparticles with high atomic number are noticed to be radiation dose enhancers. The metal, which is used, depends on the source. They can increase the photoelectric effect or Compton effect then increase the amount of dose that reaches the tumors. It uses in vivo in the tumors or utilize in dosimeter to make it more sensitive to dose. Radio-sensitization observed by several authors points out gold nanoparticles (AuNP) as the best candidates to be used as flagships of radionanotherapy (Hainfeld, et al., 2010; Herold M, et al., 2000; Jones, et al., 2010; Rahman, 2010; Zhang, et al., 2009). From dosimetric point of view, the use of Au nanoparticles in radiotherapy was motivated by the fact, that probability of photoelectric increases due to the presence of high Z (Au□79) material inside the polymer gels and hence increases the absorbed dose (Marques, et al., 2010). Ag and Au nanoparticles have attracted immense attention because of their potential application in different fields such as chemical and biochemical sensing, biological imaging, medical diagnostic and therapeutic. These two nanoparticles have great optical properties due to excellent Surface Plasmon Resonance (SPR) (Lee and El-Sayed, 2006). Thus, in this current research, Ag nanoparticles were embedded in PAGAT polymer gel dosimeter and irradiated with gamma ray (γ-ray) to investigate the characteristic dosimerty of Ag nanoparticles as the particle, which have the same specifications as Au. Also, Porcel et al. (2010) studied Pt nanoparticles as radiation sensitizer in radiotherapy cancer treatment which Pt nanoparticles count as high Z element (78) and it shows great enhancement of biological efficiency of radiation and increase the lethal dose to kill DNA in cancerous cell. Therefore, Pt nanoparticles were also used as another enhancer in this research with PAGAT polymer gel for dosimetry application.

1.2 Problem Statement

Gel dosimeter is a method, which can measure absorbed dose in 3D with high spatial resolution. Moreover, the initial aim of studying polymer gel is to prepare more effective device to detect the absorbed dose in 3D with high dose resolution so that it will be possible to map two doses with slight differences accurately (Lepage, et al., 2001a). As the polymer gels are tissue equivalent, it can be utilized as phantom of body to investigate the effect of metal nanoparticles as dose enhancer and increase the contrast in MRI. Au nanoparticles as dose enhancer were studied by several authors and were found to be excellent particles for reaching this aim. But growing requirement in this field leads to the investigation of other metal nanoparticles to enhance the dose and measurement of 3D dose distribution more accurately with lower price, which can be used as contrast agent in radiology, dose enhancer in radiotherapy and dosimetry of the ionizing radiation.

1.3 Significant of Study

Gel dosimeter is always considered as the best dosimeter for measuring 3D dose distribution .The first step to synthesize this kind of dosimeters easily and then use it widely is to prepare them on normal atmosphere, which is called normoxic. After that it is studied to increase the sensitivity of the polymer gel by adding some kind of metal nanoparticles like gold (Au). It can make them more accurate. As the polymer gels are tissue equivalent, they can be used as a phantom to assess the effect of dose in the presence of nanoparticles inside the body so as to increase the effect of dose in treatment in radiotherapy. The significant of this study is to establish two types of metal nanoparticles (silver and platinum) inside the polymer gel, which can increase the dose in body tissue and raise the contrast in MRI.

1.4 Objective of Study

The primary purpose of this study is to improve the sensitivity of polymer gel to enhance the contrast in MRI and measure the dose in 3D more accurately by adding silver and platinum nanoparticles. If these two particles can enhance the dose, it can be used in radiotherapy for treatment. More details of investigation are presented as below:

1. Synthesized PAGAT polymer gel using the Venning method and synthesized Pt and Ag nanoparticles by laser ablation method.

2. Investigation of 3D dose distribution of PAGAT polymer gel dosimeter using different concentrations of Ag and Pt nanoparticles as contrast agent for enhancing gray scales of MRI images of the radio-sensitive polymer gel for use in radiotherapy treatment planning.

3. Investigation of dose with depth in presence of Ag and Pt nanoparticles.

1.5 Outline of the Thesis

The structure of this thesis is divided into six chapters. Chapter I, deals with general introduction about research background, objectives of study, problem statement and significant of study. Chapter II, focuses on the history of polymer gel dosimetry and related literature in view of metal nanoparticles, which apply in radiotherapy treatment and dosimetric point of view. The general theory of interaction of ionizing radiation with matter, mechanism of polymerization of polymer gel dosimeter, properties of platinum and silver and general physics in Magnetic resonance imaging (MRI) are explained in chapter III. The methodology of study including materials, the experimental methods to prepare the samples are described in chapter IV. The major part of this thesis is in chapter V, which presented the result of experiment, analyzed and discussed. Chapter VI depicted the conclusion including a brief summary and suggested future work.

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