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Polymer gel dosimeter calibration using X-Ray CT imaging

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Citation

C'est lorsque l'on se perd que le voyage commence vraiment.

It's when you get lost that the journey really begins.

'Marine DE Nicola'

DEDICATIONS

I would like to dedicate my thesis to my parents, whose confidence in me has been unwavering, and to my second family uncle Djamal and all his family. Thank you for the moral and financial support that kept me afloat.

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LIST OF ABBREVIATIONS

MAGIC	Methacrylic and Ascorbic acids in Gelatin Initiated by Copper
CT	Computed Tomography
HU	Hounsfield Unit
IMRT	Intensity Modulated Radiotherapy
MAGAT	Methacrylic acid Gelatin and Tetrakis
PDD	Percentage Depth Dose
ROI	Region of Interest
SSD	Source to Surface Distance
TLD	Thermo luminescent Dosimeter
TPS	Treatment Planning System
MRI	Magnetic Resonance imaging
PAG	Polyacrylamide Gelatin
SSDL	Secondary Standards Dosimetry Laboratories
3D	Three dimensional
BANANA	bis, acrylamide, nitrous oxide and agarose
BANG	bis, acrylamide, nitrogen and aqueous gelatin
MAA	Methacrylic Acid
AA	Ascorbic Acid

ABSTRACT

The aim of this work is to study the development of a polymer gel dosimetry system called MAGIC gel (Methacrylic acid and ascorbic acid in copper-initiated gelatin) using X-ray CT (CT-scan) in order to be applied in dosimetry during the 3D simulation of a radiotherapy treatment and to determine the calibration curve describing the performance of this gel as a dosimeter capable of translating the three-dimensional distribution of the irradiation dose. The MAGIC gel was prepared from the formulation proposed by Fong et al. in the literature. (2001) with some modifications. The irradiations of the gel were carried out horizontally using a source of ^{60}Co. The tubes irradiated with MAGIC gel were evaluated using UV-Vis. The results showed a linear relation between the maximum absorbance and the -absorbed dose ranging from 2 to 14 Gy. After the synthesis and irradiation of the MAGIC gel, a scanner reading dedicated to the CAC-Setif was made for the determination of the calibration curve of this dosimeter in external radiotherapy. The doses used vary between 2 and 14 Gy. The CT protocol for obtaining the best image quality of the irradiated MAGIC gel was determined to evaluate the dose information. CT scanner parameters were 120 kV, 200 mA variation, slice thickness of 6 mm, 5 s exposure time, using a standard resolution and reconstruction algorithm for 25 x 25 cm^2. A linear relationship was found approximately between CT numbers the doses between 2 and 15 Gy. The PDD obtained based on measurements with CT was compared to the calculated PDD referenced to the laboratory SSDL based on measurements made with an ionization chamber.

Key-words: MAGIC gel, X-ray CT (CT- scan), calibration, 3Ddose distribution

Résumé

L'objectif de ce travail est de réaliser l'étude du développement d'un système de dosimétrie sur le gel polymère nommé MAGIC (Methacrylic Acid in gelatin initiated by copper) en utilisant la tomodensitométrie par rayons X (CT-scan) afin de pouvoir l'appliquer en dosimétrie lors de la simulation en 3D d'un traitement de radiothérapie, et de déterminer la courbe d'étalonnage décrivant les performances de ce gel comme dosimètre capable de traduire la distribution tridimensionnelle de la dose d'irradiation. Le gel MAGIC a été préparé à partir de la formulation proposée par Fong et al. (2001) avec quelques modifications. Les irradiations de ce gel ont été effectuées horizontalement à l'aide d'une source de ^{60}Co. Les tubes irradiés ont été évalués à l'aide d'un spectrophotomètre UV-Vis. Les résultats ont montré une relation linéaire entre l'absorbance maximale et la dose absorbée allant de 2 à 14 Gy. Après la synthèse et l'irradiation du gel MAGIC, une lecture par scanner dédié à la radiothérapie au niveau du CAC-Sétif a été réalisée pour la détermination de la courbe d'étalonnage de ce dosimètre en radiothérapie externe. Les doses utilisées variaient entre 2 et 14 Gy. Le protocole de CT pour obtenir la meilleure qualité d'image du gel MAGIC irradié a été déterminé pour évaluer les informations de dose. Les paramètres de CT scanner étaient de 120 kV, 200 mA, des coupes d'épaisseur de 6 mm, un temps d'exposition de 5 s, utilisation d'un algorithme standard de reconstruction et de résolution pour 25 x 25 cm^2. Une relation linéaire a été trouvée approximativement entre la variation du nombre CT et les doses comprises entre 2 et 14 Gy. Le PDD obtenu basé sur des mesures effectuées avec CT a été comparé au PDD calculé référencé au laboratoire SSDL basé sur des mesures effectuées avec une chambre d'ionisation.

Mots clé : gel MAGIC, X-ray CT (CT- scan), calibration, distribution de dose 3D.

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Introduction

During the last decades, radiation therapy has gone through a significant development process. Today, there are treatment techniques with an improved capability for tumor control while sparing normal tissue surrounding the tumor, using conformal radiotherapy. Dynamic treatment techniques, stereotactic multiple beams and Intensity Modulated RadioTherapy (IMRT) can be used. With the new techniques patient complications are minimized, with a greater probability of cure. These achievements are mainly based on technological improvements that make advanced and complex irradiation methods possible. Optimizations of the absorbed dose distribution can be performed using the recent developments, for example utilizing dynamic multileaf collimators and other IMRT techniques.

There is a great need for three-dimensional absorbed dose measurements, especially in large patient-mimicking volumes, to verify the complete treatment chain, including CT scanning, image transfer, TPS (Treatment Planning System) dose calculations and beam delivery.

Imaging of irradiated polymer gel dosimeters (PGD) using x-ray computed tomography (XCT) is a newly developing method for extracting information about the absorbed dose in a radiosensitive polymer gel. Using suitable compounds of gel and appropriate parameters affecting quality of the final image, this method could become more advantageous alternative of the most frequently used read out method for the purposes of polymer gel dosimetry till this time, quantitative magnetic resonance imaging (MRI). The PGD principle relies on radiation induced polymerization of dissolved monomers in gelatin. As a consequence, density (and some other physical parameters) change and that causes increase of linear attenuation coefficient. The change can be detected with X-ray CT.

CT images of irradiated MAGIC (Methacrylic and Ascorbic acid in Gelatin Initiated by Copper) gel, quantified a CT dose response for MAGIC gel and thereby established the potential of CT as an alternative to MRI and optical CT for gel read-out. The advantages of CT readout are practical ones: accessibility of CT to clinical radiation therapy environments, ease and speed of image acquisition, and a CT dose response that is stable and relatively insensitive to environmental factors.

This work was therefore initiated in order to determine calibration curves using UV and CT scan images of the absorbed dose and to investigate the feasibility of using x-ray computed tomography (CT) for performing MAGIC dosimetry.

To better understand the advantages of MAGIC gel, we start by reminding in Chapter I, backgrounds and introducing radiation dosimetry. The chapter II is dedicated to present polymer gel dosimeters in general focusing on MAGIC gel which makes point of this study.

In the experimental part, represented by chapters III and IV, we present methods and materials used in this work, then results with discussions.

Chapter I

Introduction to radiation dosimetry

Chapter I: Introduction to radiation dosimetry

I.1. Introduction to radiation and its types

Radiation is naturally present in our environment and exists since the birth of this planet. It comes from outer space (cosmic), the ground (terrestrial), and even from within our own bodies. It is present in the air we breathe, the food we eat, the water we drink, and in the construction materials used to build our homes. Radiation is energy in the form of electromagnetic waves or stream of particles. There are two forms of radiation –ionizing and non-ionizing. **[M.A.El-Ahdal.2014]**

I.1.1. Non-ionizing radiation

Non-ionizing radiation has less energy than ionizing radiation; it does not possess enough energy to produce ions (to remove electrons from atom). Examples of nonionizing radiation are visible light, infrared, radio waves, microwaves, and sunlight. These are defined as Extremely Low-frequency (ELF) waves. **[M.A.El-Ahdal.2014]**

I.1.2. Ionizing radiation

Ionizing radiation is capable of knocking electrons out of their orbits around atoms, upsetting the electron/proton balance and giving the atom a positive charge. Electrically charged molecules and atoms are called ions. Ionizing radiation includes the radiation that comes from both natural and man-made radioactive materials. **[M.A.El-Ahdal.2014]**

I.2. Main types of interactions of ionizing radiation (γ radiation) with matter

In radiotherapy dose may be deposited by electrons, γ-rays, X-rays, protons, neutrons and a number of other heavy charged particles. These particles are classed as ionizing radiation, which is any radiation that has the ability to ionize atoms of the matter through which it passes. The minimum energy required to eject an electron from an atom is approximately 4 eV. Therefore, any particle that possesses kinetic energy greater than 4 eV is considered to be an "ionizing radiation". Similarly, photons whose energies are greater than 4 eV can be considered to be "ionizing radiation". Dose is deposited through the interactions of ionizing radiation with matter,

in radiological physics there are three main types of interactions of γ-ray and X-ray photons with matter that have to be considered. These are the Compton Effect, the Photoelectric effect and pair production. These three interactions result in the transfer and deposition of energy to the medium. The probability of each of these interactions occurring is dependent on the energy of the beam and Z_{eff}, the electron-fraction-weighted-average atomic number of the atoms in the absorbing material The Z_{eff} of water is 7.42 whereas the Z_{eff} of the gel dosimeter is approximately 7.31. **[Mohammad Abdalla.2008]**

I.2.a. Photoelectric Effect

The photoelectric effect describes the interaction that occurs when an incident photon relinquishes all of its energy to an atomic electron. The electron atomic subsequently ejected from the atom, as shown in Figure1. The energy of the electron is that of the incident photon minus the binding energy of the electron. **[Boris .2012], [Mohammad Abdalla.2008]**

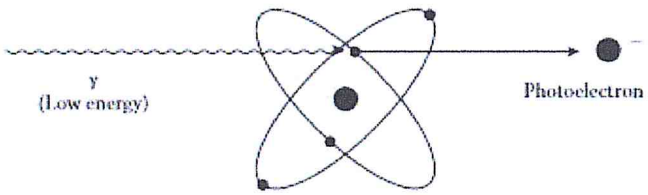

Figure I.1. The photoelectric effect **[Boris .2012]**

I.2.b. Compton scattering

The Compton Effect describes the interaction which an incident photon gives up part of its energy to a weakly bound electron, causing the electron to be ejected from the atom. The incident photon is then scattered, as shown in Figure2, with energy that is equal to that of the original quantum less the recoil energy of the electron. **[Boris .2012], [Mohammad Abdalla.2008]**

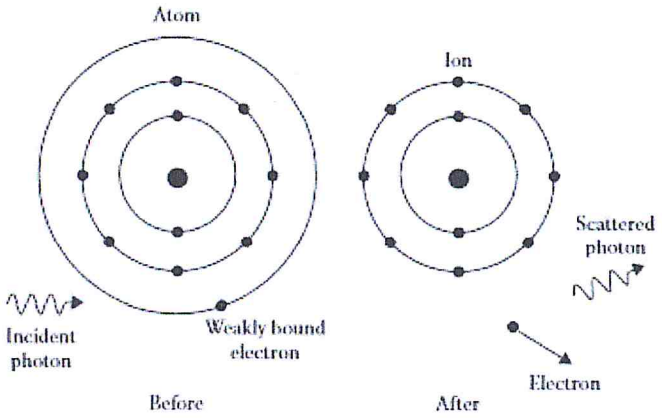

Figure I.2. Compton scattering by a weakly bound electron. **[Boris .2012]**

I.2.c. Electron–positron pair production

For water equivalent material and for photons whose energy is greater than 1.02 MeV, there is a probability that the photon will be completely absorbed through the mechanism of pair production. This phenomenon occurs when a photon passes near the nucleus of an atom and is subject to strong nuclear fields. The photon may suddenly disappear and emerge as a positron-electron pair, as shown in Figure3. Charge is conserved because the lepton pair has opposite charges. Any energy in excess of 1.02 MeV is shared between the pair. **[Boris .2012], [Mohammad Abdalla.2008]**

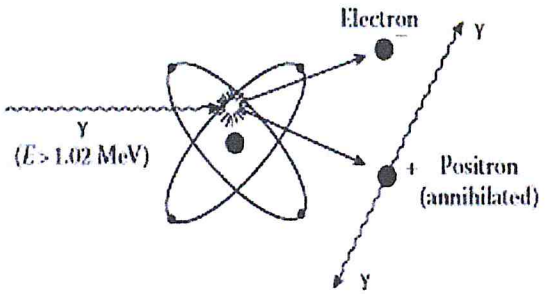

Figure I.3. Schematic of pair production.

An incident photon, passing through the Coulomb force field of an atomic nucleus, vanishes giving rise to a positron- electron pair. **[Boris .2012]**

I.3. Interaction of ionizing radiation with organic polymers

The interaction of ionizing radiation with matter results in the formation of very reactive species (free neutral radicals, cationic and anionic ions, excited molecules). These can significantly modify the molecular structure of the irradiated material. In particular, irradiation of organic polymers induces molecular chain branching, crosslinking and molecular degradation or scissioning. Chain branching and crosslinking increase the molecular weight of the polymer. Crosslinking forms an insoluble three-dimensional polymer network; while degradation or scissioning causes a reduction of the initial molecular weight. During irradiation, all these phenomena coexist and their prevalence depends on several factors, such as the initial molecular structure and morphology of the polymer and the irradiation environment. If the polymer is irradiated in presence of air, the molecular modifications are different with respect to the effects of irradiation in vacuum or in presence of an inert gas. During irradiation in air, the free radicals, produced by interaction of ionizing radiation and polymers, can also react with oxygen, giving rise to oxidative degradation, which competes with other reactions that occur in absence of oxygen. All these molecular modifications can modify the properties of the material. Studies have been devoted to understanding the mechanisms of the modification of the molecular structures and of the properties of polymers resulting from exposure to ionizing radiation. **[Giuseppe Spadaroliv .90128]**

I.4. Radiation dosimetry

Radiation processing dosimetry is used to quantify the energy deposited in a material or absorbed by a human from radiation sources. Different dosimetry systems are used for different purposes in industry and research irradiation facilities, which have different requirements for dose determinations. Radiation safety standards and issues involving the radiation protection of humans against radiation exposure have their own dosimetry metrology. Radiation dosimetry is a branch of physical science exploring different methods for the quantitative determination of energy, which is deposited in a given material by ionizing radiation. Dosimetry deals with

determinations and calculations of quantities (dose) that describe the energy absorbed in a material and to some extent its rate of deposition (dose rate).

Dosimetry determinations that are performed by exposing a dosimeter to a radiation source help in evaluating the radiation-induced effects, physical, chemical, and/or biological, on an irradiated material. To assure that the desired radiation effects are achieved and that the irradiation process is performed safely, validation and process control procedures are implemented. Process controls rely on the establishment of a relationship between the source parameters and the absorbed dose in an irradiated object. Applications of ionizing radiation in materials processing inferred from determinations made with a suitable dosimetry system having some level of accuracy and precision. Absorbed dose and dose distribution is inferred from determinations made with a suitable dosimetry system having some level of accuracy and precision. **[Diana.Adliene-51368]**

I.5. Absorbed dose measurement tools

It is well known that modern radiological quality assurance (QA) procedures require reliable and accurate dosimetry techniques. Dosimetry plays an essential role in most clinical applications involving ionizing radiation providing reliable treatment verification. There are so many dosimeters used to measure dose distributions such as ionization chambers, thermoluminescent dosimeters (TLDs), diodes, radiochromic films and dosimetry gels. **[M.Valente.2018]**

I.5.1. Traditional dosimetry

The traditional dosimeters used to measure dose distributions include radioactive ionization chambers, Silicium diode, radiography films, radio chromic films and thermo luminescent dosimeter (TLD cards).The TLD cards (also called mini ionization chambers) have some drawbacks in measuring very high doses with high dose gradients, due to their finite sizes which permit measurement of the dose only at a single point. Film batches can also offer 3D dose measurements by positioning film in multiple planes but accurate positioning of films in several layers can be a difficult and time-consuming process. Therefore, conventional dosimeters are suitable for clinical brachytherapy. **[Mohammad Abdalla.2008]**

I.5.1.a. Ionization Chamber

There are different types of ionization chambers such as the large air-walled chamber (used only in standard laboratories), the cylindrical chamber (used for the quality assurance of photon beams in radiotherapy) or the flat chambers used for the dosimetry of electrons (energy below 10 MeV) as well as low energy photons (acceleration voltage less than 100 kV).

Its principle relies on the creation of electron / ion pairs in the sensitive volume. The high voltage (~ 300 V) applied through the electrodes of the chamber makes it possible to collect the created carriers. The resulting (electron) charge current can subsequently be converted to an absorbed dose value after applying a set of correction factors. One of the advantages of the ionization chamber is that the energy required to generate a pair of ions in the air is independent of the energy of the incident radiation.

Liquid-filled chambers take advantage of the higher density of liquids to increase the energy deposited in the chamber. Nevertheless, these chambers exhibit poor stability due to the progressive pollution of the liquid by conductive ionic species. **[D.Benoit.2008]**

I.5.1.b Thermo luminescent Dosimeters

Thermo luminescent dosimeters, TLDs, are mainly composed of lithium fluoride in two major forms: powder and shaped solids. At the atomic level, the lithium fluoride atoms form a crystalline pattern which is called a crystal lattice. The electrons are shared in this lattice pattern, and when exposed to irradiation the electrons move from their atomic location to locations called traps, where they remain trapped until the dosimeter is heated. When the electrons are released through the heating process, they travel to the conduction band and then fall to their lattice ground states, emitting light. This light is amplified by a photomultiplier tube in units of electric charge and is proportional to the amount of radiation that was absorbed, expressed in cGy. TLDs may be reused but at the cost of losing the information; consequently, they can only be read out once. Benefits of TLDs are that they are nearly independent of dose-rate and have a wide dynamic range. Also TLD is nearly tissue equivalent. For patient monitoring in Radiotherapy, the typical size of a TLD shaped into a chip is 3 mm x 3 mm x 1/2 mm. **[C.Baldock.2010]**

I.5.1.c. Radiographic film

Film, also known as photographic emulsions, consists of microscopic grains of silver bromide dispersed in a gelatin layer on either or both sides of a supporting film. The reaction that takes place when a beam is incident on film is that Ag+ ions are converted to Ag atoms. A latent image is produced and now the film is ready to be developed. During the developing process all remaining ions are reduced to silver atoms and the bromine is removed. The grains that have the latent image will have the ions reduced more rapidly. What is left is opaque silver that can be measured optically. Optical density (OD) is the parameter obtained from the transmission of light through the film and which is related to dose. Some advantages of film are that it has good spatial resolution, is dose-rate independent, and is easily obtained commercially.

Some major disadvantages of film are that it has a strong dependence on the incident energy due the high Z of the material, the film batch and the processor. The high Z of film increases the probability of photoelectric absorption occurring at low energies. Furthermore, air gaps can arise when placing film in a phantom and cause inaccuracies in measuring dose. **[C.Baldock.2010]**

I.5.1.d. Silicium Diode

Diodes, also known as semiconductor dosimeters, are solid state devices that measure dose and dose rate. The most common diodes used are silicon diodes with p-i-n junctions. The p region is where holes are located, the i region is called the intrinsic region, and the n region is where excess electrons are located. The diode is used either in a reversed-bias or an unbiased state. The incident radiation creates electron-hole pairs. The electrons are then elevated to the conduction band while the holes are left in the valence band. The resulting current which is related to the dose rate is measured by an electrometer. The signal that is produced when the radiation is applied is approximately 10 times greater than that produced in an ion chamber. This is due to the small amount of energy required to produce an ion pair (W/e), the high density of the material and high atomic number (Z).Unfortunately, like film, diodes have energy dependence and therefore must be calibrated against an ion chamber. The advantages of diodes are that they have good spatial resolution and like TLDs can be placed on the skin of patients to measure localized surface doses. But unlike TLDs, diodes are able to read and display the dose in real time. The diodes are very widely used in medical dosimetry because of their robustness and their ease of use (almost immediate reading). The diodes are more sensitive and compact than the

ionization chambers. The most likely disadvantage diodes lie in the loss of sensitivity with the cumulative dose. **[C.Baldock.2010], [Benoit.2008]**

I.5.1.e. Radio chromic film

The radio chromic films consist of one or more active layers of radio-sensitive monomer (diacetylene) protected by a polyester envelope. Irradiation of these films results in a polymerization reaction which leads to a color change of the film. The intensity of the color obtained is proportional to the dose absorbed by the film. The dose is evaluated by measuring the light transmission of the film by means of a photo densitometer. These films do not require any development. The set of films offers a wide range of use ranging from 0.5 to 2500 Gy. They are equivalent tissue and are widely used in medical dosimetry. **[D.Benoit.2008]**

I.5.2. Three dimensional gel dosimetry

Over many years individuals have endeavored to measure absorbed radiation dose distributions using gels. As long ago as 1950, the radiation-induced colour change in dyes was used to investigate radiation doses in gels. Further, in 1957 depth doses of photons and electrons in agar gels were investigated using spectrophotometry. Gel dosimetry today however, is founded mainly on the work of Gore et al who in 1984 demonstrated that changes due to ionizing radiation in Fricke dosimetry solutions, developed in the 1920's, could be measured using nuclear magnetic resonance (NMR). **[Clive Baldock.2006]**

I.5.2.a. Fricke Gels

The first gel dosimeters proposed for radiation therapy were also developed by Gore et al in 1984 who added a ferrous sulphate solution into a chemical gel matrix to contain the 3D dose information which then could be measured using nuclear magnetic relaxometry (NMR) (Gore et al 1984). These gel dosimeters were based on the Fricke solution in which ferrous (Fe2+) ions are converted to ferric (Fe^{3+}) ions upon irradiation (Fricke and Morse 1927).

Both ions have a different NMR spin-lattice relaxation rate (R1 = 1/T1) which allows the use of quantitative MRI sequences to image the amount of radiation induced conversion of ferrous to ferric ions. These Fricke gel dosimeters were fairly quickly shown to suffer from spatial instabilities originating from the diffusion of ferric and ferrous ions within the gel matrix following the irradiation (Olsson et al 1992). The diffusion rate of the reporter molecules inside

the dosimeter was found to be affected by the type of gelling agent (e.g. agarose (Appleby et al 1987), gelatin (Olsson et al 1990), sephadex (Hiraoka et al 1992), polyvinylalcohol (Chu et al 2000)), the concentration of the gelling agent and other properties of the dosimeter (Baldock et al 2001a). Additionally, metal chelators such as xylenol orange (XO) succeeded in improving the spatial stability of the recorded dose distribution while also allowing for a visible inspection due to a colour change of the gel upon irradiation (Gupta and Gomathy 1974, Gupta et al 1982, Gupta et al 1985 and Appleby et al 1991). The chemical fine-tuning did not entirely eliminate the diffusion of ferric ions which still resulted in a limited timespan during which the quantitative dose distribution could be imaged with MRI. In the gel dosimetry community, the search for more spatially stable dosimeters was therefore preceded. **[J.Vandecasteele.2013]**

I.5.2.b. Polymer Gels

Polymer gel systems for the use of radiation dosimetry were first proposed as early as 1954, where Alexander et al discussed the effects of ionizing radiation on poly methyl methacrylate. Following this, Hoecker et al in 1958 investigated the dosimetry of radiation-induced polymerization in liquids, and in 1961 Boni used polyacrylamide as a gamma dosimeter. Much later in 1991, Audet et al reported changes in NMR transverse relaxation measurements of irradiated polyethelene oxide. In 1992, Kennan et al reported on NMR longitudinal relaxation studies performed on an irradiated aqueous solution of N,N'-methylene-bis-acrylamide and agarose, which showed that the relaxation rates increased with absorbed dose. In 1992 a new gel dosimetry formulation was proposed, which was based on the polymerization of acrylamide and N, N'-methylene-bis-acrylamide (bis) monomers infused in an aqueous agarose matrix. This system was given the acronym BANANA due to the use of the chemical components (bis, acrylamide, nitrous oxide and agarose). This type of gel dosimeter did not have the associated diffusion problem of Fricke gels and was shown to have a relatively stable post-irradiation dose distribution. The polymerization reaction occurred by cross-linking of the monomers induced by the free radical products of water radiolysis. In 1994 the BANANA formulation was refined by replacing agarose with gelatin and given the acronym BANG (bis, acrylamide, nitrogen and aqueous gelatin), the first of a series of new polymer gel formulations. In 1994 this formulation was patented and became commercially available through MGS Research Inc. as BANG. Subsequently, due to the naming of the commercial product, PAG (Polyacrylamide Gelatin) became the polymer gel dosimeter acronym of choice for most authors. Numerous authors

subsequently published results of work investigating different compositions and formulations of polymer gel dosimeters some of which were summarized by Lepage et al. Although polymer-type dosimeters did not have the diffusion limitations of Fricke-type gel dosimeters, there was another significant limitation in their use. Due to the nature of their free radical chemistry, polymer gel dosimeters were susceptible to atmospheric oxygen inhibition of the polymerization processes. As a result, these gel dosimeters had to be manufactured in an oxygen-free environment, such as in a glove box pumped with nitrogen gas. Along with the use of potentially toxic chemicals, this was a significant limitation in the introduction of gel dosimetry into the clinic. **[Clive Baldock.2006]**

I.5.2.b.1. Normoxic Polymer Gel Dosimeters

A significant development in the field of gel dosimetry occurred when results of using an alternative polymer gel dosimeter formulation were published by Fong et al in 2001. This new type of polymer gel dosimeter, known as MAGIC gel, bound atmospheric oxygen in a metallo-organic complex thus removing the problem of oxygen inhibition and enabling polymer gels to be manufactured on the bench-top in the laboratory. This created what was to be known as a normoxic gel dosimeter, compared with the previous PAG formulation which subsequently became known as a hypoxic gel dosimeter. The MAGIC polymer gel formulation consisted of methacrylic acid, ascorbic acid, gelatin and copper. The principle behind the MAGIC gel is in the ascorbic acid oxygen scavenger. Ascorbic acid binds free oxygen contained within the aqueous gelatin matrix into metallo-organic complexes and this process is initiated by copper sulphate Cu (II). Numerous authors subsequently published results of work investigating different compositions and formulations of normoxic polymer gel dosimeters and were recently summarized by Senden. Recent work has included the development of less toxic polymer gels. **[Clive Baldock.2006]**

The advantages to be expected with gel dosimetry compared with conventional dosimeters may be summarized in the following properties:

- Independence on radiation direction, radiation quality and dose rate for conventional clinical beams
- absorbed dose integration in the dosimeter (of utmost importance for dynamic treatment and multiple beams)

- evaluation of a complete volume
- potential for true three-dimensional dosimetry utilizing a high spatial resolution
- possibility to measure in a phantom that is equivalent to anatomical soft tissue

The gel dosimetry method may be developed further as the gel can be moulded in any desired shape and inhomogeneities can be placed within the phantom. **[Sven A.J. Back 8-100]**

I.5.3. Applications of Polymer Gel Dosimetry

Polymer gel dosimeters possess several characteristics that give them advantages over more conventional dosimetry systems. Polymer gels have the unique ability to conform to the shape of their container. This allows their use in anthropomorphic phantoms that mimic human anatomy. Additionally, gel dosimeters are capable of rendering complex dose distributions in three dimensions. While other dosimeters are capable of taking measurements at a point (ion chambers, TLDs) or in a plane (film), only three-dimensional dosimeters are capable of giving a complete picture of the dose being delivered. Three dimensional dosimeters like polymer gels are necessary to rigorously confirm the accuracy of three-dimensional treatment plans. Polymer gel dosimeters have been used to measure the dose distributions produced in a range of treatment modalities, including intensity modulated radiation therapy (IMRT), HDR brachytherapy, and proton radiotherapy. **[Vredevoogd, Kevin M.2012]**

Diagnostic radiology and radiotherapy are the most used techniques to detect and treat several pathologies like tumor and cancer diseases. According to available evidence at least 52% of cancer patients should receive radiotherapy during their treatment. The process of radiotherapy is complex and involves understanding of the principles of medical physics, radiobiology, radiation safety, dosimetry, radiotherapy planning and simulation. One of the key aspects of radiotherapy is to determine the real dose an organ or tissue is exposed. If the irradiation conditions are well defined and known in relation to the anatomy of the patient, dosimetry calculation is straightforward by means of treatment planning algorithms or Monte Carlo simulations. However, if those conditions are not fulfilled, it is also possible to measure the organ dose directly with the use of suitable phantoms and dosimeters. Among the different types of dosimeters, polymeric ones are of great interest because of their capability to mimic

soft-tissue while maintaining a specific shape presenting high dose sensitivity and ability to retain 3D spatial dose distribution for long periods of time after being exposed. **[F. Mattea.2013]**

Chapter II

MAGIC polymer gel

Chapter II: MAGIC polymer gel

II.1. Ionizing radiation-induced polymerization

Since the last quarter of the twentieth century, a growing interest in polymerization induced by ionizing radiation has been observed. Throughout this period, researchers have exposed polymeric materials to ionizing radiation and reported the occurring of crosslinking and other useful effects, e.g., miscibility of blend polymer as well as many different applications in the polymer radiation chemistry field. Ionizing radiation is a promising technology for preparation of cost-effective, efficient, safe, and high-quality polymers. The radiation techniques applied for polymerization processes could be carried out using many different types of radiation sources. Ionizing radiations (gamma rays, X-rays, accelerated electrons, and ions beam) initiate polymerization reactions by the formation of very reactive intermediates (energetic radicals).

Energetic radicals resulting from ionizing radiation hit molecules causing electrons ejection from a particular orbit, then the atom becomes ionized. As soon as their orbits begin to be ready to overlapping forming new bonds, these electrons may be much more detached by other molecules. This phenomenon occurs when the transferred energy is enough for ionization, electrons are gaining energy and transfer to an upper energy level, resulting in excitation. The ionizing potential for most molecules ranges from 7 to 15 eV, while the energies of ionizing irradiators (gamma rays, X-rays, accelerated electrons, and ions beam) range from 1 to 100 MeV, so ionization is the predominating process. Energetic radicals with ionized electron excite another molecule upon calling energy transfer. The ionized species associated with the energetic radicals created by irradiation would induce various reactions. This power in a polymerization reaction is an eco-friendly process. Polymers are covalently cross-linked by ionizing radiation at room temperature.

II.2. MAGIC polymer gel composition

The **MAGIC** gel is an aqueous solution of gelatin mixed with methacrylic acid (MAA), copper sulphates (Cu II) and ascorbic acid (AA). Gel dosimeters are mostly composed of water, gelatin and small amounts of other compounds that provide tissue equivalency, upon irradiation, it polymerizes in an aqueous gelatinous matrix as a function of the absorbed radiation dose. The gel dosimeters do not present angular dependence and have high resolution. In 2001, Fong et al. proposed the use of antioxidants to reduce the oxygen effect. Ascorbic acid (ASC) is used to

bind oxygen contained within the gel matrix in a process initiated by copper sulfate, possibiliting the gel preparation at normal atmosphere conditions.

The gelatin in polymer gel dosimeters is used to fix spatially the radiation induced polymer chains. It is a biodegradable protein with an average molecular weight. It is derived from acidic decomposed collagen from porcine skin. The charge on a gelatin molecule and its associated isoelectric point are primarily determined by the carboxyl, amino, and guanidino groups on the side chains. The generally accepted model for the gelatin molecule is a triple helix. The triple helix is stabilized by hydrogen bonds between the N-H groups of the gelatin backbone of one chain and C=O groups of a neighbouring chain.

Water acts as an intermediary in inter-chain and intra-chain hydrogen bonding (Wustneck et al 1988, Fruhner and Kretzschmar 1989).

Table II.1. Chemical composition of MAGIC polymer gel

Compound	Concentration
Gelatin	8%
Methacrylic acid	9%
Ascorbic acid	0.0352%
Copper sulphates II	0.002%
Hydroquinone	0.2%
Deionized water	82.8%

II.3. Polymer gel dosimetry principle

The PGD principle relies on radiation induced polymerization of dissolved monomers in gelatin. As a consequence, density (and some other physical parameters) change and that causes increase of linear attenuation coefficient. The change can be detected with different techniques X-ray CT, MRI or OCT. Polymer gel dosimetry involves three major steps in order to obtain a dose distribution: (a) after fabrication, the gel is poured into an anthropomorphically shaped cast and

into calibration vials; (b) the phantom is irradiated with a specific dose distribution and calibration samples are irradiated to known doses;(c)the irradiated gel dosimeter phantoms are scanned with an appropriate and optimized scanning technique (x-ray computerized tomography (x-ray CT); (d) finally the data are used to produce an image of the irradiated dose distribution.

The principles of the polymerization are expected to be similar kind to the dosimeter gel, though there is gelatin present. The gel dosimeters are gelatin-matrix-based dosimetry system that avoids the diffusion problem. In these three-dimensional dosimetry systems the gelatin matrix contains monomers that polymerize by free-radical induced chain reactions to form spatially fixed cross-linked networks. Polymer gel, which contains monomers dissolved in a gel matrix based on radiation induced polymerization and crosslinking of acrylic monomers. The solution is polymerized due to free radicals produced by radiolysis. This alters the gel dosimeter physicochemical properties in proportion to the absorbed dose causing opacity. There are different polymer gel compositions of which many were susceptible to atmospheric oxygen. Initially, the polymer gel dosimeters should be manufactured in an oxygen-free environment, because oxygen inhibits the radiation-induced polymerization.

II.4. Polymerization reaction mechanisms

Polymers are large chain molecules having a high molecular weight in the range of 103 to 107. These are made up of a single unit or a molecule, which is repeated several times within the chained structure. A monomer is the single unit or the molecule which is repeated in the polymer chain. It is the basic unit which makes up the polymer.

It is believed that the complex of Cu (II), ascorbic acid and oxygen with radiolysis of water serves as a free radical source for the initiation of the polymerization of methacrylic acid.

$O_2 + AA + Cu_2+ <=>$ (complex), irradiation => R

These free radicals (R) can react with monomers (M) and form monomer radicals (M*), (initiation), it is also likely that formation of gelatin radicals takes place. This step is followed by propagation or termination, where the propagation involves creation of polymer radicals, and the termination occur when two polymer radicals neutralize each other or a polymer radical reacts with Cu (II) and the chain propagation stops.

Initiation: R + M => M 1*

Propagation: M (n-1) + M => M n*

Termination: M n* + M n* => polymer M n* + Cu (II) => polymer + Cu (I)

The formation of a polymer by addition polymerization is an example of a chain reaction. Once a chain reaction gets started, it is able to keep itself going. **[Mohammad Abdalla.2008]**

II.5. Dosimetric characteristics (the choice of using MAGIC gel)

Based on the recommendation from previous works, the characteristics of a good polymer gel as dosimetry are: It must be tissue or water equivalent, as the dosimeter itself must not perturb the dose distribution, and it must have a linear dose response over a clinically useful range. The dosimeter must be stable for a sufficiently long period to enable irradiation and dose analysis. It must remain stable during shipment, unaffected by a variety of environmental conditions throughout the analysis period. It must be no toxicity in polymer gel MAGIC gel has less toxicity compared to other gels main composed of Acrylamide.

For a polymer gel dosimeter to be of use in radiation dosimetry, it should display water-equivalent radiological properties. The radiological properties of the MAGIC (Methacrylic and Ascorbic acid in Gelatin Initiated by Copper) normoxic polymer gels were investigated. The mass density (ρ) was determined based on Archimedes' principle. The weight fraction of elemental composition and the effective atomic number (Z_{eff}) were calculated. The electron density was also measured with 90° scattering angle at room temperature. The linear attenuation coefficient (μ) of unirradiated gel, irradiated gel, and water were determined using Am-241 based on narrow beam geometry. Monte Carlo simulation was used to calculate the depth doses response of MAGIC gel and water for 6MV photon beam. The weight fractions of elements composition of MAGIC gel were close to that for water. The mass density was found to be 1027 $\pm$ 2 kg m^{-3}, which is also very close to mass density of muscle tissue (1030 kg m^{-3}) and 2.7% higher than that of water. The electron density (ρ_e) and atomic number (Z_{eff}) were found to be 3.43×10^{29} $e.m^{-3}$ and 7, respectively. The electron density measured was 2.6% greater than that for water. The atomic number was very close to that for water. The prepared MAGIC gel was found to be water equivalent based on the study of elementary composition, mass density, electron density and atomic number. The linear attenuation coefficient of unirradiated gel was very close to that of water. The μ of irradiated gel was found to be linear with dose 2-40 Gy. The depth dose response for MAGIC gel from a 6 MV photon beam had a percentage dose difference

to water of less than 1%. Therefore it satisfies the criteria to be a good dosimeter for radiotherapy. So the choice was based on its density equivalent to tissue, less toxicity and the most important its economic interest. **[M Aljamal.2013]**

II.6. Error sources

Various sources can lead to a loss in both accuracy and precision at different levels of the measurement process (see table Annex 1). To evaluate the performance of a 3D gel dosimeter, the accuracy and precision can be evaluated at different stages of the gel dosimetry experiment.

The prescribed dose distribution as obtained from the radiation treatment planning (D_{presc}) is presented to the radiation unit (^{60}CO…) and the radiation is delivered to the gel dosimeter resulting in a delivered dose distribution in the gel dosimeter (D_{deliv}). In the gel dosimeter, a dose-dependent polymerization reaction takes place which is results in a spatial distribution of polymer (Polym). The gel dosimeter is read out by use of a non-invasive imaging technique (in our case X-ray CT). The result is a parametric map (Ω) that reflects the absorbed dose distribution. The parametric map displaying a physical quantity (CT numbers) is converted to a dose map by use of a calibration curve ($\Omega(D)$) that is obtained by passing through all of the above-mentioned stages but starting with a known dose or dose distribution (Dpresc = Dcal). When a gel dosimeter is subjected many times to a radiation treatment, there will be stochastic deviations in both radiation output (ε (D)) and spatial errors (ε (x,y,z)) that may result in both spatial and dosimetric deviations in the resulting dose distribution. The magnitude of the positioning deviations will depend on the robustness of the positioning operation. Sophisticated methods to accurately positioning the gel dosimeters apply stereotactic frames and fiducial markers.

Note that any positioning error during the irradiation of calibration phantoms may also give rise to dosimetric errors in the final dose distribution. It has been shown that in some gel dosimeters, the dose response depends on the temperature of the gel dosimeter during irradiation and thus temperature variations may introduce dose variations (ε (T)). The response of the gel dosimeter is susceptible to variations in different parameters during fabrication of the gel. Differences in the temperature treatment during (ε (T_{fabr})) and after (ε ($T_{storage}$)) fabrication of the gel dosimeter as well as variations in the concentration of the chemical components (ε (chemical concentrations)) may result in differences in the dose response.

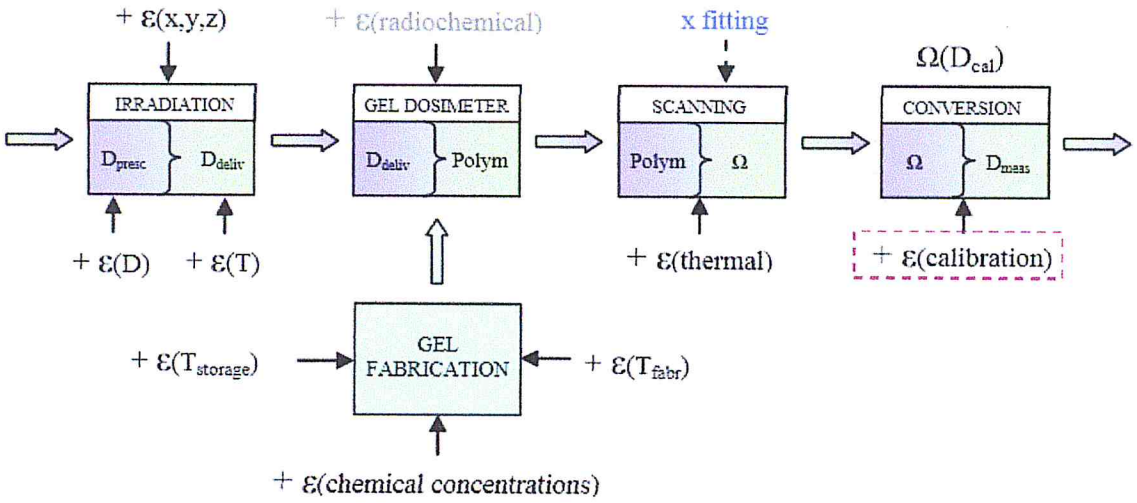

Figure II.1. Dosimetric source errors

Gel dosimetry is performed in different stages. At each stage, errors can be added, leading to a decrease in the overall precision and accuracy. **[Yves De Deene.2006]**

Most of these deviations are compensated by using calibration phantoms that are constructed from the same batch of gel. However, it may be difficult to keep the temperature history after fabrication (ε ($T_{storage}$)) similar for both the gel dosimeter phantom and the calibration vials because of the differences in phantom size. Upon irradiation, a complex set of radiation-induced chemical reactions take place. On a molecular level, these reactions are probabilistic in nature (ε (radiochemical)). In most gel dosimetry applications, the voxel-size is several orders of magnitude bigger than the molecular size. As a result, this intrinsic radiochemical noise contribution can be easily neglected. After irradiation, the gel dosimeter is scanned. During scanning, detector (thermal) noise (ε (thermal)) will be added to the measurements. The processing (fitting) of acquired data, may have a big influence on the amplification of the noise figure. Imaging artefacts may result in systematic errors. Imaging artefacts can result in dosimetric errors and in geometrical distortions. **[Yves De Deene.2006]**

Chapter III

Materials & Methods

Chapter III: Materials and methods

Introduction

This work were divided in two main steps part one at CRNA in which we have prepared, irradiate and characterize the gel tubes and the second part at Setif center (anti-cancer center) for the reading of MAGIC gel response using CT images .

II.1. Experimental setup

II.1.1. MAGIC-polymer gel synthesis

The MAGIC gel was prepared based on the formulation proposed in the literature by Fong et al. (2001) with some modifications. In the CRNA laboratory 8 g of Gelatin was mixed with 70 ml of deionized water and then heated to 50◦C using a magnetic stirrer for 30 minutes until a clear solution was obtained (white solution). At this point, 0.2 g hydroquinone solution (dissolved in 4.8 ml of water and stir slightly) was added and the solution was allowed to cool. After further cooling to 37-38◦C, solutions (0.0352 g) of the ascorbic acid (dissolved in 5 ml) and (0.002 g) of copper sulphates (dissolved in 3 ml) of water were added. The whole solution was allowed to mix for a further 1-2 min before, the 9 ml of methacrylic acid was added, and continuously stirred until the monomer was completely dissolved. The gel was dispensed into nine prepared glass vials in order to be irradiated by RX to cause the polymerization. **[N.B.15]**

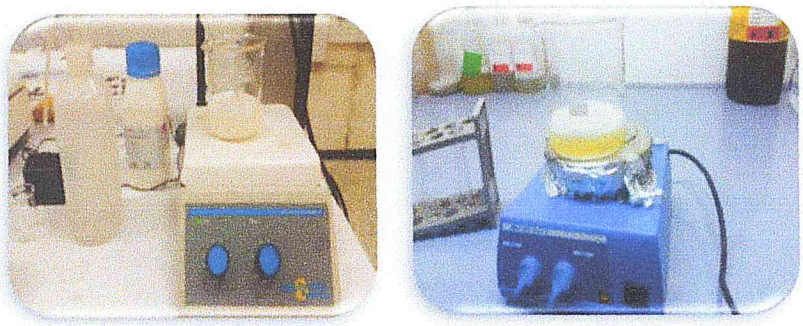

Figure III.1. Manufacturing MAGIC gel at CRNA laboratory

II.1.2. Gel irradiation with Co^{60} (CRNA)

As soon as gelation was completed the gel vials Irradiation were performed horizontally using a source of cobalt ^{60} (CRNA) 24 hours after preparation with a field size of 8 cm x 8 cm at the isocentre and at 60 cm source detector distance (SDD). The dose rate was 24, 18 cGy min^{-1}. The samples were irradiated with absolute doses of2, and 3, 4,5,6,7.5,10,15 Gy and one vial was left un-irradiated for background measurement by parallel opposed beams so that the gels received a uniform dose at 5 cm depth. Solid water phantom slabs were placed above and below the Perspex cuvette holder and the samples were placed at the midregion of the phantom as shown in Figure III.2

After being irradiated these monomers react with each other to form polymer chains.

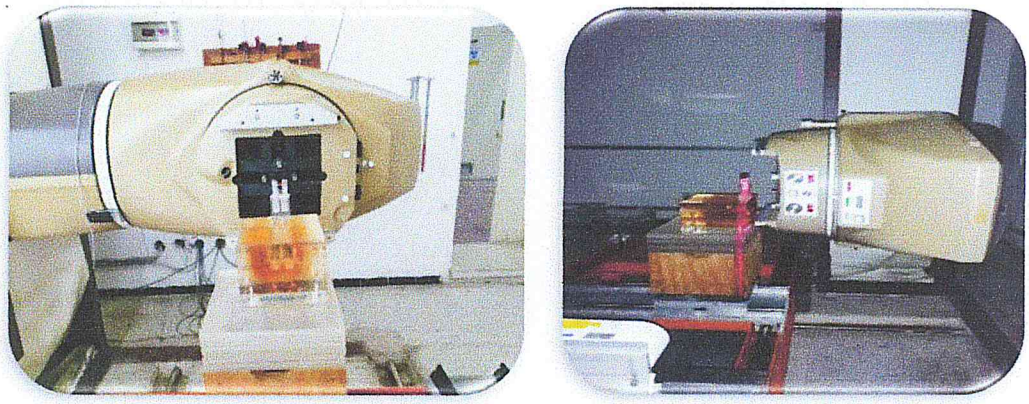

Figure III.2. Gel vials irradiation

II.1.3. Gel assessment

UV-visible gel assessment (quantification)

Spectrophotometry is a fast, cheap, sensitive and convenient measurement technique and the characterization typically has as a goal to improve the performance of the polymer gel, our gel has been quantified by UV-VIS spectrophotometry for a dosimetric application. The UV-Visible absorption spectra have been recorded using instrument Model U-3900H spectrophotometer (see figure 3) operator: spectro-photocata at CRNA. The unirradiated gel (0 Gy) was used for

baseline correction for spectrophotometer analysis. The absorption wavelength was selected in the range from 200 to 600 nm with a step of 2 nm.

Figure III.3. Instrument Model U-3900H spectrophotometer

Table III.1. UV-VIS parameters

UV-VIS parameters	
Scan Speed	600 nm/min
Slit Width	2 nm
Sensitivity	1

II.1.3.1. CT-scan gel assessment

As the polymerization process initiated by irradiation may induce a change in mass density of the polymer gel which cause change in contrast in the CT image, this imaging technique has been selected for our study. The CT imaging was performed using CT (Siemens®) scanning brand of scanner dedicated to radiotherapy service of Setif Anti Cancer Center. The parameters used for the scanning were 120kV voltage, 300mA current and 6 mm slice thickness.

Following the CT scan acquisition, the images were sent to the treatment planning system. The Reconstructed cuts were transferred to the exploitation from which the UH number (Hounsfield Unit) by targeting regions of equal size interest on each gel image. PS: The readings were taken 24 hours after irradiation.

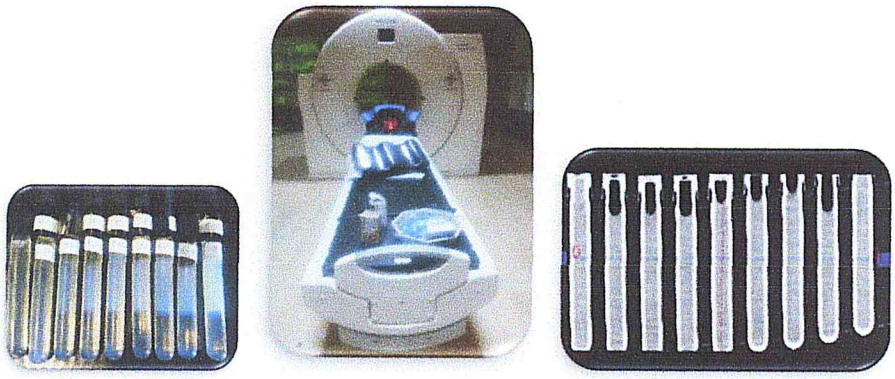

Figure III.4. Gel vials imaging with CT (Siemens®) scanner

Using this scanning protocol, the irradiated MAGIC gels of different doses were imaged to establish relation between average CT numbers and doses.

Table III.2. CT-scan parameters

CT parameters	
Tube voltage	120 kV
Tube current	200 mA
Slice thickness	0.6 cm
Acquisition time	5 sec
Matrix	256x256 pixels
FOV	25 x 25 cm2

II.1.4. CT-scan dose response of MAGIC gel

In order to verify the usefulness of the CT based gel dosimetry to measure dose, the Profile Depth Dose (PDD) and isodose curve (beam profile) of 10x10 cm2 field size photon beam from ^{60}Co source were measured. The measured PDD values of cobalt 60 photon energy were compared with that measured in water referenced to SSDL lab using a radiotherapy treatment planning computer system (TPS) which are based on measurements ionization chamber

Chapter IV

Results & Discussion

Chapter IV: Results & discussions

IV.1. Gel irradiation result

After being irradiated we have noticed that for higher doses the radiation causes opacity (condensation) of the gel, as can be seen in Figure 1

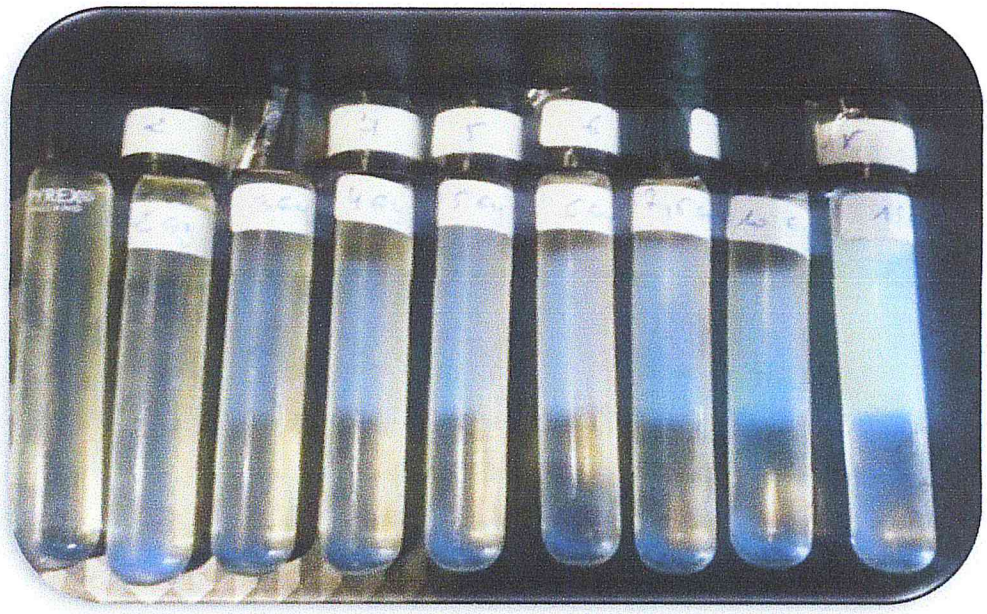

Figure IV1. Aspect of vials containing MAGIC gel which have been irradiated with ^{60}Co source to different absorbed doses using a dose rate of 24, 18 cGy min^{-1}

IV.2. UV-Visible results

IV.2.1. Absorption spectra

After examinating the irradiated vials by UV-Visible, we have traced the absorbance spectra of the gel as function of the irradiation doses and we've got the spectra as it is shown in figure 2

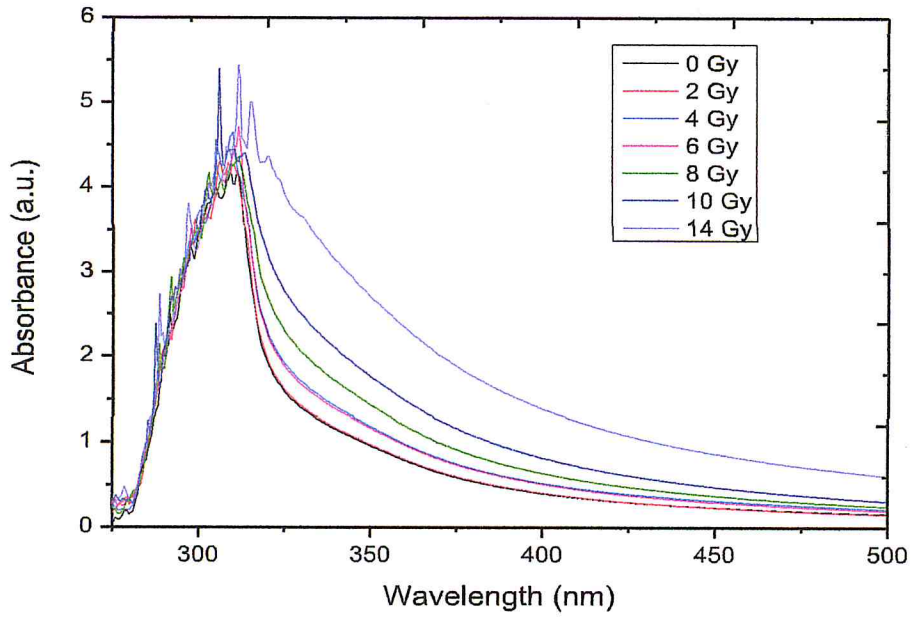

Figure IV.2. Absorbance spectra of the MAGIC gel

The spectra showa peak of absorption around 310-340 nm Due to change in gel color caused by irradiation as shown in figure 1. It is shown that the intensity of the irradiated samples increase with the increase of the dose. In fact, the observed increase in absorbance intensity with the dose stresses the enhance in the polymerization rate. This can be the results of the linear augmentation of the number of monomer unit (m) in the polymer chain confirmed by the formation of polymerized particle.

IV.2.2. Calibration UV-Visible curve

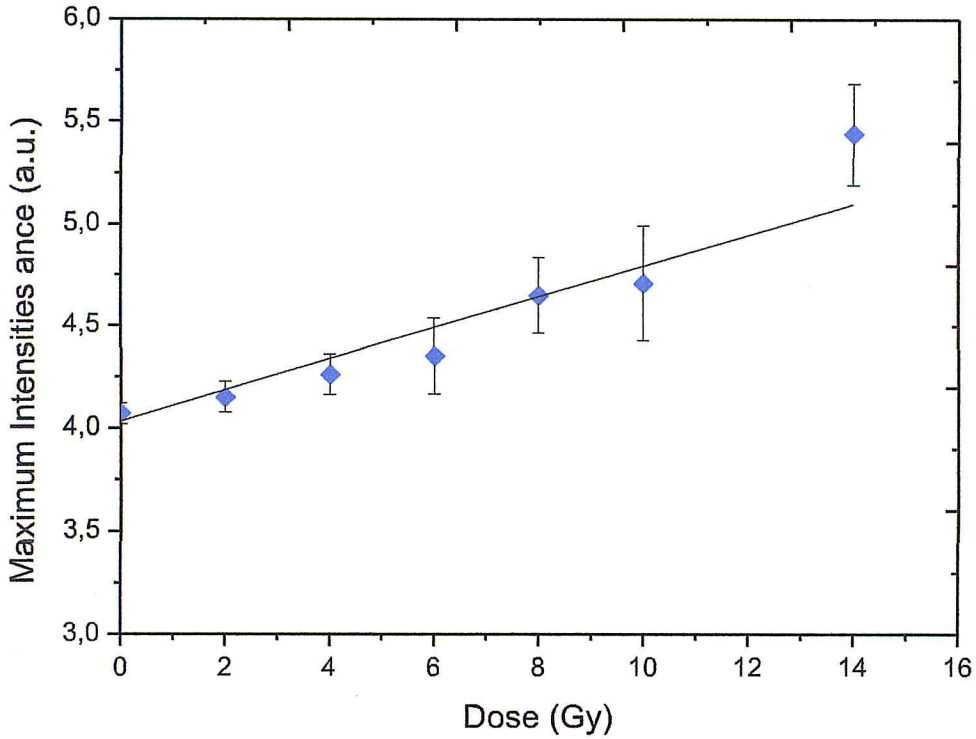

Figure IV. 3. Dose response of **MAGIC** gel at maximum intensity

The curve shows the evolution of the maximum intensity uv-visible absorption as function of absorbed dose in MAGIC gel samples. It is observed that the absorbance of the polymer gel dosimeters increase linearly with the increase of the dose. The representative curve shows a good linearity between maximum intensities and the gamma radiation doses (y=0.089X+4.180 with a determination factor of R^2 = 0.951, this linearity confirms the capability of the MAGIC gel polymer to be used as radiation dosimeter.

IV.3. CT-scan results

The irradiated MAGIC gel vialswere imaged, CT images were acquired for these tubes to determine the CT number (opacity, blackening) and dose response. Figure shows an example of the CT reading of an irradiated MAGIC vial.

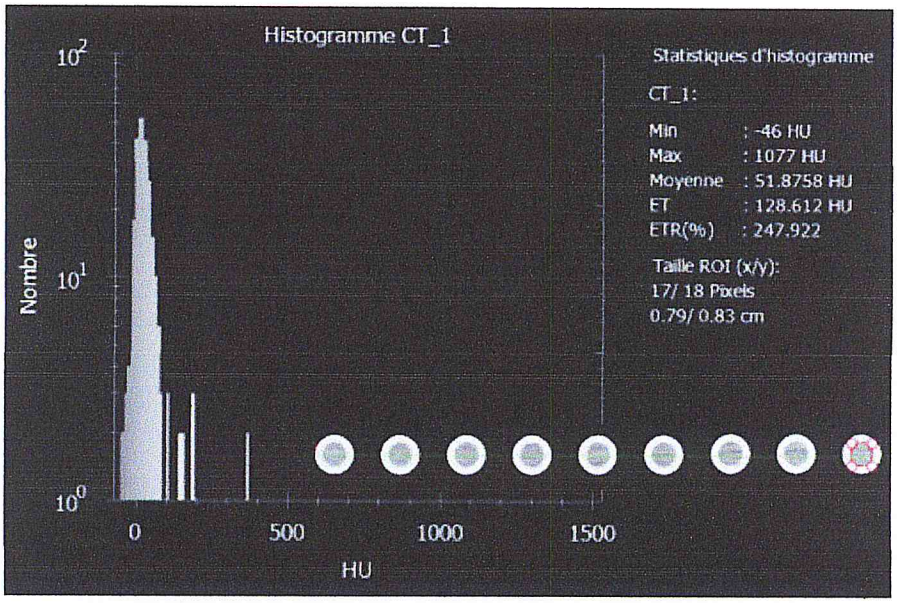

Figure IV.4. CT reading after irradiation

The contrast of the observed image in the X-ray CT image of the irradiated polymer Magic gel is the result of the change in the mass density, which occurred in the gel during polymerization. This change can be related to unit of Hounsfield (UH) using the following relation:

$$N_{CT} = \frac{\mu_{gel} - \mu_{water}}{\mu_{water}} \times 100 \qquad (01)$$

So, the evaluation of the gel response as function of the dose are presented in terms of UH for the calibration of our system.

- The dose response of Magic gel by CT-Scan

Figure IV.4 shows experimental CT slices of the scanned pack containing gel dosimeter samples. **[Clive Baldock1.2010]**

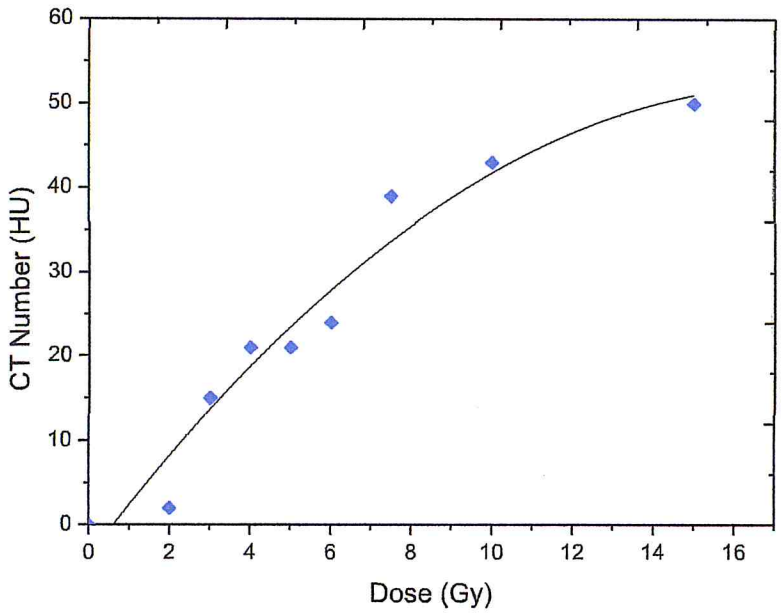

Figure IV.5. ΔN_{CT}- dose response curve for MAGIC gel

Figure 5 represents the calibration curve. This curve has been established in term of the number of CT variation (ΔN_{CT}) as a function of the absorbed dose. It is notable the increase of the Ct variation withdose in the investigated dose range. The dose response could be fitted by the following fitting equations:

(y= 4.00727x +0.513) with R2= 0.88

(y= -0.18x2 + 6.35x –3.746) with R2= 0.938

We note that evolution of CT variation as function of the absorbed dose is better for quadratic fit than linearly one.

IV.4. CT-scan dose response of MAGIC gel

The profile depth dose represents the percentage of variation as function of the depth (the penetration of the irradiation beam). The obtained PDD based on measurements with CT were compared to thecalculated PDD referenced to SSDL lab which is based on measurements using an ionization chamber.

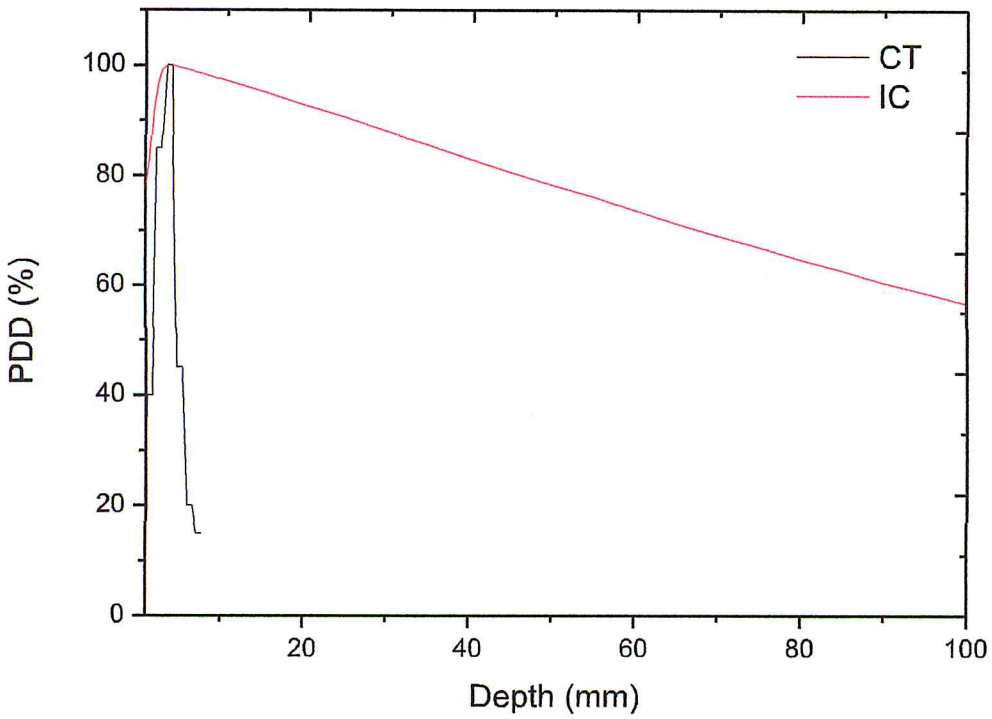

Figure IV.6. PDD for ^{60}Co

The dose distributions obtained with 3D dosimetry have been compared to those measured with the treatment planning system, which are based on measurements using an ionization chamber. The comparison has been calculated with dose in an axial slice through the isocentre. As can be seen in fig. 11, for a ^{60}Co beam, , the depth doses measured in using the gel showed only for

maximum position a good agreement with the ionization chamber masurements. As known, the radiation absorptivity, which is the absorption per unit of distance per unit dose, depends on the dosimeter material and calibration samples. Errors that compromise the accuracy may occur at different stages of the dosimetry procedure (as shown in chapter II).

We only have the maximum value comparing it with ionization chamber PDD which is at 0.5 cm unlike other values,samples suffer a self-degradationand this could be a limitation for the use of PGD. They quickly decreased and didn't follow the PDD. We can justify that huge decrease by temperature effect and storage problem. Especially that the scan and the transport of the gel from CRNA lab to the radiotherapy service (setif) has took much time therefore, the gel has lost its charactrestics becoming in kind of a liquid state. However, the maximum that corresponds to the max still also a good result .

So we can say that the temperature effect is one of the disadvantages that difficulty working with this gel, it is necessary to keep it in 4° temperature.

Conclusion

Polymer gel dosimetry remains one of the most promising tools for measuring inherently complex 3D radiotherapy dose distributions. The whole process, including preparation, characterization, calibration and comparison of the gel has been presented.

The good linearity relationship observed by UV-Vis as function of irradiation dose encourages us for other applications in radiation therapy field.The good fitting of CT response as function of absorbed dose by quadratic polynomial equation instead of linear equation with a good agreement with watanabee assumption. **[S.Brahimi-Moussa.2014, Yoichi Watanabe.2017]**

Finally, the results indicate that MAGIC gel dosimeter appears to be a promising dosimeter which makes it a suitable dosimeter in a wider range of absorbed dose measurements in dosimetry and radiotherapy. However, thefeasibility of the use of CT as a reading mean remains fragile as a dosimetric tool, due to many influential factors, particularly the temperature effect and its transport from the lab where the gel was prepared to the radiation therapy service where the imaging analysis and irradiation were realised.

Despite all constraints and obstacles that we had seen, principally the none disponibility of reading system in the same place where we have to prepare and irradiate, subject stills very interesting and we must make attention in next studies to temperature effect and storage issue. For each preparation, it is necessary to realize a calibration of gel set in order to be applied in 3D dosimetry in radiotherapy treatment.

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