

Preliminary analysis of N-vinylpyrrolidone based polymer gel dosimeter

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Abstract

This paper aims to evaluate the dosimetric characteristics of modified VIPARnd for radiotherapy dosimetry using optical investigations. The absorbance spectrum of the irradiated gel dosimeter was evaluated optically with spectrophotometer techniques and with a CMOS imaging system. The useful dose range for the peak value and the relative area under the absorbance curve is 3-20 Gy. The dose-response curve for CMOS readout has an interval of linearity from 3-20 Gy. The modified VIPARnd developed has a good dose range and good temporal stability in the spectrophotometric analysis of the intervals studied. The CMOS readout is transportable, cheaper, easier to use and an excellent alternative for dosimetry.

Keywords: *polymer gel dosimetry, optical analysis, N-vinylpyrrolidone, radiotherapy.*

1. Introduction

Recent technological advances in radiotherapy require quality assurance (QA) that needs accurate and precise dose verification. Dose measurement usually falls into two categories: absolute and relative. The reference dosimetry aims to produce accurate and consistent values relative to the primary standards, this allows the comparison of results between centers. However, to ensure safe operation of the equipment in order to verify dose calculations and verify the dose administered in vivo, measurements are required in a range of conditions covering all aspects of clinical use, not only the conditions of reference. A possibility for measuring dose distributions is gel dosimetry^[1,2].

The gel dosimeters are gelatine-matrix-based dosimetry system that avoids the diffusion problem. In these three-dimensional dosimetry systems the gelatine matrix contains monomers that polymerize by free-radical induced chain reactions to form spatially fixed cross-linked networks^[1]. 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^[1,3]. The gel dosimeters do not present angular dependence and have high resolution^[2]. Currently, there are two types of gel dosimetry systems: Fricke and polymer gel dosimeters^[1].

The Fricke dosimeter consists of ferrous sulfate in an aqueous gel solution. The irradiation induce oxidation and to conversion of ferrous ions to ferric ions modified NMR

relaxation rates^[1,4,5]. Fricke gel exhibits high ion diffusion that causes low spatial resolution. Currently, recent formulations of Fricke gel dosimeters based on PVA-GTA (Poly-vinil alcohol-glutaraldehyde) were developed^[6]. These materials are not toxic and have a lower diffusion coefficient than the standards Fricke gels. The increase in GTA concentration is a viable method to reduce the diffusion of ions into the gel matrix^[7]. The other type is polymer gel, which contains monomers dissolved in a gel matrix based on radiation induced polymerization and crosslinking of acrylic monomers^[2,8]. 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^[1,2].

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^[9,10]. The polymer gel dosimetry was studied for different types of irradiation, such as gamma rays from cobalt sources used in external radiotherapy, high-energy x-rays and high-energy electron beams produced by clinical linear accelerators and brachytherapy sources^[11]. Significant energy dependence was not found in studies of high-energy photon and high-energy electron beams^[2,10,11].

Baras et al. reported an adequate spatial resolution for dosimetry of 192Ir HDR brachytherapy source^[12]. Authors continued to investigate polymer gel dosimetry for treatments with high dose gradient, as with Intensity-Modulated Radiation Therapy (IMRT)^[13,14], Intensity-Modulated Arc Therapy (IMAT)^[15,16] and stereotactic radiosurgery (SRS)^[17,18]. Polymer gel dosimeters have also been applied to proton^[19,20] and neutron beams^[21]. To evaluate the dose distribution in polymer gel dosimeters, many imaging modalities can be used. The opacity caused by radiation provides optical contrast in polymer gels^[22,23,24]. Some researchers considered optical investigation to analyze different solution compositions and response to ionizing radiation^[25,26,27]. The use of monomer N-vinylpyrrolidone (NVP) in gel dosimetry has been developed by Pappas et al.^[25] and was named VIPA. The VIPAR polymer gel consists of NVP, N,N'-methylenebisacrylamide (BIS), gelatin and argon was used to remove oxygen. A new gel composition (VIPARnd) based on Pappas formulation described by Kozickic et al., 2007 has the NVP concentration doubled and copper sulphate and ascorbic acid added^[28-30]. The concentration of monomers in gel solutions alters the dosimetric characteristics, like dose range, sensitivity and stability^[31,32].

The purpose of this study is to evaluate basic dosimetric properties of this modified VIPARnd using infrared, UV-Vis spectrophotometer and a CMOS camera. The dose-response sensitivity, repeatability, reproducibility and post-irradiation temporal stability were investigated for a 6 MV photon beam used in radiotherapy.

2. Materials and Methods

2.1 Mechanism

Radiolysis of water under high energy radiation has been extensively studied. Water is the compound at the highest concentration within the dosimeter solution and it is the molecule that to interact most likely with radiation^[25]. Radiation ($h\nu$) causes ionization in water molecules, a phenomenon called radiolysis^[28]. The processes of the radiolysis of pure water at where The initial breakdown in

the water molecule occurs is presents according to schematic shown in Figure 1^[30].

The molecular product hydrogen is chemically inert and readily escapes, whereas the other molecular product hydrogen peroxide (H_2O_2), is retained in water and reacts with the reducing species ($e^{\cdot aq^-}$ and $H\cdot$) to produce $\cdot OH$ and corresponding species. In pure water, after a time in a given radiation field, the concentration of hydrogen peroxide becomes steady. With g-ray radiation the concentration is low and there is very little decomposition^[31].

The positive ion formed from the reaction (H_2O^+) can form the hydroxyl radical ($OH\cdot$) when dissociating into the form $H\cdot$ and $\cdot OH$. Hydrogen and hydroxyl radicals can also be formed through the excitation of the water molecule. As a result of the radiolysis reactions we have a pair of H^+ and OH^- ions and a pair of free radicals $H\cdot$ and $\cdot OH$ ^[31].

In the reactions shown in 1, 2 and 3 (Figure 2) the breaks of the carbon double bond in BIS are caused by interaction with the hydrogen ($H\cdot$) and hydroxyl ($\cdot OH$) and electron-hydrated ($e^{\cdot aq^-}$)^[33].

In the NVP reactions with the free radicals produced by the radiolysis of the water can form the radicals shown in Figure 3^[32].

The VIPARnd gel dosimeter is a hydrogel gelatin in which th10e monomers N-vinylpyrrolidone (NPV) and N, N'-methylenebisacrylamide (BIS) are dissolved. When the gel is irradiated the water molecules are ionized. From the possible reactions between NVP and BIS molecules as shown in Figure 4, several subsequent reactions can

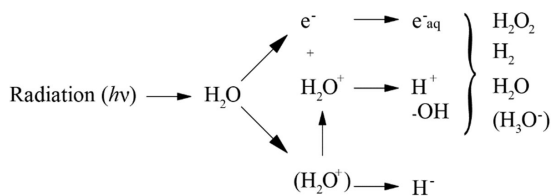


Figure 1. Process of radiolysis of water^[30-32].

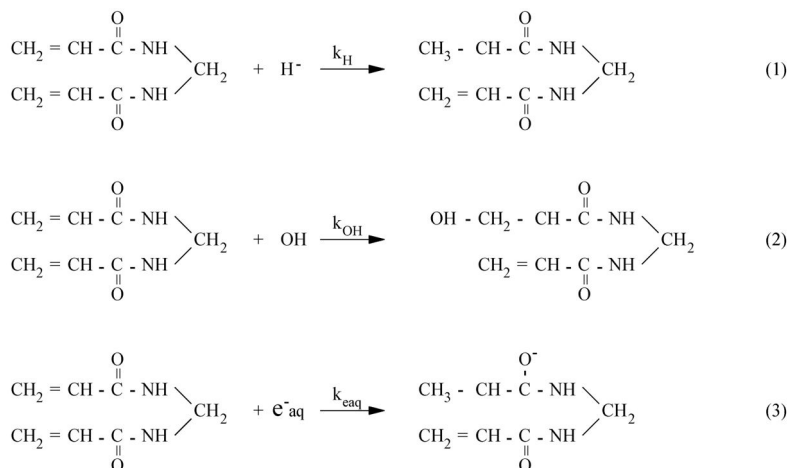


Figure 2. Breaking possibilities of BIS molecules^[33].

occur by forming a series of chromonomer radicals in the solution. The gel dosimeter obtained in this work follows the mechanistic proposal presented in Figure 4.

2.2 Gel preparation

The modified VIPARnd was prepared using NVP (purity grade $\geq 99\%$, Sigma Aldrich), N,N'methylenebisacrylamide (BIS) (Sigma Aldrich), gelatin (Type A/300 bloom, Sigma Aldrich), $\text{CuSO}_4 \cdot 5\text{H}_2\text{O}$ (purity grade $\geq 99\%$) and ASC and ultra pure deionized water (Table 1). The procedure used to manufacture the solution was the same as described by Kozickic et al., 2007. BIS was dissolved in water heated below 50°C , after gelatin was added. The solution temperature was cooled to 33°C , the NVP, ASC and $\text{CuSO}_4 \cdot 5\text{H}_2\text{O}$ were added. The solution was stirred continuously during the entire mixing procedure.

The VIPARnd formula described by Kozicki et al.^[28] exhibit high opacity with the absorbed dose. In the studied polymer gel the concentration of NVP and BIS was reduced aiming to optical analysis.

The cuvettes used as gel containers were closed with Parafilm foil and stored in a refrigerator ($\sim 6^\circ\text{C}$) to solidify. The cuvettes were made of glass with 10mm path-length. Resulting gels were clear and transparent. The major difficulty is not having air bubbles at the top of the gel container. The influence of bubbles is shown in Figure 5. This gel cuvette was placed upside down before the gel solidified, placing the air bubble on the closed end. After that, the

cuvette was irradiated to show the air influence. Oxygen presence inhibits the radiation-induced polymerization.

2.2 Characterization

The gels were irradiated 24 hours after preparation with absolute doses of 3, 5, 10, 15, 20 and 50 Gy using a Varian 21 iX linear accelerator with 6MV photons at 5.0 cm of depth in a RW3 solid water phantom. The samples were removed from the refrigerator before irradiation to stabilize at room temperature ($\sim 22^\circ\text{C}$). The irradiation was done with a field size of $10 \times 10\text{ cm}^2$, SSD of 100 cm and a dose rate of 6 Gy/min. The dose was calculated at the center of the cuvette in order to have a homogenous dose distribution.

Absorption spectra were measured for a wavelength range 280-480 nm using a Varian Cary 5000 UV-Vis spectrophotometer with temperature-controlled cuvettes, maintained at 22°C . The baseline is the empty cuvette. The absorbance measurements were carried out with 2 mm intervals between the highest dose to the lowest dose and lastly with the reference cuvette (0 Gy). For the analysis of temporal stability, the absorbance of doses 0, 5, 10 and 15 Gy was measured at three different post-irradiation times, 24 h, 192 h and 360 h.

A cheaper and simpler analysis method was also developed. The gel cuvettes were analyzed with an imaging system developed at 24h post-irradiation. This apparatus consisted of a medical light box, a CMOS camera and a MatLab routine. The cuvettes were placed on a medical

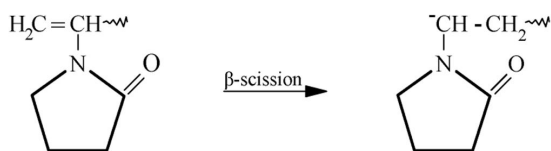


Figure 3. Breaking N-vinylpyrrolidone (NVP) molecule without oxygen fixation^[32].

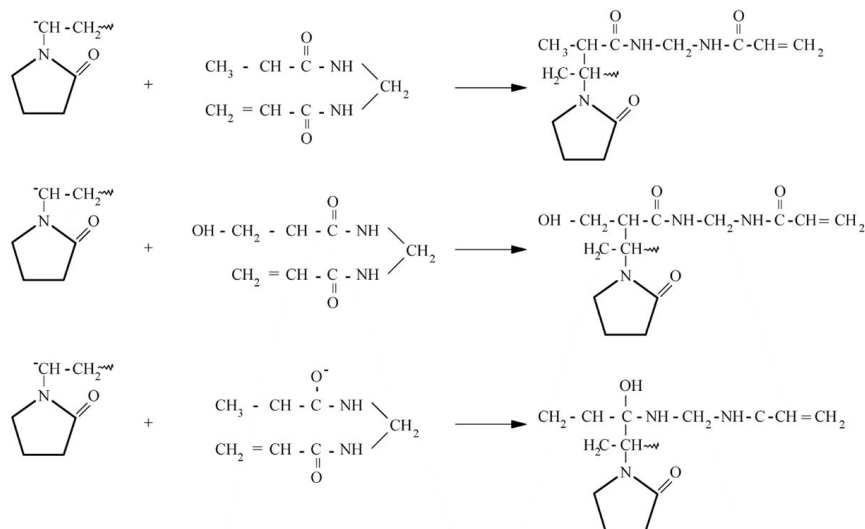


Figure 4. Proposal of possible reactions between molecules of N-vinylpyrrolidone (NVP) and N,N'methylenebisacrylamide (BIS)^[30].

Table 1. Gel composition.

Component	Concentration
Gelatin (300 Bloom)	3.5% w/v
N,N'methylenebisacrylamide (BIS)	2% w/v
N-vinylpyrrolidone (NVP)	4% w/v
Ascorbic acid (ASC)	397 mM
Copper sulfate pentahydrate ($\text{CuSO}_4 \cdot 5\text{H}_2\text{O}$)	32 mM
Pure Water	90.5% w/v

light box and photographed, without flash and no filters on automatic mode. The CMOS camera was a Nikon Coolpix P510. The file image format is JPG and this file was read in a MatLab routine that converted it into only one matrix (gray scale image). For determining the gray scale, the routine analyzes a matrix of 9×9 at the cuvette center using mean pixel intensity.

3. Results and Discussions

After being irradiated these chromonomers react with each other to form polymer chains. The radiation causes opacity in the dose absorbed in the gel, as can be seen in Figure 6a. After irradiation the dosimeters were put back



Figure 5. Air bubble in an irradiated modified VIPARnd gel cuvette.

in the refrigerator for storage. The gel opacity reduces the pixel intensity as seen in Figure 6b.

High energy radiation produces ionization and excitation in polymer molecules. These energy-rich species undergo, dissociation, abstraction and addition reactions in a sequence, leading to chemical stability. As a result, a polymeric material may undergo cross linking reaction, leading to increase in the molecular weight or undergo chain scission reaction, leading to decrease in the molecular weight. Generally, both processes occur simultaneously but in given radiation conditions, one of them is the predominant process^[24].

Due to the high fraction of cross-linking agents the final structure of the polymer is a three-dimensional network. The degree of polymerization is directly proportional to the absorbed dose. And because of the gelatinous matrix the polymer aggregates cannot diffuse into the gel structure preserving the spatial information of the dose absorption^[30].

The visible and ultraviolet absorption spectra of the gel with different doses are shown in Figure 7.

Data shows that the modified VIPARnd had a maximum absorbance at 300-320 nm depending on the absorbed dose. The relative absorbance of peak values was obtained by subtracting the maximum absorbance of the reference cuvette with 0 Gy. This data has sigmoidal compartment (Figure 8). The linear regression of data are also reported in Figure. 8, showing good linearity of response in the dose range of 3-20 Gy, with $R^2 = 0.9780$.

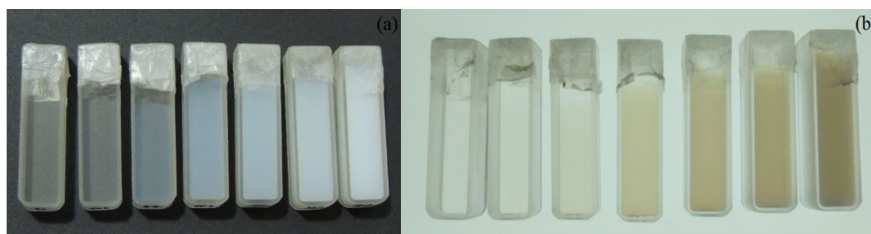


Figure 6. (a) Represents cuvettes of irradiated modified VIPARnd. The dose absorbed from left to right is 0, 3, 5, 10, 15, 20 and 50 Gy and (b) Image acquired by the imaging system developed. The dose absorbed from left to right is 0, 3, 5, 10, 15, 20 and 50 Gy.

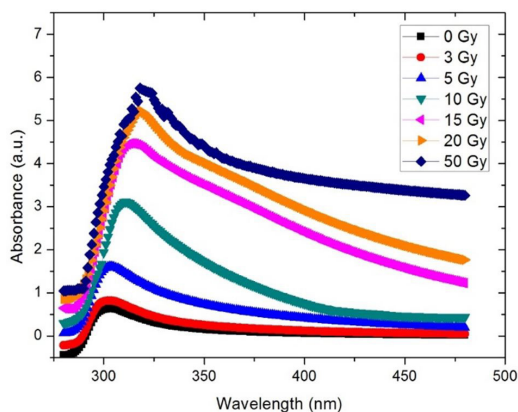


Figure 7. Visible and Ultraviolet absorption spectra for modified VIPARnd at 24h after irradiation measured in Gray (Gy).

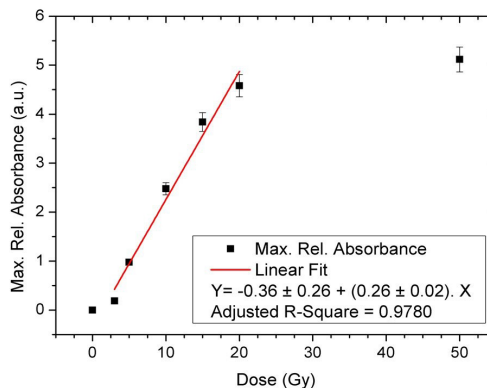


Figure 8. The relative maximum UV-VIS absorbance as a function of absorbed dose. Error bars indicate the standard deviation of the relative maximum absorbance values measured in Gray (Gy).

The absorbance values for UV-VIS spectra in Figure 4 increase for any wavelength with increasing dose, as is characteristic of opacity gel^[26,34]. Therefore, the areas under the spectra were investigated by curve integrated as shown in Figure 9. The area values were obtained by subtracting the area of the reference cuvette with 0 Gy. The same sigmoidal compartment was observed, and the linear regression of data showing good linearity of response in the dose range of 3-20 Gy, with $R^2 = 0.9802$.

The picture obtained by the imaging system developed was converted into gray scale and value of pixel was registered. The inverse of gray values (1/pixel value) was calculated and normalized by subtracting the value of the reference cuvette with 0 Gy. In Figure 10, the relative inverse of gray values (1/pixel value) obtained through a MatLab routine is reported versus dose. The plotted data show similar compartment of spectroscopy data analysis, and the linear regression of data showing good linearity of response in the dose range of 3-20 Gy, with $R^2 = 0.9849$.

Radiation causes gel opacity that can be detected by optical investigations. The aim of this paper was to analyze the modified VIPARnd using two different optical techniques, spectrophotometry and an CMOS imaging device. The dose-response for spectrophotometric data was analyzed for absorbance peak and area under absorbance curve. The inverse of pixel intensity in function of dose was analyzed for the CMOS imaging device.

Temporal variation of the relative peak absorbance to the modified VIPARnd at 24h, 192h and 360h post-irradiation with 0, 5, 10 and 15 Gy is plotted in Figure 11.

It was previously shown that the time between irradiation and gel readout doesn't affect the system dose characteristics for N- vinylpyrrolidone based polymer gel^[23]. The data remains practically constant for at least two weeks after irradiation.

The repeatability and reproducibility of gel response were 0.5% and 5% respectively.

4. Conclusions

The interval of linearity for high-energy photon (6 MV) extends from 3 Gy up to 20 Gy for analyses using the spectrophotometric technique and from 3 Gy up to 20 Gy for data obtained with the imaging device developed. The data for the area under absorbance curve presented the highest sensitivity for dose-reponse.

Absorbance measurements with different post-irradiation time indicated a temporal stability for the gel dosimeter up to at least 360 h, though the spectra shape has varied with the absorbed dose and also over the time after irradiation. However, the trends of the observed changes are similar, independently of the dosimeter composition.

The modified VIPARnd polymer gel presented could be helpful for dosimetry of beams used in radiotherapy. The CMOS readout is a transportable, cheaper, easier to use and we suggest as future work the analysis of spatial resolution this setup for two-dimensional (2D) measurements as an alternative for dosimetry of small fields in radiotherapy.

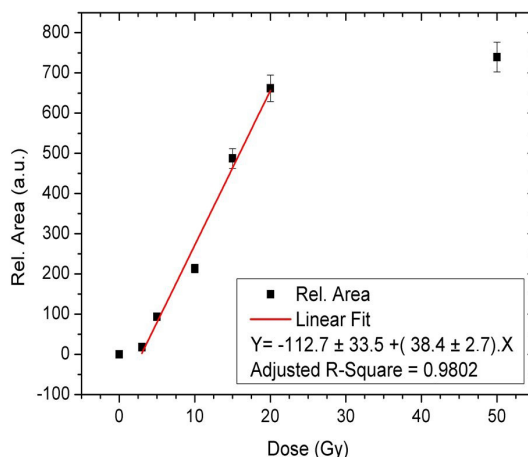


Figure 9. The relative area under the UV-VIS spectra as a function of absorbed dose. Error bars indicate the standard deviation of the relative area values measured in Gray (Gy).

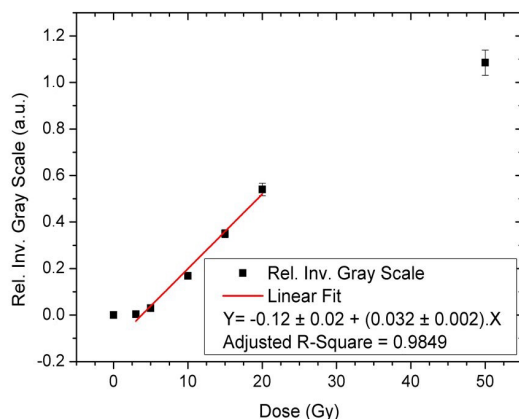


Figure 10. Relative inverse gray value as a function of absorbed dose. Error bars indicate the standard deviation of the relative inverse gray values measured in Gray (Gy).

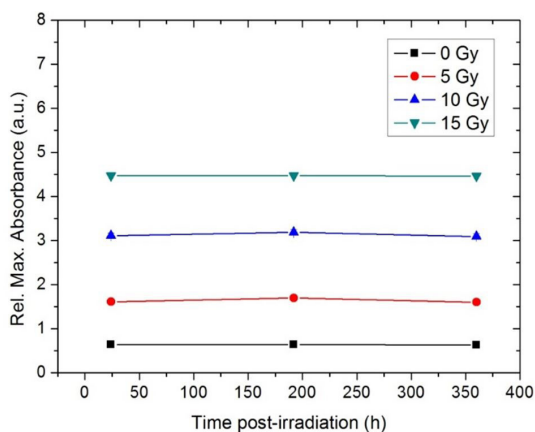


Figure 11. Variation of maximum UV-VIS absorbance at different times post-irradiation measured in Gray (Gy).

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