

Simulation of Water Equivalency of Polymer Gel Dosimeters with GAMOS

Türkan Özbay^{1,2}, Ayşegül Yurt^{2,3}, İsmail Özsoykal²

¹Izmir University of Economics, Vocational School of Health Services, İzmir, Turkey

²Dokuz Eylül University, Department of Medical Physics, İzmir, Turkey

³Dokuz Eylül University, Vocational School of Health Services, İzmir, Turkey

Address for Correspondence: Türkan Özbay, **E-mail:** turkan.ozbay@ieu.edu.tr

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ABSTRACT

Purpose: Polymer gel materials are used as dosimeters to provide three dimensional radiation dose distribution. An ideal gel dosimeter should offer radiological properties equivalent to water. This study aims to determine and compare the water equivalencies of various new polymer gel mixtures using Geant4-based Architecture for Medicine-Oriented Simulations (GAMOS) software program.

Methods: To do this, percentage depth dose curves of the simulated polymer gel mixtures were compared with water curves. Different combinations of materials from 2 main classes were used in the mixtures: (1) Sodium Carboxymethyl Cellulose (SODIUM CMC), Polyvinyl Alcohol (PVA) and Polyethylene Glycol (PEG) as gel matrix, (2) 2-Hydroxyethyl Methacrylate (HEMA), 1-Vinyl-2-Pyrrolidinone (VP) and Diethylene Glycol Dimethacrylate (DEGDMA) as monomer. Each mixture also contained ultra-pure water and an antioxidant material called Tetrakis Hydroxymethyl Phosphonium Chloride (THPC).

Results: Percentage depth doses of gel mixtures and water was calculated for 6, 18 MV and 50, 70, 100, 150 kVp X rays. The mixture sample showing radiological behavior nearest to water over all energies was observed to be 31a (Water, PEG, HEMA, THPC), while the largest difference is observed in 11a (Water, SODIUM CMC, HEMA, THPC).

Conclusion: As a result, simulated gel formulations were found to be radiologically water equivalent.

Keywords: Polymer gel, Dosimeter, Monte Carlo, GAMOS, Simulation

INTRODUCTION

There are a number of radiation measurement tools used in clinical practice based on radiation therapy and imaging applications, each with relative advantages and disadvantages. One tool, gel dosimeters, are produced from radiation-sensitive chemicals, which after irradiation, are polymerized as a function of the absorbed dose. For this reason, compared to other equipment, such as ion chambers and film dosimeters, they are able to deliver more precise three dimensional (3D) dose distributions and display better dosimeter performance. Gel dosimeters can also be used as phantoms since they are mostly composed of water equivalent materials. Two commonly used gel materials are Fricke and polymer gels. Polymer gel dosimeters exhibit more reliable results compared to Fricke type gel dosimeters, since they are unaffected by ferric ion diffusion, which may cause relatively misleading dosimetry (1).

Polymer gel dosimeter consists of monomers dissolved in the gel matrix and polymerized by free radicals resulting from the

radiolysis of water molecule. The amount of polymerization occurring in the polymer gel is related to the absorbed dose. The gel matrix preserves the spatial integrity of the polymers, and dose distribution can be determined by imaging modality such as magnetic resonance imaging (MRI) (2-4), computed tomography (CT) (5,6), ultrasound (7,8), optical computed tomography (9-11) and vibration spectroscopy (12-14). Due to its nature, polymer gel dosimeter provides high spatial resolution, precision and 3 dimensional accuracy, making it a valuable tool for analyzing 3D dose distribution and confirming complex treatment plans. The importance of gel dosimetry in modern radiation therapy has been previously emphasized (1-14).

Many authors have expressed concern about the excess toxicity of acrylamide found in commonly used polyacrylamide gel (PAG) dosimeters. Acrylamide is a serious neurotoxin and it is thought to be a human carcinogen and teratogen, and is easily absorbed through the skin (15). For this reason, considering the radiological

water equivalency of the polymer gel dosimeters, it is crucial to develop the research and use of less toxic monomers with a similar chemical structure to acrylamide. This simulation based study aims to focus on alternative gel compositions, based on different monomers, which is both safe and reliable.

The rapid development of computers allows modeling of the interaction of ionizing radiation with matter and dose information stored in matter to be determined with great accuracy within reasonable time (16). In order to simulate radiation transport, various algorithms and various codes have been developed. Monte Carlo method is a methodology that is used to solve mathematical and physical problems using a simulation technique. Another important application of Monte Carlo-based simulation programs used for many purposes, such as detector response, image reconstruction and evaluation of image quality, is to assess tissue equivalence of materials as dosimetry or phantom material (17,18).

There are many Monte Carlo codes that are successfully used in radiotherapy simulations. The most popular ones are: BEAMnrc (19), MCNP (20), PENELOPE (21) and GEANT4 (22). These codes use certain programming languages such as C++, which makes it difficult for researchers who are not familiar with such programming languages. On the other hand, there are some software programs acting as an interface between the user and the main programs mentioned above. These secondary software programs use the primary code of software but not requires any knowledge of programming language from the user. GAMOS software is one of them. It is built based on GEANT4 and widely used for simulation studies in medical physics (23, 24).

The purpose of this study is to construct necessary irradiation setup geometries in GAMOS and compare the percentage depth dose curves (PDD) of different polymer gel materials and water for diagnostic and radiotherapy X-ray energies. After these comparisons, gel mixtures with the radiological properties closest to water will be identified for the experimental part of the study, and will be produced for testing.

METHODS

Polymer gel mixtures

There are two categories of gel mixtures in this simulation study. In the first category are the combinations of two main components; gel matrices and monomers which are mixed in two different amounts (a and b) with water and an antioxidant material called Tetrakis (Hydroxymethyl) Phosphonium Chloride (THPC) (Table 1). In these mixtures, CMC, PVA and PEG were used as gel matrices, while HEMA, VP and DEGDMA were used as monomers. The percentages of gel mixtures used in this study were determined based on the most commonly used values in the literature (1, 25, 26). A second category was created to observe the influence of the crosslinking agent on the water equivalency of the mixture. In this category, VP and DEGDMA were also used as crosslinking agents, replacing half the amount of monomer. The polymer gel mixtures were combined in two different amounts, a and b (Table 2).

Table 1. Composition of polymer gel mixtures without crosslinking agents.

Component Type	Component Name	Amount [%] [a]	Amount [%] [b]
Liquid Medium	Water	88,85	85,85
Gel Matrix	1. SODIUM CMC ^a		
	2. PVA ^b	5	5
	3. PEG ^c		
Monomer	1. HEMA ^d		
	2. VP ^e	6	9
	3. DEGDMA ^f		
Antioxidant	THPC ^g	0.15	0.15

^aSodium Carboxymethyl Cellulose

^bPolyvinyl Alcohol

^cPolyethylene Glycol

^dHydroxyethyl Metacrylate

^eVinyl Pyrrolidinone

^fDiethylene Glycol Dimetacrylate

^gTetrakis Hydroxymethyl Phosphonium Chloride

Table 2. Composition of polymer gel mixtures including crosslinking agents.

Component Type	Component Name	Amount [%] [a]	Amount [%] [b]
Liquid Medium	Water	88,85	85,85
Gel Matrix	1. SODIUM CMC ^a		
	2. PVA ^b	5	5
	3. PEG ^c		
Monomer	1. HEMA ^d		
	2. VP ^e	3	4.5
	3. DEGDMA ^f		
Crosslinking Agent	1. DEGDMA 2. VP	3	4.5
Antioxidant	THPC ^g	0.15	0.15

^a Sodium Carboxymethyl Cellulose

^b Polyvinyl Alcohol

^c Polyethylene Glycol

^d Hydroxyethyl Metacrylate

^e Vinyl Pyrrolidinone

^f Diethylene Glycol Dimetacrylate

^g Tetrakis Hydroxymethyl Phosphonium Chloride

Monte Carlo simulation

GAMOS v.5.1.0 software program was used for the simulations in this study. The hardware equipment used in this study included two work stations with Intel Xeon® CPU E5630 4x2x(HT) 2.53 GHz and 64 bit processors with 24 GiB random access memory. Geometry was designed such that there was 100 cm distance between the source and the target. Initial target material was a cube of water with dimensions 100x100x100 cm³. Afterwards, gel mixture volumes of 10x10x10 cm³ dimensions were placed in the center of the target surface corresponding to the source. A 10 cm thick tungsten collimator block was placed between the source and the target to obtain a radiation field of 10x10 cm² at the target surface (Figure 1). Source, block and target were placed coaxially in the geometry. Each mixture volume was divided into 100 consecutive voxels in the direction of primary beam in order to give 100 consecutive (i.e. per mm) dose values along the depth of 10 cm. Every mixture was irradiated at 50 kVp, 70 kVp, 100 kVp, 150 kVp, 6 MV and 18 MV photon energies and PDD were recorded in text files.

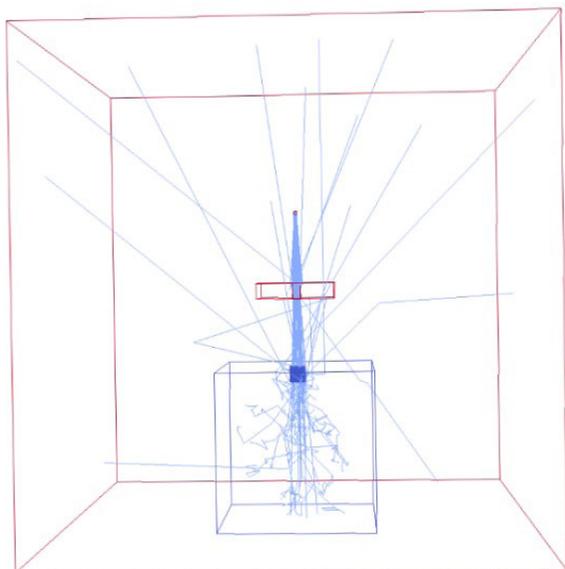


Figure 1. Source, block and target geometry in GAMOS with 10x10 cm² irradiation field and source to target distance of 100 cm.

In the geometry file, for means of simplification of the simulation, source model was represented in the form of energy spectrums generated with 1x10⁸ histories. The energy spectrums for the photon beams at kVp range were obtained by using SPEKTR 3.0 program (27) while 6 MV and 18 MV spectrums were obtained from the linear accelerator (Primus, Siemens) system database. Any particle in the simulation has been tracked until losing its energy down to 990 eV, which was a default value set by GAMOS.

Validation of the GAMOS Code

Prior to obtaining the results, a test was carried out on the constructed model to validate its operation. This test included the PDD values obtained from the measurement made for 6 MV photon beams. The measurements was made in a water phantom using a calibrated ionization chamber. Then a similar setup was modelled in GAMOS and PDD values were obtained. In both classes of PDDs, depth of the maximum dose (d_{max}) and ratio of the 20 cm depth dose to the 10 cm depth dose ($TPR_{20,10}$) values were recorded and compared.

Comparison and analysis of data

PDD data was obtained for all polymer gel mixtures and water at all energies. Each single PDD dataset consisted of 100 consecutive

relative dose values, normalized to the maximum dose. Water equivalency of the mixtures was examined over the comparison and analysis of these datasets, as shown in equation 1;

$$D(z) = \frac{M(z) - W(z)}{W(z)} * 100 \tag{1}$$

where D(z) represents the percentage difference between the relative dose values of polymer gel mixture (M) and water (W) at depth z ranging from 1 to 100 mm. Third quartiles (Q3) of these datasets were determined for each polymer gel mixture in order to compare the behaviour of different mixtures at a single energy, and also the behaviour of a single mixture at different energies.

OriginPro 2017 (OriginLab, Northampton, MA, USA) was used to plot D(z) against depth (z) for radiologically the least and the most water equivalent polymer gel mixtures at different energies.

RESULTS

Validation test of the constructed model in GAMOS yielded reasonable results as shown in Table 3. Table 4 shows the Q3 values of the difference between water and polymer gel mixtures without any crosslinking agent. The mixture sample showing radiological behavior nearest to water over all energies was observed to be 31a coded sample, while the largest difference is observed in 11a coded sample. These samples' PDDs and difference relative to water are plotted against depth in Figure 2.

Table 3. Validation of the model constructed in GAMOS.

	Measurement	Simulation	Difference [%]
D_{max}^a (cm)	1.6	1.6	-
$TPR_{20,10}^b$	0.67	0.657	1.9

^adepth of maximum dose

^btissue to phantom ratio

In the second category of mixtures with crosslinking agents, sample 311a exhibited the radiological behaviour nearest to water, while the largest difference has been observed in 111a (Table 5). On the other hand, Figure 3 shows the results of radiologically the least (111a) and the most (311a) water equivalent mixtures, which include crosslinking agent.

Table 4. Q3 values of the difference between water and polymer gel mixtures without crosslinking agent.

Gel Mixtures	Group 1				Group 2				Group 3									
	11a	11b	12a	12b	13a	13b	21a	21b	22a	22b	23a	23b	31a	31b	32a	32b	33a	33b
50 kVp	4,8	4,1	3,7	2,5	4,6	4	3,2	3,7	4,1	5,4	3,1	3,9	1	1,7	2	3,1	1,1	1,7
70 kVp	3,8	3,5	3,2	2,5	3,8	3,3	1,7	2,1	2,3	3,2	1,8	2,2	0,3	0,7	1	1,7	0,4	0,7
100 kVp	3	2,7	2,6	2,1	3,2	2,9	1	1,3	1,7	2,3	1,2	1,5	0,2	0,4	0,6	1	0,2	0,4
150 kVp	2,5	2,5	2,1	1,9	2,6	2,3	0,9	1	1,4	1,6	1	1,1	0,3	0,4	0,3	0,7	0,2	0,3
6 MV	0,8	0,8	0,7	0,7	0,8	0,8	0,2	0,2	0,4	0,2	0,2	0,2	0,3	0,2	0,2	0,2	0,2	0,4
18 MV	0,7	0,8	0,6	0,6	0,6	0,6	0,1	0,2	0,2	0,2	0,2	0,2	0,2	0,2	0,3	0,2	0,2	0,2

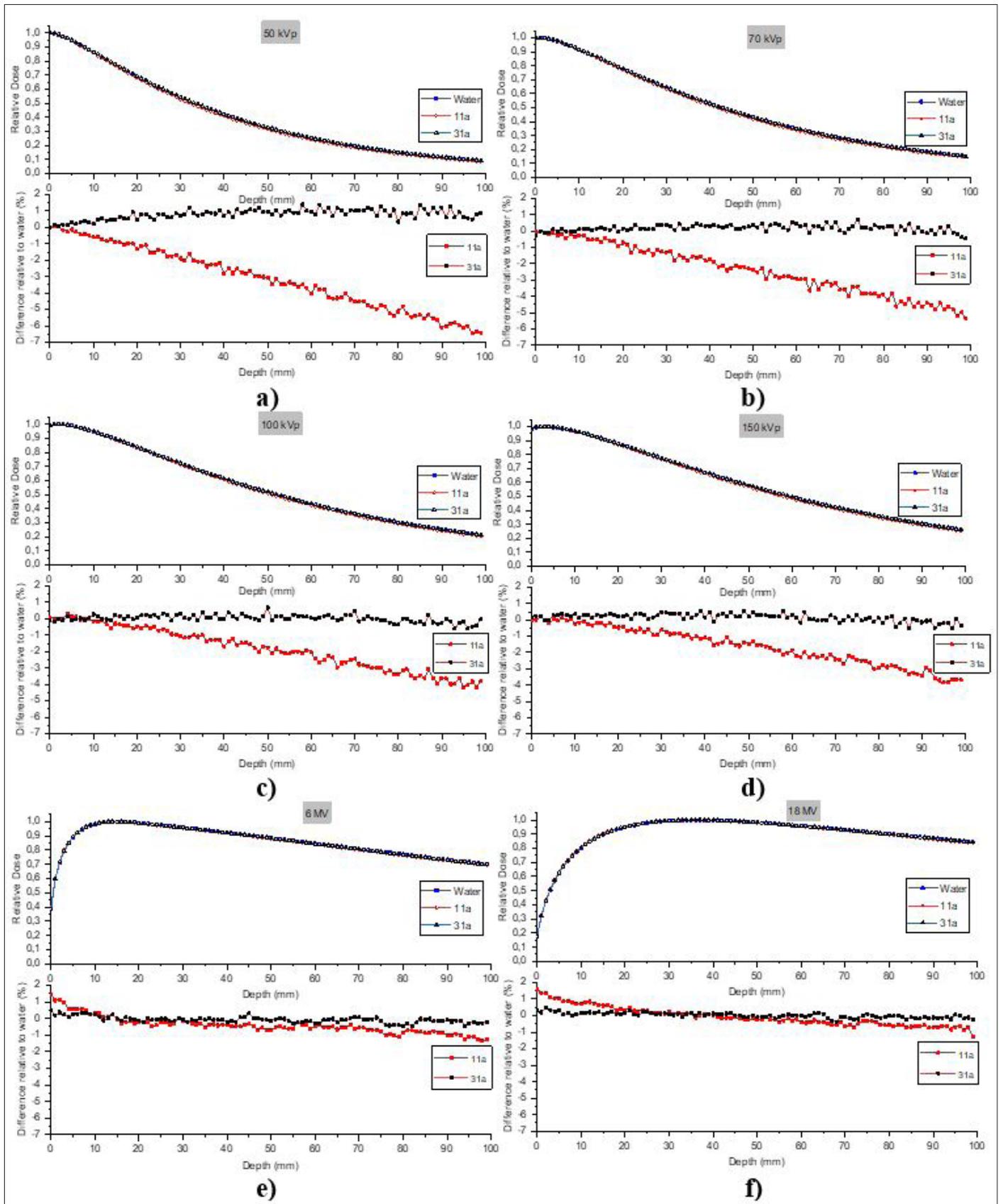


Figure 2. PDD and relative difference of mixtures without crosslinking agent (11a and 31a) according to water at (a) 50 kVp, (b) 70 kVp, (c) 100 kVp, (d) 150 kVp, (e) 6 MV ve (f) 18 MV.

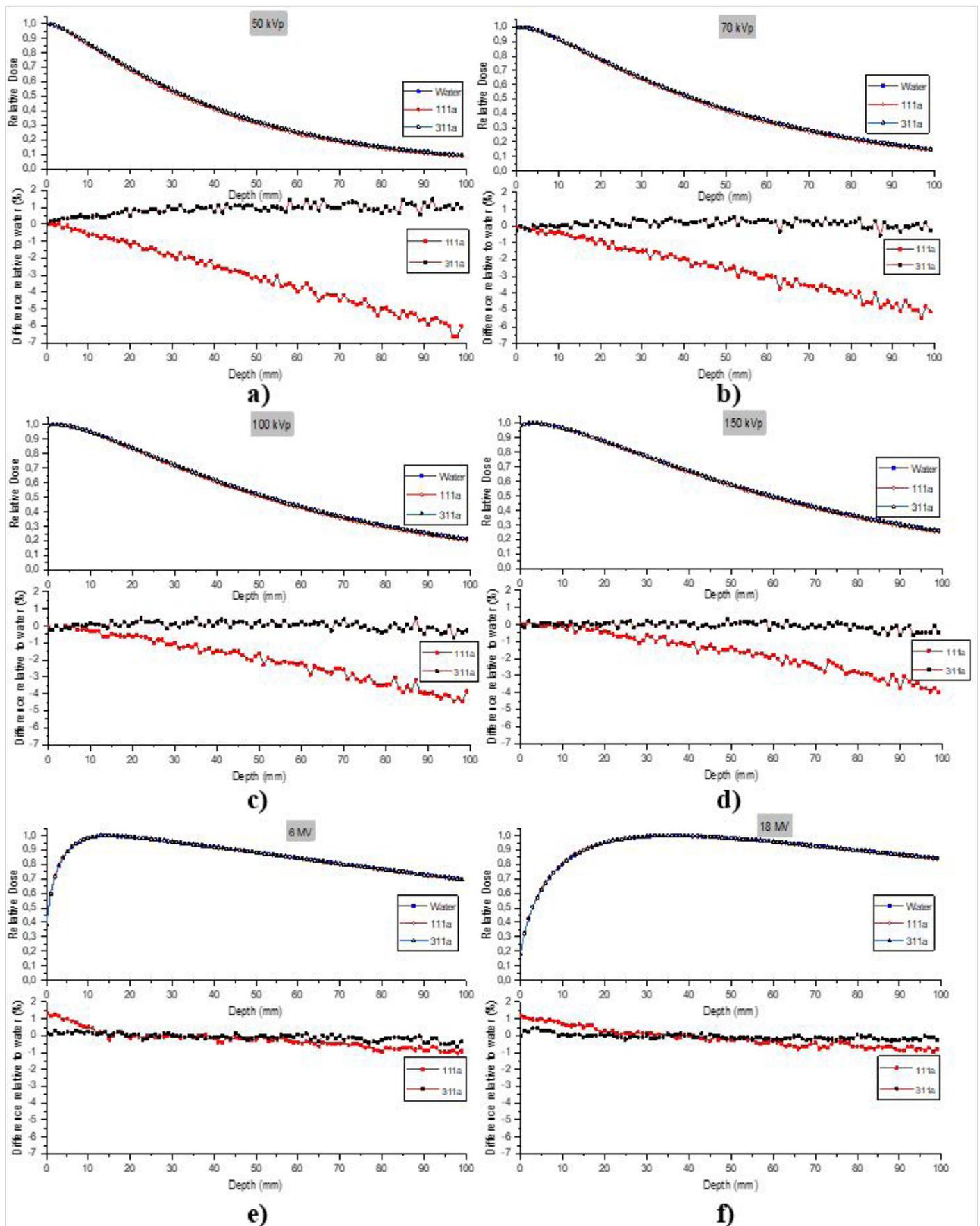


Figure 3. PDD and relative difference of mixtures with crosslinking agent (111a and 311a) according to water at **(a)** 50 kVp, **(b)** 70 kVp, **(c)** 100 kVp, **(d)** 150 kVp, **(e)** 6 MV ve **(f)** 18 MV.

Table 5. Q3 values of the difference between water and polymer gel mixtures with crosslinking agent

Gel Mixtures		Group 1 [Gel matrix: SODIUM CMCa]											
		111a	111b	112a	112b	121a	121b	122a	122b	131a	131b	132a	132b
a]	50 kVp	4,6	3,9	4,1	3,3	4,1	3,3	3,7	2,5	4,6	4	4,1	3,3
	70 kVp	3,8	3,5	3,5	2,9	3,4	2,9	3,2	2,5	3,8	3,3	3,4	2,9
	100 kVp	3,1	2,8	2,7	2,5	2,8	2,5	2,6	2,1	3,2	2,9	2,8	2,5
	150 kVp	2,6	2,3	2,3	2	2,4	2	2,1	1,9	2,6	2,3	2,4	2
	6 MV	0,7	0,8	0,7	0,7	0,8	0,6	0,7	0,7	0,8	0,8	0,8	0,6
	18 MV	0,7	0,2	0,2	0,1	0,2	0,1	0,6	0,6	0,6	0,6	0,2	0,1
Gel Mixtures		Group 2 [Gel matrix: PVAb]											
		211a	211b	212a	212b	221a	221b	222a	222b	231a	231b	232a	232b
b]	50 kVp	0,5	2,0	2,3	1,8	2,9	1,6	4,1	5,4	3,1	3,9	2,9	1,6
	70 kVp	0,6	2,1	2,3	0,6	1,5	0,7	2,3	3,2	1,8	2,2	1,5	0,7
	100 kVp	0,7	2	2,1	0,4	1	0,3	1,7	2,3	1,2	1,5	1	0,3
	150 kVp	0,6	2	2	0,3	0,8	0,4	1,4	1,6	1	1,1	0,8	0,4
	6 MV	0,5	1,1	0,8	0,3	0,2	0,5	0,4	0,2	0,2	0,2	0,2	0,5
	18 MV	0,7	0,3	0,2	0,1	0,1	0,1	0,2	0,2	0,2	0,2	0,1	0,1
Gel Mixtures		Group 3 [Gel matrix: PEGc]											
		311a	311b	312a	312b	321a	321b	322a	322b	331a	331b	332a	332b
c]	50 kVp	1,1	2,0	2,6	1,1	1,5	1,1	2	3,1	1,1	1,7	1,5	1,1
	70 kVp	0,3	2,1	2,7	0,5	0,6	0,4	1	1,7	0,4	0,7	0,6	0,4
	100 kVp	0,3	2	2,3	0,4	0,4	0,4	0,6	1	0,2	0,4	0,4	0,4
	150 kVp	0,2	2	2,3	0,4	0,4	0,4	0,3	0,7	0,2	0,3	0,4	0,4
	6 MV	0,2	0,3	1	0,4	0,2	0,5	0,2	0,2	0,2	0,4	0,2	0,5
	18 MV	0,2	0,1	0,2	0,2	0,3	0,2	0,3	0,2	0,2	0,2	0,3	0,2

^a Sodium Carboxymethyl Cellulose

^b Polyvinyl Alcohol

^c Polyethylene Glycol

DISCUSSION

An ideal polymer gel dosimeter must provide accurate dosimetric results at both low and high energy radiation. This is possible with a composition that displays water equivalent characteristics when it is exposed to low and high energy particles.

Mixtures with the initial digit of 1 are those that contain SODIUM CMC, the chemical with the highest mass density of all the mixture components, 1.6 g/cm³. In addition, SODIUM is the element with the highest atomic number of all the mixtures. This increased the probability of photoelectric absorption in mixtures 11a and 111a at energies within the kVp range, resulting in a lower penetration ability of the beam. For this reason, when compared to water, lower doses with a percentage difference up to 7% were observed with increasing depth. However, this is not the case for higher energies, where the interactions are dominated mostly by Compton scattering.

On the other hand, polymer gel mixtures with PEG as gel matrix, having the initial digit of 3, displayed significantly more water equivalent characteristics for all energies and depths, due to PEG's lower mass density, 1.13 g/cm³.

When the difference with respect to water is considered for the mixtures 31a and 311a, it is observed that the maximum values of percentage difference are 1.4% and 1.5%, respectively, occurring at 50 kVp for both mixtures. This can be interpreted as the crosslinking agents used in this study do not have a significant role on the water equivalency of the mixture. However, the effect of this materials will be examined further in the experimental part of this study, since they may have an influence on the polymerization process and therefore dose response of the gel.

Similar studies reviewed in the literature were carried out by using EGSnrc and BEAMnrc codes for simulation. In contrast, the current study is the only one to utilize GAMOS code.

Venning et al. (2005b) studied on MAGIC, MAGAS and MAGAT dosimeters, and investigated their water equivalency over PDD curves. The largest differences with respect to water at 6 MV have been reported as 3.5 %, 3.5 % and 3 % for MAGIC, MAGAS and MAGAT, respectively (28).

In studies carried out by Gorjiara et al., PRESAGE gel dosimeter was examined in the same respect. The results reported between an energy range of 50 kVp and 6 MV showed that the gels with

lower halogen content were superior to original PRESAGE with the maximum difference of 4 % versus 12 %. In this study, maximum differences with respect to water were at 50 kVp, and observed to decrease with the increasing beam energy, which is parallel to the findings in our study (29, 30). Another simulation study carried out by Gorjiara et al. showed that genipin gel exhibits a better performance, with a maximum difference of 0.8 % at 6 MV (31).

Our results indicate that 31a-b and 33a-b gel formulations show the best water equivalent dose response for the 70–150 kVp, 6 and 18 MV x-ray beams. In addition, group 3 compositions including crosslinking agents were found to have a better water equivalency, except for 311b, 312a and 322b.

This is a simulation study in which the actual measurement setup has been modeled by reasonable assumptions. However, GAMOS, while modeling radiation interactions with matter very accurately, cannot account for any chemical changes that occur in the matter due to radiation exposure and may yield different results. This study has compared various chemical composite materials for their physical properties. It should and will be followed by another study including actual irradiation and dosimetric evaluation procedures to overcome this limitation.

CONCLUSION

This study focused on the polymer gel mixtures which can be used as water equivalent dosimeters in radiology and radiotherapy departments. For this purpose, 54 different gel mixtures were composed and examined in a simulated medium.

Density and atomic number of the components play important roles in attaining this equivalency, as indicated by this study. Some of the results for the simulated gel mixtures yielded differences of

up to 7% compared to water, while other results were significantly closer. In the next step of this study, samples displaying the closest radiological properties to water (maximum percentage difference <2%) will be manufactured in laboratory conditions and tested experimentally before being recommended for use in clinical practice.

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