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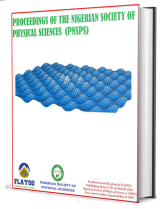

Optimization of HEMA composition in polymer gel dosimeters for cost-effective radiation measurement

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ABSTRACT

Polymer gel dosimeters (PGDs) are potential tools for radiation measurement in radiotherapy treatment planning systems (TPSs) and nuclear energy monitoring. Optimization of monomer composition is an essential aspect of PGD formulation because it helps achieve optimal dosimetric performance. In this study, 2-hydroxyethyl methacrylate (HEMA), in combination with N,N'-methylene-bis-acrylamide (Bis), was optimized to improve dose-evaluation efficiency and cost-effectiveness. The results show that the HEMA-free formulation containing 2% (w/w) Bis had the lowest sensitivity. When Bis and HEMA were both added, sensitivity increased linearly with increasing total monomer percentage (%T). However, when the weight fraction (WF) of Bis was fixed and the co-monomer percentage (%C) of HEMA increased from 0 to 60%, sensitivity decreased beyond the optimal value because of exhaustion of the vinyl groups in Bis. The mass density of the HEMA–Bis PGDs (≈ 1.04 g cm^{-3}), their electron density ($\rho_e = 3.42 \times 10^{23}$–$3.43 \times 10^{23}$ cm^{-3}), their number of electrons per unit mass ($n_e = 3.28 \times 10^{23}$–$3.30 \times 10^{23}$ g^{-1}), and their effective atomic number ($Z_{\text{eff}} = 7.3624$–7.4176) are comparable to those of water and muscle tissue, indicating water and tissue equivalence. These findings not only yield a more efficient HEMA-based PGD but also improve cost-effectiveness, thereby increasing affordability for healthcare applications.

Keywords: HEMA, Polymer gel dosimeter, Monomer optimization, Tissue equivalence, Radiological properties.

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1. INTRODUCTION

Monomers are essential components of polymer gel dosimeters (PGDs). Upon irradiation, they polymerize to indicate the absorbed-dose distribution for radiotherapy treatment planning systems (TPSs) [1, 2]. Monomer weight fraction (WF), together with the concentration of the radiolytic by-products of water, controls radiation-induced polymerization [3].

Because monomer composition influences not only the radiation sensitivity of PGDs but also their tissue equivalence and radiological features, such as mass density (ρ), electron density (ρ_e), and effective atomic number (Z_{eff}), optimizing the monomer WF can improve PGD efficiency, polymer-network stability [4], and thermal stability [5]. It can also reduce production costs, thereby improving the affordability of PGD-based TPS applications.

The optimization of monomer composition for efficient three-dimensional (3D) radiation-dose measurement has attracted con-

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siderable research interest, and several studies have been reported [6–9]. In these studies, the monomer WF is typically varied while the other gel-formulation components are kept constant [7, 8], and the total monomer percentage (%T) generally varies up to 8% (w/w) [9]. In addition, the effect of varying the WF of either the linear monomer or the cross-linker monomer in PGDs has been investigated [7, 10]. Based on monomer composition, PGDs reported in the literature can be grouped into three categories: (i) formulations containing both cross-linker and linear monomers [11–13], (ii) formulations containing only a cross-linker monomer, such as N,N'-methylene-bis-acrylamide (Bis) [14], and (iii) formulations containing only linear monomers, such as methacrylic acid (MAA) [15, 16]. However, most previous studies have focused on Bis and monomers other than 2-hydroxyethyl methacrylate (HEMA), despite the importance of HEMA as one of the less toxic monomers [17].

In this study, the effect of varying %T from 2% to 8% is investigated by simultaneously increasing the WFs of both Bis and HEMA. In addition, the effect of fixing the WF of Bis while increasing the WF of HEMA is examined to determine the optimal formulation while maintaining the tissue equivalence of the HEMA PGDs.

2. MATERIALS AND METHODS

2.1. PGD PREPARATION

HEMA PGD samples were prepared using the procedure described in a previous study [5]. Two batches, each containing four different samples, were prepared from deionized water (89.0%–92.0%), gelatin (6.0%), Bis (2.0%–3.6%), HEMA (0.0%–4.8%), and tetrakis(hydroxymethyl)phosphonium chloride (THPC) (7.5 mM). In the first batch, the WF of Bis was kept constant while that of HEMA was varied, causing the %C of Bis to decrease from 100% to 40% while the %C of HEMA increased from 0% to 60%. In the second batch, %T was varied from 2% to 8% by simultaneously increasing the WFs of both HEMA and Bis.

The PGD solutions were prepared in a fume hood. The procedure involved sequential addition of the reagents, which were weighed using a beam balance with an accuracy of $\pm 10^{-4}$ g. Deionized water was first heated to 48 °C, after which Bis was added and allowed to dissolve completely. Gelatin was then added gradually with continuous stirring until it dissolved fully. To avoid excessive heating of Bis, heating was stopped while stirring continued. As the temperature of the solution decreased, maltose solution was introduced at 35 °C, followed by HEMA and, finally, THPC at 27 °C.

The final mixture was stirred for 10–15 min at room temperature (T_R) to ensure uniformity. It was then poured into Perspex cuvettes with dimensions of 1 cm × 1 cm × 4.5 cm and covered with parafilm to prevent further oxygen penetration. The prepared samples were stored in a refrigerator maintained at 4–6 °C for gelation.

2.2. PGD IRRADIATION AND DOSE EVALUATION

For each sample, seven cuvettes were prepared: one served as a control sample, while the remaining six were irradiated at 22 ± 0.5 °C using a linear accelerator (LINAC). The PGDs were positioned in a water tank containing a cuvette holder [18] and irradiated with doses ranging from 5 to 30 Gy at an average dose rate of 540 Gy min^{-1}, delivered in a single fraction per exposure [18, 19].

The irradiated samples were analyzed using a Shimadzu UV-1800 ultraviolet–visible (UV–Vis) spectrometer. An unirradiated sample was used as the reference sample while the samples were scanned over the wavelength range of 200–800 nm. The change in absorbance (Abs.) was recorded at the wavelength of maximum absorbance for each sample. Abs.–dose response curves were plotted and fitted to sigmoidal dose-response curves using OriginPro 2018. Sensitivity was determined from the slope of the Abs.–dose response curves [20].

2.3. DETERMINATION OF TISSUE EQUIVALENCE AND RADIOLOGICAL FEATURES

Tissue equivalence is an important property that enables the use of PGDs in TPS for accurate dose delivery to target tumours while minimizing exposure to healthy tissues. The key parameters that constitute tissue equivalence include mass density, elemental composition, and radiological properties such as electron density, number of electrons per unit mass, and effective atomic number [21].

2.4. MASS DENSITY

The mass density of the HEMA PGDs was determined by measuring the mass of three PGD-filled cuvettes from each sample. The mass of the empty cuvette was subtracted to obtain the net mass of each sample. The average mass was then divided by the volume of the cuvettes:

$$\rho = \frac{m}{v}, \tag{1}$$

where ρ is the mass density, m is the mass, and v is the volume of the PGDs.

2.5. ELEMENTAL COMPOSITION

The elemental compositions of the HEMA-based PGDs were analyzed using field-emission scanning electron microscopy (FESEM) coupled with energy-dispersive X-ray spectroscopy (EDX), hereafter FESEM–EDX analysis. The samples were first freeze-dried, after which elemental analysis was performed. The technique provides the weight percentages or weight fractions of the constituent elements, except for hydrogen, which cannot be detected because of its low atomic number. To estimate the elemental WF of hydrogen in the PGDs, an approximation approach was used, as described in [22]. In this approach, it was assumed that all water content of the PGDs was removed during freeze-drying and that the hydrogen WF in the residual samples was negligible. Therefore, the hydrogen WF in each PGD sample was taken to equal that of the removed water.

The elemental WF of each element was compared with those of water and biological tissues to evaluate the overall tissue equivalence of the PGDs. These values were subsequently used to calculate the radiological properties of each sample.

2.6. ELECTRON DENSITY

Electron density is a radiological property representing the number of electrons per unit volume of a sample. It is calculated using

equation (2):

$$\rho_e = \rho N_A \sum_i w_i \left(\frac{Z_i}{A_i}\right), \tag{2}$$

where ρ_e is the electron density, N_A is Avogadro's number, and w_i, Z_i, and A_i are the WF, atomic number, and mass number of the ith element, respectively [23].

2.7. NUMBER OF ELECTRONS PER UNIT MASS

The number of electrons per unit mass represents the number of electrons per unit mass of a sample. It is also called the effective electron density [23] and is calculated using equation (3):

$$n_e = \frac{\rho_e}{\rho}. \tag{3}$$

2.8. EFFECTIVE ATOMIC NUMBER

The effective atomic number represents the overall atomic number of a compound or mixture and accounts for the contribution of all constituent elements in a sample. It is calculated using Mayneord's formula [23]:

$$Z_{\text{eff}} = \left(\sum_i^n a_i Z_i^{2.94}\right)^{1/2.94}, \tag{4}$$

where a_i is the relative electronic fraction of the ith element.

3. RESULTS AND DISCUSSION

3.1. RADIATION SENSITIVITY

Following irradiation of the PGDs, radiation-induced polymerization occurred proportionally with absorbed dose. The Abs.–dose response curves for batches 1 and 2 are presented in Figure 1.

Figure 1 presents the Abs.–dose response curves for HEMA PGDs irradiated with doses ranging from 5 to 30 Gy. In Figure 1(a), absorbance increased linearly with dose, with R^2 values of 0.9400, 0.9723, 0.9158, and 0.9840 for the HEMA 0%, HEMA 20%, HEMA 40%, and HEMA 60% samples, respectively. In Figure 1(b), the corresponding R^2 values were 0.9982, 0.8832, 0.9851, and 0.9670 for the 2%, 4%, 6%, and 8% T samples, respectively. The slopes of the linear graphs represent the radiation sensitivities of the corresponding dosimeter samples [1]. The radiation sensitivity of batch 1, with fixed WF of Bis and varying WF of HEMA, and that of batch 2, with varying %T by simultaneous increase in the WFs of Bis and HEMA, are presented in Figure 2.

As shown in Figure 2, when the WF of Bis was fixed at 2% (w/w) and that of HEMA increased from 0% to 3% (w/w), the %C of HEMA increased from 0% to 60%, while that of Bis decreased from 100% to 40%. The orange curve shows that sensitivity decreased with increasing HEMA %C and decreasing Bis %C. This decrease is attributed to the reduced availability of Bis cross-linking sites due to the exhaustion of its vinyl groups [11].

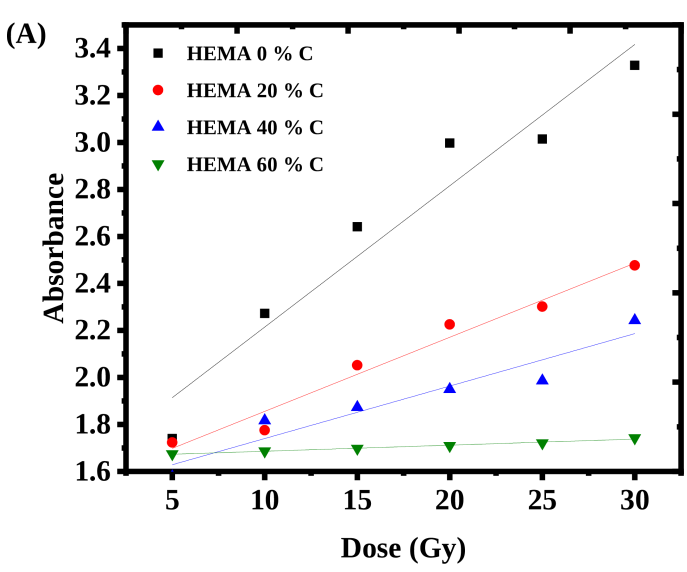
Conversely, when the WFs of Bis and HEMA were simultaneously increased such that %T increased from 2% to 6%, sensitivity increased. This can be attributed to the increased availability of pendant vinyl groups for cross-linking of the linear monomer as radiation-induced polymerization proceeded. Similarly, when the %C of Bis was slightly reduced from 3.6% to 3.2% and that of HEMA doubled from 2.4% to 4.8%, while %T remained 8.0% (w/w) of the whole PGD sample, sensitivity still increased. This suggests that sufficient cross-linking sites remained available in the composition, as each Bis molecule has two vinyl groups available for cross-linking [24]. In addition, because the radiation-induced polymer network involves HEMA, which produces only linear polymer chains, the presence of an adequate proportion of the cross-linker monomer Bis, with two vinyl groups, enhances the cross-linked polymer chains in 3D and increases cross-linking density. This, in turn, produces higher optical density and absorption per unit dose in UV–Vis spectrometry. Accordingly, samples in which both HEMA and Bis increased in an appropriate proportion showed higher sensitivity than samples in which the %C of HEMA increased while that of Bis decreased.

Therefore, to improve radiation sensitivity, both monomers should be varied simultaneously to maintain an adequate balance between chain propagation of the linear monomer and cross-linking facilitated by the cross-linker. The observed increase in sensitivity from about 0.002 to about 0.063 $\text{cm}^{-1}\ \text{Gy}^{-1}$ with increasing %C of Bis is consistent with earlier findings by Maryanski *et al.*, who reported that, in NMR studies of acrylamide (AAm)-based PGDs, increasing Bis or its %C in the total monomer component enhanced the maximum transverse relaxation rate ($R_2^{\max}$) from 0.8 s^{-1} at 33% C to 11.8 s^{-1} at 83% C [25]. The $R_2^{\max}$ increased significantly when the WF of the cross-linker exceeded approximately 30% C, with optimal performance observed around 50% C [6, 15].

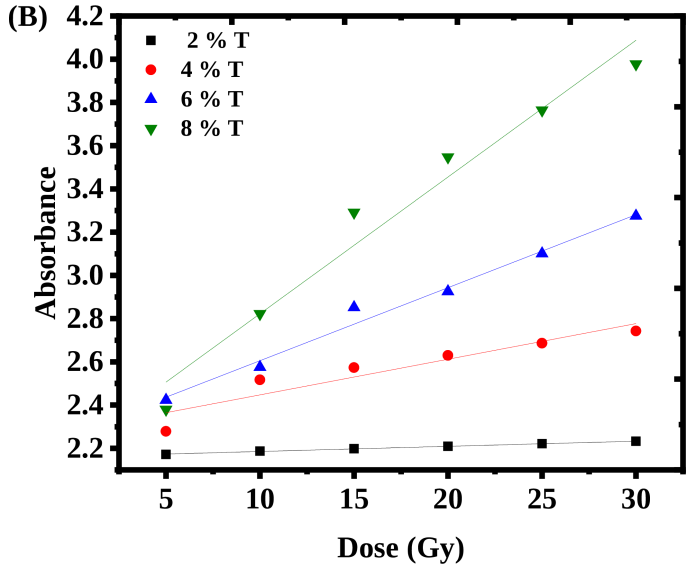
Because Bis is relatively toxic [26], increasing both Bis and HEMA in the present PGD formulation increases formulation toxicity, although it improves radiation sensitivity. In contrast, fixing the WF of Bis while increasing that of HEMA improves safety but reduces radiation sensitivity. Therefore, an optimum formulation must balance sensitivity and safety. The optimal monomer composition is proposed to occur at the intersection of the two sensitivity curves in Figure 2, where the total co-monomer content is approximately 4.7% T. At this point, HEMA constitutes 57.5% C (≈ 2.7% of the total formulation), while Bis constitutes 42.5% C (≈ 2.0% of the total formulation). This composition provides improved radiation sensitivity without substantially increasing toxicity.

The sensitivities and optimal monomer %C in the present study differ from those reported by Maryanski *et al.* for AAm and Bis co-monomers [25]. This difference is due to the different principles used to evaluate the PGDs. In their study, sensitivity was determined from changes in nuclear relaxation rate, whereas in the present study optical changes were used. This distinction reflects the differences in the physical quantities and variables involved, and it applies even when the same formulation is evaluated using two or more techniques. For example, NIBMAGAT gel dosimeters exposed to radiation doses ranging from 0 to 30 Gy exhibited a sensitivity of 0.016 $\text{cm}^{-1}\ \text{Gy}^{-1}$ using UV–Vis readout but 0.0775 $\text{s}^{-1}\ \text{Gy}^{-1}$ using NMR [27].

From an economic perspective, for a PGD-filled humanoid phantom with an internal volume of approximately 1 L, HEMA at 1%–4% (w/w) costs about USD 2–8, while Bis at 1%–4% (w/w) costs approximately USD 2.8–8.4. Thus, the combined cost of Bis and HEMA ranges from USD 4.8 to USD 16.4. However,



(a) Fixed WF of Bis and increasing WF of HEMA.



(b) Simultaneous increase in the WFs of Bis and HEMA.

Figure 1. Abs.–dose response curves of HEMA PGD samples: (a) fixed WF of Bis and increasing WF of HEMA; and (b) simultaneous increase in the WFs of Bis and HEMA.

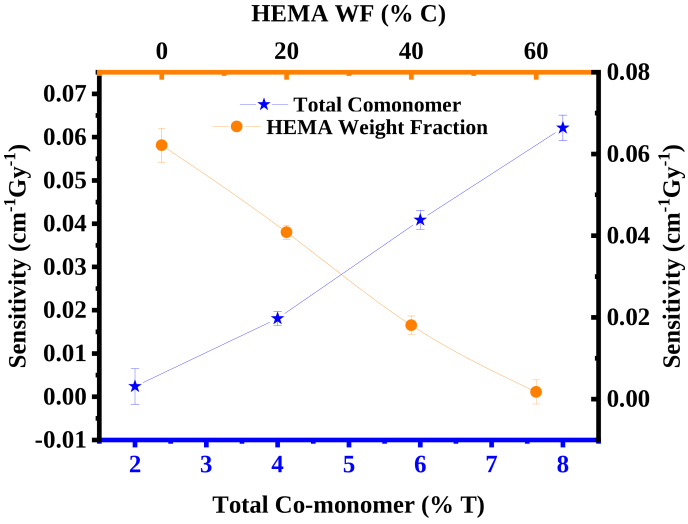


Figure 2. Variation in radiation sensitivity for the two batches of HEMA PGD samples as a function of monomer composition.

using the optimum WF of 2.7% (w/w) HEMA and 2.0% (w/w) Bis, corresponding to 57.5% C HEMA and 42.5% C Bis, results in an optimal sensitivity of 0.03 cm^{-1} Gy^{-1}, and the total cost of the two monomers does not exceed approximately USD 8.8. Because adding monomers beyond their optimal WF wastes resources, this optimization reduces waste, improves affordability, and contributes to economic efficiency.

3.2. MASS DENSITY

Table 1 presents the mass densities of HEMA PGDs with varying HEMA %C in comparison with those of water and muscle tissue.

Table 1 shows that the densities of the HEMA PGDs with varying HEMA %C are very close to those of water and muscle tissue. The obtained values are also comparable to those reported for other PGDs, such as PAG (1.035–1.042 g cm^{-3}) and MAG (1.046–1.050 g cm^{-3}). This close agreement confirms the tissue equivalence of the developed PGDs [28].

3.3. ELEMENTAL COMPOSITIONS

The elemental weight fractions of the HEMA PGDs, alongside those of water, muscle tissue, and other PGDs such as PAG and MAG, are presented in Table 2.

Table 2 shows that the weight fractions of oxygen and hydrogen in the HEMA PGDs are comparable to those of muscle tissue when rounded to two decimal places. However, they are lower than the corresponding values in water because water contains only oxygen and hydrogen. Similarly, the WFs of carbon and nitrogen in the HEMA PGDs, when rounded to two decimal places, are consistent with those reported for other PGDs, such as PAG and MAG.

The close agreement between the elemental WFs of the HEMA PGDs and those of water and muscle tissue indicates that these PGDs are suitable surrogates for the human body because they can attenuate radiation in a similar manner [15]. Furthermore, their similarity to PAG and MAG suggests that they can perform similarly to these two PGDs in clinical radiotherapy applications.

3.4. RADIOLOGICAL PROPERTIES

The electron density, number of electrons per unit mass, and effective atomic number of the HEMA PGDs were determined as described in Section 2 and are presented in Table 3.

Table 3 shows that the ρ_e values of the HEMA PGDs range from 3.42×10^{23} to 3.43×10^{23} cm^{-3}. These values are close to that of water (3.34×10^{23} cm^{-3}) and to those of radiochromic genipin gel dosimeters, which vary from 3.33×10^{23} to 3.43×10^{23} cm^{-3} [23]. Similarly, n_e ranges from 3.28×10^{23} to 3.30×10^{23} g^{-1}, which is comparable to that of water (3.34×10^{23} g^{-1}) and close to the range of 3.31×10^{23}–3.37×10^{23} g^{-1} reported for

Table 1. Mass densities of HEMA PGDs with varying HEMA %C alongside the densities of water and muscle tissue.

Sample	HEMA 0% C	HEMA 20% C	HEMA 40% C	HEMA 60% C	Water	Muscle
ρ (g cm^{-3})	$1.0379 \pm 1.0 \times 10^{-5}$	$1.0380 \pm 1.0 \times 10^{-5}$	$1.0380 \pm 1.0 \times 10^{-5}$	$1.0381 \pm 1.0 \times 10^{-5}$	1.0000	1.0400
Ref.	–	–	–	–	[28]	[28]

Table 2. Elemental weight fractions of the HEMA PGDs.

Element	Weight fraction							
	HEMA 0% C	HEMA 20% C	HEMA 40% C	HEMA 60% C	Water	Muscle	PAG	MAG
H	0.0979	0.0954	0.0944	0.0931	0.1120	0.1020	0.1065	0.1069
C	0.0647	0.0782	0.0885	0.0777	–	0.1230	0.0620	0.0771
N	0.0217	0.0292	0.0241	0.0411	–	0.0350	0.0218	0.0139
O	0.8134	0.7958	0.7883	0.7870	0.8880	0.7298	0.8096	0.8051
P	0.0010	0.0011	0.0030	0.0009	0.0000	–	–	–
S	0.0000	0.0002	0.0000	0.0003	–	–	–	–
Cl	0.0012	0.0001	0.0017	0.0000	–	–	–	–
Ref.	–	–	–	–	[28]	[6]	[28]	[28]

Table 3. Radiological features of the HEMA PGD samples with varying HEMA %C in comparison with water and muscle tissue.

Sample	ρ_e (cm^{-3})	n_e (g^{-1})	Z_{eff}	Ref.
HEMA 0% C	3.43×10^{23}	3.30×10^{23}	7.4003	–
HEMA 20% C	3.42×10^{23}	3.28×10^{23}	7.3627	–
HEMA 40% C	3.42×10^{23}	3.29×10^{23}	7.4176	–
HEMA 60% C	3.43×10^{23}	3.29×10^{23}	7.3624	–
Water	3.34×10^{23}	3.34×10^{23}	7.4170	[29]
Muscle tissue	3.36×10^{23}	3.36×10^{23}	7.4200	[30]

radiochromic genipin gel dosimeters [23]. In addition, Z_{eff} of the HEMA PGDs varies from 7.3624 to 7.4176, which is consistent with those of water (7.4170) and muscle tissue (7.4200) and is comparable to values reported for radiochromic genipin gel dosimeters [23].

These results indicate that, despite variations in the WF of HEMA for optimization purposes, the dosimeters exhibit radiological properties (ρ_e, n_e, and Z_{eff}) comparable to those of water and muscle tissue. Therefore, they are suitable for use in TPS.

4. CONCLUSION

HEMA PGDs prepared by varying both Bis and HEMA monomers showed increased radiation sensitivity with increasing total monomer WF. However, when the WF of Bis was fixed while that of HEMA increased, sensitivity decreased. Because increasing both Bis and HEMA improves radiation sensitivity but raises formulation toxicity, an optimal monomer composition is proposed to balance high radiation sensitivity and safety. The optimum formulation identified in this study consists of 2.7% (w/w) HEMA (equivalent to 57.5% C) and 2.0% (w/w) Bis (equivalent to 42.5% C), at which the cost of monomer components does not exceed approximately USD 8.8, compared with a potential cost of up to USD 16.4 at higher WFs. This optimization not only improves the efficiency and safety of the PGDs but also significantly improves their affordability. Consequently, it offers economic benefits for both individuals and healthcare systems.

DATA AVAILABILITY

No external data were used for this manuscript.

DECLARATION OF COMPETING INTEREST

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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LIST OF ABBREVIATIONS

Abbreviation	Definition
%C	Co-monomer percentage
%T	Total monomer percentage
3D	Three-dimensional
AAm	Acrylamide
Abs.	Absorbance
Bis	N,N'-methylene-bis-acrylamide
EDX	Energy-dispersive X-ray spectroscopy
FESEM	Field-emission scanning electron microscopy
HEMA	2-Hydroxyethyl methacrylate
LINAC	Linear accelerator
MAA	Methacrylic acid
n_e	Electrons per unit mass
PGD	Polymer gel dosimeter
Ref.	Reference
THPC	Tetrakis(hydroxymethyl)phosphonium chloride
TPS	Treatment planning system
T_R	Room temperature
UV–Vis	Ultraviolet–visible
WF	Weight fraction
Z_{eff}	Effective atomic number
ρ_e	Electron density

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