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Water-equivalence of gel dosimeters for radiology medical imaging

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HIGHLIGHTS

- Water-equivalence for radiology applications was evaluated for gel dosimeters.
- Fricke and polymer (NIPAM, PAGAT and itaconic acid) gel dosimeters investigated.
- Experimental, theory and Monte Carlo approaches were applied.
- Variations less than 3% were found for radiology energy range (up to 130kVp).
- Results suggest promising performance of gel dosimetry for radiology applications.

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ABSTRACT

International dosimetry protocols are based on determinations of absorbed dose to water. Ideally, the phantom material should be water equivalent; that is, it should have the same absorption and scatter properties as water. This study presents theoretical, experimental and Monte Carlo modeling of water-equivalence of Fricke and polymer (NIPAM, PAGAT and itaconic acid ITABIS) gel dosimeters. Mass and electronic densities along with effective atomic number were calculated by means of theoretical approaches. Samples were scanned by standard computed tomography. Photon mass attenuation coefficients and electron stopping powers were examined. Theoretical, Monte Carlo and experimental results confirmed good water-equivalence for all gel dosimeters. Overall variations with respect to water in the low energy radiology range (up to 130 kVp) were found to be less than 3% in average.

1. Introduction

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 many different types of dosimetry systems commonly used for clinical QA, such as ionization chambers, thermoluminescent dosimeters (TLDs), optically stimulated luminescent (OSLs), scintillators, diodes, radiochromic films and dosimetry gels (Hill et al., 2014; Kron et al., 2016)

International dosimetry protocols, TRS-398 IAEA and TG-51 AAPM, are based on determinations of absorbed dose to water. In fact, three decades ago, major efforts were carried out by primary laboratories focused on shifting dosimetry standards from air Kerma or exposure to absorbed dose to water. Nowadays, dose determinations must be referred to absorbed dose to water at a reference depth in a homogeneous water phantom (Huq et al., 2001). Ideally, phantom's material should be water equivalent; in other words, it should have the same absorption and scatter properties as water. Therefore, dosimetry systems directly based on in-water measurements would be convenient alternatives since they could avoid or minimize the uncertainties derived from

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conversions of absorbed dose between different materials. In this context, gel dosimeters appear as the most suitable option to design and implement dosimeters capable of performing direct measurements of absorbed dose in aqueous media. Furthermore, among the different available dosimetry techniques, only gels are capable of 3D dose mapping. There are two main types of dosimetry gels, namely Fricke gel (Schreiner, 2006) and polymer gel (Baldock et al., 2010), and while the latter presents the advantage of having long-term post-irradiation stability, Fricke gel dosimeters require less complicated manufacturing procedures and significantly cheaper chemicals reagents.

In terms of radiation-matter interaction, two different materials can be considered to be exactly equivalent if the corresponding fundamental physical quantities, like cross section and stopping power, are identical within the energy range of interest. Clearly, fundamental physical quantities influence the energy and angular distributions of primary and secondary particles inside the phantom and, consequently, the corresponding dose distributions. In that regard, water-equivalence for some gel dosimeters at radiotherapy energy ranges has been widely investigated and confirmed. However, there are much less studies focused on radiological properties of gel dosimeters at low energies for radiology imaging (Pantelis et al., 2004; Brown et al., 2008; Gorjiara et al., 2011), which may be useful for applications like computed tomography or mammography, among others.

The main goal of this study is to evaluate the water-equivalence of different gel dosimeters, such as Fricke gel and several polymer gels, within radiology energy range. To this aim, reported formulations for Fricke and three types of polymer gel dosimeters based on N-isopropylacrylamide (NIPAM), polyacrylamide (PAGAT) and itaconic acid (ITABIS) were carefully studied by direct experiments as well as theoretical and Monte Carlo approaches.

2. Materials and methods

In this study, theoretical, Monte Carlo and experimental approaches were employed. Gel dosimeters were produced in laboratory facilities following typical reported protocols, and then irradiated with X-rays with a beam quality corresponding to radiology applications obtaining their respective dose-response curves. Finally, irradiated and non-irradiated samples were scanned along with water samples by means of computed tomography (CT).

2.1. Gel dosimeters

Extreme thorough and precise protocols were used during the manufacturing and manipulation of the gel dosimeters, in order to minimize the differences between their physical and radiological properties with the ones used in the MC simulations. The different types of gel dosimeters used in this study were prepared using Milli-Q water and gelatin from porcine skin, type A 300 Bloom purchased from Sigma Aldrich.

Fricke gel dosimeter mainly consists of a solution of ferrous sulfate containing xylenol orange (XO) in Agarose or gelatin gel. In this study, the sensitive material was manufactured based on the method described elsewhere (Valente, 2007). Briefly, Fricke solution was prepared by using 1.38% w/w of sulfuric acid, 0.04% w/w of XO, 0.06% w/w of ferrous sulfate and 3.00% w/w of gelatin. NIPAM gel dosimeters were manufactured according to the protocol described by (Mattea et al., 2013) with 5.00% w/w of N-isopropylacrylamide (97%), 3.00% w/w of N,N′-methylenebisacrylamide (BIS) (99%) and 5.00% w/w of gelatin. PAGAT gel dosimeters were manufactured based on the method published by (Venning et al., 2005) by using 5.00% w/w of gelatin, 3.00% w/w of BIS (99%) and 3.00% w/w of acrylamide (AAm) (99%). ITABIS gel dosimeters were manufactured using 3.00% w/w of itaconic acid (99%) and 1.50% w/w BIS (99% purity) according to the protocol described by (Mattea et al., 2015) employing gelatin and a phosphate based buffer. Additionally, 10 mM of Tetrakis-phosphonium chloride (THPC) (80% w/v solution in water) was added to the studied dosimetric materials to minimize polymerization inhibition due to dissolved oxygen in the dosimeters. In addition, because oxygen may affect the overall performance of the polymer gel dosimeters, the prepared PAGAT, NIPAM and ITABIS materials were carefully stored in a nitrogen atmosphere. All gel dosimeter samples were stored at 4 °C. Table 1 reports the corresponding elemental compositions, calculated as weight fractions, for all materials used in this study.

Table 1
Chemical composition, expressed in terms of weight fractions*, of the different investigated gel dosimeters: FRICKE, NIPAM, PAGAT and ITABIS.

Element	Z Atomic number	FRICKE	PAGAT	NIPAM	ITABIS
Hydrogen	1	0.65916	0.65005	0.64992	0.64677
Carbon	6	0.00951	0.03003	0.03831	0.03047
Nitrogen	7	0.00232	0.00884	0.00891	0.00897
Oxygen	8	0.32809	0.31096	0.30272	0.31261
Sodium	11	0.00001	–	–	0.00161
Phosphor	15	–	0.00006	0.0006	0.00113
Sulfur	16	0.00089	0.00003	0.0003	0.00003
Chlorine	17	–	0.00006	0.0006	0.00060
Iron	26	0.00001	–	–	–

* The symbol – indicates values below 0.00001 w/w.

Mass densities (ρ) were evaluated by means of successive and precise volume and weight measurements, obtaining the results reported in Table 2.

Gel dosimeter samples were irradiated with photon beams produced by a 3 kW Siemens Kristalloflex generator (Valente et al., 2016) with electrical current and accelerating voltages within 5–60 mA and 20–60kVp, respectively. Dose rate was measured by means of calibrated ionization chambers: farmer type (PTW-Freiburg 30013) and pinpoint (PTW-Freiburg 30006) obtaining a (635.4 ± 0.8) cGy/min dose rate at sample's position accounting for all corrections and uncertainties. Besides, the X-ray spectrum was directly measured by means of a Cd-Te XR-100 Amptek® detection system, which properly processed the acquired signal by means of the DPPMCA software provided by the manufacturer. This information was used as an input for further studies based on Monte Carlo simulation. Finally, readout from the samples was performed by optical absorbance (A) measurements by means of a Shimadzu® UV-1800 spectrophotometer, and then absorbed dose (D) was estimated by using Eq. (1):

$$\Delta A = m \cdot D + n \quad (1)$$

where m and n are the slope and offset, obtained from dose-response calibration curve linear regression, respectively.

2.2. Characterization by computed tomography

Radiological properties of the different gel dosimeters were assessed by means of a CT (Siemens® Somaton) scanning with different settings with the aim of characterizing the properties of the samples within a wide range of radiological applications. The relative electron density (ρ_e) can be accurately obtained from CT values (Hounsfield index). Thus, radiological and morphological sample properties can be easily acquired.

A package containing 8 vials of irradiated and non-irradiated

Table 2
Gel dosimeter mass density and dose range.

Gel dosimeter	ρ [g cm^{-3}]	Dose range [Gy]
FRICKE	1.035 ± 0.009	0–30
PAGAT	1.038 ± 0.011	0–15
NIPAM	1.022 ± 0.008	0–15
ITABIS	1.051 ± 0.015	0–300

Fig. 1. Pack containing irradiated and non-irradiated vials of Fricke, PAGAT, NIPAM and ITABIS dosimeters along with water and aluminum samples for CT scanning.

samples of gel dosimeters along with water and aluminum samples, properly distributed in the corresponding styrofoam cells was prepared for CT scanning, as shown in Fig. 1.

Acquisition mode was set to “standard abdomen” and dedicated images filtered reconstruction based on Siemens AMPR technique was used. Sample pack was scanned with different tube voltages, namely 80, 110, and 130 kVp. Additionally, CT spectra were measured by means of energy-dispersive X-ray CdTe detector (XR-100 AMPTEK) provided with PX5 multichannel processor, positioned on the head zone of the patient couch. The detector's window (Be) was directly pointed to the rotating X-ray source and the CT was operated at different accelerating voltages during 5 min.

2.3. Theory and Monte Carlo for radiology properties

In this study, the molecular cross section $\sigma(E)$ of compounds and mixtures was evaluated by means of the additivity approximation, that is, the sum of the atomic cross sections of the elements involved in the compound or mixture. Thus, for a chemical compound A_aB_b, whose molecules consist of a atoms of the element A and b atoms of the element B, the number of electrons per molecule was obtained as $ZM = aZ(A) + bZ(B)$ and the molar mass was calculated as $AM = aAm(A) + bAm(B)$; where $Z(X)$ and $Am(X)$ are the atomic number and molar mass of element X, respectively. In this context, generalized oscillator strength (GOS) models (Inokuti, 1971) may be directly applicable to compounds and mixtures, since the oscillators may pertain to either atoms or molecules. The mean excitation energy of a compound were estimated from Bragg's rule. Then, the compound GOS were approximated as the sum of the atomic GOSs of the atoms according to Eq. (2):

$$Z_M \ln (I_M) = \sum_j f_j Z_j \ln[I_j] \tag{2}$$

where f_j and I_j indicate the quantity of atoms per molecule and the mean excitation energy, respectively for the j-atomic constituent, and $Z_M = \Sigma_j f_j Z_j$.

Similarly, relative electron density (ρ_e) and effective atomic number (Z_{eff}) were assessed by means of Eq. (3) (Sellakumara et al., 2016):

$$\rho_e = \rho\ N_A \left[\frac{\sum_j \alpha_j Z_j}{A_j}\right] \quad Z_{eff} = \left[\sum_j \alpha_j Z_j{}^x\right]^{\frac{1}{x}} \quad \alpha_j = \frac{\frac{f_j Z_j}{A_j}}{\sum_j \left(\frac{f_j Z_j}{A_j}\right)} \tag{3}$$

where N_A is Avogadro´s number and $x = 3.5$ is used (Khan, 1994).

As known, stopping power is the most relevant property for water-equivalence in dosimetry applications, whereas photon mass absorption coefficient (μ/ρ) is considered as the main transport quantity for water-equivalence in the context of radiology imaging applications. Bethe theory (Bethe, 1932) was applied to calculate the collision stopping power ($(S/\rho)_{col}$) in terms of the mean excitation energy I and accounting for the density effect, whereas radiative stopping power ($(S/\rho)_{rad}$) was calculated in terms of Bremsstrahlung according to (Seltzer and Berger, 1985). Finally, total stopping power ($(S/\rho)_{Tot}$) obtained as the sum of collision and radiative terms, was obtained for the different materials within the energy range of interest. The photon mass attenuation coefficients of the different compounds were calculated following the standard additivity rule, by combining the values for the elements present in the gel in terms of their weight fraction. Uncertainties were carefully evaluated by means of standard error propagation theory and one standard deviation was used to report the corresponding uncertainties.

Ratios of stopping powers and photon mass absorption coefficients were assessed by means of the additivity rule using the tables.exe routine in the PENELOPE Monte Carlo main code (Sempau et al., 2003). The specific geometry setups were exactly equivalent to the experimental configurations and incident spectrum was taken from experimental measurements performed directly on the X-ray generator and the CT scan. Particle transport parameters were set to the default configuration, except for absorption energy cutoff that was fixed at 1 keV for photons, electrons and positrons. Simulations were carried out by a standard PC i7 core and required 3×10^4 s for achieving statistical deviations lower than 3%. A dedicated subroutine was adapted to perform CT reconstructions by means of detecting transmitted photon fluence in a collimation grid as performed in the CT. Experimental CT DICOM® images were used to determine the simulation setup by means of a voxel-defined geometry. Air, water and PMMA (vial walls)

Table 3
Relevant physical quantities estimated for the different gel dosimeters relative to values for liquid water.

Quantity	FRICKE	PAGAT	NIPAM	ITABIS
Z_{eff} /$Z_{eff,\ water}$	0.96 ± 0.03	0.95 ± 0.03	0.94 ± 0.03	0.96 ± 0.03
ρ / ρ_{water}	1.035 ± 0.009	1.038 ± 0.011	1.022 ± 0.008	1.051 ± 0.015
ρ_e / $\rho_{e,\ water}$	1.03 ± 0.03	1.04 ± 0.03	1.02 ± 0.03	1.05 ± 0.03
I / I_{water}	1.00 ± 0.02	1.03 ± 0.02	1.00 ± 0.02	1.02 ± 0.02

materials were taken from PENELOPE database, whereas the materials of gel dosimeters were defined by user routines with their corresponding composition. For the simulations, 180 angular projections with a 1° shift were obtained. These projection images were further used for CT image reconstruction by means of typical back-projection algorithms, thus obtaining the corresponding ratio between CT reconstruction index for gel dosimeters and for liquid water.

3. Results

First, theoretical approaches were used to evaluate radiological physical quantities for the different gel formulations using water as a reference. This information was also useful to prepare corresponding databases for the required radiation-matter interaction properties necessary in Monte Carlo simulations.

Electron density (ρ_e) relative to water, effective atomic number (Z_{eff}) and mean excitation potential (I) were obtained for every studied material, as shown in Table 3.

Fig. 2 reports the photon mass absorption coefficient and total stopping power for the different gel dosimeters in terms of the corresponding values for water, estimated by PENELOPE.

Gel dosimeters were properly stored for 24 h after their manufacturing, then irradiated and analyzed providing the corresponding dose-response curves reported in Fig. 3.

3D distributions, expressed in terms of Hounsfield units (HU), were obtained after image acquisition and tomography reconstruction. Fig. 4 shows typical CT and Monte Carlo simulated slices of the scanned pack containing gel dosimeter samples.

HU were averaged on a $8 \times 8 \times 8\,mm^3$ region of interest (ROI), thus avoiding vial walls. The obtained results are summarized in Tables 4, 5 for experimental and Monte Carlo approaches, respectively.

4. Discussion

Application of theoretical approaches for fundamental physical properties as well as radiological quantities provided reliable estimations for all studied gel dosimeters. The obtained values for relative electron density (ρ_e), effective atomic number (Z_{eff}), mean excitation potential (I), photon mass absorption coefficient (μ/ρ) and total stopping power ($(S/\rho)_{Tot}$) show differences lower than 5% with respect to water within the investigated radiology energy range. As reported, all gel dosimeters showed a linear trend for the dose-response curve within

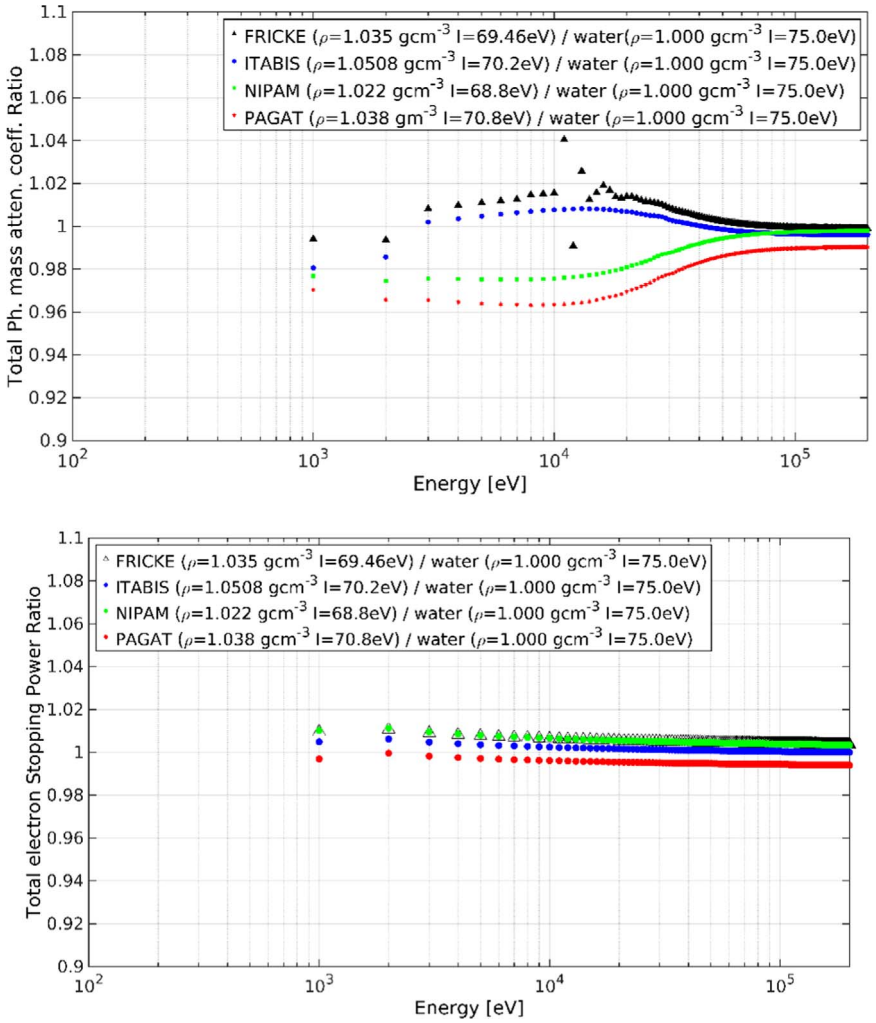

Fig. 2. Photon mass absorption coefficient (top) and total stopping power (bottom) relative to water for different gel dosimeters obtained by PENELOPE.

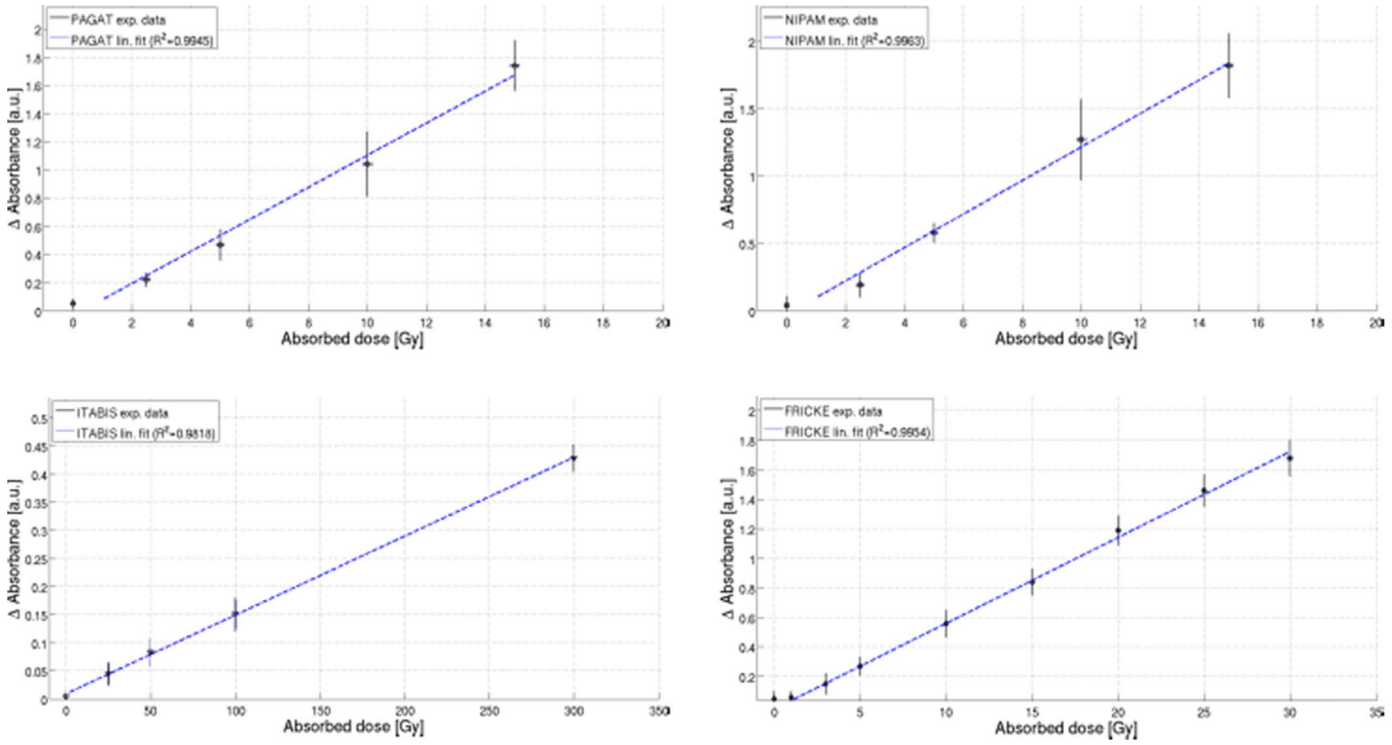

Fig. 3. Dose-response curve for PAGAT (top left), NIPAM (top right), ITABIS (bottom left) and Fricke gel (bottom right) dosimeters along with their corresponding linear fit. Error bars indicate 95% of confidence.

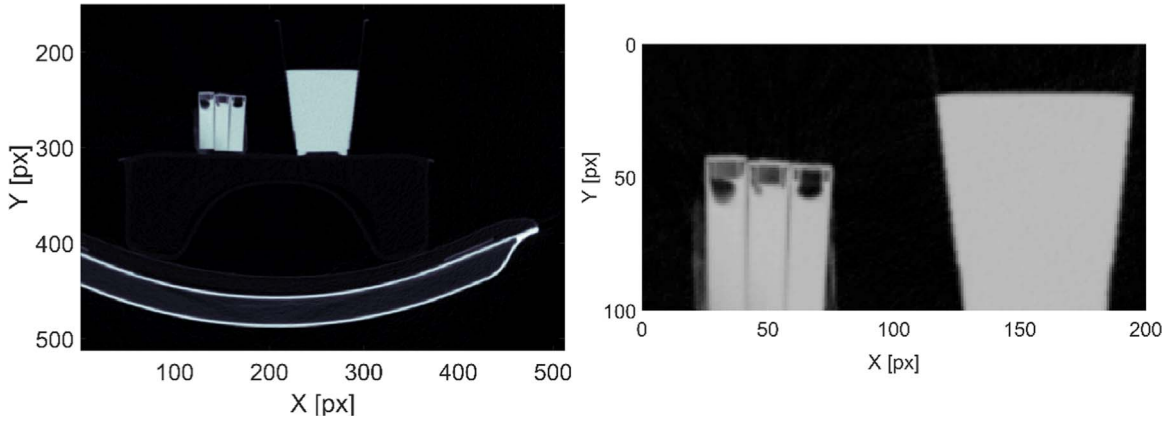

Fig. 4. Experimental (left) and simulated (right) CT slices of the scanned pack of vials along with water as reference at 110 kVp.

Table 4
Experimental HU relative to water for the different irradiated and non-irradiated gel dosimeters.

kVp	FRICKE (0 Gy)	FRICKE (15 Gy)	PAGAT (0 Gy)	PAGAT (15 Gy)	NIPAM (0 Gy)	NIPAM (15 Gy)	ITABIS (0 Gy)	ITABIS (100 Gy)
80	1.04 ± 0.01	1.02 ± 0.02	1.02 ± 0.02	1.01 ± 0.02	1.03 ± 0.02	1.02 ± 0.01	1.02 ± 0.03	1.02 ± 0.03
110	1.04 ± 0.01	1.03 ± 0.02	1.03 ± 0.02	1.01 ± 0.02	1.03 ± 0.02	1.03 ± 0.01	1.03 ± 0.02	1.03 ± 0.03
130	1.01 ± 0.01	1.02 ± 0.02	1.02 ± 0.02	1.01 ± 0.02	1.05 ± 0.01	1.02 ± 0.02	1.02 ± 0.02	1.02 ± 0.03

Table 5
Monte Carlo HU relative to water for the different gel dosimeters.

kVp	FRICKE (0 Gy)	PAGAT (0 Gy)	NIPAM (0 Gy)	ITABIS (0 Gy)
80	1.03 ± 0.06	1.04 ± 0.06	1.05 ± 0.06	1.04 ± 0.07
110	1.03 ± 0.05	1.03 ± 0.06	1.04 ± 0.06	1.04 ± 0.06
130	1.01 ± 0.04	1.01 ± 0.05	1.04 ± 0.04	1.03 ± 0.04

the corresponding dose range, also confirming that optical analysis states as a reliable and accurate technique for dose-response characterization of all of the different gel dosimeters hereby investigated.

During CT scanning, the presence of water samples were useful as references to establish accurately the corresponding HU. Furthermore, the inclusion of a high electron density material (aluminum), which may affect in a non-negligible manner the image reconstructed morphology, was also useful to verify the CT calibration curve as well as to study the use of different reference material for signal normalization

purposes.

The use of directly measured spectra as input for the Monte Carlo simulations was essential to reproduce almost exactly the experimental configurations, thus providing the Monte Carlo approach the capacity to generate more realistic and reliable CT reconstructed gel dosimeters images. Comparisons between HU relative to water Monte Carlo and experimental approaches show good agreements, thus remarking the suitability of the developed Monte Carlo technique.

5. Conclusions

Different gel dosimeters were carefully studied in terms of water-equivalence within the radiology energy range. Theoretical models based on additivity rules were adapted to estimate different physical and radiological properties. Dedicated Monte Carlo subroutines, adapted from the PENELOPE main code, demonstrated to be capable of describing different relevant properties regarding water-equivalence. CT scanning provided a reliable method to obtain direct information about radiological properties of gel dosimeters within radiology framework. In summary, theoretical, experimental and Monte Carlo approaches provided proper characterizations for the hereby-investigated materials confirming their high degree of water-equivalence. Moreover, excellent agreement was found among theory, Monte Carlo simulations and experiments, observing deviations with respect to water lower than 5% for all investigated properties. Although some works have previously reported about water-equivalence of some gel dosimeter types for radiotherapy applications, no literature is available about water-equivalence at low energy, which highlights the relevance of this work reporting the water-equivalence performance for four different gel dosimeters.

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