

## Radiological properties and structural morphology of different MAGAT gel dosimeters formulation

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### ABSTRACT

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**Keywords:** MAGAT gel dosimeter, radiological properties, polymer gel morphology, zinc oxide nanoparticle, methylene blue.

**Background:** : One of the most important qualities of a radiation dosimeter is tissue-equivalent, which represents radiation absorption and scattering that is similar to that of human tissue. The goal of this study is to determine the radiological and structural properties of several MAGAT gel dosimeter formulations prior to irradiation. This will provide a basis on how radiation properties altered prior to irradiation, which will later affect radiation absorption behaviour. **Materials and Methods:** Radiological properties of different MAGAT gel formulations (MAGAT, MAGAT doped methylene blue, MAGAT doped zinc oxide nanoparticles, MAGAT dope methylene blue and zinc oxide nanoparticles) were measured from density, linear attenuation coefficients, mass attenuation coefficients and CT values. The results were compared to soft tissue and water as references. The gel's morphological structures were investigated using electron microscopy for better understanding of gel's absorption mechanism. **Results:** The mass density of all MAGAT gel formulations differed by up to 2.5% from that of water and are very close to that of muscle tissue. This is due to the high gelatin and monomer concentration used in MAGAT gel formulation. The CT-values, furthermore, are within the soft tissue range. The MAGAT doped with methylene blue and ZnO NPs demonstrated the greatest increase in linear attenuation value, as well as having a nano spider-web morphological structure with a large surface area, which may have increased gel sensitivity and attenuation coefficients of MAGAT gel. **Conclusion:** Different MAGAT gel formulations were found to exhibit similar radiological properties to soft tissues and water.

### INTRODUCTION

As dose measurement conversion system (dosimeter) is to provide high accuracy and dependability for the demanding applications, a promising development in the radiotherapy verification system is the introduction of 3D radiation-sensitive gels (polymer gel dosimeter) <sup>(1)</sup> that are made of monomers, crosslinkers, gelatin, antioxidant, and water. Upon irradiation of polymer gel dosimeters, it will polymerize as a function of the absorbed dose. As the polymer gel dosimeters allow the integration of dose within the dosimeter, evaluation of a complete volume at once, equivalence of anatomical soft tissue, and enablement of true 3D dosimetry <sup>(1-3)</sup>, it is to be on the advantage side compared to the current ones. For a polymer gel dosimeter to be suited in radiation dosimetry, water-equivalent radiological properties that are of physical density, relative electron density, effective atomic number, and absorption and scattering of radiation should be exhibited <sup>(4-6)</sup>.

Radiosensitive nanoparticles play an important

role in the betterment of tumour dose that aims to deliver effective radiation doses to remove cancer cells without surpassing normal tissue tolerance. There are a number of published studies on the significance and influence of nanoparticles on dose amplification in polymer gel dosimeter application <sup>(7-10)</sup>. The fact that photoelectric chance increases when metal is presence inside a biological target leads to an increased dose absorption has driven the nanoparticle application in radiotherapy (radionanotherapy) <sup>(11)</sup>. This process takes advantage of the high mass-energy absorption coefficient ( $\mu_{en}/\rho$ ) of these materials relative to tissue. As the deposited dose from ionizing radiation is proportionate to attenuation coefficient, the nanoparticles are high likely to increase the local tumor dose <sup>(12)</sup>. Despite that, with regard from extensive studies comprising radiation dose enhancement using gold nanoparticles, bismuth, and platinum in polymer gel dosimeter <sup>(7-12)</sup>, several studies have been conducted to investigate the probable dominance of other high atomic numbers, Z nanoparticles mainly the metal oxide particles. Thus,

the comparative valuation of the outcomes of dose enhancement in clinical radiotherapy has been scarce. Few decades ago, metal oxide nanoparticles which structures exhibit amazing novel and improved physical, chemical, and biological properties have attracted interests especially the Zinc Oxide nanoparticles (ZnO NPs) that is known to be one of the most substantial metal oxide nanomaterials given its versatile medicinal use, biocompatibility, and biomedical functions<sup>(13)</sup>.

When the nanoparticles are integrated into the polymer gel, there are a various degree of interactions shown chemically and physically. It has become the motivation of this work to investigate the radiological properties and structural changes due to the addition of a high atomic number of ZnO NPs in the MAGAT gel dosimeter. This measurement must be carried out prior to the irradiation process as it is crucial to acquire a complete insight of the gel composition interactions before the dose response measurement. Even so, no quantifiable data on the changes in structural polymer gel dosimeter was found in the scientific literature preceding the irradiation process. Therefore, this study aims to present a set of radiological properties measurement of MAGAT gel dosimeter doped with different additives and structural properties prior to irradiation. This is to justify on how the radiation properties change before the irradiation takes place which would affect the polymerization process. An advanced understanding of the microstructural of polymer gel will be offered by the outcomes of this study. This is vital to the future development of highly sensitive and hence clinically viable, polymer gel dosimetry systems.

## MATERIALS AND METHODS

### Fabrication of MAGAT gel dosimeter

The diverse formulations of Methacrylic Acid Gelatin and Tetrakis (hydroxymethyl) phosphonium chloride (MAGAT) gel dosimeter consisted of methacrylic acid (MAA) (Acros, Organics), gelatin (250 bloom, Bovine) (Sigma Aldrich), purified water, tetrakis (hydroxymethyl) phosphonium chloride (THPC) (Sigma Aldrich), methylene blue (StainPur® (C.I. 52015) and zinc oxide nanoparticle of size 20 nm (MK Nano). The MAGAT gel was prepared under standard atmospheric according to Razak *et al.*,<sup>(14,15)</sup>. The gelatin is first blended with purified water in a mixing beaker and continuously stirred at approximately 48 °C until a completely dissolved and uniform solution is achieved. Before adding the monomer and THPC into the solution, the solution is cooled and stirred continuously until the gel becomes a clear solution. Then, the methylene blue is added before adding the ZnO that is prepared by blending zinc oxide nanoparticle with methacrylic acid (MAA).

Towards the end of the preparation, the mixture is continuously stirred for 20 minutes to allow all ZnO nanoparticles to dissolve until a homogeneous solution is obtained. All the MAGAT formulations are then poured into 4 ml tissue-equivalent polystyrene cuvettes with internal measurements of 1 cm × 1 cm × 4.5 cm (width × length × height) and are covered by caps. All gels are to be wrapped with aluminum foil to prevent any preliminary polymerization from the ambient light. The sample vials are stored at 4°C before irradiation. In this study, the concentrations of MAGAT gels are according to Razak *et al.*,<sup>(14)</sup> are shown in table 1.

Table 1. The MAGAT gel formulations.

Formulation	Concentration				
	Gelatin	MAA	THPC	ZnO (20 nm)	Methylene Blue
MAGAT	6%	6%	10mM		
MAGAT + ZnO NPs	6%	6%	10mM	50mg/ml	
MAGAT + Methylene Blue	6%	6%	10mM		10mg/ml
MAGAT + Methylene Blue + ZnO NPs	6%	6%	10mM	50mg/ml	10mg/ml

## MATERIALS AND METHODS

### Density measurements

The density measurement was performed by using the density meter (Anton Paar: DMA™ 35 Basic portable density meter). The measurements were made in the energy laboratory (school of physics, USM). All the measurements of the density for each formulation are listed in table 2.

Table 2. Linear attenuation coefficient, mass attenuation coefficient, and mass density.

Formulation	Linear Attenuation Coefficient $\mu$ (cm <sup>-1</sup> )	Mass Attenuation Coefficient $\mu/\rho$ (cm <sup>2</sup> /g)	Density g/cm <sup>3</sup>
MAGAT	0.2016	0.1978	1.0190
MAGAT + ZnO NPs	0.2210	0.2156	1.0250
MAGAT + methylene blue	0.2055	0.2018	1.0180
MAGAT + methylene blue + ZnO NPs	0.2340	0.2285	1.0240
Muscle <sup>(16),b</sup>	0.2420	0.2326	1.0400 <sup>(16)</sup>
Kidney <sup>(16),c</sup>	0.2180	0.2076	1.0500 <sup>(16)</sup>
Brain, grey/white matter <sup>(4)</sup>	0.2140	0.2058	1.0400
Evaluated Xcom value for water <sup>(17)</sup>	0.2066	0.2066	1.0000

<sup>a</sup>density at 23 °C; b,c The mass attenuation coefficients tabulated by King *et al.* (16) are converted to linear attenuation coefficients using densities of 1.04 and 1.05 g/cm<sup>3</sup> from ICRP (1975)(18) for muscle and kidney, respectively.

### Linear attenuation coefficient & mass attenuation coefficient measurements

The linear attenuation coefficients of the MAGAT

gel dosimeter are measured at 59.5 keV using a gamma source (Am-241) by using a NaI(Tl) detector for gamma spectrometry. The detector is shielded with a lead housing to lessen the scattered radiation from the source or the background. The Multi-Channel Analyser (MCA-3 Series) / P7882 (supplied by FAST COMTEC) is used to measure the attenuation of samples with the samples placed between the NaI (Tl) detector and gamma source. The real-time, which is the total time for a detector to count, is set to 600 seconds where the photopeak, full width at half maximum (FWHM) and net area of photopeak are recorded using MAESTRO software (MAESTRO-Pro Ortec Ametek; Advanced Measurement Technology, Tennessee, USA). The linear and mass attenuation coefficients are calculated by using Beer Lambert's law (equations 1 and 2).

$$I = I_0 e^{-\mu x} \quad (1)$$

Where;  $I$  is the intensity of photons transmitted across some distance,  $I_0$  is the initial intensity of photons,  $\mu$  is the linear attenuation coefficient, and  $x$  is the thickness of the absorber;

$$\text{mass attenuation coefficient} = \mu/\rho \quad (2)$$

#### CT-number measurement

The CT-number measurements were obtained from a Toshiba Aquilion Computed Tomography (CT) scanner at Advanced Medical and Dental Institute (IPPT), Universiti Sains Malaysia. The MAGAT gel samples were placed on top of the CT scanner couch and scanned using the standard brain protocol routine at 120 kV and 300 mAs.

#### Field emission scanning electron microscopy (FESEM) with energy dispersive X-Ray spectroscopy (FESEM-EDX)

In the freeze-drying process, the items were put under high vacuum until the frozen liquid sublimates to solid and dried components. The freeze-dried MAGAT gel samples were then cut into 5 mm-thick slices for microstructural imaging using FESEM (Model FEI Nova NanoSEM 450) with energy dispersive X-ray spectroscopy (EDX) system (Model JSM - 6460 LV). To attain the cross-sectional view of the sample surface, the magnifications used were 5000 $\times$ , 50000 $\times$ , and 100000 $\times$ .

## RESULT

#### The mass density, linear attenuation coefficient, and mass attenuation coefficient

The evaluated linear attenuation coefficient, mass attenuation coefficient, and density for the four dissimilar formulations of MAGAT gel dosimeter

were weighed against the reference values of muscles, kidney<sup>(16)</sup>, brain (grey/white matter)<sup>(4)</sup>, and the evaluated value of water<sup>(17)</sup> at the equal geometry of 59.54 keV. Table 2 shows the linear attenuation coefficient, mass attenuation coefficient, and density acquired by the Xcom database and experimental value of different MAGAT gel formulations. The radiation properties of materials towards ionizing radiation were linked with the density where the density of four formulations of MAGAT gel were close to water density with 1.8% - 2.5% difference to water.

The linear attenuation coefficient for MAGAT was equal to 0.2016 cm<sup>-1</sup>, with a difference of 2.4% compared to water, and as the zinc oxide nanoparticle was added, the value increased to 0.221 cm<sup>-1</sup>, which was 6.96% greater than the original MAGAT value. For the formulation of MAGAT doped with methylene blue, the linear attenuation value increased to 0.2055 cm<sup>-1</sup>, with a 0.532% difference compared to water. As for the formulation of MAGAT doped with methylene blue and ZnO, the linear attenuation showed the highest value of 0.234 cm<sup>-1</sup>, which was 13.26% greater than MAGAT. Nevertheless, the linear attenuation value of all four MAGAT gel formulations was within the range of soft tissue materials (brain, kidney, and muscle). These value increments are paramount in gel dosimetry as it may lead to an increase in gel sensitivity towards the radiation by absorbing more radiation energy.

#### CT-number measurement

While for the CT number, this measurement aims to compare the CT number of MAGAT gel doped different additives with the CT number of soft tissue equivalent. According to table 3, the values of the CT number of four different MAGAT gel dosimeters were located within the range of soft tissue (25 HU–45 HU). The CT number for MAGAT (25.7 HU) and MAGAT-doped Methylene Blue (25.9 HU) both gels are within the range of kidney or brain (white matter). However, for MAGAT-doped ZnO (45.5 HU) and MAGAT-doped Methylene Blue and ZnO (41.7 HU), both gels are within the range of muscles or brain (grey matter). The results show that the four MAGAT gel formulations are tissue-equivalent materials.

**Table 3.** CT-number of different MAGAT gel dosimeter and several tissue-equivalent materials.

Material	CT-number
MAGAT	25.7
MAGAT + ZnO NPs	45.5
MAGAT + Methylene Blue	25.9
MAGAT + Methylene Blue + ZnO NPs	41.7
Muscle <sup>(19)</sup>	+35 to +55
Kidney <sup>(19)</sup>	+20 to +45
Brain, grey matter <sup>(20)</sup>	35 to 60
Brain, white matter <sup>(20)</sup>	25 to 38
Water <sup>(21)</sup>	0

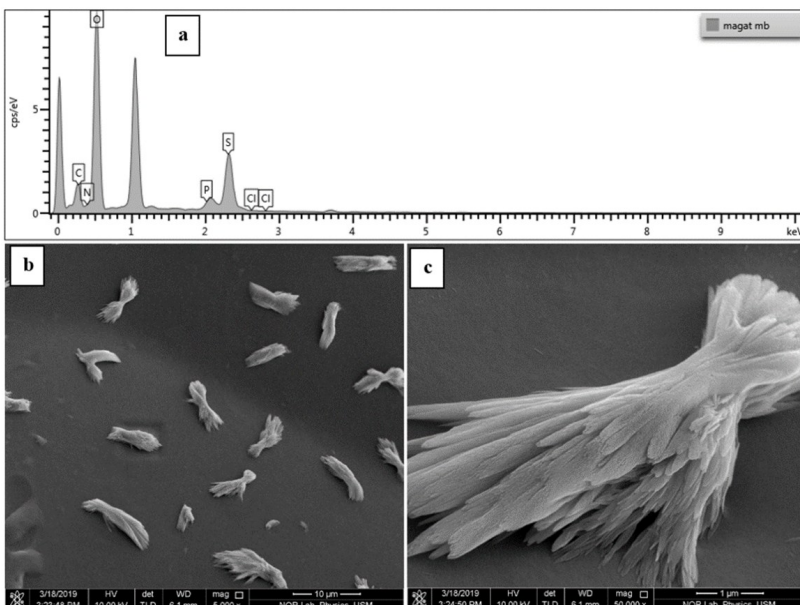
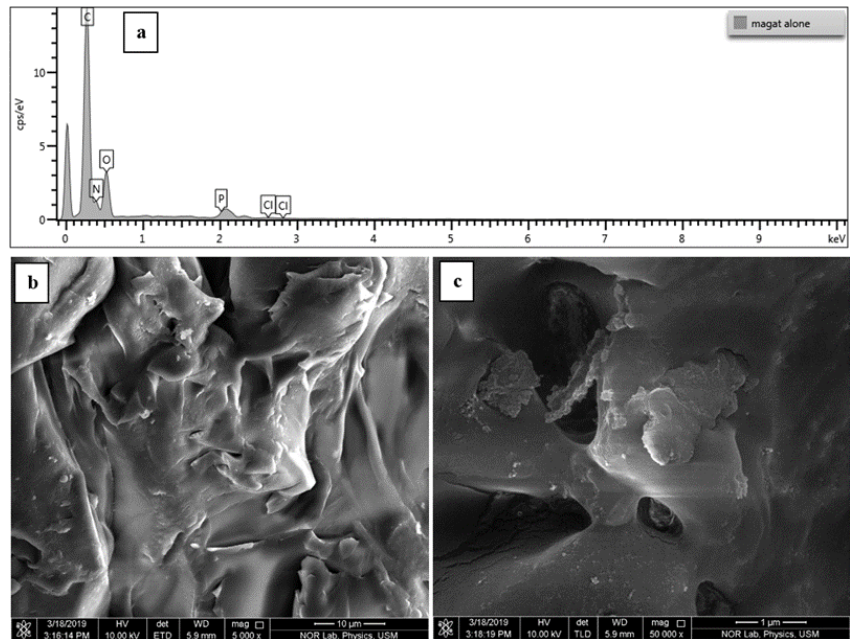
**Morphological structure of different types of MAGAT gel dosimeter**

Electron microscopic (FESEM) micrographs and elemental data (EDX) of all four MAGAT gel dosimeters are shown in figures 1 through 4. The EDX data in figure 1 (a) shows that MAGAT gel dosimeter contained elements of C, N, O, P, and Cl which are familiar elements in MAGAT while the FESEM micrographs in figure 1 (b) and 1 (c) show its wavy surface (with hills and valleys) that is ordinary for a cut surface of MAGAT. The EDX data in figure 2 (a) shows that the MAGAT-added ZnO specimen comprises of extra peaks for Zn that verify the presence of Zn in the formulation. The EDX of figure 3 (a) shows the presence of S which is an element of methylene blue that is known to absorb gamma radiation that steers to its degradation.

The fascinating morphological structure can be

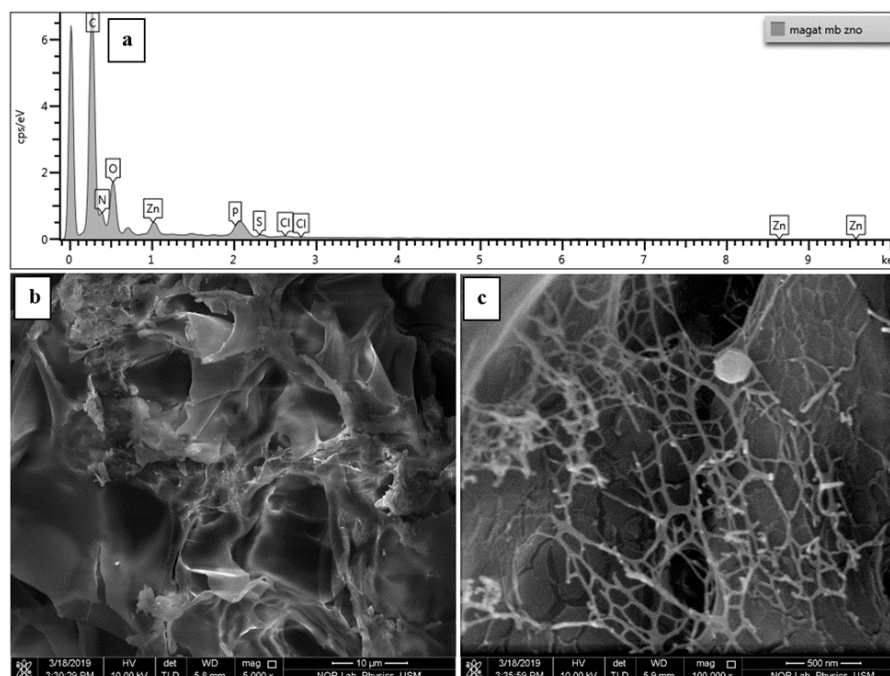
seen in the FESEM micrographs of figure 3 (b) and 3 (c) where a shape resembling tree trunk fibers with diameters of 40-500 nm and lengths of 2-10 μm were present in the MAGAT added with methylene blue sample. Figure 4 (a) shows the presence of Zn, O, and S in the EDX data. When both methylene blue and zinc oxide were added in MAGAT formulation, a flake-like morphology was produced as shown in the FESEM micrograph in figure 4 (b) and when magnified to 100000x, an intriguing spider-web morphology was revealed as shown in figure 4 (c). As the ZnO NPs used in this study were made of rod shape, an interesting morphological rod-like structures were apparent in the FESEM micrographs of figure 2 (b) and 2 (c) which may suggest that Zn content in MAGAT could have formed the ZnO rod crystals with the diameter of 50-3000 nm and length of 1-12 μm during the fabrication process.

**Figure 2.** FESEM-EDX micrographs of MAGAT added with ZnO (a) EDX data shows the presence of Zn and O elements, and the micrographs at (b) 5000x, and (c) 50000x.



**Figure 3.** FESEM-EDX micrographs of MAGAT added with methylene blue (a) EDX data, and the FESEM micrographs at (b) 5000x, and (c) 50000x.

**Figure 4.** FESEM-EDX micrographs of MAGAT added with methylene blue and ZnO (a) EDX data, and the FESEM micrographs at (b) 5000× and (c) 10000×.



## DISCUSSION

The radiological properties tabulated in table 2 and 3 demonstrate a good agreement between all MAGAT gel dosimeter formulations with those of water/soft tissue. The most significant property to consider when utilising a dosimeter in megavoltage X-ray or electron beams is radiological water equivalence<sup>(22)</sup> due to the prevalence of the Compton Effect across the therapeutic energy range<sup>(23)</sup>. Therefore, the equality of different formulations of MAGAT gel dosimeter in terms of mass density, CT number, and attenuation coefficient with water/soft tissue obtained within this study is vital to dosimetric measurement as the addition of different additives might modify the MAGAT gel dosimeter radiation absorption and scattering properties. Hence, all of the MAGAT polymer gel dosimeters formulations investigated are suitable for dosimetry purposes in the therapeutic energy range.

When developing normoxic polymer gel dosimeters, the mass density should ideally be as close to unity as possible. Table 2 shows the mass density of all MAGAT gel formulations differed by up to 2.5% from that of water and are very close to that of muscle tissue. This resulted in differences with water up to 2.5% through the Compton dominant energy range for absolute attenuation, energy absorption, and collision stopping power coefficient ratios<sup>(24)</sup>. This is due to the high gelatin (6%) and monomer concentration (6%) employed in the MAGAT gel formulation. As indicated by Ashikin *et al.*,<sup>(14)</sup> and Mahbod *et al.*,<sup>(25)</sup> these concentrations are utilised as an optimum concentration where they are able to capture and store larger radiation dose information while producing the highest sensitivity of dose-response upon irradiation.

Table 2 shows that the attenuation coefficient of four MAGAT gel dosimeter formulations differ significantly. In comparison to the other MAGATs formulation, the MAGAT doped with methylene blue and ZnO NPs demonstrated the greatest increase in linear attenuation value. The importance of this increment is that, when the polymer gel is irradiated, more energy will be deposited due to higher generations of photoelectrons resulted from the presence of ZnO NPs in the polymer gel which may lead to an increase in gel sensitivity. The sensitivity enhancement in the polymer gel dosimeter is essential to reduce a certain amount of absorbed dose to the patient while also being able to reduce random errors in radiotherapy treatment<sup>(11)</sup>. These results are in good agreement with those reported by Mustaqim *et al.*,<sup>(26)</sup> and Banaee *et al.*,<sup>(27)</sup> which showed that when the ZnO was added into the polymer gel formulation, the polymer gel's sensitivity increased. This can be explained by the attenuation and absorption data attained from Hubbell's physical data<sup>(28)</sup>, where the value of attenuation coefficient for Zn is noted to be higher than the value for water which signifies that the impact of the high atomic number of Zn corresponds to higher attenuation properties of radiation within the MAGAT gel dosimeter.

At present, the radiological properties of polymer gel dosimeters are mainly determined on a macro-scale<sup>(29)</sup>. Therefore, there is still an incomplete understanding of the absorption mechanism prior to irradiation in polymer gel dosimeter. In this work, our study also focuses on elucidating the micro-network structure of the MAGAT gel dosimeter. Several significant features of polymer gel reveal that they are associated to the attenuation properties of various MAGAT gel

formulation as mentioned previously.

As shown in figure 1 to figure 4, the SEM micrograph show the gel's intrinsic matrix structures which indicate that different additives led to diverse structural networks of the MAGAT gel dosimeter. The MAGAT gel doped with ZnO & methylene blue (shown in figure 4 (c)) indicates a fascinating morphological structure with the presence of a large surface area of the nano spider-web which possibly have enhanced the gel sensitivity, and later increase both linear and mass attenuation coefficients (as indicate in table 2). The presence of Zn, O, and S in the EDX data of figure 4 (a) might suggest that the nano spider-web (30-50 nm diameter range) could probably be a complex compound of ZnO and methylene blue which lead to the idea on this novel finding. This is also probably due to the competition between the gelation process, the cross-linking, and rearrangement processes<sup>(30)</sup> which occur with different MAGAT compounds. Such new knowledge can be useful in improving the polymer gel dosimetric performance for better accuracy and sensitivity of the radiotherapy dose verification system. This can later offer a solution to minimize the dose errors in the patients, especially in the therapeutic dose range.

## CONCLUSION

The four MAGAT formulations exhibited radiological properties similar to water or soft tissue materials. The discovery of novel morphological structures may lead to future works in understanding the role of these morphologies in playing a fundamental role in polymer gel attenuation. The addition of ZnO nanoparticles with methylene blue confirming increment of attenuation shown that the capability of this formulation to record better dose information.

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