Effect of Particle Size, Filler Loadings and X-ray tube voltage on the Transmitted X-ray Beam Intensity by Tungsten Oxide – Epoxy Composites

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Abstract

The effect of particle size, filler loadings and x-ray tube voltage on the transmitted x-ray beam intensity by WO_3-epoxy composites has been investigated using the mammography unit and a general radiography unit. Results indicate that nano-sized WO_3 has a better ability to attenuate lower x-ray energies (22 – 35 kV) when compared to micro-sized WO_3 of the same filler loading. However, the role of particle size on transmitted x-ray beam intensity was negligible at the higher x-ray energy range (40 – 120 kV).

Keywords: micro-sized WO_3-epoxy composites, nano-sized WO_3-epoxy composites, x-ray attenuation, filler loading.

1. Introduction

Use of nano-particles in designing advanced materials have attracted much attraction amongst the researchers because of superior physical and mechanical properties that can be achieved. For instance, recent studies have shown that nano-sized filler reinforced polymer composites provided much improvement in chemical, physical and mechanical properties by virtue of better dispersion of the nano-particles within the polymer matrix (Chapman and Mulvaney, 2001; Karim et al., 2002; Ohno et al., 2002; Schmidt and Malwitz, 2003). The main improvement of nano-particles as a filler assembly over conventional materials is the maximization of the surface/volume ratio of the fillers (Fabiani et al., 2010). For example, nano-particles improved the electrochemical capacitance of α-$Ni(OH)_2$ in alkali solutions as compared to micro-sized particles of the same hydroxide due to the greater surface/volume ratio of the nano-particles (Jayalakshmi et al., 2006).

Additionally, this size-effect also becomes one of the virtues in designing materials for shielding of ionizing radiations. Some x-ray technologists believe that this effect will improve the x-ray attenuation ability of the composite since nano-sized fillers are able to disperse more uniformly within the polymer matrix with less agglomeration as compared to micro-sized fillers (Botelho et al., 2011; El Haber and Froyer, 2008; Steinhart, 2004). A recent study by Botelho et al. (2011) found that nanostructured copper oxide (CuO) is more effective in attenuation of lower x-ray beam energy (26 and 30kV) and no significant variation in x-ray attenuation at higher x-ray beam energy 60 and 102 kV). Kunzel et al. (2012) also provided similar results, which show that the x-ray absorption is higher for a nanostructured CuO compound compared to the microstructured counterpart for low energy x-ray beams (25 and 30 kV) for all CuO concentrations incorporated into polymeric resin.

In general, the attenuation of photons (x-rays/gamma-rays) is dependent upon three factors: density, elemental composition, and thickness of the absorbing material (Sprawls, 1993). Even though lead (Pb) or lead compounds provide good x-ray shielding properties, their usage as the shielding materials has increasingly become a sensitive issue due to its hazardous nature. As a result, much research has been focusing on developing new x-ray shielding materials which are safer and easier to handle (Robert, 2005; Spinks and Wood, 1976).

The aim of this work was to develop new x-ray shielding materials based on tungsten oxide-epoxy composites with either nano-sized or micro-sized fillers. The effect of filler size on the x-ray attenuation in the diagnostic imaging energy range (22 – 120 kV) has been investigated. The filler size effect in x-ray shielding ability is discussed.

2. Experimental Procedure

2.1. Samples preparation

Nano-sized (<100 nm) and micro-sized (~20 µm) tungsten oxide (WO_3) were used as filler for synthesizing WO_3-epoxy composites. The former were obtained from Sigma-Aldrich and the latter (FR251) from Fibreglass and Resin Sales.

To prepare WO_3-epoxy composite samples, WO_3 powder was added into the FR251 epoxy resin (Bisphenol-A diglycidyl ether polymer) before the FR251 hardener (Isophoronediamine) was mixed into it. The ratio of epoxy resin to hardener used was 2:1. The mixing of WO_3 powder in epoxy resin was done through gentle stirring using a stirring machine at constant speed for 15 minutes to ensure uniform dispersion of the powder in epoxy matrix. The well-mixed mixture was then cast in a 4 cm x 6 cm rectangular silicon rubber mould with a thickness of 7 mm and was allowed to set overnight at room temperature. The list of prepared samples with different weight percentages of WO_3 are shown in Table 1.

2.2. Measurement of x-ray mass attenuation coefficient

This work was done using two different sets of equipment. For the lower x-ray energy range, a mammography unit (brand: Siemens AG, model: 2403951-4 G.E Health Care) was used while a general diagnostic x-ray machine (brand: Shimadzu, model: Circlex 0.6/1.2 P364DK-100SF) was used for the higher x-ray energy range. The initial x-ray intensity (I_o) was determined by directly measuring x-rays with the DIADOS diagnostic detector connected to DIADOS diagnostic dosimeter (PTW-Freiburg, Germany) in the absence of the sample. The dosimeter is a universal dosimeter for measuring simultaneous dose and dose rate for radiography, fluoroscopy, mammography, dental X-ray and CT with a sensitivity of 0.01 microRoentgen (µR). Meanwhile the transmitted x-ray intensity (I) was taken with the sample placed on the detector. The distance between the x-rays' tube and the detector was set to 86 cm since this is the maximum distance that can be adjusted for the mammography unit, and the x-ray beam was well collimated to the size of the sample.

The exposure was set at 10 mAs to obtain meaningful readings for this type of detector. The range 22 – 49 kV of x-ray tube voltage was selected from mammography unit since the machine can go only within this range for lower x-ray energy. On the other hand, the range of x-ray tube voltage (40 – 120 kV) was chosen from the general diagnostic x-ray machine because this range is the normal range of x-ray tube voltage used in general diagnostic imaging purposes.

The transmitted x-ray beam intensity (*I*) for each sample at each x-ray tube voltage was determined directly from the dosimeter reading. For each composite, the measurements were performed three times to obtain an average value. The performance of micro-sized and nano-sized WO_3-epoxy composite was compared from the graph of *I* as a function of filler loading for each x-ray tube voltage.

3. Results and discussion

Three different anode/filter combinations (Table 2) were used for low energy range x-rays transmitted from the mammography machine, since the combination was operated by the machine itself. The x-ray beams generated by these anode/filter combinations composed mainly of the characteristic x-ray energies of molybdenum (17.5 keV and 19.6 keV) or rhodium (20.2 keV and 22.7 keV). For WO_3 of 5 wt% and 10 wt%, the exit dose reading was observed at 22 kV x-ray energy while for 20 wt% - 35 wt%, the dose reading was initiated at 30 kV x-ray energy due to the zero reading of the dosimeter at 22 kV and 25 kV x-ray tube voltages.

Results in Fig. 1(a-c) clearly show a big difference in the transmitted x-ray beam intensity between the micro-sized WO_3-epoxy composite and the nano-sized WO_3-epoxy composite at 22 kV – 35 kV generated by the mammography unit at the same filler loading. The ratio of transmitted x-ray beam intensity for the micro-sized WO_3-epoxy composite (I_m) to transmitted x-ray beam intensity for the nano-sized WO_3-epoxy composite (I_n) was in the range 1.3 – 3.0. The ratio (I_m/I_n) was larger at these energy ranges (22 – 35 kV) and was decreasing as the x-ray energy increased ($\geq$ 40 kV) as can be seen in Fig 1(d, e). In contrast, the ratio (I_m/I_n) is ≈ 1.0, for the diagnostic x-ray energy range (40 kV -120 kV) generated by the general radiography unit, which suggests $I_m \approx I_n$. Thus, the size effect of WO_3 particles was negligible at the higher energy range (Fig 2).

Besides that, Fig.3 showed that even though the x-ray energy selected from the mammography unit, and general radiography unit is same (40 kV), the value of *I* transmitted by the nano-sized WO_3 epoxy composite is higher for the x-ray photon generated by the mammography unit. A similar result was also obtained for micro-sized WO_3 epoxy composite. This outcome proved that the general radiography unit comprised of a continuous spectrum of x-rays energy, which is having lower equivalent energies as compared to the characteristic x-ray energies produced from the mammography unit (17.5 keV - 22.7 keV).

Generally, photoelectric absorption dominates at lower photon (x-ray) energies range. A photon is completely absorbed by the atom of the material, and a photoelectron is ejected in the process. The ejected photoelectrons may undergo single- or multiple-scattering events with neighboring atoms, which can alter the mass attenuation coefficient of an element relative to the bulk material when considered over a small range of x-ray energies together with slight fluctuations in the probability of emission of Auger electron and fluorescent photons. The probability of the photoelectric interaction is approximately dependent on Z^3/E^3 where Z is atomic number of the absorbing material and E is the photon energy. Furthermore, the number of W particles/gram in the nano-sized WO_3-epoxy composite is higher than that for the micro-sized

WO_3-epoxy composite. Thus, the probability of an x-ray with lower energy to interact and to be absorbed may be higher for nano-sized WO_3-epoxy composite rather than micro-sized WO_3-epoxy composite.

As photon energy increases, the probability of Compton scattering increases and hence the attenuation by the material decreases since this interaction was weakly dependent on Z and E. Hence, the probability of an x-ray photon with higher energy to interact and to be absorbed may be similar for nano-sized and micro-sized WO_3-epoxy composite.

To support our attenuation results for low x-ray photon energies, we have repeated the procedure to measure transmitted x-ray beam intensity of a WO_3 compact disc of the same mass for 20 wt% (69% porosity) and 35 wt% (66% porosity) of WO_3 loading in the epoxy composite sample. The results in Fig.4 (a, b) clearly show that the WO_3 compact disc has the lowest *I* when compared to both micro-sized and nano-sized WO_3-epoxy composite of the same mass since WO_3 particles have been compressed closer together instead of being dispersed in the epoxy matrix.

Hence, the results obtained in this work are in good agreement with the work of Kunzel et al.(2012) which showed that nano-sized fillers are superior to micro-sized fillers for attenuating lower x-ray energy but for the higher energy range, the effect is the same (Künzel and Okuno, 2012).

4. Conclusions

The results of this work showed that for the same WO_3 loading, nano-sized WO_3-epoxy composite has better attenuating ability in the lower x-ray energy range (22 – 35 kV) when compared to the micro-sized WO_3-epoxy composite. However, the role of particle size in x-ray shielding was insignificant at the higher x-ray energy range (40 – 120 kV).

References

Botelho, M.Z., Künzel, R., Okuno, E., Levenhagen, R.S., Basegio, T., Bergmann, C.P., 2011. X-ray transmission through nanostructured and microstructured CuO materials. Applied Radiation and Isotopes 69, 527-530.

Chapman, R., Mulvaney, P., 2001. Electro-optical shifts in silver nanoparticle films. Chemical Physics Letters 349, 358-362.

El Haber, F., Froyer, G., 2008. Transparent polymers embedding nanoparticles for x-rays attenuation (Review). Journal of the University of Chemical Technology and Metallurgy 43, 283-290.

Fabiani, D., Montanari, G.C., Krivda, A., Schmidt, L.E., Hollertz, R., 2010. Epoxy based materials containing micro and nano sized fillers for improved electrical characteristics, 2010 International Conference on Solid Dielectrics. IEEE, Potsdam, Germany, pp. 1-4.

Jayalakshmi, M., Mohan Rao, M., Kim, K.B., 2006. Effect of particle size on the electrochemical capacitance of a-Ni$(OH)_2$ in alkali solutions. International Journal of Electrochemical Science, 324-333.

Karim, A., Amis, E., Yurekli, K., Krishnamoorti, R., Meredith, C., 2002. Combinatorial methods for polymer materials science: Phase behavior of nanocomposite blend films. Polymer Engineering and Science 42, 1836-1840.

Künzel, R., Okuno, E., 2012. Effects of the particle sizes and concentrations on the X-ray absorption by CuO compounds. Applied Radiation and Isotopes 70, 781-784.

Ohno, K., Koh, K.M., Tsujii, Y., Fukuda, T., 2002. Synthesis of gold nanoparticles coated with well-defined, high-density polymer brushes by surface-initiated living radical polymerization. Macromolecules 35, 8989-8993.

Robert, R.D., 2005. High density composites replace lead. Ecomass Technologies.

Schmidt, G., Malwitz, M.M., 2003. Properties of polymer-nanoparticle composites. Current Opinion in Colloid and Interface Science 8, 103-108.

Spinks, J.W.T., Wood, R.J., 1976. An introduction to radiation chemistry, 2nd edition ed. Wiley-Interscience, New York.

Sprawls, P., 1993. The Physical Principles of Medical Imaging, 2nd edition ed. Aspen Publishers, Gaithersburg, Md.

Steinhart, M., 2004. Introduction to Nanotechnology. By Charles P. Poole, Jr. and Frank J. Owens. Angewandte Chemie International Edition 43, 2196-2197.

Table 1

List of prepared samples with different weight fractions of filler (WO_3) and epoxy resin.

Composite by weight fraction (wt %)	
Filler (WO_3)	Epoxy resin
5	95
10	90
20	80
30	70
35	65

Table 2

Anode/filter combination operated by mammography machine.

X-ray tube voltage (kV)	Anode/filter combination
22	Mo/Mo[1]
25	
30	
35	Mo/Rh[2]
40	
45	Rh/Rh[3]
49	

[1] Mo/Mo stands for molybdenum anode/molybdenum filter, [2] Mo/Rh stands for molybdenum anode/rhodium filter, and [3] Rh/Rh stands for rhodium anode/rhodium filter.

Figure captions:

1. Transmitted x-rays beam intensity as a function of filler loading of the nano-sized WO_3-epoxy composite as compared to the micro-sized WO_3-epoxy composite for x-ray energy of (a) 22 kV (5 – 10 wt%); (b) 35 kV (5 – 10 wt%); (c) 35 kV (20 – 35 wt%); (d) 49 kV (5 – 10 wt%); and (e) 49 kV (20 – 35 wt%) generated by mammography unit.
2. Transmitted x-rays beam intensity as a function of filler loading of the nano-sized WO_3-epoxy composite as compared to the micro-sized WO_3-epoxy composite for (a) 40 – 60 kV; and (b) 70 – 120 kV generated by general radiography unit.
3. Comparison of transmitted x-rays beam intensity values for all nano-sized WO_3 wt% loading in epoxy sample for 40 kV x-ray energy used by the mammography unit and the general radiography unit.
4. Transmitted x-rays beam intensity for WO_3 compact disc as compared to both micro-sized and nano-sized WO_3-epoxy composite of the same mass of (a) 20 wt% and (b) 35 wt% of WO_3 loading in epoxy sample at x-ray energy range (30 – 49 kV) generated by mammography unit.