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Characterization of the radiosensitization effect of pulsed laser ablated-gadolinium

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Abstract. Nanomedicine is a field of medical treatment research that has developed rapidly in recent years for the diagnosis and treatment of cancer. New strategies in nanomedicine are to improve cancer diagnosis and therapy at the cellular and molecular level. Materials that are often used for nanomedicine are metal-based gadolinium. This research presents a preliminary study of the radiosensitization properties of gadolinium nanoparticles colloid. Gadolinium nanoparticle colloid was synthesized using a pulsed laser ablation method in pure water. The gadolinium nanoparticle colloid was characterized using a Scanning Electron Microscope and UV-Vis Spectrophotometer. Radiosensitization properties of gadolinium nanoparticles colloid were examined based on the decomposition of x-ray irradiated methylene blue. The results show that gadolinium nanoparticle colloid consists of a spherical shape particle and has a bandgap energy of 5.76 eV. Gadolinium nanoparticle colloid possessed radiosensitization properties with a dose enhancement factor in the range of 1.51-1.54.

1. Introduction

In recent years nanotechnology-based medicines are developing so rapidly [1]. Nanomedicine is defined as nanoparticle-based medicines with particle sizes of less than 1000 nm [2]. In nanomedicine, metal-based nanoparticle material is very important because of its excellent physical, optical, magnetic, electrical and nuclear properties that can be applied to various fields. Gadolinium is one of the metals that have the potential to be developed [3]. Gadolinium has atomic numbers higher than iodine (Z = 64 and Z = 53, respectively) and has an x-ray attenuation coefficient of 2.5 times of iodine [4]. Consequently, gadolinium has potency as a radiosensitizer. That property is correlated to photoelectric effect and Auger electron production phenomena which lead to an increase in reactive oxygen species (ROS) formation and radiation absorption in cancer cells [5].

Several methods have been used for the synthesis of gadolinium nanoparticle such as precipitation [6], sol-gel [7], polyol method [8]. These methods have the disadvantage of complex preparations, low purity products which lead to less suitable for nanomedicine applications. The pulsed laser ablation is a more suitable method to prepare gadolinium nanoparticles colloid for nanomedicine applications. In this method, the metastable, pure and biocompatible colloid can be obtained by ablating high purity metal targets in liquid medium [9]. In this study, the gadolinium nanoparticle colloid was prepared by using a pulsed laser ablation method in pure water. The obtained colloid was characterized by SEM and UV Vis Spectrophotometry. The radiosensitization properties of gadolinium nanoparticle colloids were investigated in conjunction with the kilovoltage energy of radiotherapy.

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2. Method

Gadolinium nanoparticle colloid was synthesized by pulsed laser ablation method in pure water. The laser with of Nd: YAG (λ = 1064 nm), the energy of 30 mJ and a pulse width of 7 ns was used as radiation ablation. High purity gadolinium (Nilaco, 99.999%) target was immersed in 20 mL pure water. The focused pulsed laser was shot to a metal target for 180 minutes with a repetition rate of 10 Hz. The radiosensitization properties were examined by mixing gadolinium nanoparticles colloid with methylene blue solution as previously reported in the literature [10]. The mixed solution was irradiated by x-ray energy of 60 kV, 80 kV, and 100 kV. The radiation exposure experiment was carried out by setting time of exposure was 0.1 second and SSD was 100 cm. The absorbance of the methylene blue solution was measured using a UV Vis spectrophotometer at a wavelength of 664 nm for further analysis.

3. Results and Discussion

Scanning Electron Microscopy (SEM) was used to analyze size and shape particles. Figure 1 shows the SEM image of the gadolinium nanoparticle colloid. This image provides information on the presence of spherical shape particles with a size of several hundred nanometers.

Figure 1. SEM image of gadolinium nanoparticles colloid synthesized by Pulsed Laser Ablation

The formation of the gadolinium particle was confirmed by UV Vis absorbance spectrum measurement. The peak of UV-Visible absorbance was observed at wavelengths of 200 - 300 nm indicating good UV light absorption properties Furthermore, based on that UV-Vis spectrum the bandgap energy was estimated using Tauch's plot of $(\alpha h u)^2$ vs the hu as shown in the inset in figure 2. The obtained band gap energy was 5.76 eV which closed to bandgap energy of Gd(OH)₃ [11]. This result indicates that the ablation of the gadolinium target in water produced Gd(OH)₃ phase.

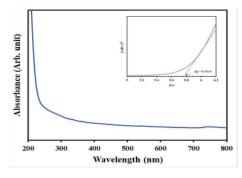


Figure 2. UV-Vis absorption spectrum and bandgap energy of gadolinium colloid

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Figure 3 shows the absorbance of un-irradiated methylene blue (MB), MB and mixed MB/gadolinium nanoparticles colloid irradiated by kilovolt x-ray at the energy of 60, 80 and 100 kV. It is observed a decrease in absorbance of MB solution and mixed MB/gadolinium nanoparticles colloid after X-ray radiation. The addition of gadolinium nanoparticles colloid into MB solution resulted in a higher decrease in absorbance. However, the energy variation does not significantly influence to decrease in MB absorbance. The decrease in absorbance of MB is due to direct interaction between x-ray and MB molecule and x-ray and water or radiolysis. Meanwhile, the decrease in absorbance of mixed MB/gadolinium nanoparticles colloid is correlated to direct interaction between x-ray and MB molecule, radiolysis of water and x-ray and gadolinium. That interaction resulted in radical oxygen species like O², OH and H which are decomposed MB and reduce MB concentration in the solution. Besides, the interaction of x-ray with high Z material of gadolinium generates secondary x-ray, photoelectron, and Auger electron, increasing the decomposition of MB. This research shows the radiosensitization of gadolinium nanoparticles colloid [10]. The percentage of MB decomposition is shown in Figure 4. The percentage of decomposition of MB solution without and with the presence of gadolinium nanoparticles colloid are 54% and 70 % respectively. This indicates an increase in the generation of radical oxygen species.

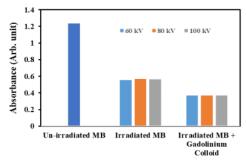


Figure 3.The absorbance of MB and MB gadolinium colloid after exposure X-ray radiation

Figure 4. Percentage of MB decomposition without and addition gadolinium colloid

The radiosensitization properties characterized by the dose enhancement factor (DEF) value of higher than one. In this research, the radiosensitization properties of gadolinium nanoparticles colloid were estimated by determining DEF using the ratio of absorbance irradiated MB to the absorbance of mixed MB/gadolinium nanoparticles colloid. Figure 5 shown DEF of gadolinium nanoparticles colloid for X-ray energy of 60 kV, 80 kV, and 100 kV. The DEF of gadolinium nanoparticles colloid was found to be 1.51-1.54 which higher than 1. Although the energy variation does not significantly influence to increase in DEF, this result suggested that gadolinium nanoparticles colloid possesses radiosensitization effect. A relatively large DEF was observed for x-rays with 80 kV energy. The result shows that gadolinium nanoparticles colloid is potential as a radiosensitizer in kilovoltage x-ray.

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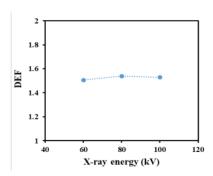


Figure 5. A dose enhancement factor of gadolinium nanoparticles colloid to X-ray radiation

4. Conclusion

Gadolinium nanoparticles colloid has been synthesized by using pulsed laser ablation of the gadolinium target in pure water. SEM image shows that gadolinium nanoparticle colloid consists of a submicron size particle. The bandgap energy of 5.76 eV indicates the formation of the Gd(OH)₃ phase. The DEF value of 1.51-1.54 allows it gadolinium nanoparticle colloid used as a radiosensitizer.

Acknowledgment

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