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Synthesis of Gadolinium Nanoparticles as a CT-Scan Contrast Agent with Pulse Laser Ablation Method

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ABSTRACT

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The use of iodine contrast agents on CT Scan clinically shows a short-term blood circulation, non-specific biodistribution and causes side effects on kidneys. Nanoparticles have a longer half-time vascular than molecular contrast agents so it can be observed for a longer time after injection. Gadolinium (Z = 64) has a higher atomic number and X-ray absorbance coefficient than iodine (Z = 53) and does not have the negative effect on kidneys. The Gd nanoparticles development as a CT Scan contrast agent has potential to give more effectiveness than iodine contrast agents. In this study, Gd nanoparticles were synthesized using pulsed laser ablation method with wavelength 1064 nm, energy 45 mJ, and pulse width 7 ns. The ablation process was carried out for 180 minutes with repetition rate of 10 Hz and 15 Hz. The formation of Gd Nanoparticles was analyzed using UV-Vis spectrophotometer and FTIR (Fourier-Transform Infrared Spectroscopy). Testing the ability of Gd nanoparticles as a contrast agent was done in the diagnosis of head and abdomen using a CT Scan GE CT Optima 580 RT type 229156-3. UV-Vis spectrophotometer analysis showed that Gd nanoparticles had high absorbance at the wavelength less than 250 nm which indicated the formation of Gd2(OH)3 compounds. The repetition rate difference in ablation process resulted in the same concentration of Gd nanoparticles with different contrasts. Repetition rate of 10 Hz produced Gd nanoparticles with HU greater than repetition rate of 15 Hz and closer to HU of iodine. The results indicate that Gd nanoparticles can be used as a CT Scan contrast agent.

Keywords : Nanoparticles, gadolinium, contrast agent CT Scan, pulse laser ablation method

I. INTRODUCTION

Computed Tomography Scan (CT Scan) is widely used in radiodiagnostics due to its relatively lower price compared to MRI and its ability to produce efficient images. Different body tissues have different mass attenuation coefficient. In result, the X-rays that pass through the human body will form contrasts in the image. CT Scan can distinguish bone and tissue easily but it is still difficult for soft tissues such as muscles, organs, and tumors because they have nearly the same mass attenuation coefficient. As a solution, contrast agents can be used on CT scan imaging. Currently, most widely used CT scan contrast agents are iodine-based molecules. However, in clinical practice it is known that iodine has a short-term existence in blood circulation, non-specific bio-distribution, and renal toxicity [1]. Nanoparticles have the potential to provide better image contrast than iodine compounds and have a longer vascular half-life than molecular contrast agents. As a result, nanoparticle contrast agents can be monitored for a longer period of time after being injected into living organisms [2].

An alternative to iodine as a contrast agent is the use of materials containing high atomic number metals [3]. Gadolinium (Z= 64, k-edge = 50 keV) has a higher atomic number and a higher k-edge than iodine (Z = 53, k-edge = 33 keV) which is more suitable with the post-filtration spectrum intensity of the energy peak that has generated during CT scan between 50-140 kVp [4]. More than that, gadolinium has been shown to have no negative effect on the kidneys [5]. High atomic number metal nanoparticles have the character of increasing the radiation dose per unit mass [6]. Thus, it is hoped that a small concentration of nanoparticles can provide more effectiveness than the use of iodine contrast agents. Various potentials of nanoparticles and gadolinium encourage us to conduct research on the synthesis of gadolinium (Gd) nanoparticles as CT contrast agents.

The formation of metal nanoparticles in the medical field needs to pay attention to purity. High purity can reduce tissue toxicity [7]. Pulsed laser ablation method in liquid medium can produce nanoparticles with high purity. This synthesis method is carried out using laser energy that is fired at the target metal in liquid medium. The greater the laser energy used, the smaller the size of nanoparticles and the greater the repetition rate used, the larger the size of the nanoparticles produced [8]. We used a pulsed laser Nd:YAG by varying the repetition rate so that we can know its effect on the effectiveness of Gd nanoparticles as a contrast agents. The effectiveness can be seen from the value of the Hounsfield Unit (HU) from the CT Scan image.

II. METHODS AND MATERIAL

The synthesis of Gd nanoparticles was carried out using a pulsed Nd:YAG laser with a wavelength of 1064 nm, energy of 45 mJ, and pulse width of 7 ns. Gadolinium metal was placed in a 25 ml glass petri dish containing 15 ml of aquadest. The synthesis used a petri dish in order to keep the space in aquadest so that the nanoparticles formed, remained in there. The Nd:YAG laser beam was directed by a convex lens (focal length 3 cm) to scrape the gadolinium metal surface at the bottom of the petri dish. Gd nanoparticles with 0.006 mg/ml concentration were made with repetition rates of 10 Hz and 15 Hz, for 180 minutes.

The Gd nanoparticles were characterized using a UV-Vis spectrophotometer at 200 nm – 800 nm wavelength and FTIR (Fourier-Transform Infrared Spectroscopy) to find out the functional groups. The next step was testing the Gd nanoparticles as a CT Scan contrast agents. At this stage, there were 3 samples, namely nanoparticles Gd 0.006 mg/ml 10 Hz, nanoparticles Gd 0.006 mg/ml 15 Hz, and iodine solution 37 mg/ml for comparison. The test was carried out by placing each sample into a 2 ml vial tube and arranged in the CT Scan's target field of GE CT Optima 580 RT type 229156-3. Scans were performed for head and abdomen diagnosis, then the images obtained were processed using RadiAnt DICOM Viewer software to determine the HU value in each sample with head and abdomen diagnosis.

III.RESULTS AND DISCUSSION

Laser ablation method could synthesize colloidal Gd nanoparticles from pure Gd plates using an Nd:YAG laser (1064 nm, 40 mJ, 7ns) in aquadest. From the synthesis that carried out, it was known that colloidal Gd nanoparticles formed had cloudy white appearance after 180 minutes of ablation. The Gd nanoparticles were analyzed using Uv-Vis and FTIR. The absorbance spectrum of Gd nanoparticles from UV-Vis analysis is shown in Fig. 1. It can be seen that Gd nanoparticles produced high absorption at a wavelength of less than 250 nm. The graph shows that the higher the repetition rate, the higher the absorbance of Gd nanoparticles.

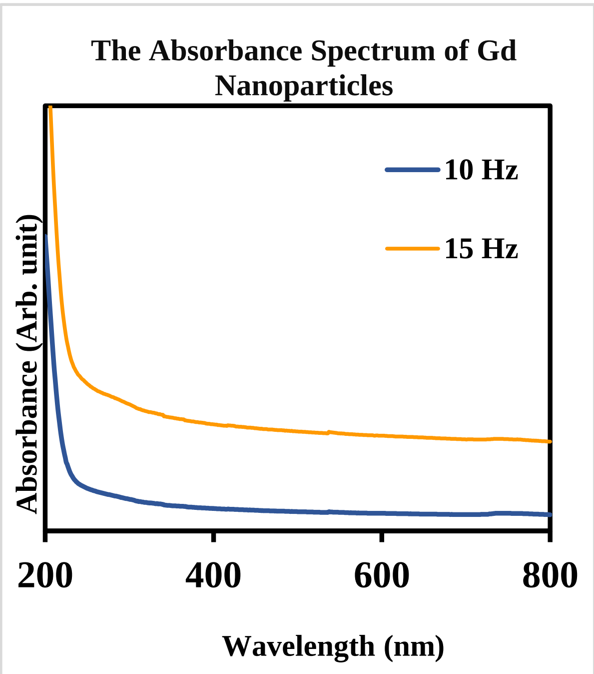
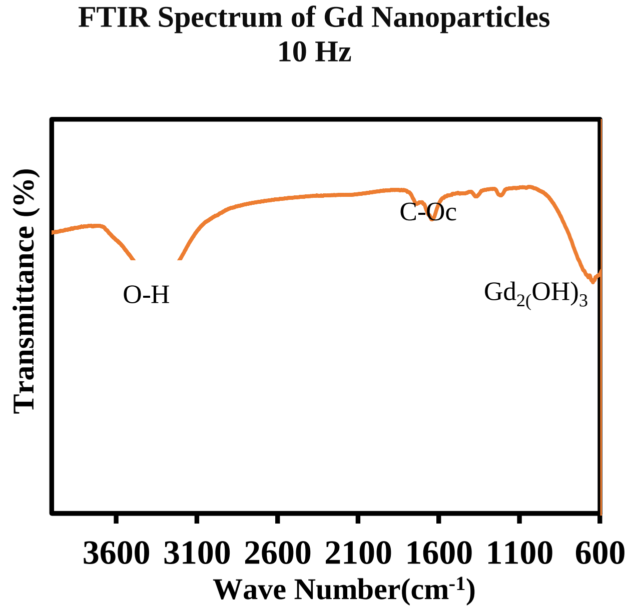
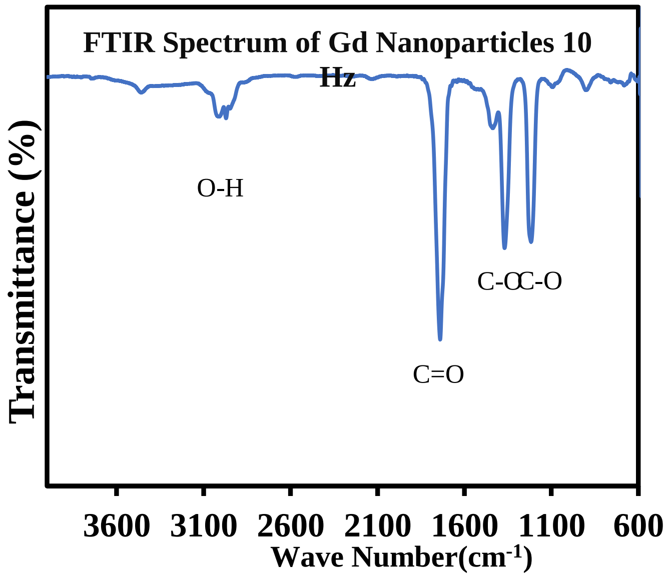


Figure 1. The Absorbance Spectrum of Gd Nanoparticles from Pulsed Laser Ablation Method in Aquadest.

Gadolinium compounds in Gd nanoparticles were analyzed by FTIR. The FTIR spectrum of Gd nanoparticles in aquadest with a repetition rate of 10 Hz and 15 Hz is shown in Fig. 2 (a) and 2 (b).



(a)



(b)

Figure 2. FTIR Spectrum of Gd Nanoparticles in Aquadest Repetition Rate (a) 10 Hz dan (b) 15 Hz

Figure 2 (a) above shows several absorption peaks, namely 643 cm^{-1}, 1648 cm^{-1}, and 3403 cm^{-1}. The absorption peak of 643 cm^{-1} indicated the presence of colloidal Gd nanoparticles. At a wave number of 1648

cm^{-1}, a functional group of C-O bonds was observed because the pH of Gd nanoparticles was 5 (acid) [9], while the peak widen at 3403 cm^{-1} it indicated the presence of H-O bonds from the H_2O molecule [10].

In Figure 2 (b) there are 4 successive peaks at the wave number of 3012 cm^{-1}; 1739,75 cm^{-1}; 1369 cm^{-1}; and 1216,75 cm^{-1}. The peak of 3012 cm^{-1} indicates the presence of O-H functional group. Wave number 1739,75 cm^{-1} shows that there is a C=O bond. C-O bonds were detected 1369 cm^{-1}, dan 1216,75 cm^{-1}. In aquadest, Gd nanoparticles tend to form $Gd_2(OH)_3$ compounds due to the presence of Gd-O-H vibration bonds [11]. When compared between Fig. 2 (a) and 2 (b) it can be seen that the 10 Hz Gd nanoparticles have fewer carbon bonds. This indicated that the purity of Gd nanoparticles 10 Hz was higher than 15 Hz.

HU values in CT Scan images were analyzed using RadiAnt Dicom Viewer software. Table 1 shows the HU values of the three samples in the head and abdomen diagnosis. It is known that the HU of Gd nanoparticles 10 Hz has a higher value than 15 Hz.

Table 1. HU Value of Gd Nanoparticles and Iodine

No	Sample	HU	
		Head	Abdomen
1.	Gd 10 Hz	26	14
2.	Gd 15 Hz	24	5
3.	Iodine	33	11

Between the two Gd samples, the HU nanoparticles Gd 10 Hz had a value close to iodine. It is known with a smaller repetition rate (result size of greater nanoparticles) can produce Gd nanoparticles with HU value close to iodine. This shows that a small Gd concentration value of 0.006 is able to have HU value that is close to iodine 37 mg/ml. This is because Gd has a higher X-ray absorbance coefficient than iodine [12].

IV. CONCLUSION

The Gd nanoparticles with high purity were successfully produced using pulsed laser ablation method in aquadest from Gd metal. Gd nanoparticles have high absorbance at wavelengths less than 250 nm. In aquadest, Gd nanoparticles tend to form $Gd_2(OH)_3$ compounds. The results showed that Gd nanoparticles with a small concentration (0.006 mg/ml) and a repetition rate of 10 Hz had a HU value close to the HU iodine value of 37 mg/ml. Thus, Gd nanoparticles can be used as contrast agents for CT scans with small concentrations.

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