

Radiosensitization Effect Of Tantalum Nanoparticles Syntesized By Pulsed Laser Ablation Method In Deionized Water

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Abstract— This study explores the potential use of tantalum nanoparticles as radiosensitizing agents to improve the efficacy of radiotherapy in targeted tissues. The nanoparticles were synthesized using the pulsed laser ablation technique in deionized water. Structural and morphological characterization was performed through Field Emission Scanning Electron Microscopy and X-ray Diffraction. To assess their radiosensitization capability, the degradation of methylene blue under exposure to X-rays and electron beams was analyzed. The results reveal that the synthesized tantalum nanoparticles are predominantly amorphous, with an average diameter of approximately 190 nm and a generally spherical shape. Furthermore, they demonstrated radiosensitizing effects, as indicated by a dose enhancement factor greater than one.

Keywords—Radiosensitizer, Nanoparticle, Tantalum, Laser Nd:YAG

I. INTRODUCTION

Nanoparticles are a noteworthy product of nanotechnology that are currently attracting significant attention (1). Nanoparticles have a small size, typically measuring less than 1000 nm (2). Metal nanoparticles possess flexible nanostructures due to their ability to regulate composition, morphology, dimensions, architecture, encapsulation, and optical characteristics during synthesis. Consequently, metal nanoparticles are extensively utilized in biomolecular detection, including cell labeling for therapeutic agent delivery, hyperthermic treatment for cancer, and as radiosensitizers to modulate mRNA expression in cells (3).

Tantalum possesses a high atomic number (Z = 73) and exhibits biocompatibility, suggesting its potential application as a radiosensitizer. High-Z metals are capable of absorbing X-ray energy and subsequently interacting with radiation within neoplastic cells. Nanoparticles can enhance the generation of reactive oxygen species (ROS) during ionizing radiation exposure through physical or catalytic mechanisms, or by facilitating the delivery of oxygen-rich substances. Physical mechanisms here refer to effects related to increased secondary electron emission and a locally enhanced physical dose. These electrons interact with and ionize oxygen-containing molecules around the nanoparticles, resulting in ROS production. Secondary electrons can also directly interact with DNA or water molecules, thereby promoting further ROS generation and enhancing tumor radiosensitivity (4).

Several methods have been utilized for the synthesis of tantalum nanoparticles, including sol-gel and precipitation approaches (5-6). These methods have the drawbacks of complex preparation and reduced purity, making them less suitable for medical applications. In this research, tantalum nanoparticles were synthesized using the PLA technique in deionized water medium. This method offers high purity, as the purity of the resulting nanoparticles primarily depends on the purity of the target material and the surrounding medium (either liquid or gas), without contamination from the reactor (7). The synthesized tantalum nanoparticles were

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characterized using FESEM and XRD spectroscopy. Their radiosensitizing properties were assessed through irradiation with a 6 MV photon and a 9 MeV electron beam..

II. EXPERIMENTAL PROCEDURE

A. Materials

The material used were tantalum, deionized water (DIW), laser Nd:YAG, methylene blue, and LINAC

B. Method to synthesis tantalum nanoparticles

Colloidal tantalum nanoparticle was synthesized using the pulsed laser ablation method in a deionized water medium. The laser used is an Nd:YAG laser ($\lambda = 1064$ nm), with an energy of 78 mJ, a pulse width of 7 nm, and a repetition rate of 10 Hz. Tantalum metal was placed in 10 mL of deionized water and irradiated with varying ablation times of 120 and 180 minutes, resulting in two colloidal tantalum nanoparticles. Figure 1 illustrates the synthesis of tantalum nanoparticles via the pulsed laser ablation method.

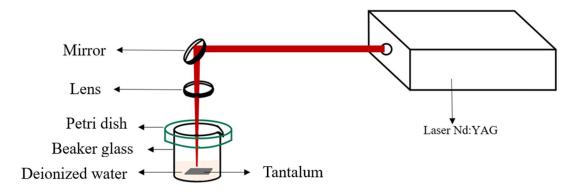
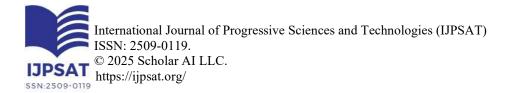


Fig. 1. Schematic illustration of tantalum nanoparticle synthesis

C. Radiosensitization Examination

The radiosensitization properties of tantalum nanoparticles was conducted by measuring the decomposition of the methylene blue (MB) solution. The synthesized tantalum nanoparticle were mixed into a 5 ml solution of 25 ppm methylene blue. The MB solution with and without tantalum nanoparticles was measured for absorbance before radiation exposure using UV-Vis spectroscopy (8). MB solutions with and without tantalum nanoparticles were exposed using photon 6 MV and 9 MeV electron beam with a dose of 2 Gy, similar to clinical therapy fractionation (9). The absorbance of the samples, both before and after irradiation, was measured using a UV-Vis spectrometer at a wavelength of 664 nm (10), and then the decomposition of MB and DEF was calculated. The schematic of the MB and mixed MB/tantalum irradiation are shown in Figure 2.

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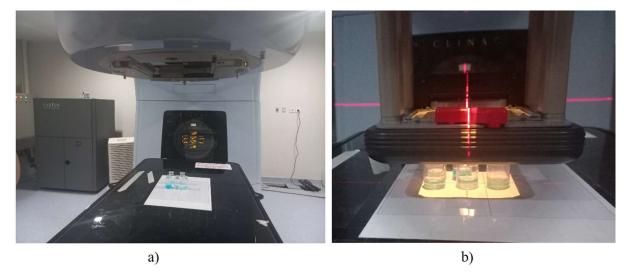


Fig. 2. Schematic of radiosensitization examination using (a) 6 MV photons and (b) 9 MeV electron beam

III. RESULT AND DISCUSSION

Tantalum nanoparticles have been synthesized using the pulsed laser ablation method. The colloid has a grayish-white color as shown in Figure 3. The increase in synthesis time makes the color of the colloid more concentrated. This is consistent with previous research conducted by Baladi and Mamoory (2010), which showed that tantalum nanoparticles have a grayish-white color (11). Therefore, tantalum nanoparticles have been successfully synthesized.



Fig. 3. tantalum nanoparticles colloid

The synthesized tantalum nanoparticle was characterized using FESEM to analyze morphology of nanoparticles. Figure 4 illustrates the FESEM image of tantalum nanoparticles together with their size distribution. Tantalum nanoparticles possess an average diameter of 190 nm and show a uniform spherical morphology. The findings align with the study of Meidanchi and Jafari (2019),

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which discovered that tantalum nanoparticles produced via the pulsed laser ablation technique show a spherical shape and uniform morphology (12).

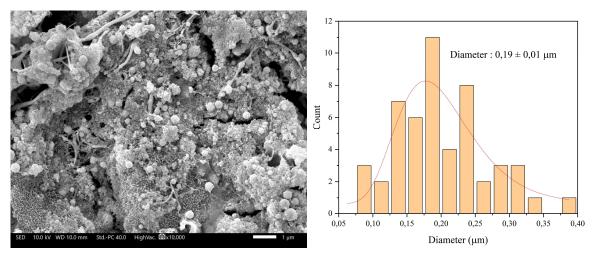


Fig. 4. FESEM image of tantalum nanoparticle colloid

The structure of tantalum nanoparticles was analyzed using XRD spectroscopy. The testing was conducted at angles from 3° to 90°. Figure 5 shows the XRD results of the formed tantalum nanoparticles. Based on the image, the tantalum nanoparticles have an amorphous structure.

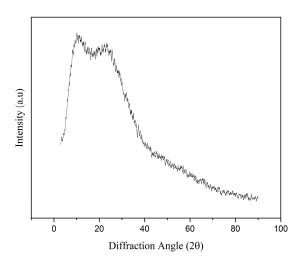


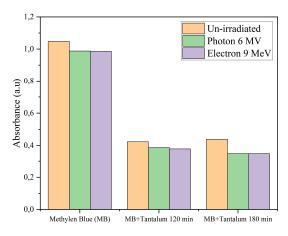
Fig. 5. XRD spectrum of tantalum nanoparticle

A preliminary investigation into the potential of tantalum nanoparticles as a radiosensitizer was conducted based on the decomposition of methylene blue (MB). The degradation of MB, with and without the addition of tantalum nanoparticles, before and after radiation exposure, was analyzed using a UV-Vis spectrometer at a wavelength of 664 nm. Figure 6 presents the absorbance spectra of MB and MB/tantalum mixtures before and after irradiation with a 6 MV photon beam and a 9 MeV electron beam generated by a linear accelerator (LINAC), with a delivered dose of 2 Gy for each beam. A reduction in absorbance was observed in both MB and MB/tantalum mixtures following irradiation. The presence of tantalum nanoparticles in the MB solution resulted in a more pronounced decrease in absorbance. However, the type of radiation used did not significantly influence the absorbance reduction. The decrease in MB absorbance can be attributed to both direct interactions between X-rays or electrons and MB molecules and indirect interactions through radiolysis. In the MB/tantalum mixture, the reduction in absorbance is primarily

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due to the direct interaction of X-rays or electrons with MB molecules, as well as the indirect interaction between the radiation and tantalum nanoparticles (13). The interaction between radiation and metal can produce secondary electrons from compton scattering interaction and photoelectric effect (4). Furthermore, electrons can directly interact with biomolecules at a localized scale or induce the production of a substantial amount of reactive oxygen species (ROS) through interactions with water molecules (14). Tantalum nanoparticles synthesized for 180 minutes resulted in a greater decrease in MB absorbance. This can occur because the increased synthesis time produces nanoparticles with a higher concentration (15). The increase in concentration causes the material density to become greater. A high density produces more secondary electrons, resulting in a greater decrease in MB absorbance (4). The decomposition of MB without nanoparticles is 5.72% and 5.91% for photon and electron beams, respectively. The decomposition of MB with tantalum nanoparticles synthesized for 120 minutes is 8.51% and 10.4% for photon and electron beams, respectively. The decomposition of MB with tantalum nanoparticles synthesized for 180 minutes is 20% for both photon and electron beams. The increase in MB decomposition indicates the formation of a large amount of ROS.



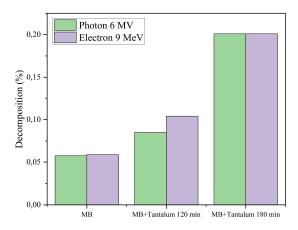


Fig. 6. Absorbance of MB and MB+Tantalum before and after iradiation

Fig. 7. Percentage of MB decomposition without and addition tantalum nanoparticles

The radiosensitization capability of a material is evaluated using the dose enhancement factor (DEF), where a value greater than one indicates an effective enhancement of radiation effects (13). DEF is calculated based on the ratio of MB decomposition with the addition of tantalum nanoparticles to MB decomposition without the addition of tantalum nanoparticles. Figure 8 shows the DEF for variations in the synthesis time of tantalum nanoparticles exposed to photon and electron beams. For tantalum nanoparticles synthesized for 120 minutes, a DEF of 1.49 and 1.76 was obtained for photon and electron beams, respectively. Whereas for tantalum nanoparticles synthesized for 180 minutes, DEF values of 3.51 and 3.40 were obtained for photon and electron beams,

respectively. Both the nanoparticles synthesized for 120 minutes and 180 minutes produced a DEF greater than one. Therefore, tantalum nanoparticles exhibit radiosensitization properties.

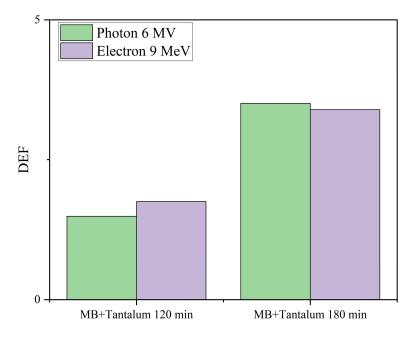


Fig. 8. Dose enhancement factor of tantalum nanoparticle colloid to photon and electron iradiation

IV. CONCLUSIONS

Tantalum nanoparticles were synthesized via the pulsed laser ablation method in a deionized water medium with varying synthesis durations. FESEM analysis reveals that the nanoparticles exhibit a spherical morphology with sizes in the nanometer range. XRD spectral analysis confirms that the synthesized nanoparticles possess an amorphous structure. Tantalum nanoparticles synthesized for 120 minutes have a DEF of 1.49 and 1.76 for photon and electron beams, respectively. Meanwhile, the tantalum nanoparticles synthesized for 180 minutes have a DEF of 3.51 and 3.40 for photon and electron beams, respectively. Therefore, tantalum nanoparticles show potential as a radiosensitizer.

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