Research Article

Evaluation of the X-Ray Absorption by Gold Nanoparticles Solutions

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The increase in the X-ray absorption due to gold nanoparticles was investigated by using aqueous solutions containing gold (Au) nanoparticles. A sample with 15 nm in size nanoparticles and 0.5 mg/mL gold concentration and a distilled water sample were used. Transmitted X-ray beams through the samples were registered with a CdTe detector and with an ionization chamber. Results show an enhancement in the X-ray absorption in the range 20%–6% for beams generated from 20 kV to 120 kV tube voltages, respectively. Results show that the use of gold nanoparticles, even at low concentrations, should result in a significant contrast enhancement for low-energy X-ray beams.

1. Introduction

X-ray radiography procedures are one of the most useful tools adopted in the early diagnosis of cancer in both time and cost related to the process of image acquisition [1]. The efficacy of these techniques relies on the image quality which depends on the X-ray absorption by the tissues that are being exposed [1]. In some of these radiological procedures, such as X-ray computed tomography, a contrast agent is injected directly into the blood stream followed by immediate imaging. The contrast agent leads to increased X-ray attenuation by the targeted tissue resulting in an enhanced image contrast [1, 2]. Currently, contrast agents used in clinical routine procedures are mainly based on iodine-containing molecules. However, such iodine-based compounds present a short-imaging time which results from its rapid renal clearance [3, 4]. In this way, biocompatible nanostructured materials are being investigated in order to improve the image contrast and to enhance the image acquisition time [3–7].

Gold nanoparticles (GNPs) have been the subject of numerous theoretical and experimental studies related to its applications as contrast agents in the X-ray imaging field [3, 5, 8]. The use of these particles in computed tomography has attractive properties such as high atomic number ($Z = 79$) and density ($\rho = 19.3$ g/cm$^3$), high X-ray attenuation coefficients, and enhanced time of blood circulation providing imaging contrast for longer time periods [5, 9].

As the radiographic image contrast depends on the radiation absorption by the target materials, the effect of gold nanoparticles in the X-ray image could be evaluated by means of X-ray spectroscopy. In this work we report the spectral changes on X-ray beams transmitted through a gold nanoparticle aqueous solution registered with a X-ray spectrometer. We chose this detector due to its good performance in the diagnostic energy range [10–12]. Results presented here account for the effect of gold nanoparticles at a low concentration on the X-ray attenuation compared to distilled water for broad beam spectra. The effects on the air-kerma and subject contrast by the gold nanoparticle solution relative to water are also evaluated.

2. Materials and Methods

2.1. Nanoparticles Samples. In this work we used aqueous solutions of highly monodispersed gold nanoparticles with
15 nm diameter (Nanoprobes). The sample was composed of 0.5 mg/mL gold concentration (0.05% wt) in distilled water. A pure sample of distilled water was also used for comparison. Both samples were placed into a plastic container with rectangular base shape and internal thickness of 2 cm.

2.2. X-Ray Spectroscopy. The knowledge of the X-ray spectra is an important tool for investigations of the dose delivered to the patient and the image quality [1]. In this work, the X-ray spectra were generated by a Philips equipment MG 450 model, equipped with a tungsten anode tube with a Be window with 2.2 mm thickness. The anode angle is of 22°. A XR-100T-CdTe detector (Amptek Inc., Bedford, MA, USA) with 9 mm² nominal active area and 1 mm nominal thickness was used to detect the radiation beam. The detector has a 100 μm Be window and is cooled by Peltier cells. Output pulses were processed by a Digital Pulse Processor (DPP) PX4.

Transmitted spectra through the above-mentioned solutions have been recorded with the detector positioned in the center of the radiation field at 520 cm from the focal spot as illustrated in Figure 1. A lead collimator with aperture of 8 mm was positioned at the window of the X-ray tube and before the sample. An EXVC tungsten collimator housing and a collimator with 1000 μm aperture and 2 mm nominal thickness was used in front of the detector. Alignment between the focal spot and CdTe detector was performed with a laser device. Rise Time Discriminator (RTD) was switched off in the DPP system. Pile-up was maintained less than 2% in all the acquisitions. The transmitted X-ray spectra were registered for beams generated in the voltage range between 20 and 120 kV. Energy calibration of the X-ray spectra was performed with the gamma and X-rays emitted by 241 Am, 133 Ba, and 152 Eu radioactive sources [13]. Photon mass attenuation coefficients were obtained from data provided by NIST [14]. The corrected X-ray spectra were used to calculate the air kerma and the mean energy for each X-ray spectra [15].

The relative air kerma was calculated as follows:

\[
\text{RK} = \frac{\text{Kerma}_{\text{Au}}}{\text{Kerma}_{\text{W}}},
\]

where Kerma_{Au} is the air kerma produced by the beams transmitted through the gold nanoparticle solution and Kerma_{W} to those produced by the beams transmitted through the water sample.

Total signal due to the beams transmitted through the samples was calculated by integrating the photon fluence spectra over all the energy range as follows:

\[
\Phi = \int_{E_{\text{min}}}^{E_{\text{max}}} \Phi(E) \, dE.
\]

The subject contrast (Cs) was evaluated according to the following relation:

\[
C_s = \frac{(\Phi_W - \Phi_{\text{Au}})}{\Phi_W},
\]

where \(\Phi_W\) is the photon fluence (photons/cm²) transmitted through the water sample and \(\Phi_{\text{Au}}\) is the photon fluence calculated from the spectra transmitted through the gold nanoparticle solution sample. The value of Cs is a number between 0.0 and 1.0 [2]. We attempt to constrain the first-order behavior of the data performing a nonlinear least squares fitting of an exponential function of the type \(C_s = Ae^{-Br} + C\) with a Levenberg-Marquardt algorithm [16].

Grayscale intensity images were generated by considering the photon fluence spectra transmitted through water and through the gold nanoparticle solution for 20 kV and 25 kV tube voltage X-ray beams. The images were built up with the aid of a computational code based on the Gnuplot graphing utility.

3. Results and Discussion

Measured transmitted X-ray spectra through a sample of distilled water and a gold solution containing nanoparticles with 15 nm are illustrated in Figure 2.

The X-ray beams transmitted through pure distilled water were normalized to the unity at the maximum of the
Figure 2: Comparison of the experimental X-ray spectra transmitted through a solution of gold nanoparticles with 0.05% wt Au concentration and a pure water sample. The data were registered for gold nanoparticles with 15 nm in size and X-ray beams generated at 20, 25, 80, and 100 kV tube voltages.

spectra. As stated in Section 2.1, both samples have 20 mm thickness, and the gold concentration in the solution was 0.5 mg/mL (0.05% wt). According to Figure 2, it is possible to observe that the X-ray absorption is clearly higher for the gold nanoparticles solution mainly for the 20 kV and 25 kV X-ray beams. The air kerma values calculated from these beams transmitted through the pure water sample are about 17% higher than those calculated for the beams transmitted through the 0.05% gold nanoparticle solution for 20 kV. For the 25 kV X-ray beams, this difference was 13%. In the case of beams measured at 80 and 100 kV tube voltages, the difference in the X-ray absorption is about 7% and 6%, respectively.

Figure 3 shows the air kerma values produced by the beams transmitted through the sample of 15 nm nanoparticles with 0.05% Au concentration and through the water sample. In this figure, the air kerma values are presented as a ratio of the values measured for the beams transmitted through the gold nanoparticle solution and those measured for the beams transmitted through the water sample. The values presented in this figure are due to measurements performed with an ionization chamber and calculated from the corrected X-ray spectra. A good agreement among the air kerma values measured with the ionization chamber and calculated from the corrected X-ray spectra is observed over all the energy range. Results based on the ionization chamber measurements show that for the 20 kV beam the X-ray absorption is about 20% higher than for the beam transmitted through the gold nanoparticle solution when compared to that transmitted through the pure distilled water sample. For the 25 kV beam the absorption by the nanoparticle solution was about 13% higher than that relative to distilled water. In the case of the 30 and 35 kV X-ray beams, this difference was 10% and 9%, respectively. For beams generated with tube voltages from 50 kV to 120 kV, this difference varies from 10% to 4%.

Figure 4 brings the grayscale images produced with the beams transmitted through the distilled water sample and the gold nanoparticles solution presented in Figure 3, for
Figure 3: Comparison of the relative air kerma values measured with an ionization chamber and calculated from the corrected x-ray spectra. The data were registered for beams generated in the voltage range from 20 to 120 kV.

20 kV and 25 kV tube voltages. The image contrast in this figure is presented as a function of the X-ray beam energy. The upper and the bottom parts of Figure 4 concern in the beam transmitted through the water sample while the middle part refers to the beams transmitted through the gold nanoparticles solution. The ticks on the intensity axis in Figure 4 indicate the boundary between the gray intensity produced by the beams transmitted through water and those transmitted through the gold nanoparticles solution. These figures show that the maximum contrast occurs in the ranges of 16 to 18 keV for the 20 kV beams and 18 to 22 keV for the 25 kV X-ray beams.

Figure 5 brings the subject contrast calculated from the photon fluence of the beams transmitted through the solution containing nanoparticles and the water sample. Results show that the subject contrast enhancement due to the gold nanoparticles varies from about 17% to 2% for beams with a mean energy ranging from 17 keV to 63 keV.

4. Conclusion

X-ray absorptions by samples of gold nanoparticles solutions and distilled water were investigated in this work. A solution with 0.5 mg/mL gold nanoparticle concentration and a water sample were used in order to evaluate the X-ray absorption and its effect on the air kerma values and subject contrast. We conclude, based on the experimental results, that the X-ray absorption for a solution with 0.05% gold nanoparticle concentration increases from 20% to 6% for beams generated between 20 kV and 120 kV, respectively. Also, a significant subject contrast enhancement was observed for low-energy beams. Effectively, targeted tissues with these electron-dense nanoparticles, even at a very low concentration, are able to
display significant enhanced X-ray absorption resulting in a higher image contrast or absorbed dose.

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