The Size Effect of Powdered Scintillator on High-Resolution X-ray Imaging System

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1. Introduction

Digital X-ray imaging uses a converter which emits visible light (indirect) or electrons (direct) when absorbing X-ray. Indirect system uses a scintillator as a converter with light detector [1-4]. A scintillator is a substance that emits visible light when it reacts with Xray or other radiations. The structure and optical properties of scintillators affect the quality of X-ray images. Powdered-scintillator screen for X-ray image is used a lot in industry, medical diagnostics and various fields [5-9]. In such a scintillator, the optical light generated from the scintillator is optically diffused by other grains, resulting in light spread, which reduces the spatial resolution of X-ray image [10-12]. However, if nanoparticle scintillators are used for powderedscintillator screen, high spatial resolution can be achieved due to low light spread [13, 14]. In this experiment, powdered zinc tungstate $(ZnWO_4)$ scintillators were prepared by the solid-state reaction. ZnWO₄ is of great interest as ultraviolet, X-ray, χ -ray, electron beam and proton beam scintillator [15-19]. For above reasons, ZnWO₄ has the unique the characteristics about luminescent property, high density, high chemical stability, short decay time, low afterglow, and economical price [16, 20-25].

Meanwhile, my research team developed the highresolution X-ray inspection system (HRXIS) [26]. This system consists of a carbon nano-tube (CNT) based miniature x-ray tube, a scintillator, an optical lens, and a scientific complementary metal-oxide-semiconductors (sCMOS) detector. The concept of the developed system is that transmits the X-ray in the form of an object attached to a scintillator, and then magnifies the image formed on the scintillator screen using an optical lens, then acquire images on the sCMOS detector. This system not only can acquire X-ray images with high spatial resolution, but also inspect the internal defect of micro-sized small objects.

In this study, the zinc oxide nanoparticles and tungsten oxide nanoparticles used in the solid-state reaction were synthesized using anodization method. Anodization method is that if a platinum relative electrode (-) and a metal wire (+) are added to the electrolyte and then a voltage of 10-100 V are applied, oxidation and etching occurs on the metal wire resulting in metal oxide nanoparticles [27]. ZnWO₄ nanoparticles were fabricated through the solid-state reaction using asprepared zinc oxide nanoparticles and tungsten oxide nanoparticles. The solid-state reaction was carried out at

different temperatures for fabricating the various sizes of $ZnWO_4$ powders. The different sizes of $ZnWO_4$ powders were fabricated as thin screens on silicon glasses. These scintillator screens were applied to the HRXIS for obtaining X-ray image. Finally, it was evaluated whether high spatial resolution of X-ray images can be obtained when nanoparticle scintillators are applied to high-resolution X-ray imaging.

2. Materials and Methods

2.1 Materials

We purchased zinc wires and tungsten wires from Goodfellow (Huntingdon, UK). Potassium chloride (KCl) powder, ammonium fluoride (NH₄F) powder and polyethylene glycol (PEG, average molecular wight 200) were purchased from Sigma-Aldrich (St Louis, Missouri, USA).

2.2 Characterization of ZnWO₄

The zinc oxide nanoparticles and the tungsten oxide nanoparticles were synthesized by anodization method. The electrolytes were made by adding 1M KCl and 1M NH₄F respectively to deionized (DI) water. The zinc oxide nanoparticles were synthesized by applying 15 V to zinc wires in the electrolyte of 1M KCl. In case of tungsten oxide nanoparticles, the voltage of 80 V was applied in 1M NH₄F. Both nanoparticles were washed with DI water and were dried at 60°C in air. Both nanoparticles with 1:1 atomic ratio of zinc and tungsten were added in DI water with 0.2 wt% PEG and mixed well with the sonication and vibrating ball milling, and then this aqueous solution was dried at 60°C in air. A solid-state reaction was implemented in electric furnace at different temperatures with 700, 800, and 900°C for 1 hour.

The nano-structures of zinc oxide nanoparticles and tungsten oxide nanoparticles were observed by a transmission electron microscope (TEM, Titan cubed G2 60-300, FEI, Hillsboro, OR, USA). The structures of ZnWO₄ powders were observed by FE-SEM. The crystalline structures of ZnWO₄ powders were analyzed by high-resolution X-ray diffraction (HR-XRD, SmartLab, Rigaku, Tokyo, Japan) with an analysis 20 range 10-90°.

2.3 Fabrication of ZnWO₄ scintillator screen

The ZnWO₄ scintillator screens on the 150 μ m silicon glass (1.5 cm x 1.5 cm) were made by drop casting method. ZnWO₄ powders, PEG, and DI water were prepared to make ZnWO₄ paste. After adding ZnWO₄ powder into DI water with 0.2 wt% PEG, this paste was ball-milled for 1 hours. Finally, ZnWO₄ paste was put on silicon glass and made to a 10 μ m thin screen by adjusting the concentration of ZnWO₄ powders. The ZnWO₄ scintillator screens were applied to HRXIS for obtaining X-ray images and evaluating the spatial resolution of the X-ray images.

3. Result and Discussion

3.1 Nanoparticles synthesized by anodization

The Zinc oxide nanoparticles and tungsten oxide nanoparticles were prepared by anodization method. Figure. 1(a) is the image of zinc oxide nanoparticles acquired by TEM. This figures demonstrate that the zinc oxide nanoparticles are spherical and sized in 20-40 nm. The TEM image of tungsten oxide nanoparticles is shown in Figure. 1(b). This images show that tungsten oxide nanoparticles are in the form of nano-plate with a width of 20-60 nm.



Fig. 1. Metal oxide nanoparticles synthesized by anodization method, (a) TEM image of zinc oxide nanoparticles, and (b) TEM image of tungsten oxide nanoparticles

3.2 Structure of ZnWO₄ scintillator

The ZnWO₄ powders were fabricated by solid-state reaction at different temperature at 700, 800 and 900°C. The structure morphology of ZnWO₄ powders are showed in Figure. 2(a)-(c) by FE-SEM. The ZnWO₄ powders were sized through FE-SEM. As a result, the average sizes of powders fabricated at 700, 800, and 900°C were 176.4 nm, 626.7 nm, and 2.127 µm respectively. The ZnWO₄ powders fabricated at 700°C and 800°C were formed in the wavelength band of light or smaller. However, the size of the powders fabricated at 900°C were formed to micro-sized particles larger than the wavelength of light. Figure. 2(d) shows the XRD patterns of the ZnWO₄ powders with an analysis 2θ range 10-900. Comparing the peak value of the XRD pattern with the standard card (PDF ICDD card #01-078-0251), these powders were crystallized into pure monoclinic ZnWO₄.



Fig. 2. FE-SEM image of ZnWO₄ fabricated at (a) 700°C,
(b) 800°C, and (c) 900°C, (d) XRD pattern of ZnWO₄.

3.3 Evaluation of X-ray imaging performance in HRXIS

The ZnWO₄ scintillator screens were prepared on silicon glass by drop casting method. Fabricated scintillator screens were applied to HRXIS (Figure. 3). This system consists of a miniature X-ray tube, a 10x optical lens, and a sCMOS detector. The miniature Xray tube was produced directly in our lab [28]. This Xray tube is composed of carbon nano-tube(CNT) based electron beam emitter, the tube voltage is 50 kVp and the current is 0.35 mA. This X-ray tube is equipped with focusing lens. The focusing lens is a cylindrical form composed of permanent magnets. The focal spot of the miniature X-ray tube using focusing lens was 337-390 um [26]. A 10x optical lens and a sCMOS detector were used to magnify and obtain images on scintillators. The pixel size of the sCMOS detector is 6.5 um. When the image on scintillator is magnified by a 10x optical lens, the effective pixel size is 0.65 um.



Fig. 3. Applying ZnWO₄ scintillator screen to HRXIS

The modulation transfer function (MTF) was measured to evaluate the spatial resolution according to the size of $ZnWO_4$ powders. The MTF is a general method to analyze the spatial resolution of X-ray image [13, 26, 29-30]. A 0.5 cm thick tungsten block was placed on the fabricated $ZnWO_4$ scintillator screen and the X-ray image for edge of tungsten block were obtained using HRXIS. The edge spread function (ESF) was determined through the obtained X-ray image, and the ESF was converted to MTF. The MTF curve of each scintillator screen is shown in Figure. 4. The spatial frequency (lp/mm) was evaluated at 10% of the MTF. The spatial frequency of the scintillator screens with an average particle size of 176.4 nm, 626.7 nm, and 2.127 μ m were 278.2, 204.1, and 124.1 lp/mm. As a result, high spatial resolution of X-ray images can be acquired with a scintillator screen composed of nano-sized particles.



Fig. 4. MTF curve of scintillator screen composed of $ZnWO_4$ powders with different averaged sizes of (a) 176.4 nm, (b) 626.7 nm, and (c) 2.127 μ m.

4. Conclusion

The ZnWO₄ scintillator powders were fabricated by a convenient way using the anodization method and the solid-state reaction. These ZnWO₄ scintillator powders were fabricated in the form of scintillator screens and applied to high-resolution X-ray imaging for evaluating spatial resolution of X-ray image. The result showed that the spatial resolution would be optimized if the average diameter of the particle was less than the wavelength of light in powdered-scintillator screen. These results proved that the ZnWO₄ nanoparticle scintillator can be utilized for obtaining X-ray images with high spatial resolution.

A commercialized powered scintillator is micro-sized particles larger than the wavelength of light [14]. However, the result of this experiment showed that scintillators with the size of 100-250 nm generate less light diffusion, thus improving the spatial resolution of the X-ray images. This study will have an impact on a lot of research about powdered-scintillator screens. This result can be applied not only to ZnWO₄ scintillator, but also to other scintillator materials, so this experiment will be of sufficient help to study other nanoparticle scintillator materials.

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