

## Thermoluminescence (TL) response of silica nanoparticles subjected to 50 Gy gamma irradiation

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### Abstract

Thermoluminescence (TL) response of silica nanoparticles synthesized by the sol-gel method is presented here. Transmission electron microscopy (TEM) was used to find out the size and morphology of the pure silica nanoparticles. By using an appropriate amount of tetraethylorthosilicate, ethanol, deionized water and ammonia solution, silica samples were synthesized. To determine the best TL response of silica, samples were irradiated with 50Gy gamma rays. The effect of size dependency towards TL yield indicates that decreasing the particles' size of silica, increases the TL yield.

**Keywords:** Thermoluminescence, sol-gel, nanoparticles

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## INTRODUCTION

In their early research work about nanoparticle, Stober et al [1] discovered a method known as sol-gel technique for preparing silica monodisperse in micron size that lead to overwhelming further research work in silica nanosize area. They used aqueous alcohol solutions of silicon alkoxides in the presence of ammonia as a catalyst. Nano-sized particles have one dimension that is less than 100 nano-meters in size. Because of their unique properties, they are useful as a catalyst, sensors, coating materials, tunable lasers, and memory devices. Nano-material is getting more interest and attention rather than bulk substance for research work in the area of nanotechnology [2]. In the sol-gel method, the prepared phosphors in the form of powders are mostly homogeneous and purer than the phosphor obtained via other conventional solid-state methods and have been widely applied to produce nanoscale materials. In order to synthesize the silica nanoparticle, many researchers have opted sol-gel as their method due to its advantages [3-5]. By taking the method into account, the size, shape and structure of the silica particle can be controlled due to the variable amount of materials used to produce the silica [6-8].

Thermoluminescence (TL) dosimeter is used to detect the amount of radiation exposure to workers of a radiation source. TL measurement is crucial for deciding the best TL dosimeter. Based on the TL mechanism where electrons escape from traps upon heating and return to the stable state, the sensitivity or the probability of electron escapes from the traps can be enhanced by giving temperature to them. So, different temperature will gives different TL sensitivity of the phosphor. In the previous research work on TL measurement, particular attention has been given to the silica material in the form of optical fiber, thin film and bulk [9-12]. In this paper,

nanosized TL particles are investigated to determine the effect of the size towards exposed doses.

## EXPERIMENTAL

### Synthesis of SiO<sub>2</sub> nanoparticles

The materials used to synthesize the SiO<sub>2</sub> are tetraethyl orthosilicate (TEOS) (96%) as precursor, ethanol (98%) acts as the solvent, deionized water, and ammonia (28%) acts as the catalyst agent. The set of 2 samples of SiO<sub>2</sub> were prepared by varying its amount of catalysis agent. SiO<sub>2</sub> sols were prepared by mixing 0.25 mol of ethanol and water for 10 minutes before TEOS (0.05 mol) was added to the mixture. While stirring for another 20 minutes by using magnetic stirrer (>150 rpm), 2 ml of ammonia solution were added dropwise into the mixture until white turbid suspension was formed. The sample was stirred continuously for 24 hrs at room temperature. The suspension was then dried in an oven at 80 °C for 24 hrs before undergoing calcination process at 600°C for 2 hrs. The final product, SiO<sub>2</sub> powder, was then grinded in a porcelain mortar. The schematic diagram for synthesizing SiO<sub>2</sub> particles is shown in Fig. 1. The same processes were repeated by using 8 ml of ammonia.

### Sample characterization and TL measurement

The X-ray diffraction pattern was obtained to see the nature of SiO<sub>2</sub> by using X-ray diffractometer (Siemens Diffractometer D5000). To identify the size of particles for both samples, Transmission Electron Microscope (TEM, JEM-2100, 200kV) was used. To verify the effect of particles size on the luminescence response, both samples were exposed to  $\gamma$ -ray for 50 Gy and by using Gamma cell 220E facility at Universiti Kebangsaan Malaysia with different annealing temperature from 100°C to 400°C for 1 h. The TL response was

measured using HARSHAW 4500 TLD reader at a heating rate of  $25^{\circ}\text{C s}^{-1}$ .

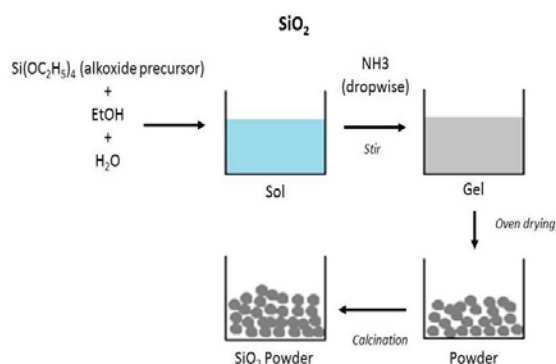


Fig. 1 Schematic diagram for the preparation of  $\text{SiO}_2$ .

## RESULTS AND DISCUSSION

### XRD analysis

The X-ray diffraction patterns confirmed the amorphous nature of the prepared  $\text{SiO}_2$  samples as shown in Fig. 2. The broad peak shows a complete amorphous structure. There is no diffraction peak observed except for a broad band centered at  $22^{\circ}$  which is the characteristic peak for amorphous  $\text{SiO}_2$ . The results is also compared with the JCPDS file for  $\text{SiO}_2$  and it reveals no impurities peak for  $\text{SiO}_2$  [4, 13].

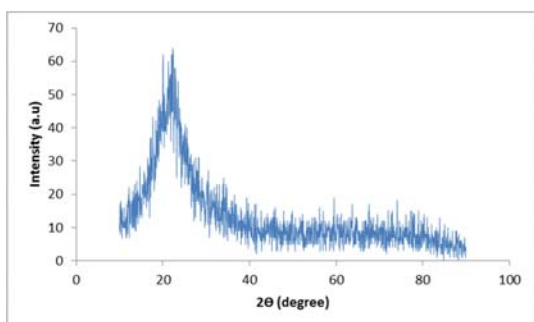


Fig. 2 XRD analysis of  $\text{SiO}_2$  nanoparticles.

### TEM analysis

To investigate the morphology and size of particles in nano scale, high-resolution Transmission Electron Microscope (TEM) is used to punctual electron diffraction analysis of the  $\text{SiO}_2$  nanoparticles. By changing the concentration of ammonia solution in  $\text{SiO}_2$  synthesis, the effect of particle's size can be varied. From Fig. 3(a), the image shows the average particles's in the range of  $<400\text{nm}$  for the sample of 2ML ammonia whereas, a sample with 8ML ammonia solution, its size is up to  $934\text{nm}$  Post analysis is done by Digital Micrograph (GATAN) version 3, year 2015. The particles are not well dispersed due to agglomeration. It can be overcome by increasing the sonication time.

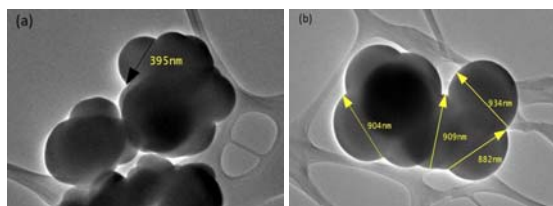


Fig. 3 TEM image of silica nanoparticles with (a)2ML and (b) 8ML of ammonia solution.

### Effect of catalyst on size particles

One of the processes involved in the sol-gel method is hydrolysis reaction where acids or bases are used as catalysts. Catalyst will increase the rate of hydrolysis and condensation process in a sol-gel process that leads to a faster kinetics. The particle size strongly depends on hydrolysis kinetics. According to Singh L.P et al [14], the size of silica particle size increases with decreasing the amount of ammonia but reverse effect is obtained by Rao et al [2]. In this work, we discovered that the  $\text{SiO}_2$  particle size was increasing with the increasing of ammonia concentration, in agreement with Rao et al [2].

### Thermoluminescence measurement

In early stage for TL identification, annealing procedure should be carried out to remove any previous exposure. Different annealing temperature from  $100^{\circ}\text{C}$  to  $400^{\circ}\text{C}$  were done for 1 hr each by using oven annealing (Harshaw) connected to readout system to identify the suitable temperature for the dosimeter material. The samples were then exposed to 50 Gy of  $\gamma$ -ray and was measured using the Harshaw 3500 TLD reader to see the best intensity. Based on the TLD reader, a sample with  $100^{\circ}\text{C}$  annealing temperature as shown in Fig. 4 and 5 exhibits a higher TL intensity for samples from both 2 and 8 ml of ammonia. The same time temperature profile was used for TLD measurement on both samples(13.3s reading cycle and  $25^{\circ}\text{C s}^{-1}$ ). However, the response for 2 ml ammonia is higher than 8 ml ammonia.

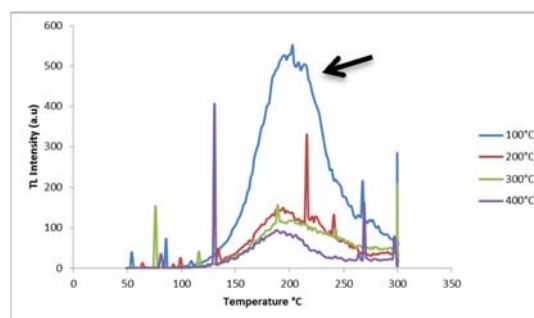


Fig. 4 TL intensity of  $\text{SiO}_2$  with 2 ml ammonia under different annealing temperature indicates higher intensity for  $100^{\circ}\text{C}$ .

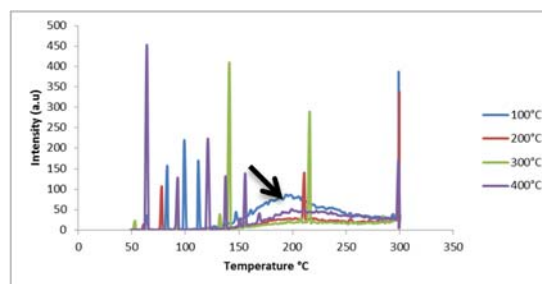


Fig. 5 TL intensity of  $\text{SiO}_2$  with 8 ml ammonia under different annealing temperature indicates higher intensity for  $100^{\circ}\text{C}$ .

## CONCLUSION

The  $\text{SiO}_2$  particles size was measured by TEM. The results shows that the size of  $\text{SiO}_2$  particles with a lower concentration of ammonia is smaller than higher concentration used in synthesizing of  $\text{SiO}_2$  powder. Based on the XRD analysis, amorphous nature of samples is confirmed since there is no crystalline phase occur. For TL identification,  $100^{\circ}\text{C}$  annealing temperature was chosen to be the best annealing temperature for  $\text{SiO}_2$  nanoparticle because of the high TL intensity. Based on the TL intensity, it also shows that smaller particles give high TL yield than a larger particle. The present result also supports that the particles's size affects the TL yield but taking ethanol (solubility agent) to be varied in the silica synthesis [15].

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## REFERENCES

- [1] Stober, W., Fink, A. (1968). Controlled growth of monodisperse silica spheres in the micron size range. *Journal of Colloid and Interface Science*. 26, 62-69.
- [2] Rao, K. S., El-Hami, K., Kodaki, T. Matsushige, K. Makino, K. (2005). A novel method for synthesis of silica nanoparticles. *Journal of Colloid and Interface Science*. 228, 125-131.
- [3] Dabbaghian, M. A., Babalou, A. A., Jannatdoust, E. (2010). A parametric study of the synthesis of silica nanoparticles via sol-gel precipitation method. *International Journal of Nanoscience and Nanotechnology*. 6(2), 104-113.
- [4] Tabatabaei, S., Shukohfar, A., Aghababazadeh, R., Mirhabibi, A. (2006) Experimental study of the synthesis and characterization of silica nanoparticles via the sol-gel method. EMAG-NANO 05: Imaging, Analysis and Fabrication on the Nanoscale. *Journal of Physics: Conference Series* 26. 371-374
- [5] Kao, M. J., Hsu, F. C. Peng, D. X. (2014). Synthesis and characterization of SiO<sub>2</sub> nanoparticles and their efficacy in chemical mechanical polishing steel substrate. *Advances in Materials Science and Engineering*. 2014, 691967, 8 pages.
- [6] Zhang, J. H., Zhan, P. P., Wang, Z. L., Zhang, W. Y., Ming, N. B. (2003). Preparation of monodisperse silica particles with controllable size and shape. *Journal of Material Research*. 18(3), 649-653.
- [7] Gao, H., Yang, J. (2010). Nanoscale silicon dioxide prepared by sol-gel process. *Modern Applied Science*. 4(9), 152-156.
- [8] Martinez, J. R., Palomares-Sanchez, S., Ortega-Zarzosa, G., Ruiz, F., Chumakov, Y. (2006). Rietveld refinement of amorphous SiO<sub>2</sub> prepared via sol-gel method. *Materials letters*. 60, 3526-3529.
- [9] Pacchioni, G., Skuja, L., Griscom, D. L. (2000). Defects in SiO<sub>2</sub> and related dielectrics: science and technology. Dordrecht: Kluwer Academic Publishers.
- [10] Ho, C. K. F., Pita, K., Ngo, N. Q., Kam, C. H. (2005). Optical functions of (x)GeO<sub>2</sub>:(1-x)SiO<sub>2</sub> films determined by multi-sample and multi-angle spectroscopic ellipsometry. *Optics Express*. 13(3), 1049-1054.
- [11] Park, R. L., Champagnon, B., Levelut, C., Martinez, V., David, L., Faivre, A., Flammer, I., Hazemann, J. L., Simon, J. P. (2008). Density and concentration fluctuations in SiO<sub>2</sub>-GeO<sub>2</sub> optical fiber glass investigated by small angle x-ray scattering. *Journal of Applied Physics*. 103(9), 094917.
- [12] Devine, A. B., Duraud, J. P., Dooryhee, E. (2000). Structure and imperfections in amorphous and crystalline silicon dioxide. New York: Wiley.
- [13] Bajpai, N., Tiwari, A., Khan, S. A., Kher, R. S., Bramhe, N., Dhole, S. J. (2013). Effects of rare earth ions (Tb, Ce, Eu, Dy) on the thermoluminescence characteristics of sol-gel derived and  $\gamma$ -irradiated SiO<sub>2</sub> nanoparticles. *Luminescence*. 29, 669-673.
- [14] Singh, L. P., Agarwal, S. K., Bhattacharyya, S. K., Sharma, U., Ahalawat, S. (2011). Preparation of silica nanoparticles and its beneficial role in cementitious materials. *Nanomaterials and Nanotechnology*. 1(1), 44-51.
- [15] Shafiqah, A. S., Amin, Y. M., Nor, R. M., Bradley, D. A. (2015). Effect of particle size on the thermoluminescence (TL) response of silica nanoparticles. *Radiation Physics and Chemistry*. 117, 102-107.