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Synthesis of SiO_2 Nanostructures Using Sol-Gel Method

H.N. Azlina[a,*], J.N. Hasnidawani[a], H. Norita[a] and S.N. Surip[b]

[a]Department of Manufacturing and Materials Engineering, Faculty of Engineering, International Islamic University of Malaysia (IIUM), Jalan Gombak, 53100 Kuala Lumpur, Malaysia

[b]Faculty of Applied Sciences, Universiti Teknologi MARA, 40450 Shah Alam, Malaysia

Sol-gel method is the simplest method and has the ability to control the particle size and morphology through systematic monitoring of reaction parameters. The objective of this research is to synthesize silica nanostructures by sol-gel method and to characterize the synthesized silica nanostructures. Silica nanoparticles were synthesized via the sol-gel method using Tetraethyl orthosilicate as a precursor. The acetic acid and distilled water were used as the catalyst and the hydrolyzing agent. Varied parameters of the study were the aging time in the range of 2 to 6 h and the calcination temperature in the range of 600–700 °C. The obtained silica nanopowder was characterized using FESEM, and Nano-Particle Size Analyzer. The results show that the silica nanospheres were successfully synthesized by using sol-gel method with the optimum parameters of 700 °C of calcination temperature and 2 h of aging time. The average size of silica nanoparticles was in the range of 79.68 nm to 87.35 nm.

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1. Introduction

The dynamic development of nanotechnology has been observed especially in the area of materials engineering. Excellent examples are various nanoparticles, where the current research is focused on their design, fabrication, characterization and applications. The properties and specific functions of nanoparticles may be controlled by their shape and size at the nanometer scale. There are several methods that have been put forward for synthesis of these materials, namely chemical vapor condensation, arc discharge, hydrogen plasma-metal reaction, and laser pyrolysis in the vapor phase, micro-emulsion, hydrothermal, sol-gel, etc. [1, 2]. The properties of metal nanoparticles depend largely on their synthesis procedure. Recently, scientific and research community have shown their great interest on metal oxide nanostructures and their applications due to their safe, easy, environmental friendly and cheap synthesis procedure. In addition, metal oxide nanoparticles have novel electronic, optical, magnetic, chemical catalytic and mechanical properties originating from the high surface to volume ratio and quantum size effect [3, 4]. This shows that metal oxides have a great market interest and industry demand.

2. Materials and methods

2.1. Materials

Silica nanostructure was synthesized using sol-gel method. There are two parameters that have been playing important roles in this study which are the aging time and the calcination temperatures. For synthesis the tetraethyl orthosilicate, TEOS was added to acetic acid, CH_3COOH with distilled water as the solvent. The aging time in the experiments was set to 2, 4 and 6 hours. The resulting colloidal sol then was centrifuged and washed with ethanol and then it was centrifuged again. Then, the resulting precipitate was dried at 60 °C for 1 day. The second parameter, the calcination temperature was varied between 600 °C and 700 °C at a holding time of 1 hour and 30 minutes in order to produce white silica nano powder.

TABLE I

Reagents compositions.

Reagent	Amount	Number of moles
TEOS	18 ml	0.086
Distilled water	6.4 ml	0.360
Acetic acid	36 ml	0.600
Ethanol	20 ml	–

TABLE II

Aging time and calcinations temperature of sol-gel reactions.

Sample	TEOS [ml]	Acid [ml]	Distilled water [ml]	Aging time [h]	Calcination temper. [°C]
A	18	36	6.4	2	600
B	18	36	6.4	4	600
C	18	36	6.4	6	600
D	18	36	6.4	2	700
E	18	36	6.4	4	700
F	18	36	6.4	6	700

Synthesis of silica nanostructures by sol-gel method in this research work included the use of several materials such as tetraethyl orthosilicate ($Si(OC_2H_5)_4$), acetic acid (CH_3COOH), distilled water (H_2O) and ethanol (C_2H_5OH). Tetraethyl orthosilicate was used as the precursor. Acetic acid and distilled water were used as the

*corresponding author; e-mail: noorazlina_hassan@iium.edu.my

catalyst and the hydrolyzing agent, respectively. Ethanol was used to remove the initial materials that provide no reaction from the surface of the formed particles. TEOS and water were mixed in the following molar ratios of H_2O:TEOS, 4:1. The compositions of each reagent used to prepare the silica nanostructure are shown in Table I, while the parameters of preparation of the silica nanostructure by sol-gel method are shown in Table II. These varied parameters include (i) reaction period (aging time) and (ii) calcination temperature.

2.2. Nano particle analyzer

The Malvern Zetasizer analyzer measures particle and molecule size from a nanometer to several microns using dynamic light scattering, zeta potential and the electrophoretic mobility using electrophoretic light scattering and the molecular weight using static light scattering. For this experiment, Malvern Zetasizer series version 6.2, MAL 1017843 model was used. The average particle size analysis of SiO_2 powder was conducted and the distilled water was used as the dispersant medium. The sample preparation was conducted during 3 days. At day one, the 40 ml of distilled water was mixed with 0.1 g of sample and was stirred for 1 day. Then, on the next day, 1 ml of liquid on the surface was taken with a pipet and mixed with 20 ml of distilled water for 1 day. On the next day, average particle size analysis was conducted. The sample preparation was conducted for 3 days, so to make sure that there was no agglomeration occurring and the single particles could be detected.

2.3. Field emission scanning electron microscopy

Field emission scanning electron microscope (FESEM) was used to study the morphology and particles of the prepared SiO_2 sample. It is a widely used technique in the characterization of nanomaterials and nanostructures. This study was carried out with JEOM JSM-6700F field emission scanning electron microscope. In order to view the sample, the thin coating was first applied to the SiO_2 powder sample by using gold to avoid occurrence of charging. The FESEM used current of 10 μA. The observations were done at a magnification of $\times$1000.

3. Results and discussion

After undergoing sol-gel process, silica nanopowder was produced. A typical sol-gel reaction is hydrolysis and condensation of TEOS as a precursor of silica. TEOS and water were mixed in the following molar ratios of H_2O:TEOS, 4:1. During the sol-gel process, TEOS was first hydrolyzed to silicic acid as shown in Eq. (1). Then, condensation reaction has led to formation of Si-O-Si bonds as shown in Eq. (2). The overall sol-gel reactions employed in the production of the SiO_2 by sol-gel method are shown below

$$Si(OC_2H_5)_4 + 4H_2O \rightarrow Si(OH_4) + 4C_2H_5OH, \qquad (1)$$

$$Si(OH_4) \rightarrow SiO_2 + 2H_2O. \qquad (2)$$

Nanoparticle size values of silica at different calcination temperatures and different aging times were examined by average particle size distribution. By varying the reaction time, the silica particles were produced with various sizes and shapes due to uncompleted core creation. After a few hours of reaction, core creation is completed and the layers around the particles are continuing to be created. As reported in Table III, the values for average nano particle size of the silica were increasing with the increasing time of sample aging. It shows that at calcination temperature of 600 °C, the particle sizes of 2 h aged sample ranged from roughly 2.81 μm to 3.29 μm, those aged for 4 h ranged approximately from 2.94 μm to 3.29 μm and those at 6 h between 3.14 μm and 3.28 μm.

TABLE III

Particle size of silica powder (based on micrographs).

Particle size of SiO_2 powder [μm]			
Calcination temperature [°C]	Aging time [h]		
	2	4	6
600	2.81–3.29	2.94–3.29	3.14–3.28
700	2.59–2.99	2.99–3.21	3.28–3.39

TABLE IV

Average size of silica nanoparticles (nanoparticles size analyzer).

Average size of SiO_2 nanoparticles [nm]			
Calcination temperature [°C]	Aging time [h]		
	2	4	6
600	$\pm$79.68	$\pm$147.6	$\pm$156.7
700	$\pm$87.35	$\pm$153.9	$\pm$162.9

Table IV and Fig. 1 are summarizing the average size distribution of synthesized SiO_2 nanoparticles obtained using nanoparticles size analyzer. The results show that samples calcined at 700 °C are slightly greater than those calcined at 600 °C. For instance, 2 h aging and calcination processes at 700 °C has produced relatively bigger average particle size than that of samples calcined at 600 °C, with the particle sizes of 87.35 nm and 79.68 nm respectively. This is in good agreement with Abdel-Latief et al., who have stated that the particle size increases and has a higher degree of agglomeration as the calcination temperature increases [5]. However, although the particles have increased in size, the dispersion of smaller particles can still be observed at higher calcination temperatures.

Field emission scanning electron microscopy was used to study the morphology of the prepared silica samples. FESEM evaluation of powders reveals that the powders consist of fine particle agglomerates, the particles appear to adhere to each other, forming aggregate of

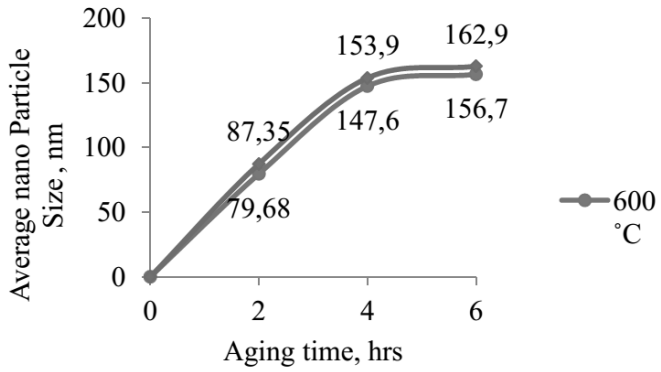

Fig. 1. Average nano particle size of SiO_2 powder.

particles, which result in irregular arrangements. These results show that at higher calcination temperatures and longer aging times, a bigger particle size has been produced. FESEM micrographs of these particles are shown in Fig. 2 and Fig. 3, which demonstrate that the spherical silica nanoparticles have been successfully synthesized via this method. These micrographs illustrate the surface morphology of samples aged during 2 h, 4 h and 6 h at calcination temperatures of 600 °C and 700 °C.

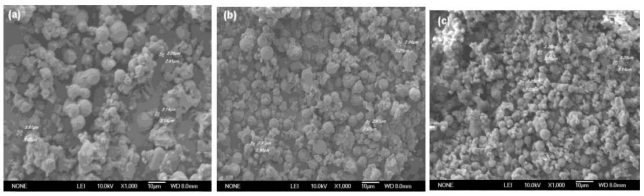

Fig. 2. Micrographs of silica nanostructures at magnification ×1000; for calcination temperature of 600 °C and various aging times. (a) Aging time 2 h. (b) Aging time 4 h. (c) Aging time 6 h.

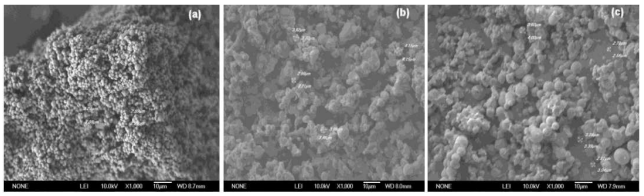

Fig. 3. Micrographs of silica nanostructures at magnification ×1000; for calcination temperature of 700 °C and various aging times. (a) Aging time 2 h. (b) Aging time 4 h. (c) Aging time 6 h.

4. Conclusions

The silica nanostructures were successfully synthesized using sol-gel method. FESEM analysis shows that the spherical structures of silica were successfully obtained. Nano-particle size analysis shows that the average particle size of silica was in the range of 79.68 nm to 87.35 nm. The optimum conditions were found to be at calcination temperature of 700 °C and 2 h aging time.

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