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EFFECT OF pH LEVEL ON THE FORMATION OF SILICA AEROGEL SYNTHESIZED FROM RICE HUSK ASH

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Abstract

Silica aerogel has been synthesized from rice husk ash via water glass solution by using sol-gel method. The synthesized silica aerogel at different pH level are characterized by X-ray diffraction (XRD) to analyze the structure formation of silica aerogel. The function group of the products are characterized by Fourier Transform Infrared Spectroscopy (FTIR). The particle sizes of silica aerogel are characterized by scanning electron microscopy (SEM). XRD result shows that the silica aerogel particles prepared at pH 7 has the capability to extract complete SiO_2 from rice husk ash with a great quantity of amorphous silica. SEM result indicates that the particle size of the silica aerogel increases at pH 5 and pH 9 and decrease at pH 7. The results show that the synthesis of silica aerogel from rice husk ash has porous structure and the particle size of silica aerogel depend on the effect of pH level.

Keywords: rice husk ash, silica aerogel and sol-gel method

Introduction

Aerogel is a synthetic porous ultralight material derived from a gel, in which the liquid component for the gel has been replaced with a gas. Aerogels have a porous solid network that contains 90% of air pockets taking up the majority of space within the material. The lack of solid material allows aerogel to be almost weightless. The term aerogel was introduced by Kistler in 1931 to define gels in which the liquid is replaced by a gas without the collapse of the polymer network. Nicknames include frozen smoke, solid smoke, solid air, solid cloud, blue smoke owing to its translucent nature and the way light scatters in the material (Tadjarodi A. et al., 2012). Aerogels possess remarkable properties such as high specific surface area and low thermal conductivity (Chen Q., 2017).

There are three most common types of aerogels such as silica, carbon and metal oxides. Among them, silica aerogel is most often used experimentally and in practical applications. Silica aerogels exhibit many unique properties, which include very high inner surface area, low sound velocity, high optical transparency, low refractive index, extremely and low thermal conductivity. Because of their special properties, silica aerogels have been widely investigated in various fields, find its applications in aerospace and aeronautic, thermal insulation, acoustic barrier materials, and more recently in catalyst products (Guangwu L., & Yangang L., 2016).

Silica aerogels contain more than 90% air and less than 10% solid silica. An aerogel is made by the so called “sol-gel process”. The first precursor used to obtain an aerogel was sodium silicate, resulting in the so called water glass solution. During this process, organic compounds containing silica undergo a chemical reaction producing silicon oxide (SiO_2). This mixture is a liquid at the creation of the reaction, and becomes more and more viscous as the reaction proceeds. When the reaction is completed, the solution loses its fluidity and the whole reacting mixture turns into a gel. This gel consists of a three-dimensional network of silicon oxide filled

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with the solvent. During the special drying procedure, the solvent is extracted from the gel body leaving the silicon oxide network filled with air. This product is called silica aerogel (Rakesh P. Patel, 2009).

Most commonly used raw materials for preparation of silica aerogels are tetraethylorthosilicate (TEOS), tetramethylorthosilicate (TMOS), and sodium silicate solution (water glass). These raw materials are more expensive and also carcinogenic. Therefore, for large-scale industrial production and for drug delivery applications, it is preferable to replace these conventional inorganic-based raw materials with an inexpensive, biocompatible, nontoxic and readily available bio-source as the raw material. Rice husk, an agricultural waste from rice mill industry, has a high silica content (~80 - 90%) and is easily available in large quantity (Kumar R. S., 2013).

In order to reduce the cost of fabrication and realize the commercial production of silica aerogels, green and cheap silica sources are very necessary. Rice husk ash are industrial wastes released by factories and thermal power plants, which consist of high content of silica. If these wastes could be converted into highly valuable silica aerogels, the environmental pollution problem caused by a stack of rice husk ash will be reduced greatly. Horticultural waste (e.g. waste leaves) could be another silica sources with 13% - 48% of silica content (Yanru L. et al., 2018).

In this study focus on the effect of pH level on the formation of silica aerogel by extracting pure silica from rice husk ash (RHA). This synthetic method provides economic benefits for the mass production of silica aerogel because it uses low-cost raw materials. The outermost layer of paddy rice is composed of about 20 wt% paddy grain, and silica comprises 20 wt% of the husk. Rice husk ash (RHA), which is produced as a by-product of combustion, contains amorphous silica. The silica aerogel has been synthesized according to the water glass solution and the effect of pH on the structure and size of silica aerogel nanoparticles was investigated.

Materials and Methods

Synthesis of Silica Aerogel from Rice Husk Ash

The rice husk (RH) was collected from a local rice mill, Yakhainekone, Mawlamyine, Mon State. Unwanted fine dust materials were removed through air-blowing separation technique and then washed properly to remove the physically adhered impurities using tap water. After successive washing, RH was dried in an air at room temperature for 72 hr to get clean and dry RH. The percentage of moisture content was measured by heating 1 gram of rice husk for 1 hr at 110℃ (ASTM D3172-89). The amount of volatile materials was evaluated by heating 1 gram of the sample for 7 min at 950℃ (ASTM 3175-02). Its ash content was determined by heating 1 gram of the sample for 4 hr at 750℃ (ASTM 3174-02) which is shown in Table 1. The rice husk was firstly dried at 700℃ for 6 hr, and grounded to pass the 150 μm sieve. The energy dispersive X-ray fluorescence (EDXRF) analysis of rice husk ash (RHA) was listed in Table 2. The rice husk ash (RHA) contains 77.34% silica, 15.98% potassium and small amount of others, such as Ca, Mn, Fe, Zn, etc. The silica aerogel was synthesized by using rice husk ash as precursor is shown in Figure 1. Five grams of RHA was mixed with 150 mL of 1 mol/L NaOH aqueous solution. The mixture was heated to 60℃ for 1.5 hr with magnetic stirring. Finally, the water glass solution was obtained by filtering out the rice husk ash. The synthesis of aerogels was started with the gelation of water glass under different pH levels such as pH 5, pH 7 and pH 9 by using sulfuric acid drop by drop. The prepared gel was aged at room temperature for 24 hr. To remove sodium sulfate obtained from the neutralization stage, the aged gel was washed three times, each time kept in deionized water for 4 hr. Then, the pretreated silica gel was directly dried at 40℃ for 2 days to yield the silica aerogel powder (Ban G., 2019). The reaction occurred as the following equation.

SiO_2 (Ash)	+	2 NaOH (Caustic soda)	$\longrightarrow$	Na_2SiO_3 (Sodium silicate)	+	H_2O (water)		
Na_2SiO_3 (Sodium silicate)	+	H_2SO_4 (Sulfuric acid)	$\longrightarrow$	SiO_2 (Silica)	+	Na_2SO_4 (Sodium sulfate)	+	H_2O (water)

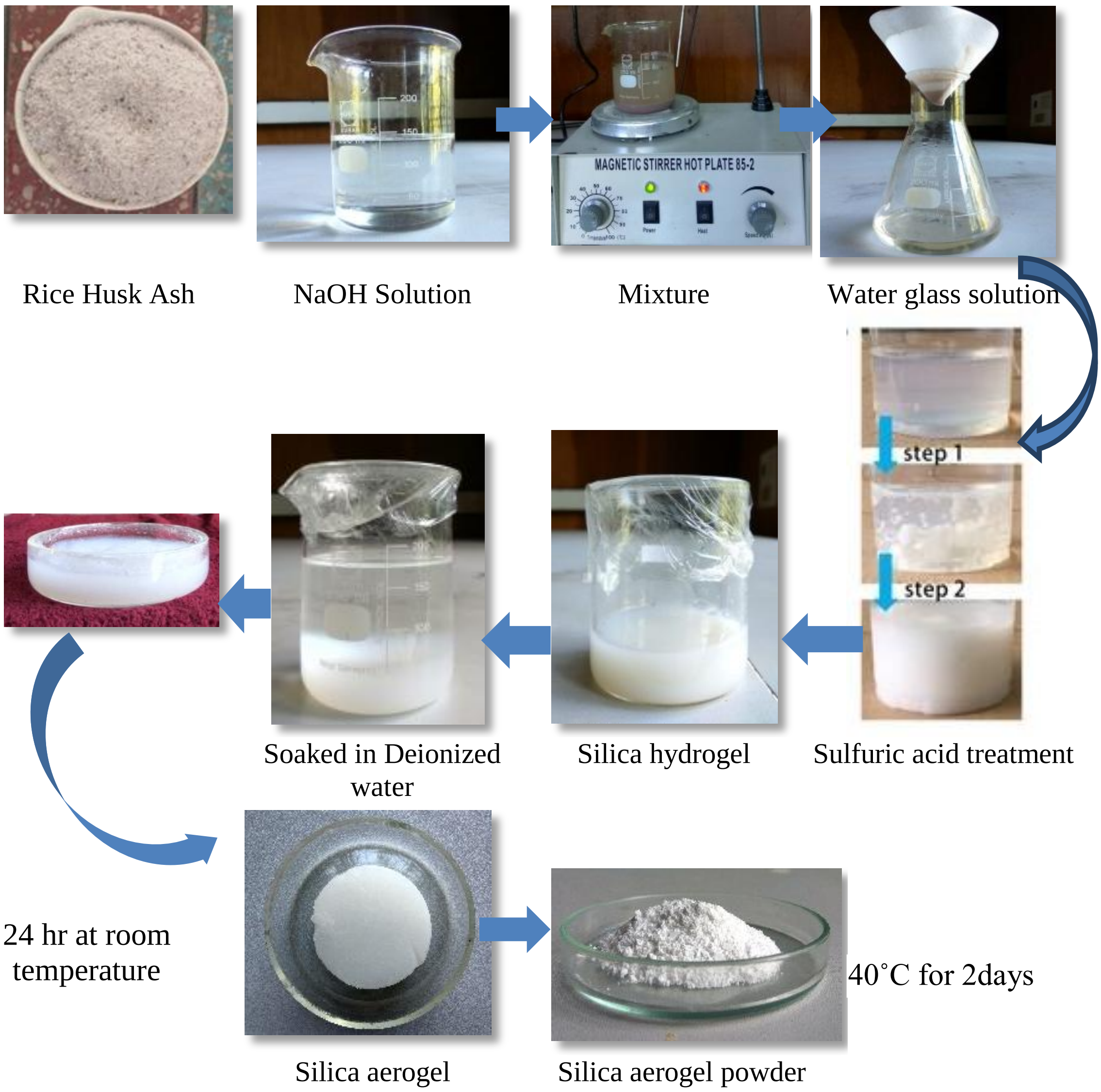

Figure 1 Synthesis of silica aerogel powder from rice husk ash

Table 1 Physical characterization of rice husk

Sample	Moisture	Volatile matter	Ash
Rice Husk (%)	10	65	25

Table 2 Elemental Analysis of rice husk ash

Elements	Si	K	Ca	Mn	Fe	Zn	Cu	Rb	Sr
Weight Percent (wt%)	77.344	15.989	3.526	1.446	1.326	0.170	0.092	0.068	0.040

Results and Discussion

Fourier Transform Infrared Spectroscopy Analysis

The FTIR spectrum of silica aerogel powder at different pH level is presented in Figure 2. The absorption peaks at 3450 cm^{-1} and 1641 cm^{-1} are due to stretching and bending vibrations of water molecules. The absorption peaks at 1091 cm^{-1} correspond to the Si−O−Si asymmetric stretching vibrations. The absorption peak at 471 cm^{-1} and the 800 cm^{-1} are corresponding with O-Si-O bending vibrations and the Si-O-Si symmetric stretching vibrations. The FTIR spectrum show almost the same patterns at different pH level such as pH 5, pH 7 and pH 9. Thus, it is confirmed that one can produce silica aerogel from rice husk ash.

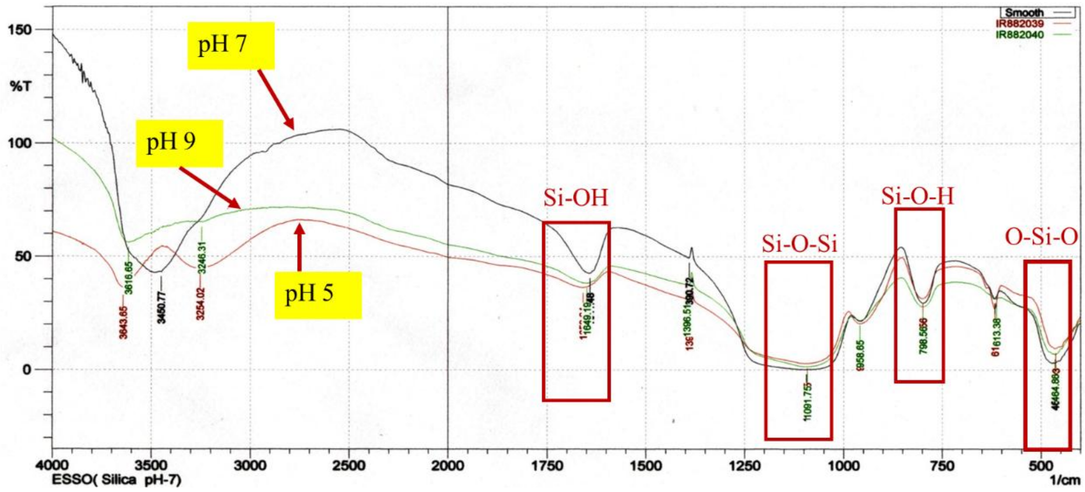

Figure 2 FTIR spectrum of silica aerogel at different pH levels

Characterization of Silica Aerogel Powder by XRD

The silica aerogel powder (SA) were synthesised at three different pH levels of pH 5, pH 7 and pH 9 to investigate the effect of pH on the quality of extract silica aerogel and particle size. The gelation time was greatly influenced by the pH level. Therefore, the gelation time decrease when the pH level increase. The XRD spectrum of product sample at pH 5, the chemical reaction is not completely and have more residual of Na_2SO_4 as shown in Figure 3(a). But the pH 7 and pH 9 , there is no chemical residual are found. The completely gelation occurred at these pH level since act as amorphous structure. The SA processing at pH 7 and pH 9, the spectrum appears as broaden with at 2θ = 22°, which indicating amorphous structure as shown in

Figure 3(b) and Figure 3(c). According to the Figures, the peak at 32˚ and 34˚ show that a few amount of Na_2SO_4 residual impurities trapped in the pores of the gel network after washing deionized water. The synthesis of silica aerogel prepared at pH 7 and pH 9 have the capability to extract more SiO_2 from rice husk ash.

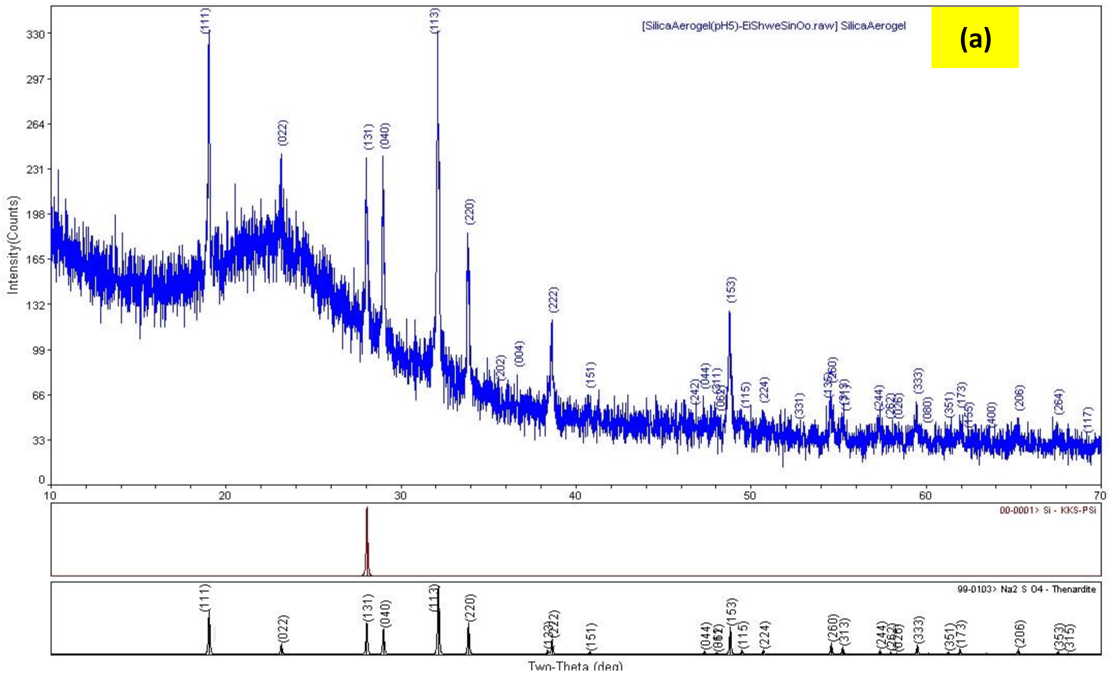

Figure 3 (a) XRD spectra of silica aerogel powder at pH 5

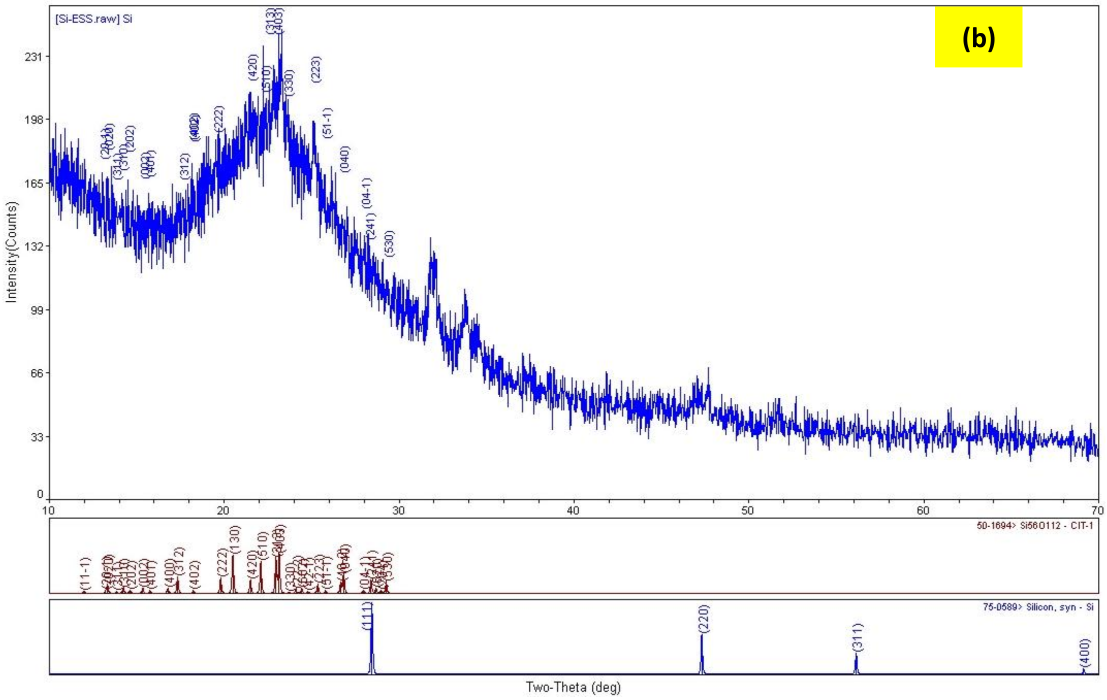

Figure 3 (b) XRD spectra of silica aerogel powder at pH 7

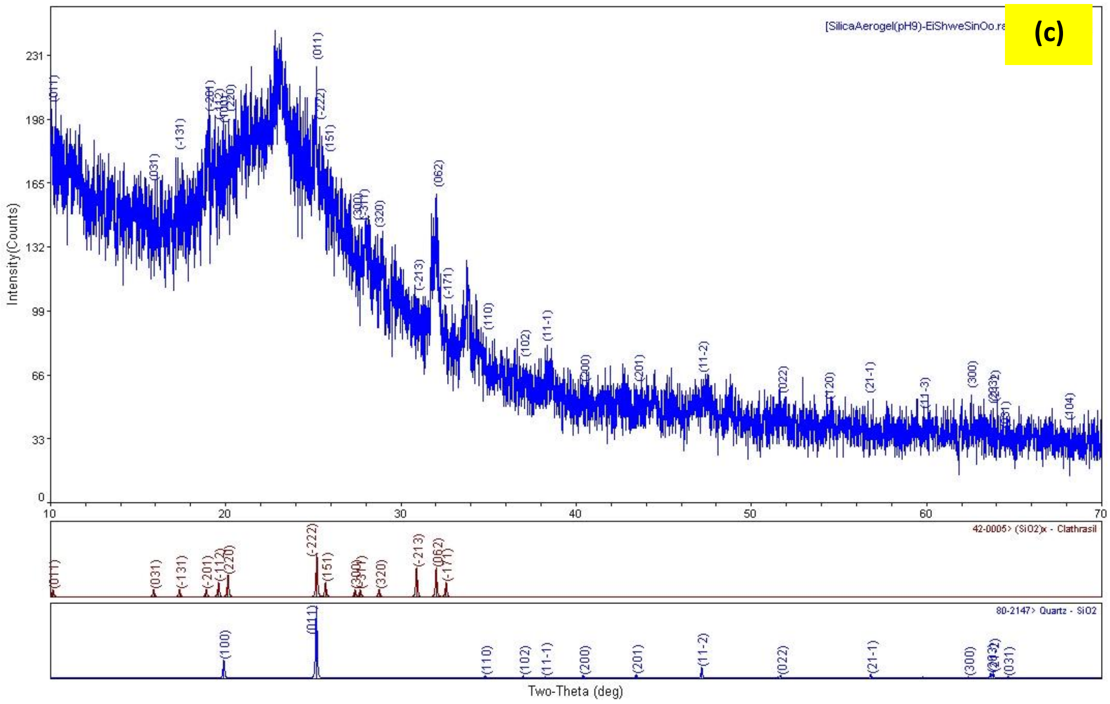

Figure 3 (c) XRD spectra of silica aerogel powder at pH 9

Characterization of Silica Aerogel Powder by SEM

The scanning electron microscope (SEM) images of silica aerogel powder synthesized at three pH levels are shown in Figure 4. The sample synthesized at pH 5 is found as flakes and is not well disperse as particles to be analyzed. At pH 7 and pH 9, the particles are although aggregate, individual particle can be distinguished and interconnect with each other to form a porous network. The average particle sizes are shown in Table 3. According to the Figure, silica aerogel had a uniform size varying from ~ 88 nm to ~ 75 nm in size at 7 and 9 pH levels. It can be concluded that the size of the silica aerogel increased at pH 5 and decreased at pH 7 and pH 9. At pH 7, silica aerogel showed nano particles in size and good morphology than others.

Table 3 Average particle size of silica aerogel

Product Sample	Average particle size
PH 5	90 nm (flakes)
pH 7	75 nm
pH 9	88 nm

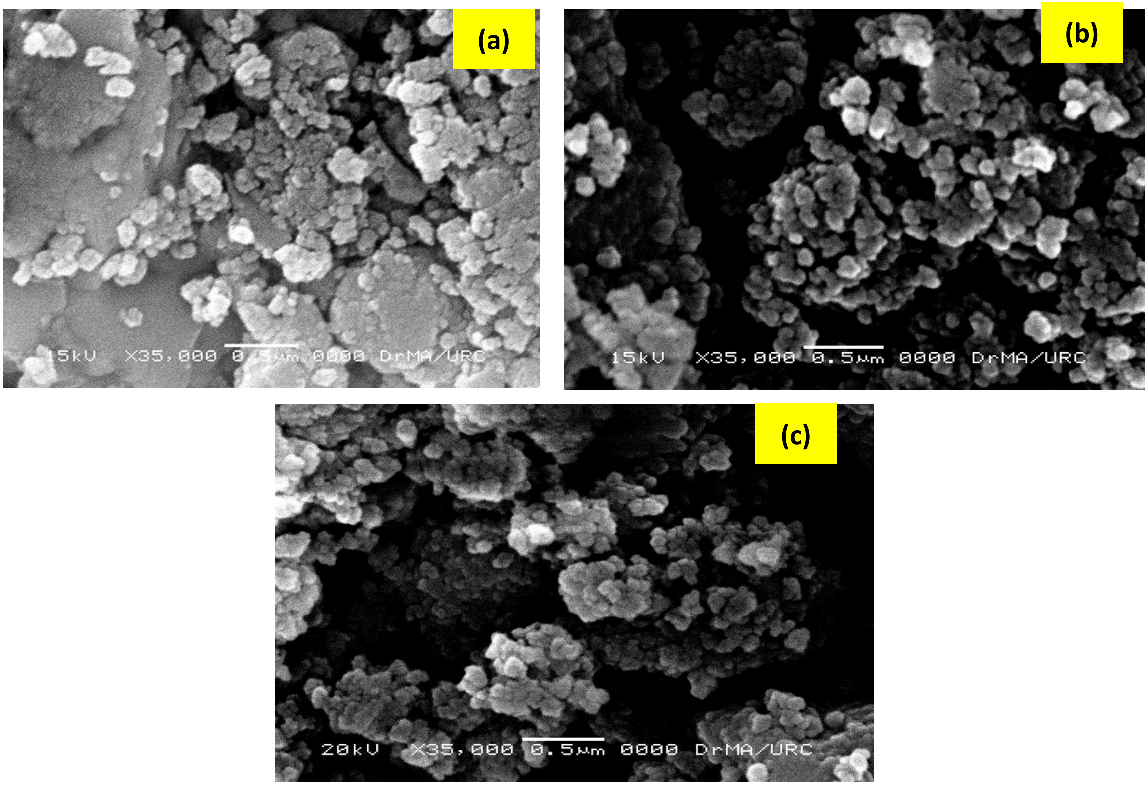

Figure 4 SEM micrographs of silica aerogel powder at (a) pH 5 (b) pH 7 and (c) pH 9

Conclusion

The silica aerogel was synthesized from rice husk ash via water glass solution by sol-gel method. Rice husk, a biomass resource left after threshing rice, is considered as economical raw material for large-scale production of silica aerogel due to its high silica content. In this study, fabrication of silica aerogel from rice husk ash under a relatively simple process was conducted with an eco-friendly, re-usable, cost-effective and safe approach compared to the conventional method. Silica aerogel was prepared by amorphous silica extracted in the form of sodium silicate from rice husk ash, and the silicate was then neutralized to form silica gel by the sol–gel process. The smallest nanoparticles size of silica aerogel synthesized using pH 7 and pH 9 showed more aggregate than large particles which are synthesized using pH 5. The silica aerogel synthesized at pH 7 showed porous structure and the smallest particle size compared with others. Therefore, the adjustment of pH level plays important role in the formation and sized silica aerogel.

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