Preparation of xerogel SiO_2 from roasted iron sand under various acidic solution

A. H. Ramelan[1], S. Wahyuningsih[2], Y. A. Ismoyo[2], H. P. Pranata[2] & H. Munawaroh[2]

[1]Electronic Materials and Energy Research Group, Faculty of Mathematics and Natural Sciences, Sebelas Maret University
[2]Inorganic Materials Research Group, Faculty of Mathematics and Natural Sciences, Sebelas Maret University
Jl. Ir. Sutami 36A, Kentingan, Jebres, Surakarta 57126, INDONESIA

E-mail: aramelan@mipa.uns.ac.id

Abstract. Xerogel SiO_2 had been prepared from roasted iron sand through variation of Na_2CO_3 addition and sol-gel process under various acidic solution. Roasting treatment was carried out on the compositional variation of iron sand:Na_2CO_3 = 1:2; 1:1 and 2:1 at 1100 ^oC. While the sol-gel process was conducted at room temperature and neutralized using HCl 0.1 M and 6 M. The color characteristics of roasted iron sand shown light brown, dark brown and dark gray of the compositional variation of iron sand:Na_2CO_3 = 1:2; 1:1 and 2:1, respectively. In addition, the levels of thoughness increased by increasing the ratio of sand in the composition of the mixture. The best composition of roasted treatment was at a variety of iron sand:Na_2SiO_3 = 1:2 with 57.72% had been dissolved in hot water. The addition of Na_2CO_3 will influence the Na_2SiO_3 formation, because of the increase of Na_2CO_3 capable produced the iron sand decomposition product. Na_2SiO_3 gel had been produced after it was neutralized with certain amount of HCl solution. The neutralization was more effective if using high concentration of HCl because of the formation of gel SiO_2 will be easier occurred. The results of SiO_2 had been identified by the FTIR spectra, which an absorption spectra of Si-O-Si asymmetric stretching at 1098.51 cm^{-1}, symmetric stretching of Si-O-Si at 804.35 cm^{-1} and the bending O-Si-O at 469.69 cm^{-1}. The result of SiO_2 content by XRF analysis is about 85.15%.

1. Introduction

Nanomaterials are experiencing a rapid development in recent years due to their existing and/or potential applications in a wide variety of technological areas such as electronics, catalysis, ceramics, magnetic data storage and structural components. Silica as a nanomaterial has been developed due to the nature of nanosilica which has high porosity and surface area so it can be used widely in the field of materials such as filler, pharmaceuticals, catalysis and chromatography [1]. Silica (SiO_2) itself is generally an abundant

element and are contained in a variety of natural resources, in an organic source such as rice husk ash which contains SiO_2 reach 86.90 to 97.30% [2, 3] and inorganic sources such as sand.

Indonesia is a country with abundant natural resources which one of them is sand. Silica is usually synthesized from quartz sands because silica content in the quartz sand high enough. Tuban sand contained 81.7% SiO_2 [4] while Bancar sand had a SiO_2 content of 69.3% [5]. Industries usually convert silica (SiO_2) in the sand to form sodium silicate (Na_2SiO_3) from melted quartz sand with sodium carbonate at a temperature of 1300 °C [6]. Sodium silicate is easily soluble in water, so that the metal oxide compounds can be separated by adding water that will dissolve the solid sodium silicate while the metal oxide remains solid form insoluble [7]. Sodium silicate is then made into xerogel through sol-gel method [8]. Sol-gel process was done by adding hydrochloric acid (HCl) to a solution of sodium silicate which was alkaline until the solution has a neutral pH [1].

This paper reported the synthesis of xerogel SiO_2 from iron sand using sol-gel method in order to widen the industrial uses of iron sand and optimized the synthesis of xerogel SiO_2.

2. Experimental methods

2.1. Preparation of iron sand

The composition of the chemical major elements of iron sand from Glagah beach was obtained from the chemical analysis determined by X-ray Fluorescence (XRF). Firstly, iron sand was treated to minimize the content of impurities in the sand then it characterized using XRF to determine the change of the content.

2.2. Pyrometallurgy process

Iron sand was added with sodium carbonate (Na_2CO_3) with the ratio 1:2; 1:1; 2:1 (w/w) and roasted at 1100 °C for 2 hours. Each 10 g roasted iron sand then dissolved in hot water.

2.3. Sol-gel process

Sol-gel process of 50 mL sodium silicate was carried out by neutralization using hydrochloric acid (HCl) in concentration of 0.1 M and 6 M. The results of sol-gel allowed to stand overnight then washed with water and dried. Synthesized xerogel analyzed using XRF and analysis of functional groups using Fourier Transform Infra Red (FTIR).

3. Results and discussions

Visually, Glagah iron sand form relatively uniform smooth black granules with slightly shiny like glass sparkle. Identification of compounds or minerals contained in iron sand was analyzed using X-Ray Fluorescence (XRF). The XRF analysis of iron sand can be seen in Table 1.

The XRF analysis shows that major elements correspond to a high proportion of Fe_2O_3 (about 48%); SiO_2 (about 18%); small content of CaO, Na_2O, TiO_2, Al_2O_3, MgO and traces amount of another elements. After iron sand was treated to minimize the content of impurities, it is known that the content of SiO_2 increased up to 38%.

Table 1. XRF analysis of iron sand

Formula	Content (%)	Formula	Content (%)
Fe_2O_3	48.88	SO_3	0.68
SiO_2	18.97	Cl	0.58
CaO	7.71	Nd_2O_3	0.27
Na_2O	6.62	V_2O_5	0.24
TiO_2	4.72	Pr_6O_{11}	0.15
Al_2O_3	4.35	Cr_2O_3	0.06
MgO	3.45	ZnO	0.06
P_2O_5	1.27	CeO_2	0.05
K_2O	1.01	SrO	0.05
MnO	0.75	SnO_2	0.04

Xerogel SiO_2 can be synthesized either by using organometallic or by the alkaline attack of the sand. In this work, the manufacturing process of silicates by dry way, called furnace process, is used. This corresponds to the fusion of silica and sodium carbonate together at high temperature in a furnace. Iron sand roasting process carried out at a temperature of 1100 °C for 2 hours with the ratio of iron sand:Na_2CO_3 = 1:2; 1:1; 2:1. Roasted iron sands was obtained with the characteristics that can be seen in Figure 1.

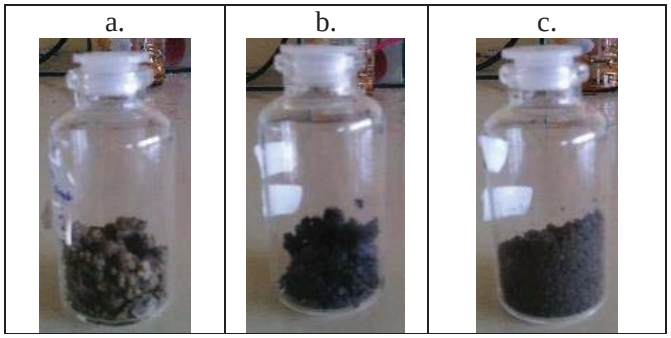

Figure 1. Roasted iron sands with the compositional variation of iron sand:Na_2CO_3 = 1:2 (a); 1:1 (b) and 2:1 (c)

In general, roasted iron sands generated molten that hardens like a rock and hard enough to be destroyed, if it compared with one another, the more addition of sand will obtain more harden solid. The color characteristics of roasted iron sands shown light brown, dark brown and dark gray of the compositional variation of iron sand:Na_2CO_3 = 1:2; 1:1 and 2:1, respectively.

At decomposition, Na_2CO_3 presence would disturb the mineral composition which Na ions trapped in the network and reduce the number of bridges/bonds between tetrahedral. Na^+ cations have an effect on the size of the hole/cavity and is expected to the formation of clusters [10]. Therefore, more or less Na_2CO_3 were added to the iron sand affects the decomposition of sand that affected the differences in color characteristics and thoughness as the result of roasting.

Table 2. Results of roasted iron sands dissolution

Compositional ratio iron sand:Na_2CO_3 (w/w)	**(1:2)**	**(1:1)**	**(2:1)**
Rendement mass (g)	4.228	8.127	9.171
Dissolution efficiency (%)	57.72	18.73	8.29

In Table 2 shows that the most soluble roasted iron sands is the variation of iron sand:Na_2CO_3 = 1:2 with dissolution efficiency reach 57% from 10 g roasted iron which has been dissolved. These additional variation is the most effective ratio in decomposing minerals in the sand and generates more sodium silicate seen from the dissolution efficiency.

Sol-gel process in this work has been done by adding a variation of 0.1 M HCl and 6 M to neutralize the alkaline solution of sodium silicate and the results can be seen in Figure 2.

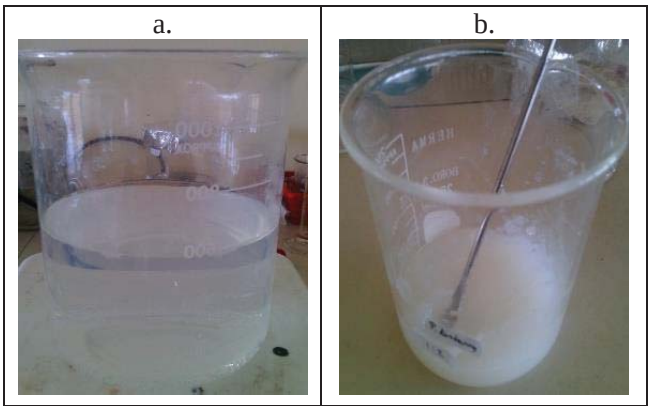

Figure 2. Neutralization results of sodium silicate with 0.1 M (a) and 6 M (b) HCl

The neutralization of sodium silicate solution with 6 M HCl is more effective because at pH 9 xerogel SiO2 is formed and requires only small addition of aqueous HCl. While the use of 0.1 M HCl less effective because at neutral pH only formed very little gel floating in solution and the neutralization required lots aqueous HCl. The use of concentrated HCl produces H^+ ions with large numbers but in a minimal presence of solvent so it would be more effective in catalyzing sodium silicate in the sol-gel process to form xerogel. Content analysis of xerogel is performed by XRF and can be seen in Table 3.

Table 3. XRF analysis of xerogel

Formula	**Content (%)**	**Formula**	**Content (%)**
SiO_2	85.15	V_2O_5	0.16
Al_2O_3	3.41	Fe_2O_3	0.06
K_2O	2.98	ZrO_2	0.02
Cl	2.62	BaO	0.01
P_2O_5	2.33	CuO	0.01
SO_3	1.81	ZnO	0.01
CaO	1.39		

Table 3 shows the final content of SiO_2 reach 85% with other contents as impurity. To clarify the final results, functional groups analysis of xerogel is done using FTIR and can be seen in Figure 3.

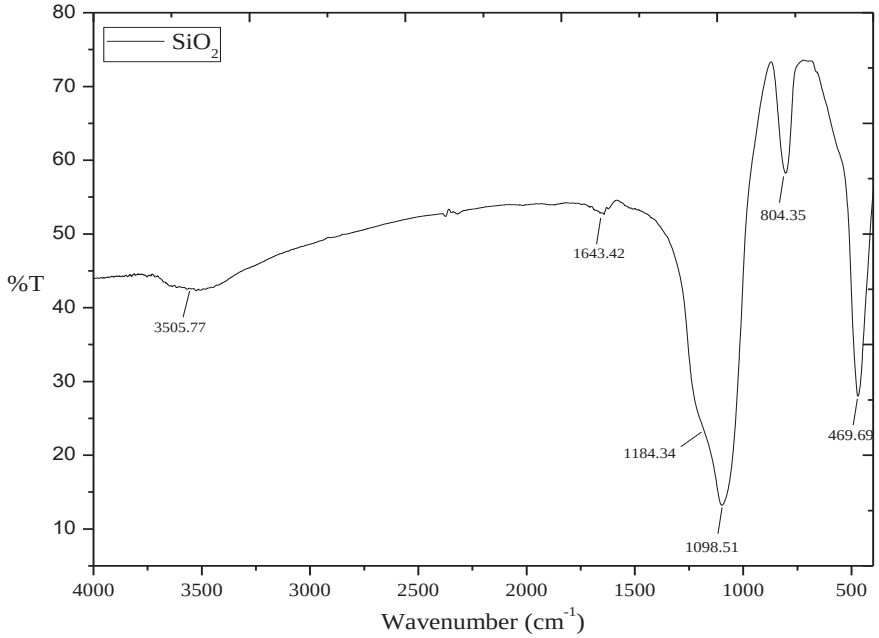

Figure 3. Infra red spectra of xerogel

The results of SiO_2 had been identified by the FTIR spectra, which an absorption spectra of Si-O-Si asymmetric stretching at 1098.51 cm^{-1}, symmetric stretching of Si-O-Si at 804.35 cm^{-1} and the bending O-Si-O at 469.69 cm^{-1}. These absorption characteristics in accordance with the research of Musić et al. (2011), which identifies amorphous SiO_2.

From this evidence, it can be assumed that the xerogel is relatively pure SiO_2 because no other group absorption was analyzed. Impurities were detected on XRF analysis only physically interact and bond does not occur so that does not affect the functionality of the compound of SiO_2. Meanwhile, it appears quite weak absorption at wavenumber 1643.42 and 3505.77 cm^{-1} is the vibration profile of –OH stretching –OH bending which indicates relatively hygroscopic properties of amorphous SiO_2 produced.

In conclusion, xerogel SiO_2 can be synthesized from iron sand with the final result contain 85.15% SiO_2. The more addition of Na_2CO_3 will increase the decomposition of iron sand, with the optimum ratio of sand:Na_2CO_3 = 1:2. While the formation of xerogel is more effectively done with a high concentration of HCl.

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