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Study of Acid Leaching in the Preparation of Silicon from Lapindo Mud

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***ABSTRACT**: Silicon was prepared by metallothermal reduction of high purity silica obtained from Lapindo mud. Silica was extracted as sodium silicate from Lapindo mud using NaOH solution and followed by titration with HCl to produce silica alcogel. Then it was washed and dried to obtain silica xerogels. The silica was characterized using X-ray diffraction and X-ray fluorescence. Silicon was prepared from metallothermal reduction of the silica at 650 °C for 3 hours using magnesium (99% purity) as a reducing agent. The product was then subjected to two stages of acid leaching sequence, first leached with HCl:HF solutions followed with HF:CH_3COOH solutions. The product of each leaching step was characterized using X-ray diffraction. The structure of the silica was amorphous and its purity was found to be at 98.1%. The results showed that the reduced xerogels contained silicon, Mg_2SiO_4, MgO and Mg_2Si. First leaching step were proved capable to remove Mg_2SiO_4 and MgO phases but both of leaching step unable remove Mg_2Si in reduction product. The final product showed silicon was the major phase whereas Mg_2Si was the minor phase.*

Keywords: *Acid leaching, Lapindo mud, Metallothermal reduction, Silicon.*

I. INTRODUCTION

Worldwide concern over the rising cost of fossil fuels in the recent years has increased interests in fossil fuel alternatives. Solar photovoltaic power generation (PV) have found many applications in the industry, especially in the aerospace industry, but the high cost associated with the materials used in fabricating the solar cells becomes the limiting factor to the widespread use of solar photovoltaic power. Currently silicon in both of monocrystalline and polycrystalline form is the dominant semiconductor material used in the fabrication of most commercially available solar cells and commanding over 90% of the market share of all PV technologies [1].

Metallurgical grade silicon with a purity of 98% has been generally prepared from carbothermal reduction of quartz (SiO_2) in electric-arc furnace at high temperature of about 1900 °C [2]. The purity of silicon can be increased by using purer starting materials, by improving the furnace construction, and by optimizing the production process. In order to achieve its economic feasibility, the high temperature used in the production of silicon should also be reduced. For this reason, this study's motivation was to find alternative reaction routes to produce high purity silicon that required low reaction temperature.

Lapindo mud is an active spurt of hot mud from a drilling location of Lapindo Brantas Inc. in Sidoarjo Regency, East Java, Indonesia, which constitutes as a national disaster. The source of hot mud is from the leakage in the drilling wells of the exploration activities of oil and gas. This hot mudflow has inundated agricultural fields, industrial and residential area, public facilities and others. The mud has been found to be rich in silica by Fadli [3] while Munasir had also successfully synthesized amorphous silica from Lapindo mud with 97.8% purity [4]. Therefore, it could be inferred that Lapindo mud has the potential to be used as raw materials for preparing silicon.

The synthesis of high purity silicon from natural substances has been performed by several researchers. High purity silica obtained from natural materials is reducible to silicon using metals like Mg, Ca, Ba and Al as reducing agent [5-10]. Using metal as reductor, presents a new set of problems. Because the metal used as reductor is still present in the final product therefore reducing the purity of the final silicon product. The presence of impurities in silicon wafer is known to have lower performance and lifetime if used as the semiconductor in photovoltaic system. So the aim of this work is to study of acid leaching in the preparation of silicon from Lapindo mud.

II. EXPERIMENTAL

1. Synthesis of high purity silica

Silica was extracted as sodium silicate from Lapindo mud using NaOH solution. Lapindo mud was sifted then stirred in NaOH 6 M solution for 5 hours at 90 °C. The sodium silicate extract was then titrated with HCl until the pH reached 8 to produce silica alcogel. It was washed and dried in 120 °C for 10 hours in order to obtain silica xerogels. The washed (WS) and unwashed (US) silica was characterized using X-ray diffraction (XRD) and X-ray fluorescence (XRF).

2. Silicon preparation

Metallothermal reduction process was conducted during the preparation of silicon. 6.18 g high purity silica and 5.00 g Mg powders were pounded homogeneously to obtain fine powder using agate mortar. The mixture was pyrolyzed in a tube furnace at 650 °C for 3 hours with a heating rate of 10 °C/min. A portion of the obtained brownish powder (Si-unleach) was analyzed by XRD.

3. Leaching of reduction products

The product from the previous step was leached in two steps sequence for purification. The first leaching step was designed to remove magnesium compounds and other impurities present by using a mixture of (4:1 v/v) HCl:CH3COOH solution in order to ensure the effective dissolution of magnesium compound (Si-leach 1). The second leaching step was designed to remove silicates and other impurities that may be present using a mixture of (1:9 v/v) HF:CH3COOH (Si-leach 2). The leaching process was carried out at 70 °C for one hour. After each steps, the leached slurry was filtered through Whattman filter paper and washed with water followed by leaching in deionized water for an hour and dried at 100 °C. The dried residue recovered from each step was then characterized using XRD.

4. Material characterization

The crystalline phases of the material were determined using X-ray Diffractometer (XRD Shimadzu 6000) using Cu filter (λ= 0.15 nm) under operating condition of 40 kV and 30 mA. X-ray Fluorescence Spectrometer (XRF PANalytical MiniPal 4) was used to determine the chemical compositions of the material under the operating conditions of 7 kV under air and helium atmosphere.

III. RESULTS AND DISCUSSION

Chemical analysis of unwashed and washed silica from Lapindo mud was carried out using XRF. Table 1 compares the chemical composition (oxide basis) of Lapindo mud, unwashed (US), and washed (WS) silica. From the chemical compositions data, Lapindo mud was found containing approximately 50.1% SiO2 and oxides of aluminium, potassium, calcium, barium, manganese and titanium (46.8%). In exception for SiO2, all other compounds present in the mud may be considered as impurities and it was also known that the brown colour of Lapindo mud was attributed to the presence of these impurities. Comparing the chemical compositions data of the WS and US, the rinsing process was found to be effective in removing NaCl. Because the decreasing number of impurities, the silica content of the washed silica raised from about 39.4% to 98.1%.

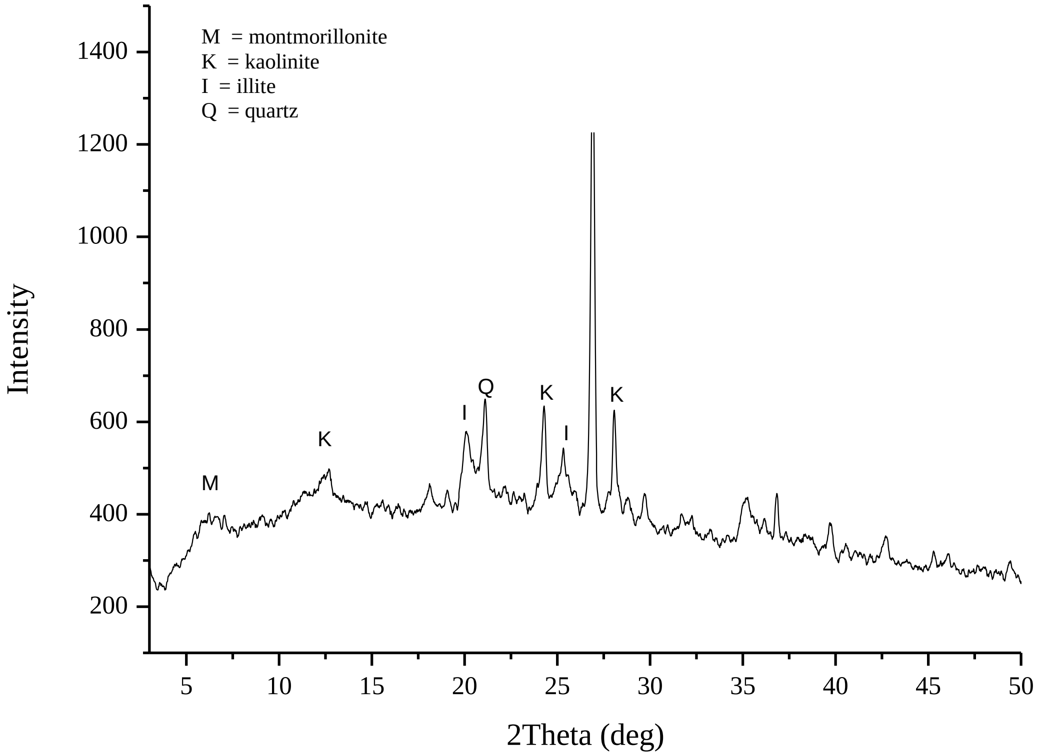

Fig. 1. X-ray diffratogram of the Lapindo mud

Table 1. Chemical composition of materials (%wt)

Sample	SiO_2	Al_2O_3	Cl	CaO	K_2O	TiO_2	MnO	BaO
Lapindo mud	50.11	14.39	-	10.44	4.82	3.93	7.74	3.90
US	39.38	-	48.86	0.67	0.44	0.06	-	-
WS	98.10	-	-	0.68	0.17	0.07	-	-

As shown in Fig. 1, montmorillonite is the major mineral in Lapindo mud and kaolinite is the second most abundant mineral. Small amounts of quartz and illite are also present. High purity silica was prepared by acid neutralization of sodium silicate obtained from Lapindo mud. HCl was used during the neutralization process. Fig. 2 compares the XRD patterns of US and WS. The US exhibits the presence of NaCl and Na2SiO3, as shown in Fig. 2 (a). However, those crystalline peaks were completely absent in the washed gel (Fig. 2 (b)) and was amorphous in nature. A rather broad peak in 2θ angle of 22° which is the characteristic pattern of amorphous was also observed.

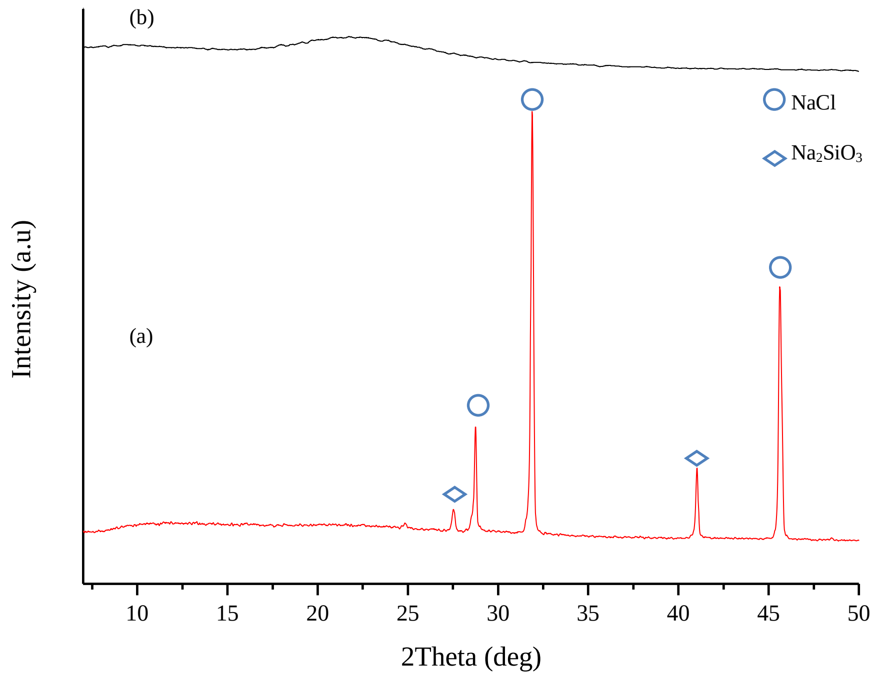

Fig. 2. XRD patterns of silica (a) unwashed and (b) washed.

The XRD patterns of Si-unleach are shown in Fig. 3 (a). The result clearly exhibits that the Si unleach contains several magnesium compounds such as Mg2SiO4, MgO, and Mg2Si. The XRD patterns of the leached powder of the one step and two steps leaching process are shown in Fig. 3 (b) and (c) respectively. Qualitatively, it can be seen by comparing the XRD patterns of Si-leach 1 and Si-leach 2, the leached silica lacked the peak intensity of the Mg2SiO4 phase, had decreased MgO and Mg2Si peak whilst the peak intensity of silicon appeared to be increased. Leaching using HCl and CH3COOH were found to be able to remove Mg2SiO4 and MgO phase from reduction product, because the chloride and acetate salts of magnesium have high degree of dissolution in aqueous medium. Both steps were unable to remove Mg2Si found in the product. It was because that the activation energy for the dissolution of Mg2Si was very high. The formation of Mg2Si can be mitigated by increasing the leaching temperature.

Table 2 compares the mineral composition of reduced and leaching products. The percentage of mineral composition calculated by comparing the peak intensity of the mineral to the total intensity of product. The composition of silicon reached 20.6% in the reduced products and increased to 94.4% in the first leaching product but decreases to 79.9% in the second leaching product. It was because silicon have high degree of dissolution in HF solution.

Tabel 2. Comparison of the mineral phase available in Si-unleach, Si-leach 1, and Si-leach 2

Sample	Phase (wt %)			
	Silicon	**Mg_2Si**	**MgO**	**Mg_2SiO_4**
Si-unleach	20.6	5.99	39,8	33,7
Si-leach 1	94.4	3,45	2,13	-
Si-leach 2	79.9	13.1	6.99	-

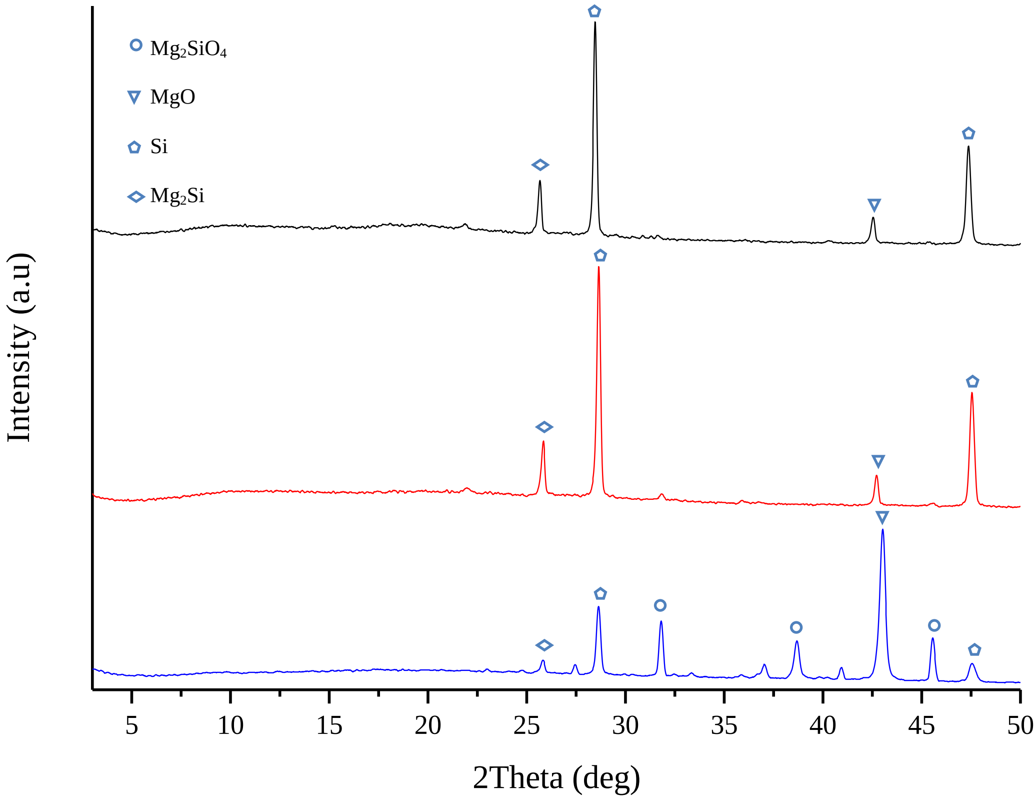

Fig. 3. XRD patterns of (a) Si-unleach, (b) Si-leach 1, and (c) Si-leach 2

IV. CONCLUSION

Silicon has been successfully prepared from Lapindo mud as raw materials using metallothermal reduction process. The reduction of silica was carried out with magnesium as the reducing agent under low temperature. Silica of high purity (98.1%) was obtained from Lapindo mud. Two leaching steps were carried out to obtain a high purity silicon. The final products have silicon as its major composition and Mg2Si as the minor phase.

V. ACKNOWLEDGMENT

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