



Impurities removal process for high-purity silica production from diatomite

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ABSTRACT

This study presents a novel chemical process for the extraction of high purity silica from diatomite. The process involves leaching of raw diatomite with acid prior to alkali extraction, solubilization of treated diatomite with alkali solution to form sodium silicate solution, and precipitation of silica gel by adding sulfuric acid. Besides, the process includes washing of silica gel with hot solution of 6 M hydrochloric acid for three hours. The morphology and the chemical composition of the raw diatomite, the derivative products, and the final purified silica were determined by scanning electron microscopy, X-ray fluorescence, X-ray diffraction, and inductively coupled-plasma optical emission spectroscopy. The results indicate that raw diatomite contains over 80 wt% of amorphous micro-porous silica (SiO₂) with particle size, 10–35 μm. We were able to enrich the content of silica in diatomite to 90.2 wt% by step-wise acid leaching. Moreover, in this study, boron (B) was removed from silica slurry using mannitol as a complexing compound. The elimination ratio of boron based on this method was about 70%. Incorporation of mannitol in the final stage of the acid washing of silica gel led to a silica product with lower B content. The final purified silica is 99.99 wt% (four nines) pure.

1. Introduction

Making solar-grade silicon with 99.9999 wt% (six nines) and achieving precise impurities control is a crucial step in silicon based solar cell industry. Initially, the natural silicon materials (silicon dioxides or silicates) are reduced with carbon in electric arc furnace (Pizzini, 1982). The reduced silicon called metallurgical silicon (MG-Si) is 99 wt% (two nines) pure with major impurities of iron (Fe), aluminum (Al), titanium (Ti), vanadium (V), boron (B), and phosphorus (P). To arrive at the six nines level, the required purity for photovoltaic, the MG-Si usually has to go through multiple purification processes. There are various purification techniques, for instance the acid leaching, the reactive gas blowing, and the slagging processes etc. However, these processes require a considerable amount of energy, and generally involve toxic and corrosive reactions (Pizzini, 1982; Bathey and Cretella, 1982; Istratov et al., 2006; Bathey et al., 1992; Reznichenko, 2016; Andrews and Clarson, 2015; Bye and Ceccaroli, 2014). From the viewpoint of the production cost and simplicity, before carbothermal reduction, a pre-purification of the natural Si-sources deems preferred

as it could be done by chemical processes at lower temperatures. Furthermore, pre-purification enables boron (B) and phosphorus (P) impurities to be initially removed from silicon dioxides (or silicate) by acid leaching. Since the content of boron and phosphorus elements in silicon is stricter than other impurities due to boron and phosphorus are used as doping element in solar cell. In addition, physicochemical properties of boron and phosphorus are very similar with silicon, which lead to remove them from silicon very difficult. Thus, pre-purification makes it possible to produce up-graded metallurgical silicon (UMG-Si, ~ 99.999 wt%) through reduction (Istratov et al., 2006; Andrews and Clarson, 2015).

Currently, silica is produced by autoclaving the quartz sand (sand stone) with strong alkali solution to form silicate solution, and subsequently forming silica gel by adding acid to lower the pH to < 10. (Iler, 1979; Seki et al., 1987; Tsugen et al., 1993; Iosef, 1995; Guo et al., 2003; Wason, 1976). The purification of the sandstone is generally preceded by a grinding-step in a ball mill or jet mill machine for several hours (Loritsch and James, 1991; Veglio et al., 1999; Du et al., 2011; Krauskopf, 1956; Okamoto et al., 1957; Rimstidt and Barnes, 1980;

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Rimstidt, 1997; Mora-Fonz et al., 2007). The grinding is essential to minimize the size of the silica particles (from millimeter to micrometer scale) for better chemical reaction during purification process. Purity of the silica-precipitate obtained by this method is 99% (two nines) (Seki et al., 1987; Tsugen et al., 1993; Guo et al., 2003; Wason, 1976; El Bouseily et al., 1975; Bessho et al., 2009; Fedorockova et al., 2015; Rohani et al., 2016). Since, purity of the refined silica is still much below the solar-grade, various efforts are currently under way by the researchers to increase the purity. Besides the non-uniform physical distribution of quartz sand source in the world, grinding procedure of quartz sand is a energy-consuming method, and also may introduce contamination from the milling container. In this connection, research to explore different natural silica source were conducted (Bessho et al., 2009; El Bouseily et al., 1975; Rohani et al., 2016; Muhammad et al., 2016; Meradi et al., 2009).

Among the solutions, diatomaceous-earth has been suggested as one of the most promising, stable, and steady sources to extract pure silica for the production of SOG-Si (Bessho et al., 2009). Diatomaceous earth (or diatomite) is a siliceous sedimentary rock mainly consists of fossilized diatoms, a kind of Algae. It is chemically composed of mainly SiO₂ (silica) ranging from 80 to 95%, and can be an economically viable raw material for the production of pure silica (Bessho et al., 2009). Silica contained in diatomite is mainly amorphous in type with an average diameter of silica particles less than 50 µm featuring high porosity, low thermal conductivity, high melting point, and chemical inertness. The high solubility of amorphous silica under basic conditions enables silica to be extracted from diatomite at low temperature. Moreover, no grinding or crushing processes are required as the size of silica particles is nearly small enough to react sufficiently with chemicals. Diatomite has numbers of industrial applications, it has been used for filtration absorbents, filler, and insulation material due to its micro-porous structure and above mentioned chemical stability. In this work, we propose a simple and low-cost chemical process to obtain high-purity silica from diatomite collected from Algeria. Diatomite was subjected to acids leaching and alkali solubilization processes in order to access the effectiveness of each individual stage in reducing the mineral content of diatomite silica. In addition, this paper examines application of mannitol, poly-hydroxyl alcohol, as a complexing compound for removing B from silica gel.

2. Experimental

2.1. Materials and chemicals

Natural diatomite source material was collected from mine of Algeria. The standard solutions for inductively coupled plasma optical spectroscopy were obtained from Sigma-Aldrich. Hydrochloric acid (HCl, 37%), Nitric acid (HNO₃, 65%), Sulfuric acid (H₂SO₄, 98%) Hydrofluoric acid (HF, 46%), Sodium Hydroxide (NaOH, 99%) and Mannitol polyhydroxy alcohol were obtained by Wako Chemicals. All the chemicals used in this study were of analytical reagent grade.

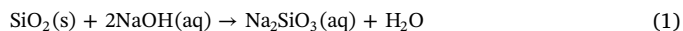
2.2. The purification of diatomite

2.2.1. Step (1): HNO₃ acid leaching

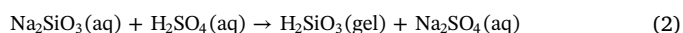
Shown in Fig. 1 is the flow sheet of the complete purification procedure. At first, an acid leaching stage was used to remove small quantities of impurity minerals prior to silica extraction from diatomite. 10 g of raw diatomite sample were leached in 2.2 M HNO₃ acid solution at 60 °C for three-hours under constant stirring. HNO₃ quantity was added gradually during the three-hours of leaching. The dispersion was filtered, and then the diatomite residue was washed several times with water. The residue was used for the extraction of silica.

2.3. Step (2): preparation of silica gel

10 g of leached diatomite sample (i.e., SiO₂ (s)) was transferred to a Teflon covered beaker containing 40 ml of 2.5 M NaOH solvent. The solution was kept at elevated temperature of 80 °C for two-hours under magnetic stirring to dissolve the silica (i.e., SiO₂ (s)) producing sodium silicate (Na₂SiO₃) solution as of reaction (1). The Eq. (1) is the general solubilization reaction of SiO₂ in aqueous NaOH.



To remove residual SiO₂, obtained Na₂SiO₃ solution was centrifuged several times, and then collected in a Teflon beaker. After that Na₂SiO₃ (aq) solution was cooled down to room temperature, then solution was titrated with 3.5 M H₂SO₄. The precipitation experiment has been carried out under constant stirring to avoid any coagulation. The experiment has been progressed as of reaction 2. Silica gel (Si (OH)₄) started to precipitate during the precipitation experiment when pH of the solution decreased below 10 (reaction 2). Due to constant stirring, formed silica gel eventually makes silica-slurry.



2.3.1. Step (3): HCl acid washing

To obtain higher purity, the silica-slurry (Si (OH)₄) was then dispersed in 80 ml of 6 M HCl solution, and stirred for three hours at 60 °C. The obtained acid leached solution was used for the next purification step.

2.3.2. Step (4): Removal of boron

For additional B elimination, we added 1 g of mannitol poly-hydroxyl alcohol to the HCl acid solution, and subsequently 100 ml of 0.25 M NaOH solution was added to the gel. The slurry was then centrifuged for 15 min at 2500 rpm. The clear supernatant was discarded, and the washing step was repeated with deionized water.

2.3.3. Step (5): second-time purification

The precipitated silica was subjected to the second-time purification. The slurry resulting from this stage was transferred into a beaker and dried at 80 °C for 12 h. Major advancements in the present method were incorporation of: (1) an initial acid leaching, (2) mannitol compound to eliminate B from silica gel, and (3) final washing method of the silica slurry.

2.4. Characterization

The morphology of the raw diatomite and purified diatomite were observed under JEOL JSM5000 scanning electron microscopy (SEM). The Crystalline phases was identified by PANalytical X'Pert PRO system at θ-2θ mode using CuK_α radiation with generator voltage of 45 kV, and current of 40 mA. The mineralogical analysis of diatomite was performed using inorganic material-database, AtomWork (Xu et al., 2011) and MAUD (Materials Analysis Using Diffraction) software which is based on Rietveld method combined with Fourier analysis. Si (2 wt%) was added as internal standard to determine the amorphous content. Elemental analysis of the raw diatomite, derivative products, and the final purified silica was carried out using inductively coupled plasma optical emission spectroscopy (Shimadzu Inc. 81,000, Japan). Up to 23 elements including B, P, Al, Fe, and Ca were measured. Prior to the ICP analysis, samples were prepared in fume cupboard as follows: A small amount of solid sample (50 mg) was crushed in small alumina mortar and then transferred into a wide-mouth Teflon vial. HF acid (400 µl, 46–48 v/v. %) and hot HNO₃ acid (200 µl) were added successively to the diatomite sample. Then the vial was placed on a hotplate for 12 h for the digestion of silica. To prevent any evaporation of B-element, one mg of mannitol was added to the digest. The digest was then diluted to 5 ml with pure water and kept at stand-still for 24 h at room

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