

PURIFICATION OF TABUK SILICA USING CHEMICAL ATTACK AND THERMAL TREATMENT

R. BOUSBIH^{a,b,*}, I. HADDADI^b, H. BEN ZINA^a, N. ALATAWI^a,
H. EZZAOUIA^b

^a*Department of Physics, Faculty of Science, University of Tabuk, Kingdom of Saudi Arabia*

^b*Photovoltaic Laboratory Research and Technology Centre of Energy, Borj-Cedria Science and Technology Park, BP 95, 2050 Hammam-Lif, Tunisia*

We investigate a purifying process of silica which plays a critical role as a starting material in the industry of glass and high-grade silicon. This method is a subsequent combination of thermal treatment and acid leaching. First, Tabuk natural sand samples were submitted to rapid thermal annealing in an infra-red furnace under O₂ atmosphere at fixed temperature 900 °C during 1 h (to weaken impurity –Si bands). Subsequently, the samples were soaked in an aqueous acid solution composed of hydrofluoric acid and nitric acid concentration. This last acid leaching step was intended to remove the gettered impurities during the annealing step. Chemical compositions of the treated samples were analyzed by energy-dispersive X-ray diffraction (XRD) and energy dispersive X-ray (EDX) spectroscopy. Surface morphology was determined by scanning electronic microscopy (SEM). The result indicates that all undesirable impurities in silica were successfully removed.

(Received July 4, 2020; Accepted September 22, 2020)

Keywords: Sand, Silica, Purification, Annealing thermal, Acid leaching, XRD, EDX

1. Introduction

Silica is the natural form of silicon dioxide (SiO₂), which is a white or colorless crystalline compound found mainly as quartz, sand, flint, and many other minerals. Silicon is the second most abundant element in the Earth's crust. It's extracted from the quartz which is a crystalline form of silica [1]. The production of silicon by the thermal reduction of silica with carbon is an industrial process used since the beginning of the 20 century [2,3]. Unluckily, the silicon produced using this method contains many undesirable impurities that limit its use in a photovoltaic application which remains a key issue for industrial production [4,5]. Many work try to purifying the Silica to produce high purity silicon with different techniques such as Acid leaching technique [6], ultrasound process with surface cleaning, reserve flotation technique, and carbothermic reduction [7]. Khalifa and al proposed an innovative technology using thermal treatment and acid leaching process that leads to a reduction in all metallic impurities improving the purity in silica from 99.7% to 99.9% [8, 9].

In this study, we used a combined treatment based on a chemical attack by HF and HNO₃ followed by thermal heating to improve the quality of purified silicon.

The structural property of the sample was examined using energy dispersive X-ray diffraction (XRD), energy dispersive X-ray (EDX) spectroscopy, and surface morphology by scanning electronic microscopy (SEM).

* Corresponding author: bousbih2014@gmail.com

2. Experimental

In this work, we report on a process of silica purification intended to be used in photovoltaic applications. Silica powders used in this study as the starting material are natural sand from the region of Tabuk in Saudi Arabia. Before purification process samples were ultrasonically cleaned for 10 min in deionized water, alcohol, successively, and then dried in air at room temperature. Therefore, after the cleaning process, samples were purified with chemical treatment HF and HNO₃ to remove metallic impurities located at the surface of silica grains followed by thermal heating at in infrared furnace at a temperature of 900 °C.

Chemical composition of the treated samples was analyzed by energy-dispersive X-ray (EDX) spectroscopy and dispersive X-ray diffraction (XRD), and the surface morphology by scanning electronic microscopy (SEM).

3. Results and discussion

3.1. Energy dispersive X-ray (EDX)

The energy dispersive X-ray technique was used to identify the elemental composition of silica. Fig.1 (a) and (b) shows EDX spectra for untreated Silica and purified Silica, respectively. The chemical composition of the silica before purification (red sand) Fig. (1.a) shown that many undesirable impurities were present with different concentrations like Magnesium (Mg), aluminum (Al), Irons (Fe), and Calcium (Ca) [10,11]. Whereas it can be seen from Fig. (1.b) (white sand) that these undesirable impurities are successfully eliminated after purification processes. These metal impurities introduce deep levels in silicon, recombining the minority carriers, making their diffusion length decrease, and impacting the solar cell efficiency.

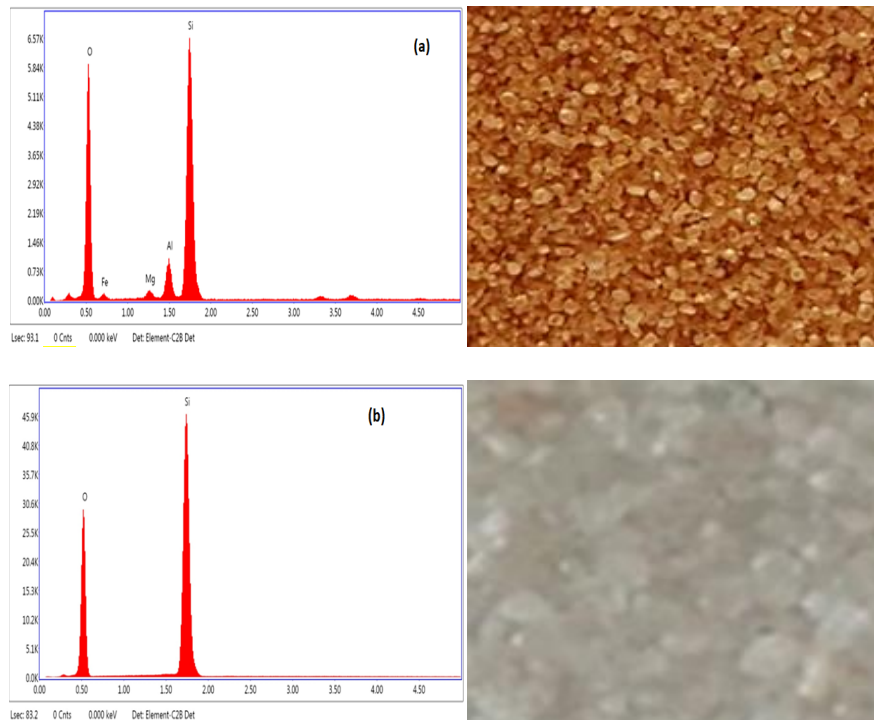


Fig.1.EDX spectra and photograph (a) untreated sand and (b) purified sand.

3.2.X-Ray diffraction

The structural property of the sample was examined by X-Ray diffraction XRD technique using a Bruker D8 advance X ray diffractometer with CuK α radiation ($\lambda_{\text{CuK}\alpha} = 0.15406 \text{ nm}$)

operating at current and tension of 40mA and 40kV, respectively. The pattern before purification shows the presence of two phases rhombohedra (Card no. 00-024-0072) [12] and hexagonal (Card no. 100-046-1045) which are specified for hematite and quartz with a lattice parameter of an axis about 5 (Å), b axis 5 (Å), c axis 13,7 (Å) respectively. The alpha-beta and gamma were 90 degrees and 120 degrees (Fig 2.a) calculated from as well as, the pattern visualizes the appearance of the most common peaks related to SiO_2 at $2\theta = 20.52^\circ$ and 26.86° , besides the peaks related to other impurities like Fe_2O_3 and Al_2O_3 . However, after purification, the pattern (Fig. 2, b) shows the existence of the SiO_2 peaks only which means the excellent purification of the sand. The crystal phase of this type of silica hexagonal with a lattice parameter of a-axis about 4.9 Å, b-axis- 4.9 Å, and c-axis 5.4 Å respectively [13]. The alpha, beta, and gamma were 90, 90 and, 120 degrees, respectively. As well as the calculated density 2,65 g/cm³ and the volume of cell $113,01 \times 10^6 \text{ pm}^3$.

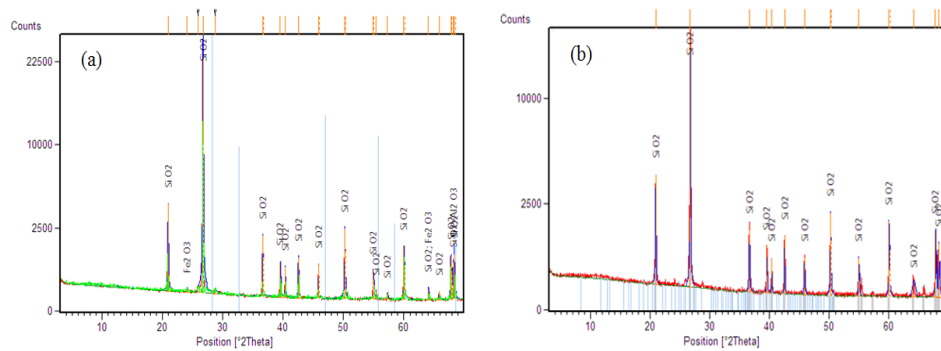


Fig.2. (a) XRD spectra untreated sand and (b) XRD spectra purified silica sand.

3.3. Scanning electronic microscopy:

Fig (3.a) shows scanning electronic microscopy images for silica surface morphology extracted before the purification process. The micrograph is observed to show the agglomeration of silica with irregular particle shapes, which varies in sizes and forms. We can notice from Fig (3.b) for the purified silica, that the size of the silica particle decreases and the form of the particle changes after the purification process, which can be explained by step 1: the thermal annealing and step 2: acid leaching effect. The first purification step leads to a gathering of impurities from the volume to the surface by rapid thermal annealing and the second one (the chemical step) aims to remove metallic impurities located at the surface of silica micro-grains [8].

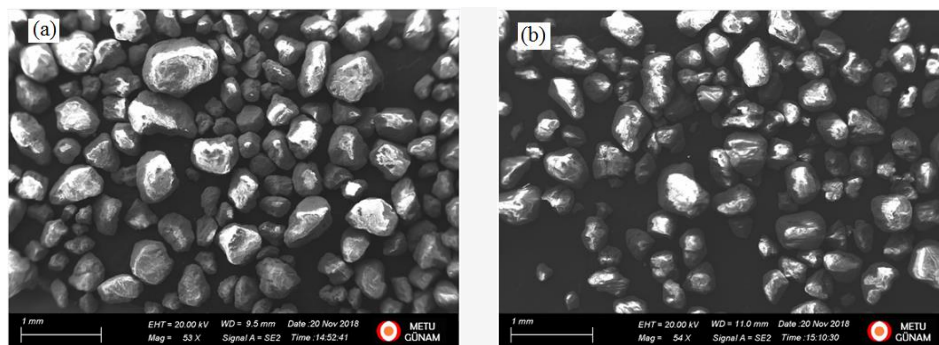


Fig.3. SEM images (a) untreated silica and (b) purified silica.

4. Conclusions

We have studied a purifying process which is a combination of thermal treatment and acid leaching. First, samples of Tabuk natural sand were submitted to rapid thermal annealing. Subsequently, the samples were soaked in an aqueous acid solution. The structural property of sample was examined by X-Ray diffraction. The pattern visualizes the appearance of the most common peaks related to SiO_2 at $2\theta = 20.52^\circ$ and 26.86° , besides the peaks related to other impurities like Fe_2O_3 and Al_2O_3 . However, after purification, the pattern shows the existence of the SiO_2 peaks only which means the excellent purification of the sand. EDX characterizations show that the purification process removed almost major impurities (Fe, Ca, Al, and Mg). We conclude that this purification's method is very interesting to produce high purity silicon.

Acknowledgments

The author would like to acknowledge university of Tabuk for the financial support under research project number S-1439- 0271.

References

- [1] Jun-Ming Hong, Bing Lin, Jie-Shan Jiang, Bor-Yann Chen, Chang-Tang Chang, *Journal of Industrial and Engineering Chemistry* **20**, 3667 (2014).
- [2] V. Raman, K. Parashar, S.R. Dhakate, *Journal of Sol-Gel Science and Technology* **25**, 175 (2002).
- [3] A. A. Popovich, P. A. Nikiforov, D. V. Onishchenko, A. K. Tsvetnikov, V. G. Kuryavyi, *Theoretical Foundations of Chemical Engineering* **42**(5), 603 (2008).
- [4] B.S. Xakalashe, M. Tangstad, *Silicon processing: from quartz to crystalline silicon solar Cells*, Southern African Pyrometallurgy 2011, Edited by R.T. Jones & P. den Hoed, Southern African Institute of Mining and Metallurgy, Johannesburg, 6-9 March (2011)
- [5] Yang Liu, Jian Kong, Yanxin Zhuang, Pengfei Xing, Huayi Yin, Xuetao Luo, *Journal of Cleaner Production* **224**, 709 (2019).
- [6] A. D. Farmer, A. F. Collings, G.J. Jameson, *Ultrasonics Sonochemistry* **7**, 243 (2000).
- [7] Tingting Jianga, Xinyi Xua, George Z. Chena, *Silicon prepared by electro-reduction in molten salts as new energy materials*, **47**, 46 (2020).
- [8] M. Khalifa, R. Ouertani, M. Hajji et al., *Hydrometallurgy* **185**, 204 (2019).
- [9] M. Khalifa, M. Hajji, H. Ezzaouia, *Impurity removal process for high-purity silica production by acid leaching*, published by EDP Sciences (2012).
- [10] I. M. Joni, L. Nulhakim, M. Vanitha, C. Panatarani, *Characteristics of crystalline silica (SiO_2) particles prepared by simple solution method using sodium silicate (Na_2SiO_3) precursor* *Journal of Physics: Conf. Series* 1080– 012006 (2018).
- [11] T. Yoshikawa, K. Morita, S. Kawanishi, T. Tanaka, *Journal of Alloys and Compounds* **490**, 31 (2010).
- [12] Primary reference: Smith et al, ICDD Grant-in-Aid, (1973).
- [13] D. Luo, *Study on the preparation of solar grade Silicon by Metallurgical Method*, January 27th (2017).