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Improvement of Impurities Removal from Silica Sand by Using a Leaching Process

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The silicon solar grade destined for the photovoltaic applications required a high purity (>99.9999%), its cycle of production consists firstly of enriching the silica by acid leaching.
In the present work, we have tried to enhance the extraction rate of impurities from Algerian silica sand by means of sequential leaching process using several solvents like HF, HCl, H_2SO_4, NaOH. The study of leaching effect is followed by DRX and Backscattered Electron images analysis.
Efficiency of the leaching process is improved and the removal of impurities is largely enhanced using a sequential leaching process with an appropriate solvent combination.

1. Introduction

The photovoltaic manufacturing of cells requires the use of a very pure silicon (purity > 99.9999 %), the purification of the silicon begins with an enrichment of the silica, until a purity superior to 98 % to prepare it for the next stage which is the carbo-reduction to obtain a silicon solar grade at 99.95 % of purity (desindes, 2000), then a purification by chemical or pyro-metallurgical way for obtaining a silicon solar grade (Braga et al, 2008).
The most annoying elements in the use of the quartz for the manufacturing of the photovoltaic cells are first of all the boron and the phosphor because of their difficulty of removal (by the technology of the metallurgical refinement. The maximal value is below 1 ppm for each (Istratov et al, 2006).
The calcium, the aluminum and other metal oxides are also undesirable impurities in silica. To release an easy physical elimination of these impurities, it is better that these elements exists separately in minerals with a sufficient size grading, hence the importance of characterization and analysis of the undesirable elements as well as their distribution in minerals.
The sand containing more than 500 ppm of iron and more than 800 ppm of aluminium is ready to be used in the industry of glass (Banza et al., 2006; Baoqi et al, 1995). However in the photovoltaic field the required technological parameters are more strong (Fe ≤ 100 ppm, Al≤ 400 ppm). That why the enrichment of high purity silica represents an obligatory passage, in the complete cycle of solar silicon grade production technology SoG (kheloufi, 2009).

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After the large use of the classical processes of treatment to basic ores as gravimetric and magnetic separation, the chemical treatment became more used in the worldwide. The ore of the less reactive metals can be directly handled in order to obtain the raw metal, which can be afterward refined. The ores ore of the more reactive (alkaline, alkaline earth, certain metal transition) must be previously cleaned by chemical treatments to eliminate all the impurities, because the metal, once elaborated, cannot be any more refined. One of the perspective methods, in the quoted case, it is the enrichment by leaching method (J. F. Richardson, J. H. Harker, 2002).
The present work consists in improving the removal of impurities from the of Algerian silica quartz sand deposits of Bousaâda using the technology of leaching process (Jadamba Temuujina et al, 2003; Judge J., 1991; Kikuyama H et al, 1994).
The solution of this problematic, within sight of the advantages which it has compared to other types of leaching, knowing the significant reduction of the rate of impurities, the realization of the cycle of the process in a record time (from a few days until two weeks) (kheloufi, 2009) and also its large applicability, more precisely for high-grade ores like the case of the Algerian quartz sand used.

2. Methodology

In our work, quartz sand samples B from algerian deposit of Boussaada are used.
After characterization of the raw material by X-ray Diffraction XRD, it was washed, cleaned, filtered than dried in order to undergo a magnetic separation on the laboratory scale.
The used diffractometer is Advanced D8's Bruker Type with Kα cooper radiation with a wavelength of 1.54 In °.
Once, these samples characterized and enriched. They have been attacked by hydrofluoric acid (HF from 20%) accompanied with HCl and H_2SO_4 acids (from 15 to 25%), followed by alkaline washing with diluted sodium hydroxide (NaOH) up to 10%, in a jar-test equipped with Teflon jars (leaching process) and finally, classified by gravimetry on shaking tables.
Mineralogical research showed that this sand is composed of: Pyrite, alumina, rutile, zircon, graphite, hematite, ilmenite, leucoxene, limonite and marcasite.
Moreover, and in order to define the internal structure and morphology and the probable of existence of inclusions as well as the existence of impurities in the crystalline lattice, the imagery sound Backscattered was used.
The hydrofluoric acid (HF) was chosen for its chemical compatibility with silica (highly hydrophobic). It removes the most possible impurities, while leaving the molecular structure of silica intact. The second acid attack by hydrochloric acid (HCl) or sulfuric acid (H_2SO_4) is intended to remove the residual impurities (Fe, Al, and Ti) in solution. Finally, this acid attack is followed by an alkaline attrition using a solution at 10% of sodium hydroxide (NaOH) in order to eliminate the residual metal from the sand quartz surface.

3. Results and Discussion

The role of HF as solvent in leaching process is double: the first one is to purify or to extract impurities from silica, in this context HF is well known by its efficiency, the second one is to etch silica grains for reducing their size, this fact improves the output of leaching.

3.1 X-Ray Diffraction analysis

Table1. Impurities quantification of Bousaâda raw quartz sand samples by X-ray fluorescence before enrichment

Samples	Impurities in ppm									
	Al	Fe	Ca	Mg	K	Ti	Na	P	Cr	Zr
Average	1270	850	357	302	104	434	185	150	174	195

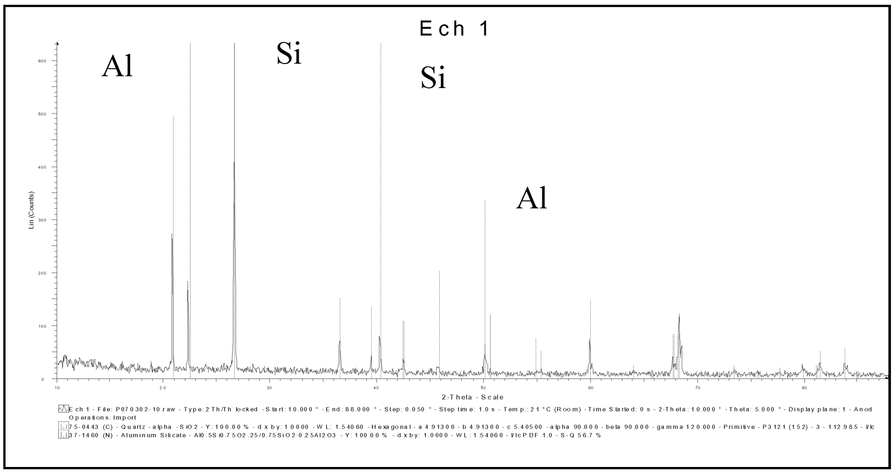


Figure 1: Raw quartz sand characterization by XRD analysis

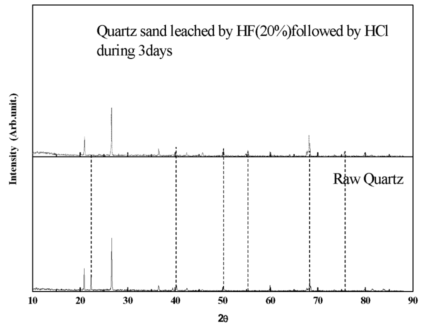


Figure 3: Quartz sand leached by HF 20% followed by H_2SO_4 during 3days

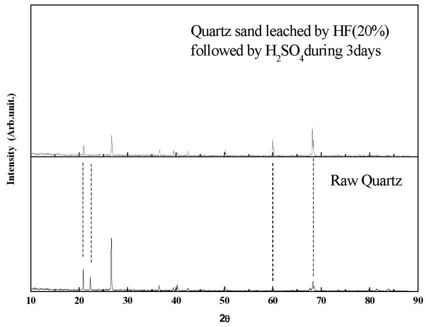


Figure 2: Quartz sand leached by HF 20% followed by HCl during 3days

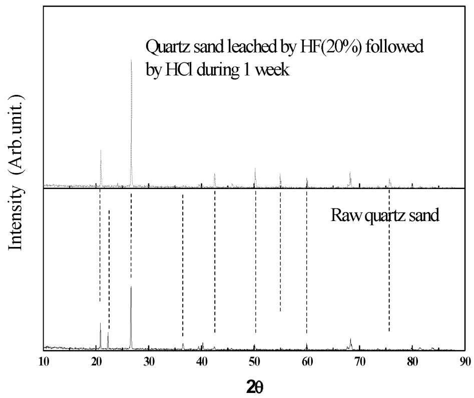


Figure 4: Quartz sand leached by HF 20% followed by HCl during 1 week

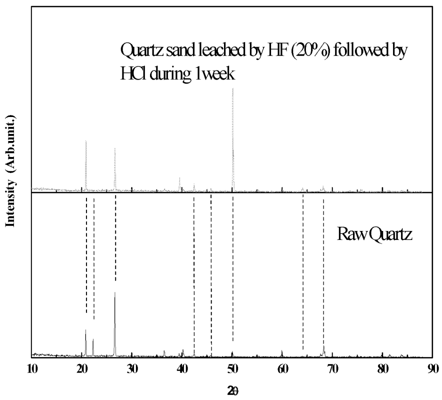


Figure 5: Quartz sand leached by HF 20% followed by H_2SO_4 during 1week

The sand no untreated presents a peak at 2θ = 22.5°which corresponds to the compound of SiO2-x Alx (fig 1),

Disappearance of the peak at 2θ = 22.5° in the case of leaching silica with HF(20%) followed by HCl leaching during 3days(fig 2), and the case of leaching by HF(20%) followed by H2SO4 during 3days (fig 3) can signifies explicitly the elimination of SiO2-x Alx compound due to the elimination of Al.

The same remark is noted in the case of leaching silica with HF followed by HCl or H2SO4 during 1 week.

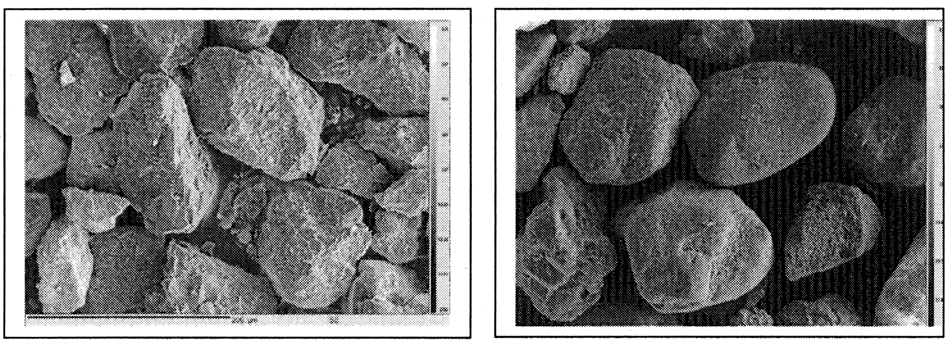
We noted an increase of the peak at 50° for the sands leached by HCl and H2SO4 during 3 days, furthermore the increase of peak at 50° for the sand leached with HCl during 1week and the increase of peak at 26,8° for the sand leached by H2SO4, this can be explained by an eventual impurities reduction

The change of intensity of peaks as well as the disappearance of some of the others are at the origin of a significant reduction of impurity or its rearrangement in the crystal lattice by the effect of the HF which reacts even with the silica, and the other acids by eliminating the maximum of impurities with a loss rate of silica after attack which does not exceed 10 %.

3.2 The Backscattered Electron images

The Backscattered Electron images (BEI) are used to locate the area of interest. Information on the atomic weight of the area of interest is available. Bright areas correspond to high atomic Z numbers and dark areas correspond to low atomic Z numbers. This analysis are carried out before and after the enrichment of raw quartz sand in order to show the improvement of impurities removal from silica sand using a leaching process (fig. 6) .

The results of (BEI) analysis (fig.6) come to consolidate again the DRX results. We can notice in these images, that the silica sand grains after enrichment by acid attack (HF, H_2SO_4 followed by attrition with NaOH) are clearer than those before enrichment. Bright areas correspond to high atomic Z numbers are eliminated. That is mean that the Heavy metals, especially iron and aluminium are significantly eliminated, that demonstrate that our aim is achieve using the leaching technology.



(a) *(b)*

Figure 6: Images SEM on quartz sand samples of Bousâada deposit (Algeria).

(a) Before enrichment

(b) After enrichment

4. Conclusion

In the present work leaching technology was proposed as an enriching method of silica quartz sand intended for solar cells manufacturing.

During prospecting the previous works, we have noted that the use of HF at low concentration aimed to improving porosity of silica, therefore, a low rate of impurities elimination is noted. So to increase the extraction rate we have opted for the high concentrations (HF at 20%)

The use of acids HCl and H_2SO_4 followed by NaOH attrition enriches more the sand. A considerable elimination of the Aluminum was well noted using the DRX analyses, where the other impurities were not detected, a change of intensity of peaks can be the origin of a reduction of the impurities.

Obtained results of DRX and Backscattered Electron images analysis allows us to confirm that the used process was effectiveness for improving removal impurities from Bousaada quartz sand silica deposit.

References

Banza A.N.,Quindt JGock, E., 2006. Improvement of the quartz sand processing at Hohenbocka,. Min. Proc.,76– 82.

Baoqi,S., Zhengbing, Ch., 1995.Chemical purification of industrial minerals. Proceedings of the XIX International Mineral Processing Congress, 2, 39, 207–211. Braga A.F., S.P. Moreira. P.R. Zampieri, J.M.G. Bacchin, P.R. Mei., 2008, New processes for the production of solar-grade polycrystalline silicon: Solar Energy Materials & Solar Cells 92, 418–424.

Désindes L., 2004, Silica ultrapure for electrometallurgy: gitology, physical and chemical characteristics of the ore quartz, Thesis presented for obtaining the rank of Doctor of the University Henri Poincaré, Nancy 1, 433, France.

Istratov A.A., Buonassisi T., Pickett , M. Heuer A.B., Weber E.R., 2006, Control of metal impurities in “dirty” multicrystalline silicon M.D. for solar cells. Materials Science and Engineering B 134 282–286.

Jadamba Temuujina, Kiyoshi Okada, Kenneth J.D. MacKenzie., 2003, Applied Clay Science, 22,87–195.

Judge J., 1991, A study of the dissolution of in acidic fluoride solutions, J. Electrochem Soc.118,1772.

Kheloufi A., 2009, Acid leaching technology for obtaining a high-purity of silica for photovoltaic area. Chemical Engineering Transactions, 17, 197-202.

Kikuyama H., Waki M., M. Miyashita, T. Yabune, and N. Miki., 1994, A study of the dissolution state and the etching reaction for HF solutions of extremely low concentration, J. Electrochem. Soc. 141,366,

Richardson, J.H, Harker J. F., 2002, Particle Technology and Separation Processes, Chemical Engineering 2, fifth edition.