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Acid leaching technology for obtaining a high-purity of silica for Photovoltaic area

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A leaching technology on samples of Algerian quartz sand deposit has been investigated in order to obtain high-purity silica (SiO_2) as raw material intended for photovoltaic application. The enrichment process was carried out by using double acid percolation leaching method (HF and HCl) for reducing the iron rate in the final product and also to decrease the final duration of the used process. In the aim to remove residual iron content from the ore surface, the leaching process was followed by an alkaline washing using NaOH and gravimetric separation. A characterization study has been carried out to establish the location, the quantity of the iron and others impurities on the ore: 850 ppm was the maximum iron contamination, whereas after the application of this technology, final iron content 96 ppm (as Fe) has been obtained. A schematic flowsheet of the process has been proposed considering the obtained experimental results and the results obtained in the literature. The experimental results have shown the technical feasibility of this process for high-purity silica sands production intended for photovoltaic area.

1. Introduction

The increase of the requirements in products for advanced technologies is responsible for an increasing demand for pure minerals and strongly conditions the strategy of the great industrial groups. In this vision, the photovoltaic field does not make of it exception, in terms of uses of high purity of silicon as a raw material for the solar panels production. However, and by anticipating the exhaustion of the natural resources to (gas and oil), the requirement for an always increasing quality and the constraints environmental imposes, the use of a new studies. For that, high purity silica was retained for the electrometallurgy technologies for its abundance and very large variety of the mineral structures containing silicon in the terrestrial sphere, and also for its great diversity of use (Désindes, 2000).

The enrichment of high purity silica represents an obligatory passage, in the complete cycle of solar silicon grade production technology SoG (Department of Geology and minerals resources Engenering, IGBt, NTNU, 2000). The continuous development of ore enrichment technologies made it possible to found highly effective processes for

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primary enrichment of materials, having a complex mineral composition like the case of silica (Katkov, 2004), and consequently, to guarantee a high quality concentrate for metallurgical industry. In our case, the presence of impurities, especially iron can limit the applicability of quartz sand intended for photovoltaic (PV) application. The sand containing (500-800 ppm) of iron is ready to be used in the industry of glass (Banza et al., 2006; Baoqi et al, 1995). However in the (PV) field the required technological parameters are more strong (Fe $\leq$ 100 ppm). In order to achieve this goal, a great interest was given to the percolation leaching technology as adequate and powerful solution of this problematic, within sight of the advantages which it has, compared to other types of leaching (heap leaching), such as a significant reduction of the rate of impurities primarily iron, a realization of the process cycle in a record time (from a few days until two weeks) (Chesné, et al.,1988) and also its vast applicability, precisely for high-grade ores as a case of the Algerian quartz sands used in this work.

2. Methodology

In our work, quartz sand samples of the deposit of Djelfa are used.
After the characterization by the X-ray fluorescence XRF, (results mentioned by elements on table 1) of the raw material. This one was washed, cleaned, filtered than dried in order to undergo a magnetic separation on the scale laboratory.
The samples of the product obtained thereafter are characterized by x-ray fluorescence the results of these analyses are mentioned in table 2.
Once, these samples characterized and enriched. They have been attacked by hydrofluoric acid (HF from 20% to 50%) and (HCl from 15 to 25%), followed by alkaline washing with diluted sodium hydroxide (NaOH) to 10%, in a jar-test equipped with Teflon jars (leaching by percolation) and finally, classified by gravimetry in shaking tables, according to the flow-sheet suggested in figure 1.
The results of characterization showed that this sand is composed of:
Pyrite, rutile, zircon, graphite, hematite, ilmenite, leucoxene, limonite and marcasite.
The Pyrite in the form of fine inclusions in sand grains proved to be the principal source of iron. For its difficulty for removal by physical techniques, the method of leaching acid by percolation (HF and HCl) was applied at various levels of pH.
HF was chosen for its chemical compatibility with silica (highly hydrophobic). It removes a maximum possible of impurities, initially iron while leaving the molecular structure of silica intact. The second acid attack by HCl is intended to eliminate the residual impurities (Fe, Al, Ti) in solution. After, the hydrogen peroxide (H_2O_2) was added in order to increase the kinetic oxidation of pyrite. The figure 2 and 3 show the iron contents in quartz sand as a function of time on the various levels of pH during leaching with and without using hydrogen peroxide.
During all the duration of the process, the temperature is maintained ambient (25°C) and the pH from 1 to 4.

Table 1: Impurities quantification of Djelfa 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

Table 2: Impurities quantification of Djelfa raw quartz sand samples by X-ray fluorescence after enrichment.

Samples	Impurities in ppm									
	Al	Fe	Ca	Mg	K	Ti	Na	P	Cr	Zr
Average	1140	820	327	285	88	425	174	150	174	195

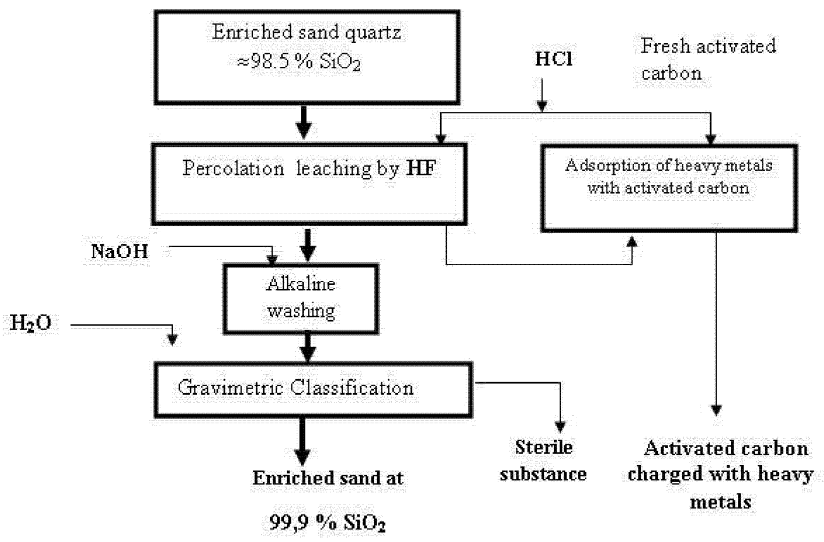

Figure1. Flow sheet of chemical enrichment of Djelfa quartz sand deposit by percolation leaching method.¶

3. Results and discussion

During the process of leaching with pH=4, without and with adding H_2O_2, any significant reduction of iron was recorded during 10 days (fig 2, 3). For a pH between 2 and 3, it was respectively reduced to 400 and 570 ppm for the same duration. However, when the process proceeds with pH=1 (HF=50%, HCl=25%), the reduction of iron decreased and reached 240 ppm (figure 3). It is noted that oxidation is less significant when the pH is above 2.

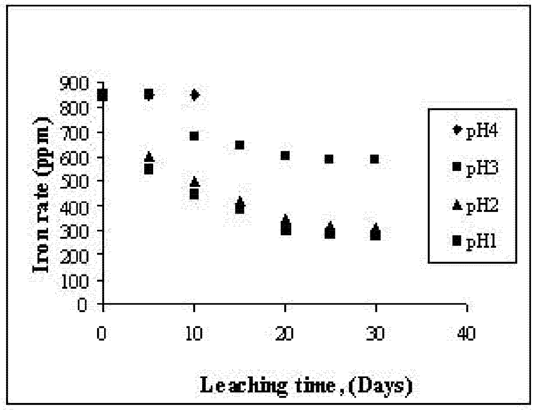

Figure 2. Iron removal from Djelfa sand quartz samples by percolation leaching without H_2O_2

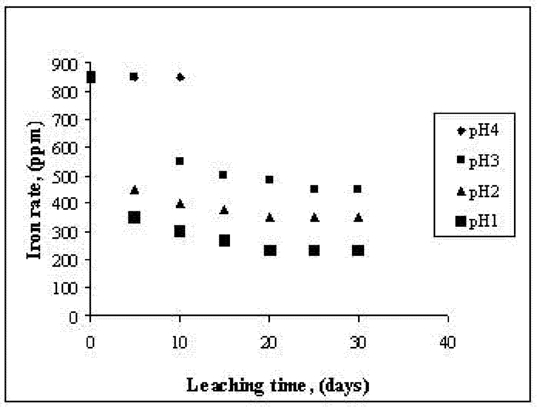

Figure 3. Iron removal from Djelfa sand quartz samples by percolation leaching with H_2O_2

The lowest values of the iron contents are obtained with pH= 1 by using H_2O_2 can be explained by the high potential redox of leached solution (500 to 600 mV). Without H_2O_2, the potential redox was lower than 500mV.

After the acid attack, washing was carried out to remove the iron of the sand surface. The water and sodium hydroxide were used as a solvent. The objective of such washing is significant to remove the fine particles and organic substances from the surface of sand. After washing, the fine sands and the iron oxide slimes were removed by gravimetric separation. Table 3 shows the residual iron contents in quartz sand after each operation: Leaching at various levels of pH, washing and classification. In spite of variability of the iron quantity in the raw material, the operation of washing carried out with diluted sodium hydroxide. This one shows that the iron contents were lowered in comparison with water washing process.

Table 3: Residual iron contents in the quartz sand from Djelfa

PH	Leaching without H_2O_2			Leaching with H_2O_2		
	1	2	3	1	2	3
Iron rate in the raw material	820	793	824	820	840	800
Iron rate after leaching	300	320	600	240	360	500
Iron rate after leaching and washing with H_2O	260	279	390	299	320	360
Iron rate after leaching and washing with NaOH	240	250	247	190	299	278

Put aside the presence of the pyrite, other ores such as the free rutile, the ilmenite, limonite and magnetite were also found in the raw material. After leaching and washing, gravimetric and magnetic experimental tests extractions were carried out.
Magnetic separation did not show any improvement with regard to iron, but gravimetric separation after leaching and washing on vibrating tables highlighted two types of quartz sand at knowing:
- Heavy and fine sand enriched containing around 446 ppm Fe. This accounts for approximately 60% of the raw material;
- Middle enriched sand product containing approximately 96 ppm Fe. This accounts for approximately 40% of the raw material.
The results are mentioned on the table 4 which represents the mass distribution of iron in various fractions of sand.

Table 4: Mass and iron distribution in various sand fractions after percolation leaching, washing and gravimetric separation.

Samples	**Quantity,**		**Fe**		**Ti**		**Al**	
	g	%	ppm	%	ppm	%	Ppm	%
Heavy and fine product	1740	58	446	53	280	66	730	64
Middle product	1260	42	96	47	140	34	400	36
Total	3000	100	190	100	-	100	-	100

By adsorption of heavy metals from the pregnant solution using active carbon, HCl will be maintained in recirculation and the requirement of fresh HCl can be reduced up to 0.4 kg/t quartz sand. The spent active carbon can be regenerated for its reuse after conventional techniques (Yehaskel, 1978).

4. Conclusion

- The acid leaching by percolation method (HF and HCl), followed by alkaline washing (NaOH) and adsorption of heavy metals with activated carbon can be used to reduce the duration of enrichment process and also of the iron rate in the final product of sands quartz used.
- The gravimetric separation after leaching and alkaline washing proved that is the optimal solution to reduce the iron contents of quartz sand samples of Djelfa deposit.
- The choice and the optimal concentrations of the selected reagents allowed obtaining high purity silica ready to be used as a raw material in the production cycle of silicon solar grade for photovoltaic application.
- From a raw sand containing 850 ppm Fe, a product containing 96 Fe ppm was obtained after twenty days by a double acid attack HF and HCl (50% and 25% respectively) followed by diluted NaOH alkaline washing up to 10% and a gravimetric separation, which coincides with the awaited results.
- The method of continues adsorption of the pregnant solution with activated carbon and re-use of the acid solution, ensure continuously the evacuation of heavy metals.

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