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Quartz sandstone as raw material for silicon solar grade elaboration

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***Abstract*— In the photovoltaic solar energy field, it is important to be interested in the role of raw material which takes a fundamental place in this vast market. In Algeria, particularly, this product continues to be imported and remain inaccessible for general use, paradoxically, the existence of a significant solar resources and a strong demand have been expressed by various sectors.**

This challenge can be achieved thanks to the implementation of new technologies by taking into account technical and financial aspects of high quality of silicon elaboration (SoG-Si). In spite of the development of alternative technologies, silicon remains the material most used today in the photovoltaic solar cells production (90%). It is obtained by reduction of the high purity silica (quartz, sand, sandstone, quartzite etc.).

Our work is dedicated to study microstructural properties of silica of El-Taref Algerian sandstones well as its enrichment as raw material for the silicon solar grade elaboration. A scanning electron microscopy (SEM) , X-rays diffraction and EPMA analysis have been used to investigate the morphological and microstructural properties of El-Taref silica.

Through this study, several silica sandstone microstructures and graphs were presented in order to determine the chemical characteristics, morphological and mechanical aspects. Finally, this raw material with content of SiO_2<98% was enriched by chemical acids leaching method in order to eliminate the impurities in the crystal lattice as well as on its surface and increase the SiO_2 content.

The obtained results have shown that a considerable elimination of impurities was well noted using the X-ray diffraction analyses, where the other impurities were no significant decreased. In addition, the scanning electron microscopy (SEM) analysis allows us to confirm that the used process was effectiveness for improving removal impurities from Algerian quartz sandstone deposit intended for silicon solar grade elaboration.

***Keywords*— Solar energy, Raw material, Enrichment, Silicon Solar grade**

I. INTRODUCTION

In Algeria The research and development activities on PV technologies represent major challenges in the short and medium term to solve the energy problems of our country with socio-economic benefits until 2030 up to 12000 MW of electricity from renewable energy, among which 2200 MW stemming from the PV. For this, it was established a multidisciplinary activity concentrated mainly on the technological development of materials intended for the PV area, to meet the needs of societal demands of the country. As such, various thematic areas and research from our point of view will be developed, notably, this activity will develop clean energy technologies (green) responding to the urgent needs of the environment protection, while exploiting the Algerian national resources from ore with high added value (silica) to the solar areas applications.

Therefore, it is imperative from now to predict the development of metallurgical treatment and enrichment processes of silica (an abundant natural resource) for poly silicon production.

In this context, the multi-crystalline silicon remains the mainly material in the manufacture of PV solar cells 90% (where it represents 40-50% of the cost of a solar cell).

In the Algerian context, in order to secure its raw material supply for PV industry, the production of solar grade silicon remains unavoidable, given the issues underlying it.

One of this kind of silica in our country is the quartz sandstone from El-Taref deposit which content 2 MT tons of exploitable reserves (sources ORGM).

The Sandstone is detrital sedimentary rocks composed of a large majority of quartz grains. They come from the consolidation of former sand. This digenesis occurs through water circulation, filing natural cement between the grains and compaction. This gradual cementing may fill all or only partly the spaces between the grains. Therefore, some sandstone retains significant porosity.

We can consider the mineralogical composition of sandstone under very different aspects:

According to the mineralogical nature of the binder. Siliceous sandstone cement, limestone, ironstone, according to the presence of exceptional mineral constituents .This distinction leads to the concept of maturity of sediments is reflected not only by the gradual disappearance of unstable constituents but also by the removal of the clay matrix, improving the classification and size by increasing the degree of rounding grain.

The macroscopic appearance of sandstone and physical property can be explained by their composition and structure. [1, 2]

Physically, they are very hard, compact, fine-grained. Chemically, they are rocks, mainly composed of silica. The relative fragility of the mineral quartz is particularly exposed to a fracturing when subjected to mechanical stress. The main mechanical and planar discontinuities cause. Their frequency and their densities depend on conditions in the immediate vicinity of grain or grains which affected the distribution is not uniform in the same geological body closely. [3.4]

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II. METHODOLOGY

In our work, Algerian quartz sandstone samples from El-Taref deposit are used. After characterization of the raw material by X-Ray diffraction, it was washed, cleaned, and filtered than dried in order to undergo enrichment at the laboratory scale. The samples with SiO2<98 % were characterized and treated. Mineralogical research showed that this kind of quartz is composed of: limonite, tourmaline, hematite and biotite and martite are the most commonly found impurities in the quartz sandstone of El-Taref.

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, limonite, 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 1 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.

III. RESULTS AND DISCUSSION

A. Electron Micro Probes Analysis EPMA

This method was used for analyze chemical composition of raw material (sand) and a major element of impurities in ppm. It is a punctual kind of analysis

We used a SX 100, the analyses results were shown in the table below (Table1)

TABLE I

TABLE 1: QUANTIFICATION BY ELECTRON MICROPROBES EPMA OF QUARTZ SANDSTONE SAMPLES FROM EL-TAREF DEPOSIT BEFORE ENRICHMENT

The samples	**Oxide**				
DataSet/Point	B_2O_3	Na_2O	MgO	Al_2O_3	SiO_2
1 / 1.	0.5	0.01	0.01	0.17	100.,61
2 / 1.	0.7	0.01	0.00	0.35	99.70
3 / 1.	0.6	0.00	0.00	0.25	99.14
4 / 1.	0.8	0.00	0.00	0.1	97.43
Average Ox wt%	**0.86**	**0.00**	**0.00**	**1.22**	**99.01**
Weight%					
DataSet/Point	B	Na	Mg	Al	Si
1 / 1.	0.00	0.01	0.01	0.00	47.03
2 / 1.	0.00	0.01	0.00	0.00	46.60
3 / 1.	0.00	0.00	0.00	0.00	46.34
4 / 1.	0.00	0.00	0.00	0.00	45.54
Average Wt%	**0.00**	**0.00**	**0.00**	**0.00**	**46.28**
Det.Lim ppm					
DataSet/Point	B	Na	Mg	Al	Si
1 / 1.	1711.00	68.00	44.00	36.00	97.00
2 / 1.	1725.00	67.00	45.00	37.00	98.00
3 / 1.	1745,00	70.00	43.00	35.00	96.00
4 / 1.	1737,00	68.00	45.00	36.00	2.00
Average Detection Limit (ppm)	**932**	**72**	**44**	**36**	**78**

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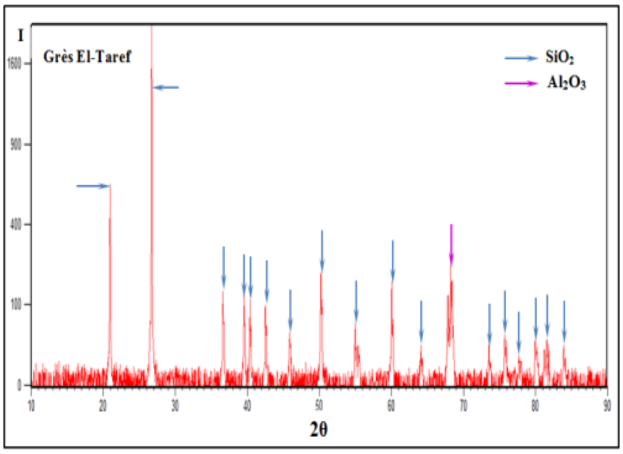
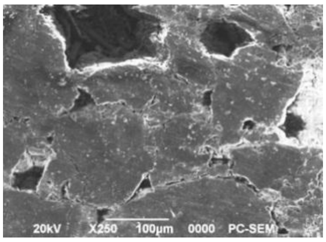
B. Ray diffraction Analyse:

We analysed the sandstone samples by the X-rays to determine the nature of the crystallized phases and also the influence of the presence of the impurities on these raw materials. On the figure1, we present the diagram of diffraction of the sandstone, which shows that it is mainly constituted by a crystalline phase of the quartz sandstone, the crystallographic characteristics of which are presented in the figure





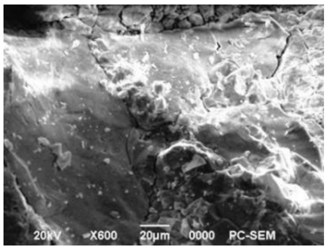


Fig. 1 X-Ray diffraction analysis of Algerian silica sandstone before enrichment

C. Micrography of Sandstone by Scanning electron microscope SEM

Fig 2.Micrograph of Sandstone X 250

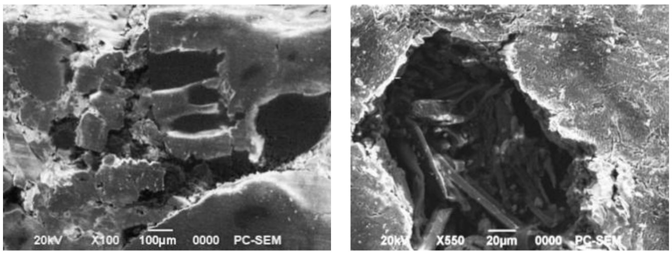


Fig 3. Micrograph of Sandstone X 500

In order to better characterize our discretion grains were observed by SEM to reveal their microstructure and subsequently connect the results of the study of their structure by XRD. Quartz is the main component in the stoneware. It represents from 50 to 95% of mineral volume [5.6.7]. It is present as almost exclusively single crystal grain of about 200 microns in diameter.

The SEM confirms the results of XRD analysis, which showed that the grains are mainly composed of crystalline silica (quartz) (Fig.2). The sandstone grows and develops through the accumulation of quartz grains but in an irregular manner (Fig.3). Between the grains there are vacuums known with deferent pore size

Inside most sandstone grains, we can observe the presence of fragments of rocks, [4] and plated with random distribution volume on the grain surface (Fig 3).

The figure.5 shows the presence of a mineral phase as the Clinoptilolite is a natural zeolite phase, it has a complex formula: ((Na, K, Ca) 2-3A_{l3} (Al, Si) $2Si_{13}O_{36}$ • $12H_2O$), it is mainly found in the range cemented. [8.9.10.11]

D. Quartz Sandstone enrichment by acid leaching process

The hydrofluoric acid (HF) was chosen for its chemical compatibility with silica (highly hydrophobic). It removes the most possible impurities; the second acid attack by hydrochloric acid (H_2SO_4) is intended to remove the residual impurities (Al,Fe, 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 sandstone quartz surface (see figure 4)[12].

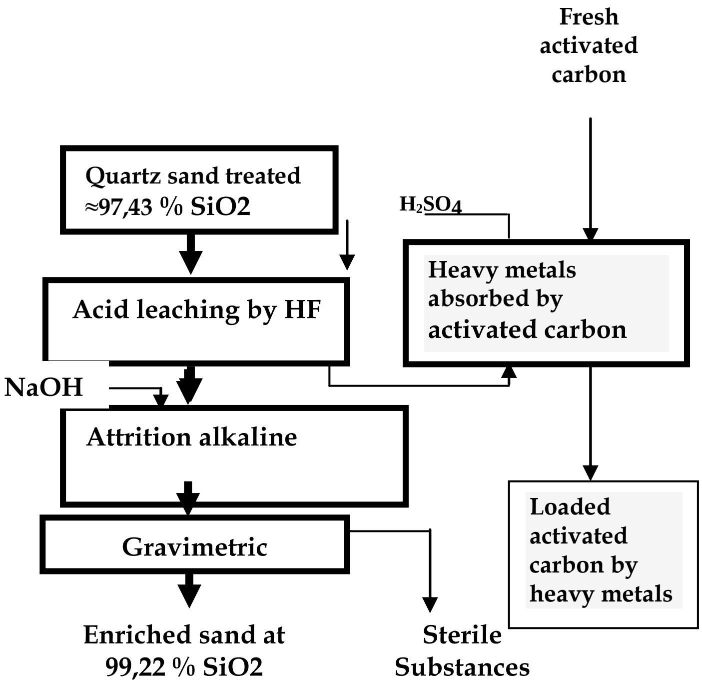


Fig 4. Flow-sheet of quartz sand enrichment of Touilila deposit by acid leaching process

The obtained result after enrichment show that the final enriched product reached the purity of 99,22% SiO_2, which is confirmed the chosen technology for increasing SiO_2 content intended for the silica elaboration. In the figure 5 the XR-Diffraction confirmed the results of acid leaching used technology.

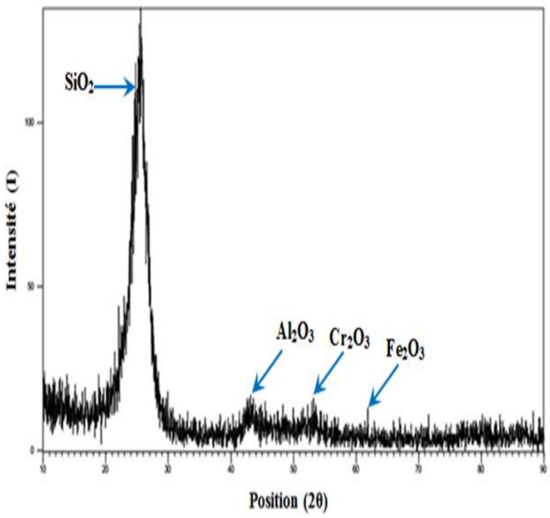


Fig 5. X-Ray diffraction analysis of Algerian silica sandstone after enrichment

IV. Conclusions

The main objective of this work was to study in depth of pure silica sandstone from Algerian El Taref deposit as raw material for national silicon grade elaboration to ensure a better quality for its future application in photovoltaic (PV) area. Within this framework and to achieve these objectives, a set of national silica sandstone samples were the subject of this study in which advanced characterization techniques (X-ray Diffraction, SEM and EPMA) were used. The samples which have not achieved the required level of purity< 98% have raised the subject of chemical enrichment by ' acid leaching method', where the concentration of acids HF, $H_2SO_4$and the leaching time played a very influential role.

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

The use of H_2SO_4acid followed by NaOH attrition enriches more the sandstone used. A considerable elimination of the Aluminum and iron were well noted using the XRD analyses, where the other impurities were no significant decreased.

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