REMOVAL OF IRON FROM SANDSTONE BY MAGNETIC SEPARATION AND LEACHING: CASE OF EL-AOUANA DEPOSIT (ALGERIA)

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Abstract: To improve the quality of raw materials from North East Algeria and their application in the field of flat glass manufacturing, the sandstone of El-Aouana (Jijel) was subjected to a physicochemical characterization. Analysis techniques such as X-ray diffraction, atomic absorption spectrophotometry and a chemical analysis of size fractions by sieving were used. According to this characterization it is noticed that sandstone feedstock contains primarily quartz (SiO_2), as well as ferriferous minerals considered as impurities during glass manufacturing.

This work concerns the removal of iron oxide from silica sandstone to obtain a material acceptable for flat glass manufacturing. For this, we have studied the silica enrichment process using a magnetic separation. Moreover, a leaching process by using hydrochloric acid has been studied under various experimental conditions; the parameters studied were: hydrochloric concentration, temperature, and time of contact. The results obtained show that the leaching studied presents a better removal of iron oxide after 150 min of treatment at temperature 90°C with a 3 mol/L, a concentrate obtained final of 99.16% SiO_2 with a content of 0.01% Fe_2O_3.

Keywords: magnetic separation, leaching, sandstone, mineral processing, characterization

1. INTRODUCTION

Silica is a major component used in various applications such as glass; ceramics and foundry manufacture (Asmatulu, 2002) and even in the production of photovoltaic cells. Despite its importance, the use of silica sand in Algeria remains limited due to their quality of the material containing harmful mineral inclusions,

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Include the case of El Aouana deposit, where the presence of impurities, mainly iron oxide, restricts the use of sandstone for the production of high-quality glass.

Therefore, physicochemical considerations, particularly the purity of sand that must meet with the strict standards of the glass makers such as 99% SiO_2, less than 0.03% Fe_2O_3 must be considered (Farmer el al., 2000). In addition, the particles size influences major the release of siliceous minerals compared to the gangue. The average size of quartz grains used in glass furnaces vary between 100 and 250 µm (Grynberg 2012).

The impurities iron oxides of silica sand can be reduced by physical and physicochemical methods, namely: attrition processes, aiming at removing iron bearing minerals from the surface of the particles (Ibrahim., et al 2013) ; separation processes (magnetic separation or flotation) for the separation of iron-rich minerals (Hacifazlioglu, 2014; Deniz et al., 2011),When the applicability of physical methods is not effective for removing chemically or physically related impurities which are not sufficiently liberated in mineral structure, other methods are then used at dissolving iron oxide and s 'is the chemical treatment (Zhang., et al 2012) or biological (Styriakova et al., 2012), however they have a rather restricted use in industrial scale due to their high operation costs and environmental hazards.

The method of froth flotation was applied to improve the quality of silica sand by using the cyclojet cell, this method removes about 80.49% iron oxide in silica sand, and Fe_2O_3 content in silica sand was decreased from 0.41% to 0.08% (Hacifazlioglu, 2014).

Various combinations of mineral processing techniques were investigated to purify silica sands from Jeddah deposit and removing iron impurities. The beneficiation tests performed including shaking table with magnetic separation reduced the iron content to 0.05% Fe_2O_3 and flotation with magnetic separation reduced the iron content to 0.1% Fe_2O_3 (Al-Maghrabi, 2004).

(Zhang., et al 2012) have recently presented a method of treatment of sand by phosphoric acid. The results obtained show that H_3PO_4 is a good agent for the removal of impurities contained in sand up to 77.1% efficiency compared with the other comparable methods that use industrial strong acids such as sulphuric (H_2SO_4), hydrochloric (HCl), and even hydrofluoric (HF) acids.

Very recently, Experimental studies examining the leaching by different acids and reach a high degree of iron removal (Tuncuk, Akcil, 2014). The highest Fe_2O_3 removal was 86.6% in the conditions of temperature 90 °C, 1M H_2SO_4 and 10% S/L ratio of solid to liquid for 120 min. Accordingly, the results obtained show that the HCl is an effective acid for removal of iron impurities in the raw material reach 86.5 % under the same conditions with a 20% S/L ratio.

Bioleaching iron from silica sand by means of microorganisms, since different bacteria capable of dissolving iron at 24 °C in 63 days (Styriakova et al., 2012).

2. MATERIALS AND METHODS

2.1. CHARACTERIZATION OF RAW MATERIAL

Characterization of sandstone ore using a variety of analytical tools focused on size distribution analysis by sieving performed on a sample of raw sandstone, an XRD mineralogical analysis aimed to determine the different mineral phases presented in the sample, and the Chemical analysis of size fractions will be the subject of oxides distribution knowledge in different particle size fractions. Therefore, a physicochemical characterization is necessary to determine the optimal mesh release and to provide the information required to conduct the sandstone enrichment tests.

A representative sample from the quarry site and then ground to 40 microns is subjected to chemical analysis by calorimeter and spectrophotometric and, the chemical composition of the sandstone is determined in the laboratory analyzes Ferphos (Iron Company and Phosphate) of Tebessa.

Other samples prepared in the same way were the subject of an X-ray diffraction analysis in the laboratory material technologies and process engineering (LTMGP) at the University of Bejaia (Algeria) using a powder diffractometer branded « X' Pert Prof Type Panalytical MPD / vertical system θ / θ PDS pass 4 x Accelerator (detector) platforms (Bracket) (sample-stage) » with Cu radiation with a wavelength $\lambda = 1.5405980$ Å at 2θ values between $10°$ and $100°$.

A representative sample of 500 g was subjected to size analysis using sieve device type RETSCH with a diameter of 200 mm $\times$ 50 mm, the particle size measurement range is from 0.045 to 4 mm on a vibratory sieve for 15 min at amplitude of 60 mm.

2.2. EXPERIMENTAL PROCEDURE

2.2.1. SAMPLE PREPARATIONS

The dimensions forming the mineral sandstone determine the liberation mesh, that is to say the dimension at which the grinding must-run to obtain a separation between minerals of different chemical elements carriers. Sample of 200 g composed of silica sand was placed in a laboratory scaled ball mill; the dry grinding was carried out for different periods (5, 10, 15 and 20 min) with a rotation speed at 200 rpm. Stainless balls were used as milling media.

2.2.2. MAGNETIC SEPARATION

The high intensity magnetic separator of laboratory working dry way is composed of three bobbins surrounding the electromagnet provided with a fluted rotor turning between the pole pieces of a magnetic circuit. The magnetic poles or pole pieces, between which turns the rotor are subjected to a magnetic induction. The ore feed is done by hopper and with a vibrating feeder of mineral material is separated from the

magnetic rotor. The magnetic particles adhere to the rotor under the influence of the magnetic force and are carried by the rotation in a low magnetic field area which is detached with a brush. The main magnetic separator parameters are the magnetic flux density which varies from 1.2 to 2 Tesla; the particle size should be less than 1 mm and the rotational speed 60 rpm (fig. 1).
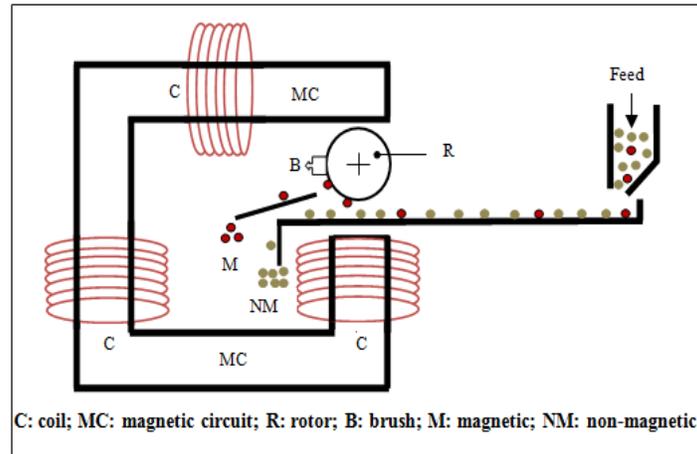

C: coil; MC: magnetic circuit; R: rotor; B: brush; M: magnetic; NM: non-magnetic

Fig. 1. Magnetic separator high intensity of laboratory (MSHI)

Samples of the size fraction (–250 + 125 µm), of 100 gm each were subjected to magnetic separation tests in order to remove the ferriferous inclusions contained in the siliceous material. The range of the current variation in the magnetic separator that was used is from 3 to 15 Amperes, and drum rotation rotor 60 rpm. The sample obtained was ground to a diameter of 40 µm and then analyzed by X-ray fluorescence the Laboratory building materials (CETIM) – Boumerdes. The sample obtained was ground to a diameter of 40 µm and then analyzed by X-ray fluorescence the Laboratory building materials (CETIM) – Boumerdes.

2.2.3. LEACHING TESTS

For the first part of the experiment, The leaching tests were carried out in a round bottomed flask (500 mL) , For each run, 200 mL of hydrochloric acid solution (prepare in water) at different concentrations (1, 2, 3, 4 and 5 mol/L) were added to the flask at room temperature. Then, 25 g of sand was added.The suspension was stirred for 1 hour and then left stand for 24 and 48 hours in an ambient temperature with occasional stirring.

The second stage included the leaching process in different ranges was realized: time (0 to 150 min) and temperature (40 to 90 °C).

The leaching reaction is based on the ability of hydrochloric acid to dissolve iron oxides; the chemical reaction during the removal of iron oxide by an attack with HCl is as follows

$$Fe_2O_3(s) + 6HCl(aq) \rightarrow 2FeCl_3(aq) + 3H_2O \qquad (1)$$
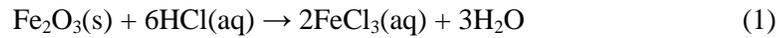
The contents of several times washed with distilled water for removing any unconsumed acid and dried at 105 °C.

The environmental risk from liquid effluents is decisive. Therefore, the potential acid is important due of the leaching of iron and titanium impurities with hydrochloric acid. To overcome this phenomenon affecting the environment, neutralization with quicklime is recommended. In contact of water with the quicklime is obtained slaked lime $Ca(OH)_2$, which causes an increase in pH. At the industrial level, the neutralization process can be realized in settling washtubs.

3. RESULTS AND DISCUSSION

3.1. CHEMICAL ANALYSIS AND X-RAY DIFFRACTION ANALYSIS

The results of the chemical analyzes and mineralogical are shown in Table 1 and Figure 2, respectively.

Table 1. Results chemical analysis of raw sandstone

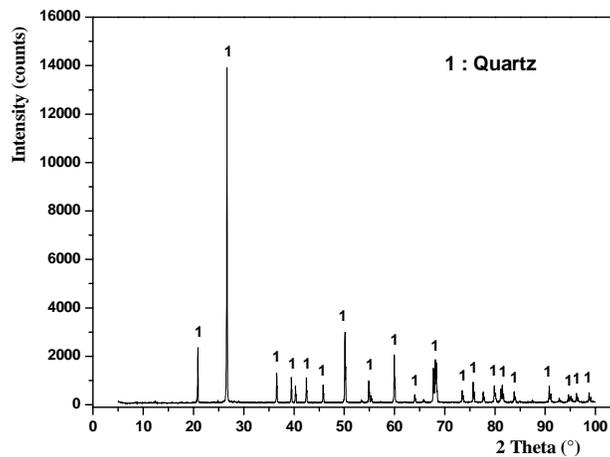| oxide	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	Na_2O	K_2O	TiO_2	SO_3	PAF
contents (%)	97.20	1.04	0.62	0.09	0.1	0.26	0.01	0.22	0.02	0.40

Fig. 2. Spectrum of a raw sandstone sample analyzed by X-ray diffraction

According to the study conducted on several samples, we found a dominance of SiO_2 content of 97.2%, the remaining oxides are divided into two categories, those that have a low weight percentage (Al_2O_3, Fe_2O_3, TiO_2, Na_2O) and those to trace (MgO, CaO, K_2O, SO_3).

Mineralogical study of as received sample revealed that the sample predominantly consists of quartz as the principal mineral and in very minor to trace amount are iron oxides (hematite), rutile and clay.

3.2. PARTICLE SIZE ANALYSIS

The results collected from the chemical analysis of size fractions reveal SiO_2 contents vary from 93 to 98% in the size fractions. As for the ferriferous inclusions contents are 0.28 to 1.20 % Fe_2O_3 showing excess iron in the raw material that does not meet the required standard ($Fe_2O_3 < 0.03$ %). Also, note that the iron oxide content increases as the particle reduction. The results of the chemical analysis of size fractions are given in Table 2.

Table 2. Results of chemical analysis of size fractions of sandstone sample

Fraction, mm	Yield ,%	SiO_2	Al_2O_3	Fe_2O_3	MgO	Na_2O	K_2O	TiO_2
> 4	15. 4	93.4	2.09	0.534	0.11	0.364	0.012	0.24
-4 + 2	18.81	96.0	1.36	0.554	0.093	0.364	0.012	0.18
- 2 + 1	08.95	97.1	0.66	0.433	0.099	0.364	0.012	0.14
- 1 + 0,5	11.33	97.8	0.49	0.351	0.081	0.350	0.006	0.16
- 0,5 + 0,25	18.78	98.4	0.26	0.283	0.080	0.350	0.006	0.12
- 0,25 + 0,125	15.99	98.2	0.32	0.374	0.065	0.357	0.012	0.11
- 0,125 + 0,063	06.98	97.6	0.87	0.523	0.073	0.350	0.012	0.16
- 0,063 + 0,045	01.29	95.6	1.66	0.734	0.093	0.364	0.018	0.29
< 0,045	02.47	94.3	3.29	1.209	0.109	0.377	0.018	0.38

3.3. SIEVE ANALYSIS

The silica sand processing is based on the nature of the accompanying minerals and gangue. Depending on these aspects and other physical, chemical and mineralogical properties, sand ore particles can be classified in three dimensions from the point of view of their treatment.

The particles +500 µm are rejected. Hence the particles of 500 µm to 106 µm, the size that must be grinding ungraded for a separation between the bearing minerals of different chemical elements and the particles <106 µm are removed by sieving. Because they represent iron and clay impurity (Ibrahim., et al 2013, Raghavan et al., 2006)

After grinding, the sandstone sample were classified according to the sieve fractions, the particle size obtained from each sample was compared with the particle size obtained from the original sample.

Figure 3 shows the corresponding changes to the grinding time and the reduced proportions to the desired liberation mesh (250 + 125 µm).

According to the grinding of the sample to a particle size below 250 µm, it is found that the liberation of valuable mineral to a desired particle size is achieved at an optimum time of 15 minutes with a mass percentage of 55%. In passing 20 minutes of time grinding, we note that the performance of fine particles almost doubled and the recovery of the desired fraction does not exceed 45%.
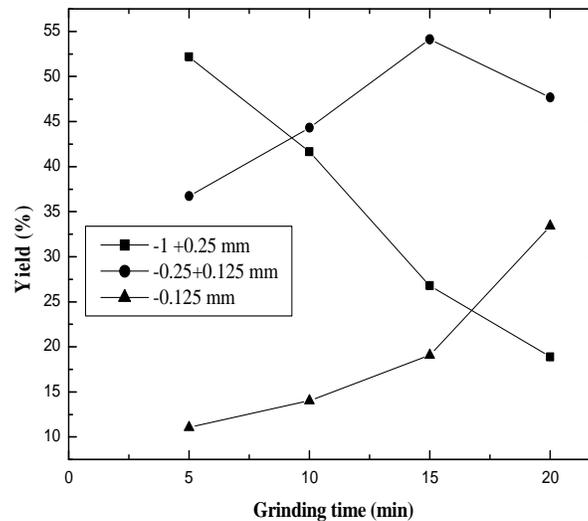

Fig. 3. Effect of the grinding time on the mass yield

A closed grinding system would further reduce particles > 250 µm, this possibility is economically feasible.

High impurities of iron and clay content was observed in the chemical analysis results from fraction < 125 µm, this fraction was reject by sieving with water. In the early experiments, a stage of desliming was carried out by quartz washing with water in a sieve of 38 µm to remove the required amount of clay impurities, tests obtained by flotation of iron impurities while pressing silica show that the iron content is approximately 0.05% or 98.8% SiO_2 and a recovery of 68.35%. Another magnetic separation – flotation in an acid medium is obtained 0.01% Fe_2O_3 and a high content of 99.3% SiO_2 with optimal recovery of 84.75% (Deniz et al., 2011).

From the chemical analysis results by FX, a significant decrease percentage of clay is noted and which is of the order of 0.12 %, The Fe_2O_3 content of the sandstone

sample were 0.6 % against to 0.28 %. The results show that washing by settling is required to remove the soft and clay minerals.

3.4. EFFECT OF INTENSITY ON THE PERFORMANCE OF THE MAGNETIC SEPARATION

The influence of the magnetic field intensity is used as an important factor in this process. The magnetic separation of the silica sand to a laboratory scale has been studied by several authors (Kheloufi et al., 2013, Sundararajan et al., 2009) due to the influence of the magnetic field strength and the grain size to reduce the rate of iron oxide.

The figure 4 depicts the effect of electric current intensity on the magnetic separation efficiency of the sandstone for iron impurity, from the obtained results by high intensity magnetic separation (MSHI), we found a significant improvement in silica content, and a remarkable reduction of impurities such as hematite and rutile was obtained in the range between 12 and 15 Amperes.With the increase in the intensity of the electric current, it is noted that the iron impurity content decreases from 0.28 to 0.10%, a difference of 0.18%. As for the content of TiO_2, regresses 0.13 to 0.07%.
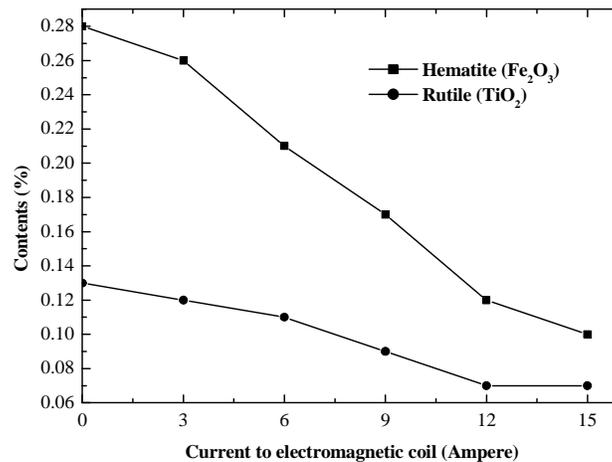

Fig. 4. Effect of current intensity the coil on contents of iron and rutile removal

The recovery of non-magnetic fraction (silica) obtained from this type of process has reached 97% against a grade of 0.1% Fe_2O_3, which shows that this type of concentrate can be used in the development of flat glass, but it is also important to apply a leaching by hydrochloric acid (HCl) in order to dissolve the ferriferous residues found in silica.

3.5. EFFECT OF HYDROCHLORIC ACID CONCENTRATIONS

In each test, the total and bivalent iron concentration in solution were measured as a function of time. During processing of the sandstone to different concentrations of HCl, there is an efficiency of the dissolution of the metal components mainly of iron with increasing HCl dose. Figure 5 shows the results of the percentage of iron removed, with a dose of 1 mol/L a slight decrease of 0.22% the Fe_2O_3 is noticed and of HCl 2 mol/L, no remarkable improvement. However, with an HCl concentration of 3 mol /L almost get the same result of 0.12 and 0.1%. The removal efficiency of iron oxide was observed that it reached 0.09 % in the first 24 h followed by a relatively maximum removal of 0.06 % in 48 h by 4 mol /L.

A significant improvement is witnessed by increasing the concentration of HCl 5 mol/L; it was observed slower removal relatively of 0.08 in the first 24 h, the maximum iron oxide content removal after 48 h was decreased from 0.28% to 0.04%.
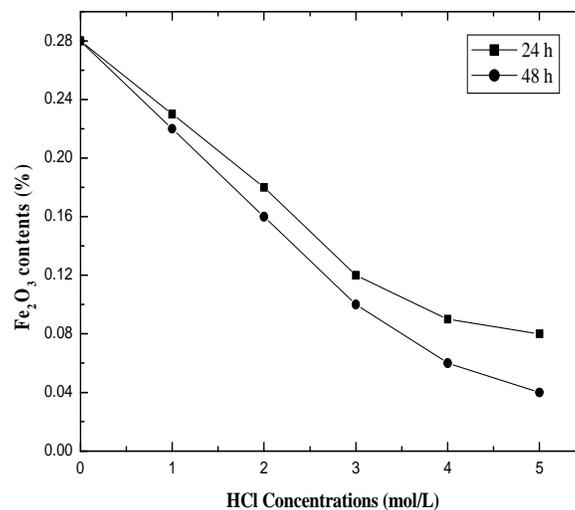

Fig. 5. Iron leaching from silica sand at 25 °C
at different concentrations of hydrochloric acid with versus time

3.6. EFFECT OF REACTION TEMPERATURES

The effects of temperature on iron impurity removed were investigated in the temperature range varied of 40–90 °C and are shown in figure 6. The leaching with temperatures from 40 °C to 65° C reduced the iron content from 0.28% to 0.12 and 0.1% respectively. However, at the highest temperature 90 °C, the considerable results obtained with iron content reduced to 0.01% for the same duration of 150 min. It is

noted that leaching efficiency to remove the iron of the sand always increased with increasing temperature.
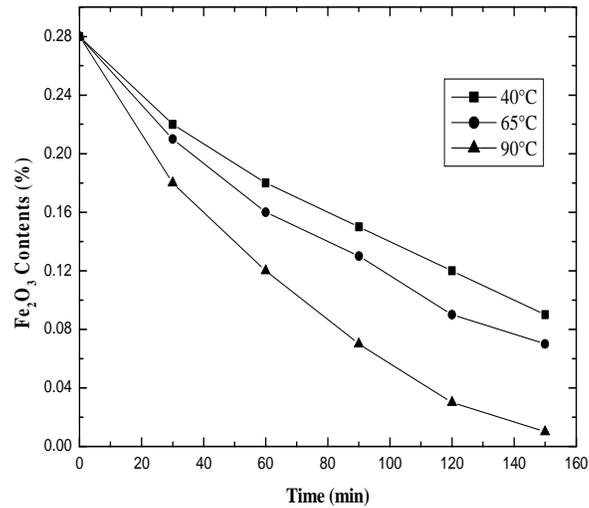

Fig. 6. Iron leaching from silica sand with hydrochloric acid (3 mol/L)
versus time at different temperatures

3.7. PROPOSED DIAGRAM FOR THE REMOVAL
OF IMPURITIES OF SANDSTONE FROM EL-AOUANA DEPOSIT

The study of the El-Aouana sandstone preparation was subjected to crushing to a size smaller than 5 mm and then classified by sieving to 250 µm before the milling stage for the recovery of particles less than 250 µm. In order to check the reliability of the mass recovery nearest to the liberation mesh size (–250 + 125 µm), a closed circuit grinding has been proposed to further reduce the particles greater than 250 µm, that possibility is economically achievable. A wet sieving was carried out to clean the surface of the lower particle -250 mm and it was removed the fraction -125 µm, then a product drying operation. The sample of the fraction –250 + 125 µm is passed through a high magnetic separator to remove iron impurities. The current standards propose a series of leaching tests for dissolution of metal components, mainly Fe_2O_3, two main solid extraction tests / liquid leaching are examined (fig. 7).
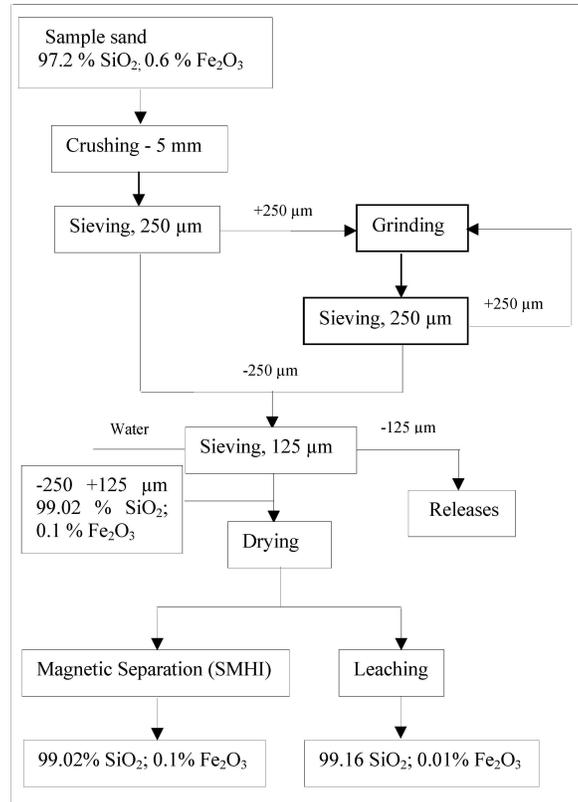
Fig. 7. Flow sheet proposed for the processing of sandstone from El-Aouana deposit

4. CONCLUSION

The study conducted on the sandstone quarry of El-Aouana (Jijel) allowed us to draw the following conclusions:

- Information from a representative sample at the physicochemical characterization confirmed the dominance of proportions, silica-rich inclusions with the inclusion of iron oxides and titanium.
- A better release of silica minerals is located in the fraction (250 to 125 µm), which reduces the levels of iron from 0.6 to 0.28% of Fe_2O_3 while eliminating fine particles < 125 µm by wet sieving.
- The non−magnetic final concentrate obtained by high intensity magnetic separation contained a Fe_2O_3 content of 0.1% does not respond the standards of quality glass.

- The tests by leaching with hydrochloric acid (3 mol/L) at temperature 90 °C had eliminated almost all of the iron oxide impurity (0.01%).The results obtained are very significant encourage the use of sandstone of El Aouana for the development of flat glass

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