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STUDY OF THE RECYCLING OF SOLID BARITE WASTES AS MAIN CONSTITUENT IN CERAMIC COMPOSITION

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Abstract: *Large amounts of Solid Barite Wastes (SBWs) materials are discarded in the Boucaïd's mine located in the North of Algeria ((Tissemsilt) producing an average of 50 wt% of barite rejects after the physico-chemical treatment. This work investigates the incorporation of solid barite waste (SBWs) as a raw material into a clay body, replacing feldspar material by up to 20 wt%. Ceramic pieces were produced at temperatures varying from 1200 to 1300°C. The technological properties of the ceramic pieces (e.g., linear shrinkage, apparent density, water absorption, and compressive strength) have been determined. The leaching toxicity of the fired pieces has also been determined. The results showed that SBWs could be used in ceramic materials (porcelain type), in the range up to 10 wt%, as a partial replacement for feldspar. Mineralogical analysis of SBWs, shows that it is mainly composed of calcite (22%), barite (25%), cerusite (13%) and hemimorphite (27%). After sintering, mullite, anorthite and quartz are the main mineralogical phases appeared at 1200 °C; when the temperature roses at 1300 °C, celcian phase appears, beside anorthite and quartz phases. The obtained ceramic samples exhibit better mechanical properties compared to the reference ceramic samples (0% SBWs). However, up to 10 wt% additions of SBWs in the ceramic bodies, micro-hardness and the Young's modulus have higher values, reaching 480 kg/mm^2 and 41.27 GPa respectively at 1300 °C. Besides, the evaluation of the release of various chemical species (Pb^{2+}, Zn^{2+}, $Ba^{2+}$and Cu^{2+}) contained in the ceramic samples indicates successful inertizationof the pollutants. The results show that solid barite wastes can be used as substituent of feldspar to produce eco- friendly ceramic materials.*

Keywords*: Barite wastes, Boucaid's mine, ceramic, mechanical properties, microstructure, environment.*

INTRODUCTION

The mining activity generates significant quantities of solid waste with no economic value, causing environmental degradation [1,2]. The magnitude of these releases risks becomes important mainly when it comes to the extraction and the processing of polymetallic deposits or industrial minerals rich in lead, zinc and barite sulfides, as in the case of a number of polymetallic and barite mines in Algeria,namely Boucaid's mine.. The sulphides displacement from discharges by the action of water and windcauses environmental pollution [3]. The mine tailings dissolution and oxidation can generate high concentrations of heavy metals, sulphates and acidity, and the chemical alteration of sulphide minerals in particular pyrite, pyrrhotite, chalcopyrite and sphalerite in mine waste deposits, generates acid mine drainage, which is a source of contamination [4], of soil, plants and water [1,4]. It was found that dissolved oxygen values in water were low and well below 5.0 mg /l, whereas the average pH values are acidic [5]. The mining waste accumulation, due to the extraction and processing of barite ore from abandoned mines, leads to the contamination of water by the heavy metals such as (As, Ba, Fe, Hg, Mn, Ni, Pb, Zn) with very high concentrations [6,7].

Barite-based ceramics ($BaSO_4$) have been widely used in various industrial fields, namely in electronic and medical applications, due to their low thermal expansion, their high melting points [8,9] and their low constant dielectric.
They are used as fluxing agents in the ceramic industry enamels and in ceramic materials. In the construction industry, they are used as a protective wall against X-rays and gamma rays [10,11]. The shielding capacity against these radiations is remarkable and can constitute a good material of protection against these rays, in order to protect the human beings against this type of ionizing rays [12]. Barium oxide is added to improve the refractive index of optical glass, and promote sintering andlower the viscosity of molten glass [13]. Recently, ceramics based on a combination of barite-kaolin derivative developed as a perfect medium, in the form of shielding for walls in diagnostic X-ray radiology [12].
The aim of this work is to study the reuse of Solid Barite Wastes (SBWs) as a raw material into a clay body, replacing feldspar material by 0, 10,20, 30 wt% of SBWs namely PCRB0, PCRB1, PCRB2, and PCRB3 respectively. Ceramic samples were produced at temperatures varying from 1200 to 1300°C. Microstructures, physical properties as porosity, density, mechanical and thermal conductivities were studied. The evaluation of the release of various chemical species (Pb^{2+}, Zn^{2+}, $Ba^{2+}$and Cu^{2+}) contained in the ceramic samples were also studied.

MATERIALS AND METHODS

The elaborated of the ceramic samples named PCRB0 is based on 50 wt% of kaolin; 20 wt% of sand and 30 wt% of feldspar. This last raw material is substituted by different amount of SBWs (10, 20 and 30 wt %) labeled, PCRB1, PCRB2, and PCRB3 respectively. The different mixtures were milled with 25 ml of distilled water during 2 hours in a planetary ball mill (Pulverisette 7 (Fritsch, Germany) using alumina balls as the grinding media. The obtained slurry was dried for 24 hours at 105° C. The mixture was manually ground for 20 minutes and sieved with 63 µm sieve. Finally the powders were pressed under uniaxial pressure (without added binder) at 55 MPa for 10 min to form pellets of 30 mmdiameter and 4 mm thickness. The pellets were calcined in air on a Nabertherm electric kiln furnace at 1200°C and 1300°C with a heating rate of 10°C/min with soaking time of 2 hours.
The chemical composition of the raw material is determined by X-ray fluorescence type PanAnalyticalPerl'X3. Mineralogical analysis of the samples and the raw materials were carried out on Bruker D2 Phaser X-ray diffractometer using CuKα radiation, a step size of 0.02 degrees 2θ, and a counting time of 10 s per step. Particle size analysis was carried out by laser particle measurement (laser Malvern MAL 1000 18 analyzer). The determination of bulk densities were achieved by the Archimedes's method in water, and absolute densities were measured by helium pycnometry (MicromeriticsAccuPyc). Bulk and absolute densities were used to calculate the total porosity of the samples. Ultrasonic Echography at 600 kHz, with an ultrasonic transmitter-receiver (Panametrics; model: 5072 PR) was used to obtain the elastic property (Young's modulus). The Vickers hardenss of the pellets was carried out on polished samples using an indentation of 9.8 N for 15 s. An average value was obtained from 10 indentations with a (microdormeter (Zwick/Roell HPE). Conductivity of the ceramic samples was determined at 10 °C using a FOX 50 Heat Flow Meter (TA Instruments) in accordance with ISO 8302:1991. The environmental assessment consisted in determining the mobility of pollutants based on the Toxicity characteristic Leaching Procedure (TCLP) established by the EPA (Environmental Protection Agency) in its Method 1311:1992. The concentrations in the filtrate were measured with an Inductively Coupled Plasma-Atomic Emission Spectrometer (ICP-AES Agilent 7500).

RESULTS AND DISCUSSION

As it is shown on Table 1, kaolin, sand and feldspar, used as raw materials for the elaboration of porcelain, contain relatively high amount of SiO_2 and Al_2O_3 which are 42.40, 95.21, 74.6 wt% and 37.8, 0.97, 12.9 wt% respectively. However, fluxes agents as Na_2O and K_2O are slightly in same amount in raw feldspar (3.7 and 4.6 respectively), TiO_2 is relatively high in the raw kaolin with 1.99 wt%. In other hand, SBWs contains a significant amounts of CaO (22.36) beside, metal oxides as

Fe_2O_3 (3.19 wt %), BaO (14.14 wt %), ZnO (1.29 wt %) PbO (1.48 wt %).
Moreover, these metal oxides could play a positive role as fluxes agents. SBWs is slightly poor in Al_2O_3 and rich in metal oxides, Hence, it could be replaced by feldspar.

Table 1. Chemical composition of the raw materials

Oxides	Kaolin (%)	Sand (%)	Feldspar	SBWs
SiO2	42.4	95.21	74.6	8.42
Al_2O_3	37.8	0.97	12.9	2.67
Fe_2O_3	0.55	0.23	1.73	3.19
MnO	0.07	/	0.04	0.09
MgO	0.05	0.01	0.4	1.61
CaO	0.26	1.32	1.08	22.36
Na_2O	0.03	0.27	3.7	0.27
K_2O	0.02	0.4	4.6	0.36
TiO2	1.99	0.01	0.3	3.71
P_2O_5	0.01	/	0.03	0.04
ZnO	/	/	/	1.29
BaO	/	/	/	14.14
PbO	/	/	/	1.48
P.F.	16.78	1.58	0.71	40.17

As presented on Fig.1, SBWs is mainly composed by quartz, dolomite, barite and calcite. However, mineralogical composition analysis, shows that it is mainly composed by quartz (4 wt%), calcite (22%), barite (25%), cerusite (13%) and hemimorphite (27%). These last mineralogical phases were not identified by XRD patterns of the reject SBWs.

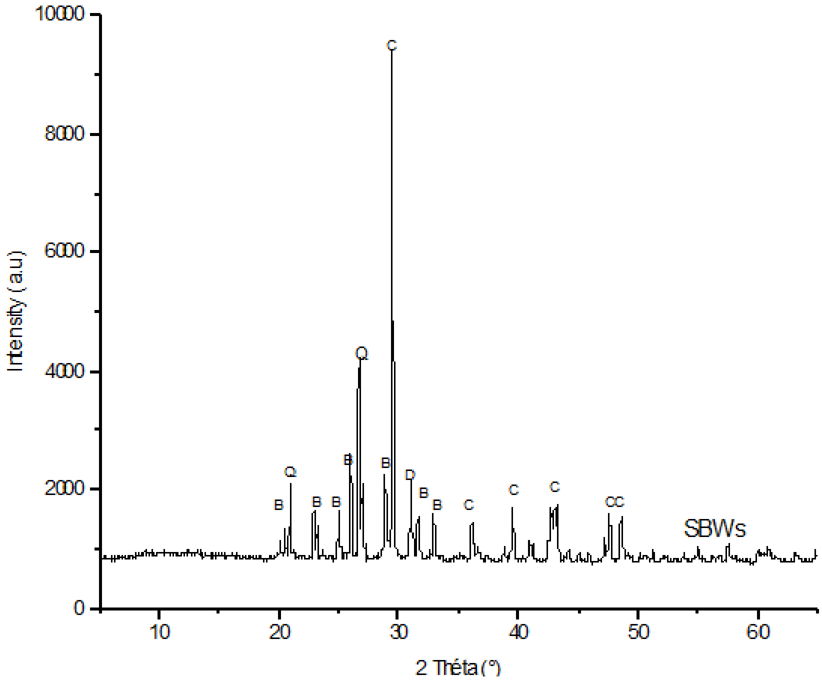

Figure 1. XRD patterns of SBWs raw material

The photos of the sintered ceramic samples based on SBWs addition are presented of Figure 2. The samples became more and more colorless compared to PCRB0, this is due to the presence of iron which plays an important role to give reddish color of the ceramic samples.

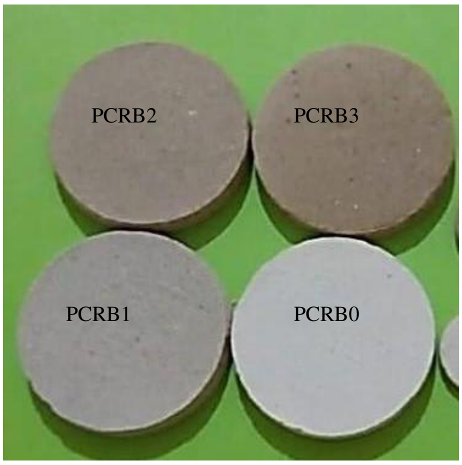

Figure 2. Photos of the ceramic samples sintered as function of *SBWs* addition sinterred at 1200°C.

The results of the sintered ceramic materials are shown on Figure 3 and Figure 4 for the samples sintered at 1200 and 1300°C respectively. Hence, mullite, anorthite, and quartz beside rutile were the main mineralogical phases found in the samples fired at 1200 °C. Indeed, Peaks of quartz and mullite decrease as function of SBWs addition. This is due to the fact that quartz participates to form in glassy phase, which is formed because of the addition of fluxing agent existing in the reject [14]. Moreover, peaks of anorthite began to rise when only 10 wt % of SBWs were added, whereas peaks of cristobalite tend to disappear as function of SBWs addition.

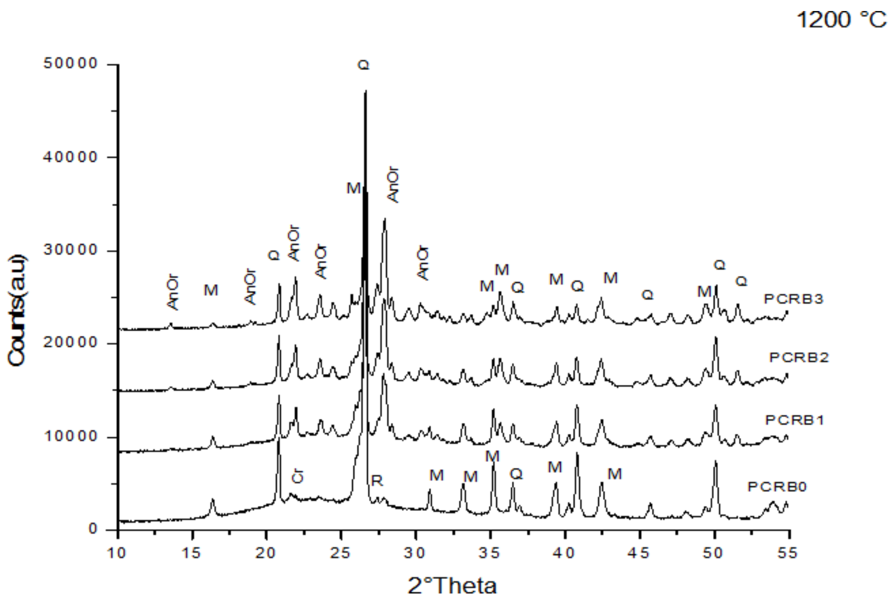

Figure 3. XRD patterns of the ceramic samples fired at 1200°C (M: Mullite, Q: Quartz, R: Rutile, AnOr: Anorthite, Cr: Cristobalite).

The behavior of the samples sintered at 1300°C (Figure 4) is different, when the firing temperature rose to 1300°C. Indeed, as the amount of reject increased, intensity of peaks of mullite and quartz decreased, it is due to the transformation of silica to vitreous phase probably promoted by the presence of fluxes agent as Fe_2O_3, CaO, PbO and ZnO, playing a role of network modification [soro, baziz]. In opposite, anorthite phase ($CaAl_2Si_2O_8$) appeared and increased as function of SBWs addition, this is due to the presence of excessive amount of CaO in the reject (22.36 wt%). A new phase correspondingto the formation of crystallite of celcian ($BaAl_2Si_2O_8$) is appeared in PCRB3, in this region of triaxial composition (SiO_2-Al_2O_3-BaO), celcian can be formed in the high silica region [15, 16], At this same temperature, rutile phase desappered as the amount of SBWs increased, it was been probably incorporated in the mullite phase [17].

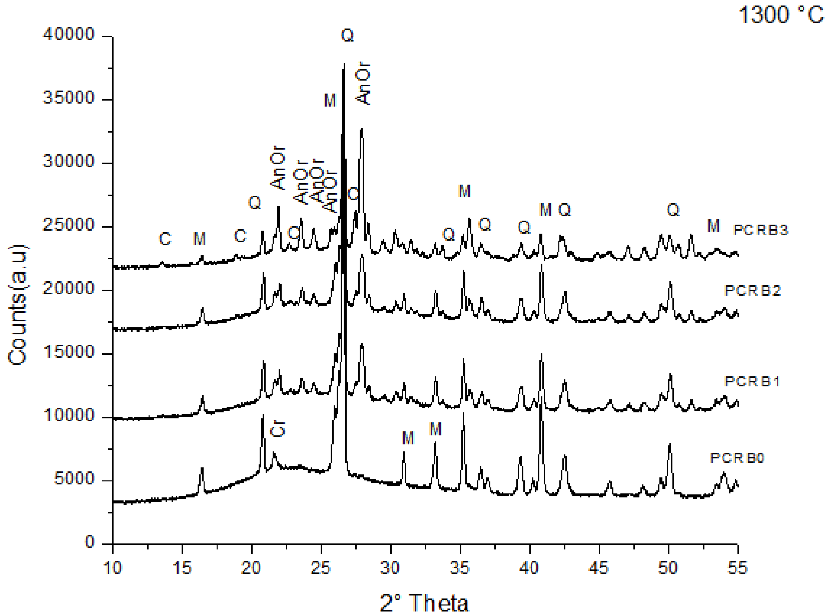

Figure 4. XRD patterns of the ceramic materials fired at 1300°C (M: Mullite, Q: Quartz, Cr: Cristobalite, AnOr: Anorthite, C: Celcian)

According to the Figure.5, Young's modulus increased as a function of the SBWs addition, and as function of the sintering temperature; when 10 wt % of SBWs were added to the mixtures, Young's modulus of the samples reached 26.9 and 41.27 respectively at 1200 and 1300°C. They are still low compared to the dense ceramic. This behavior can be justified by the appearance of the amorphous phases and mullite network at high temperature. Young's modulus may be improved with the increase in the content of mullite in the microstructure according to the mullite strengthening theory [18,19]. Thus the increase in sintering temperature induced the formation of mullite which plays an important role in improving the mechanical properties. However, when the amount of SBWs addition increased above 10 wt%, these properties tend to decrease in both firing temperatures. This is due, in one hand to the decrease of the mullite phase and in other hand to the increase of porosity (24.2%).

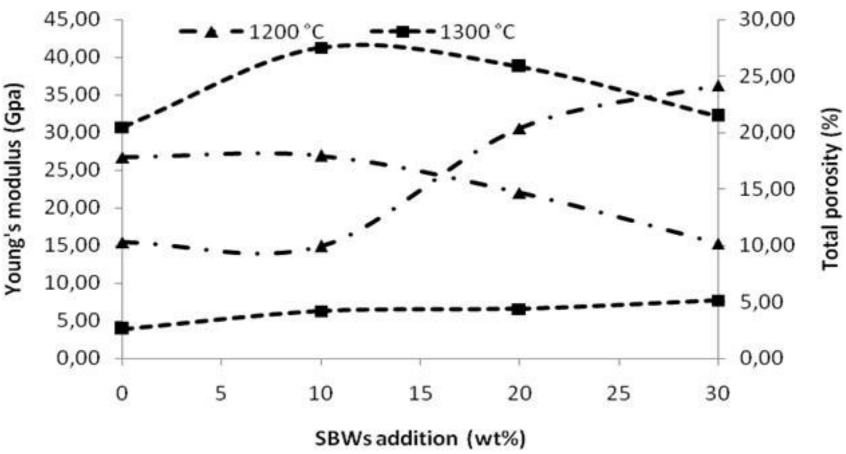

Figure 5. Young's modulus and total porosity as function of SBWs addition of the samples fired 1200 and 1300°C

Thermal behavior of the samples decreased as function of SBWs addition and as firing temperature. This property is directly linked to the porosity of the samples since the thermal conductivity of the samples diminished as the porosity increased [20]. Indeed, there is relationship between total porosity and the thermal conductivity of the ceramic samples.
This could indicate that factors such as type, size and distribution of the pores are important factors that are controlling the thermal conductivity of the ceramics that incorporate barite rejects [21, 22]. As the temperature is increased from1200 to 1300°C, thermal conductivity of the sample PCRB0 is increased from 0.376 to 0.633 W/mK. Thermal conductivity of fired clays are affected by their mineralogical compositions, consequently, their physical properties (ie: porosity and density) were affected with the increasing of the sintering temperature [22].

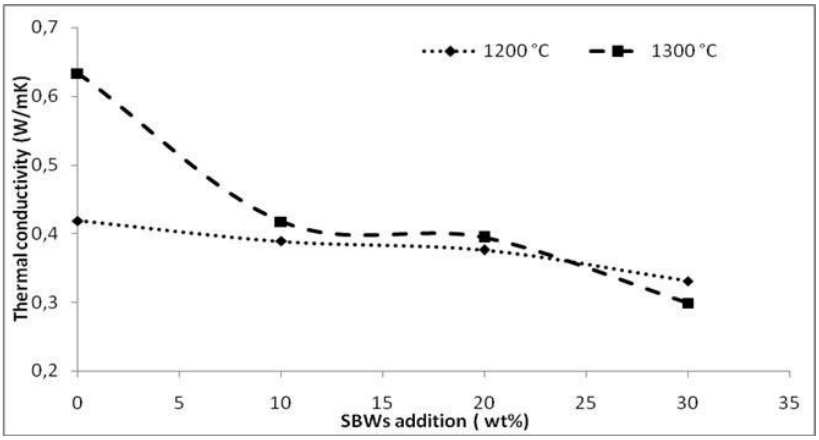

Figure 6. Thermal conductivity and total porosity as function of SBWs addition of the samples fired at 1200 and 1300°C

The environmental study carried out on the sample PCRB1 (Table 2), calcined at 1200 and 1300°C respectively, was performed to evaluate the degree of metal immobilization that can be achieved. Leaching tests [23] were applied to ground ceramic samples. The leachate concentrations have been compared with the US Environmental Protection Agency (USEPA) limits. As shown on the Table 2, the leachate concentrations of the samples fired at 1200°C is higher than those fired at 1300 °C; this is probably due to the vitrification process of the heavy metals into the ceramic matrix. However, the values met the USEPA regulated TCLP limits. Consequently, the leaching test indicated a degree of immobilization of the heavy metals suggesting that the incorporation of 10 wt % of the reject into ceramic products is an efficient inertization method.

Table 4. USEPA TCLP test results (ppm) of PCRB2 fired at 1200 and 1300°C and the maximum concentration of contaminants for toxicity characteristics

	PCRB2		
Components(ppm)	1200 °C	1300 °C	Limits US-EPA (ppm)
Pb	0.089	0.054	5
Zn	0.070	0.030	300
Ba	1.053	0.456	100
Cu	0.047	0.012	5

CONCLUSIONS AND RECOMMENDATIONS

According to the found results and their discussion, it was shown that Solid Barite Wastes contains metal oxides as well as quartz and barite. Based on its mineralogical and chemical compositions, SolidBarite Wastes is an appropriate candidate to substitute feldspar material. However, due to its chemical composition, this reject contains heavy metals that affect the environment. During sintering intensities of mullite and quartz in the ceramic samples decreased in the favor of apparition of anorthite phase which is promoted by calcite which belongs to SBWs material.

However, celcian ($BaO.Al_2O_3.2SiO_2$) phase is present only in PCRB3, when the firing temperature reached 1300 °C. Fluxing agents as Fe_2O_3, PbO, ZnO, present in SBWs material play a positive role to promote formation of glassy phase.

The compressive strength of the samples increased with the addition of up to 10 wt% of SBWs for the bricks fired at 1200 and 1300 °C respectively. The factor that determined the mechanical properties was the total porosity, since the microstructure of the ceramic samples is hardly modified with the addition of SBWs at both firing temperatures, as indicated by the XRD data study. Optimum values of compressive strength are 26.90 GPa and 41.27 GPa, they were obtained for PCRB1 ceramic sample fired at 1200 and 1300 °C, respectively. Thermal conductivities of all samples were low; they were ranged from 0.419 to 0.361 and from 0.633 to 0.299 for the ceramic samples fired at 1200 and 1300°C respectively. They are considered as insulator materials. Solid Barite Wastes (SWBs)can be considered as a material which can substitute feldspar and could be used as raw material, to produce eco-friendly ceramic materials insulators. This might be an alternative and reliable method for the disposal of this reject to preserve the environment.

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