

# Effect of firing temperature and atmosphere on sintering of ceramics made from Bayer process bauxite residue

Y. Pontikes<sup>a</sup>, C. Rathossi<sup>c</sup>, P. Nikolopoulos<sup>b</sup>, G.N. Angelopoulos<sup>a,\*</sup>,  
D.D. Jayaseelan<sup>d</sup>, W.E. Lee<sup>d</sup>

<sup>a</sup> *Laboratory of Materials and Metallurgy, Department of Chemical Engineering, University of Patras, Rio 26500, Greece*

<sup>b</sup> *Laboratory of Ceramic and Composite Materials, Department of Chemical Engineering, University of Patras, Rio 26500, Greece*

<sup>c</sup> *Section of Earth Materials, Department of Geology, University of Patras, Rio 26500, Greece*

<sup>d</sup> *Department of Materials, Imperial College London, S. Kensington Campus, London SW7 2AZ, UK*

Received 1 October 2007; received in revised form 19 October 2007; accepted 29 November 2007

Available online 10 April 2008

## Abstract

Bauxite residue, the principal waste from the Bayer process, was dried, pressed and studied for its thermal and sintering behaviour under different atmospheres, up to 1100 °C. For sintering in air and N<sub>2</sub>, shrinkage begins at 800 °C and ranges from 2.6% to 13.9%, after firing at 1000–1100 °C. Bulk density varies from 1.7 to 2.3 g/cm<sup>3</sup> whereas water absorption from 31.5% to 17.7%. The main crystalline phases identified on firing in air were hematite (Fe<sub>2</sub>O<sub>3</sub>), gehlenite (Ca<sub>2</sub>Al<sub>2</sub>SiO<sub>7</sub>) and perovskite (CaTiO<sub>3</sub>) whereas magnetite (Fe<sub>3</sub>O<sub>4</sub>) was also found on firing in N<sub>2</sub>. Microstructures are characterised by irregularly shaped, <20 μm Feret diameter, pores in a ceramic matrix with interconnected porosity. The average pore size is greater in samples fired in N<sub>2</sub>. On sintering in 4%H<sub>2</sub>/Ar, shrinkage begins at 710 °C. After firing at 1100 °C, shrinkage is 20.1% and water absorption 1%. The main crystalline phases are magnetite, wustite (FeO), gehlenite and perovskite. Microstructures are characterised by a compact heterogeneous matrix, with isolated <15 μm Feret diameter, closed pores. The grains have reacted with the adjacent phase and their shape is rounded with no sharp facets. Increased sintering temperature results in improved physical properties for all atmospheres tested and in higher average pore size when sintering takes place in air and N<sub>2</sub>. The use of magnetite-reducing sintering conditions can potentially assist in the production of a variety of ceramic compositions containing bauxite residue.

© 2008 Elsevier Ltd and Techna Group S.r.l. All rights reserved.

**Keywords:** A. Sintering; Red mud; Bauxite residue; Ceramics; Thermal behaviour

## 1. Introduction

Worldwide, the alumina industry produces approximately 70 Mt/annum of dry Bauxite Residue, BR, (“red mud”) [1]. Typically, BR is considered as a waste being discharged to sea or disposed in ponds or landfills. Only a small fraction of the produced volume is utilized in other industrial processes or applications [2–5]. This relates either to technological limitations of the proposed processes or unfavourable economics, due to the transportation cost in conjunction with the high water content of BR.

In Europe, legislation, public concern and increased awareness of social responsibility from the industrial sector have

resulted in intensified efforts towards improved utilization of BR [6]. In this direction, the use of high-pressure filter presses has been industrially implemented in Greece. Efforts are underway for the marketing of the dewatered filter-cake in the cement and heavy clay ceramic industry [3,4] as well as in other applications [7].

Utilization of BR in ceramic production has attracted much interest with more than 55 publications in the last 40 years, ~20% of the scientific output on BR utilization. However, only a few focus on the sintering of 100% BR ceramics. Knight et al. [8] report on the physical–mechanical properties of sintered ceramics from Jamaican BR, prepared from <75 μm powder and sintered at 1000–1100 °C. Apparent porosity was 40–48%, fracture toughness 0.39–0.69 MN/m<sup>3/2</sup>, modulus of rupture 17.23–27.09 MN/m<sup>2</sup>, compressive strength 42.0–83.9 MN/m<sup>2</sup> and Brinell Hardness 26.2–59.9 Kg/mm<sup>2</sup>. The high strength and toughness achieved is attributed to the formation of a glassy

\* Corresponding author. Tel.: +302610997509; fax: +302610990917.

E-mail address: [angel@chemeng.upatras.gr](mailto:angel@chemeng.upatras.gr) (G.N. Angelopoulos).

matrix and is comparable with commercial products. Prasad and Sharma [9], summarizing results on bricks made of 100% BR report that: (a) minimum sintering temperature is 1050 °C and optimum 1100 °C, (b) sintering at >1120 °C leads to dark heavy bricks, deformed and with extensive formation of glassy phase, (c) heating and cooling rates in the range of 10 °C/min are suggested, (d) soaking times of ~100 h seem adequate, and (e) the bricks deteriorate fast. di San Filippo and Usai [10] report that a 100% BR ceramic sintered at 950 °C, achieved compressive strengths similar to those of masonry bricks only when a shaping pressure 5000 kg/m<sup>2</sup> and sintering time >48 h were used. Moya et al. [11], report that sintering of 100% BR ceramics should take place at 1200 °C, where the bodies present the highest shrinkage and minimum water absorption. Finally, Perez et al. [2] also suggest sintering at 1100–1200 °C. To overcome the high shrinkage during sintering, the authors used 74% calcined BR. After sintering at 1175 °C for 4 h, porosity was 12.18% and compressive strength 171.8 MPa, values comparable to commercial heat accumulating bricks.

Despite variations in findings, probably relating to different characteristics of BR and ceramic processing, it is generally accepted that BR demonstrates moderate reactivity when heated in air and up to 1050 °C. Hematite and the developing alumina oxides, accounting usually for more than 50 wt.% of BR, react only to a limited extent, whereas sodium bearing compounds can participate in liquid phase formation. This is further supported by studies on the thermal behaviour of BR [12,13].

Considering the above, it would be beneficial to enhance the reactivity of BR and thus confer added value to the material. This could be done by exploiting the fluxing activity of FeO, where eutectics exist in the SiO<sub>2</sub>–CaO–FeO<sub>x</sub>, SiO<sub>2</sub>–Al<sub>2</sub>O<sub>3</sub>–FeO<sub>x</sub> and Al<sub>2</sub>O<sub>3</sub>–CaO–FeO<sub>x</sub> phase diagrams for ~1100 °C. So far, the use of reducing atmosphere on sintering of BR ceramics has not been investigated. To explore this possibility, the sintering of BR ceramics was studied at temperatures to 1100 °C under different atmospheres.

## 2. Experimental

BR was supplied by “Aluminium of Greece”. The chemical and mineralogical composition is provided in Tables 1 and 2. In terms of particle size distribution, BR is very fine with  $d_{10} = 0.7 \mu\text{m}$ ,  $d_{50} = 5.3 \mu\text{m}$  and  $d_{90} = 74.9 \mu\text{m}$ . The material was ground to a particle size <125  $\mu\text{m}$ , mixed with 6 wt.% water and left for 24 h in a hermetically closed plastic bag. Shaping was performed in a uniaxial hydraulic press, at 39 MPa. The samples were cylindrical, 31.0 mm in diameter and  $7.5 \pm 0.4$  mm in height. Drying took place initially at room temperature for 24 h and then in a drying oven at 110 °C till constant weight. After drying, the water content on formation

Table 2  
Mineralogical composition of BR

Crystalline phases in BR	
Major	Hematite Fe <sub>2</sub> O <sub>3</sub> , diaspor $\alpha$ -AlO(OH), gibbsite $\gamma$ -Al(OH) <sub>3</sub> , calcite CaCO <sub>3</sub> , calcium aluminum iron silicate hydroxide Ca <sub>3</sub> AlFe(SiO <sub>4</sub> )(OH) <sub>8</sub> , perovskite CaTiO <sub>3</sub> , cancrinite Na <sub>6</sub> Ca <sub>2</sub> Al <sub>6</sub> Si <sub>6</sub> O <sub>24</sub> (CO <sub>3</sub> ) <sub>2</sub> ·2H <sub>2</sub> O
Minor	Sodium aluminium silicate hydrate 1.0Na <sub>2</sub> O·Al <sub>2</sub> O <sub>3</sub> ·1.68SiO <sub>2</sub> ·1.73H <sub>2</sub> O, quartz SiO <sub>2</sub> , goethite FeO(OH)

was  $5.63 \pm 0.34\%$ . The linear shrinkage during drying was minimal. Green density was  $1.82 \pm 0.05 \text{ g/cm}^3$ . Since BR has a specific gravity of  $3.4 \text{ g/cm}^3$  this corresponds to ~46% porosity for the green bodies.

Thermal behaviour was studied by dilatometry (402ES, Netzsch) on 25 mm long and 6 mm diameter rods, taken from the cylindrical samples after green machining and polishing with abrasive papers (1200 grit). Maximum sintering temperature was 1000 °C, heating and cooling rates were 5 °C/min and the atmosphere was analytical grade air, N<sub>2</sub> or 4%H<sub>2</sub>/Ar at a flow rate of ~1.3 l/h. No soaking time was employed. To assist the interpretation of dimensional changes, BR pellets were fired at selected temperatures in an air-tight, alumina tube resistance furnace, quenched in atmosphere with forced air flow and the crystalline phases were determined by X-ray diffraction analysis, XRD (X-Pert Pro, Philips). The heating rate, atmosphere and flow rate was as in dilatometry experiments. XRD analysis was performed on powdered pellets, for Cu K $\alpha$  radiation, 40 mA and 40 kV and a  $2\theta$  range from 10°–70°.

Based on the dilatometric behaviour and theoretical expectations from ternary phase diagrams, the sintering of BR pellets was studied from 1000 to 1100 °C, with a heating rate of 5 °C/min and 60 min soaking time. Samples were left to cool inside the furnace. The firing and cooling atmosphere was also in this case, analytical grade air, N<sub>2</sub> or 4%H<sub>2</sub>/Ar, at a flow rate of ~1.3 l/h.

The physical properties of the sintered bodies were evaluated based on ASTM C373–88 and ASTM C326–82 standard protocols. The crystalline phases were determined by XRD as described above. Pore size distribution was determined by image analysis (SigmaScan Pro, Ver. 4, Jandel Scientific) on backscattered SEM images. The morphology was studied by scanning electron microscopy, SEM (JSM-6300, Jeol) on gold coated, polished samples. For microchemical analyses, an energy dispersive X-ray spectrometer, EDS (Oxford Instruments), was used. The default standards of LINK ISIS have been used.

## 3. Results and discussion

Dilatometry, Fig. 1, reveals that to 400 °C, all three atmospheres lead to similar behaviour, i.e. limited dimensional change until 250 °C and then a small contraction until 400 °C. In this temperature interval the reactions taking place correspond to removal of mechanical and crystalline water. At 300 °C, the intensities of calcium aluminum silicate hydroxide peaks are reduced; disappearing completely by 400 °C with a significant

Table 1  
Chemical composition of BR, wt. %

SiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	CaO	Fe <sub>2</sub> O <sub>3</sub>	MgO	K <sub>2</sub> O	Na <sub>2</sub> O	TiO <sub>2</sub>	LOI
7.60	16.63	11.36	42.58	0.56	0.07	3.49	5.00	12.2

Download English Version:

<https://daneshyari.com/en/article/1465481>

Download Persian Version:

<https://daneshyari.com/article/1465481>

[Daneshyari.com](https://daneshyari.com)