Construction and Building Materials 98 (2015) 257–264

Contents lists available at ScienceDirect

Construction and Building Materials

journal homepage: www.elsevier.com/locate/conbuildmat

The production of porous brick material from diatomaceous earth and Brazil nut shell ash

E. Escalera [a,c,*], G. Garcia [b], R. Terán [c], R. Tegman [a], M-L. Antti [a], M. Odén [d]

[a] Division of Materials Science, Luleå University of Technology, 97187 Luleå, Sweden
[b] Chemical Technology, Luleå University of Technology, 97187 Luleå, Sweden
[c] Chemistry Department, San Simon University, Campus, Cochabamba, Bolivia
[d] Nanostructured Materials, Department of Physics, Chemistry and Biology, Linköping University, Linköping, Sweden

HIGHLIGHTS

- Lightweight ceramic from low-cost raw materials were achieved at 850 °C.
- Diatomaceous earth with 10 wt% of BNS ash reduced the sintering temperature 100 °C.
- Addition of 10 wt% ash produces lightweight bricks with 8.5 MPa at 850 °C.
- Porous brick obtained at 850 °C and 10 wt% ash had thermal conductivity of 0.20 W/m K.

ARTICLE INFO

Article history:
Received 31 January 2015
Received in revised form 14 July 2015
Accepted 4 August 2015
Available online 24 August 2015

Keywords:
Diatomaceous earth
Brazil nut shell ash
Thermal conductivity
XRD
SEM

ABSTRACT

Diatomaceous earth was mixed with Brazil nut shell ash (BNS ash) in different amounts between 0 and 30 wt% and sintered at temperatures between 750 and 950 °C. The BNS ash contains 33 wt% K_2O and 11 wt% CaO mainly in carbonate form. The addition of BNS ash into the diatomaceous earth caused significant changes of the microstructure after sintering. The BNS ash addition produces lightweight porous bricks with acceptable strength at lower sintering temperature. The best combination of strength and porosity was achieved for a mixture of 10 wt% of BNS ash in the diatomaceous earth sintered at 850 °C. The achieved high porosity was 49%, density 1.06 g/cm^3, thermal conductivity 0.20 W/(m K) and the compressive strength was 8.5 MPa.

1. Introduction

Porous lightweight ceramics are interesting materials for a wide range of applications. The production of porous ceramics has increased worldwide in the last decades for industrial applications. The good thermal insulation properties that are connected to the porosity make these materials attractive also for use as bricks in building constructions [1,2].

Different raw materials are used to produce lightweight bricks, the most common being different clays. To the clay different pore forming materials are added to generate a desired porosity, often waste materials such as rice husks [3], kraft pulp production residues [4], and sawdust residues [5]. An alternative to clay based raw material is to use naturally occurring porous materials for production of lightweight bricks, such as for example diatomaceous earth (also known as diatomite). Diatomaceous earth is an inexpensive material consisting of silica rich particles with small particle size between 5 and 100 μm. In particular, diatomaceous earth is light in weight due to high porous inner structure with pore size about 100 nm [6]. The primary source of the diatomite is the large amount of residues from different single-cell algae living in both salt and fresh water. At present, at least 2 million tons per year of diatomaceous earth are mined worldwide [7].

Diatomaceous earth has been used to form lightweight calcium silicate bricks with good thermal insulation properties [8]. Recently, diatomite and volcanic ash were used to produce porous ceramics with excellent performance for control of humidity that can be used as a new construction material [9].

The sintering step is often a critical step in the ceramic industry when producing low-cost lightweight bricks with high strength. A low sintering temperature is desirable to reduce energy costs, but the temperature has to be high enough to achieve an acceptable strength. Therefore sintering aids are often added, so called flux materials, for example feldspars, granites

* Corresponding author at: Division of Materials Science, Luleå University of Technology, 97187 Luleå, Sweden.
E-mail address: escalera.edwin@gmail.com (E. Escalera).

http://dx.doi.org/10.1016/j.conbuildmat.2015.08.003
0950-0618/

[10] and sodium borates [9], which are rich in alkali elements. A cheap solution for flux materials is to use agricultural residual ashes containing alkalis; mainly potassium and calcium carbonates. Such residues could be ashes from sunflower husk, coffee husk and sugarcane bagasse [11–14]. Brazil nut shell ash is a residue resulting from the direct combustion of Brazil nut shells (BNS). This ash is rich in alkaline elements such as potassium and calcium. In fact, BNS ash have the potential to lower the melting points while sintering and thus it could be a cheap and attractive residual material to replace the traditional fluxing materials used in production of ceramics, namely feldspars and granites [12]. Nowadays, Brazil nut based industry is an emerging important local business in the Amazonian region. This region generates large amounts of residues from shelling of the nuts. Often these residues are used as a bio-fuel for heating and electric power generation. Thus, by direct combustion of BNS considerable amounts of ashes are generated. The combustion of BNS generates ashes in quantities of 80–150 tons over a period of approximately 6 months every year.

The purpose of this work was to produce lightweight porous ceramics with acceptable strength by sintering diatomaceous earth and Brazil nut shell ash. We also discuss the correlation between the obtained microstructures and brick relevant properties such as strength, open porosity, and thermal conductivity of the sintered specimens.

2. Experimental procedure

2.1. Materials

Diatomaceous earth (DE) raw material was obtained from the Murmutani zone, near Llica in Potosi-Bolivia. The BNS ash was provided by a processing factory of nuts, located in north-eastern part of Bolivia. The DE and BNS ash raw materials were separately ground by dry ball-milling for 30 min and then sieved through 50 mesh. The resulting powders were used for all subsequent experiments.

2.2. Characterization

Chemical analysis of both starting materials (BNS ash and DE) was done according to the modified EPA (U.S. Environmental Protection Agency) method 200.8 by Inductively Couple Plasma-Sector Field Mass Spectrometry (ICP-SFMS). Mineralogy and phase evolution of the raw materials and sintered samples were determined by X-ray diffractometry (XRD), using a PANalytical Empyrean X-ray diffractometer using Cu Kα radiation. The morphology of the raw materials and the microstructure of the sintered samples were studied by Scanning Electron Microscopy, SEM (Magellan 400, FEI Company) without conductive coating. The particle size distribution of the raw materials was determined using a laser particle size analyser (CILAS 1064) and the specific surface area was measured by nitrogen sorption using the Brunauer–Emmett–Teller (BET) method (ASAP 2010, Micromeritics). Differential Scanning Calorimetry analyses (Netzsch STA 449C Jupiter) of the samples were performed between room temperature and 1000 °C in air at a heating rate of 10 °C/min. The dilatometric behave of the samples at heating was studied by a horizontal dilatometer (Netzsch DIL 402C). The samples in the dilatometer were run from room temperature to 1000 °C in air at a heating rate of 5 °C/min. Prior to measurements, the samples were dried overnight at 110 °C.

2.3. The preparation of the sample

For the preparation of samples, the DE powder was mixed in a ball mill for 30 min with BNS ash in different weight proportions of 10%, 20% and 30%. Subsequently, the powder mixtures were uni-axially pressed into cylindrical compacts (35 mm in diameter and 35 mm in length) at 15 MPa. The compacted samples were dried overnight at 110 °C, and then sintered in a tube furnace (Nabertherm GmbH, type S) at 750, 850 and 950 °C in air at a heating rate of 5 °C/min and a holding time of 1 h at the maximum temperature. After sintering the samples were furnace-cooled. The sintered porous compacts were used to evaluate bulk density and open porosity by Archimedes' immersion technique according to ASTM C20-00 [15]. Thermal conductivity (λ) of the selected samples was calculated via the thermal diffusivity (α), bulk density (δ) and specific heat capacity (Cp). Thus, thermal conductivity was calculated using the equation $\lambda = \alpha\ \delta\ Cp$. Thermal diffusivity was measured by the flash method, using Laser Flash Apparatus (Netzsch LFA 457) according to ASTM E2585-09 [16]. For the measurements, disk-shaped specimens of 1.5 mm in thickness were coated with a thin layer of graphite. The specific heat capacity was measured with Differential Scanning Calorimetry (Netzsch DSC 404C) in argon atmosphere, according to ASTM E968-02 [17]. The strength of the sintered samples was obtained by the compressive load on the perimeter area of the cylinder until a crack was formed by using a universal testing machine (Model Mtest Quattro/100 kN) and a crossbar speed held at 0.5 mm/min for all tests. The experiments were usually performed on at least six samples of each mixture and the strength results are expressed as the mean value with the corresponding standard deviation of the measurements.

3. Results and discussions

3.1. Chemical and mineralogical analyses

Table 1 shows the chemical composition of DE and BNS ash in oxide form. The chemical components of the DE are Si, Al and Na along with small amounts of Fe, K, Mg and some other elements that do not exceed 1.0 wt%. The sodium content of 5.7 wt% Na_2O in the DE is much higher than that in diatomite from Taiwan (3.7 wt% Na_2O) [9]. The DE has a loss on ignition of 11 wt%, which is a typical value of other diatomaceous earth reported in the literature [18]. According to the chemical analysis of BNS ash, the main inorganic constituent is K followed by Ca and S. Other elements in small amounts are also present in the BNS ash. The high loss on ignition at 1000 °C for the BNS ash is 27.5 wt%. This value is related to the presence of non-burned organic particles, carbon and decomposition of carbonates that occurs with significant weight loss.

Typically, the presence of the different element concentrations in BNS ash can vary depending on soil of plantation, geographical location, and quantity of fertilizers used for the growing of Brazil nuts.

Fig. 1 shows the X-ray diffractograms of the DE and BNS ash. The X-ray diffractogram of DE powder (Fig. 1a) shows a broad hump around 2θ: 18–28° which indicates the presence of amorphous SiO_2, which represents the major phase of the DE powder. This is well corroborated with the chemical analysis that shows 71 wt% of SiO_2 (Table 1) where most of the SiO_2 content belongs to DE. The X-ray diffractogram of DE also indicates the presence of several crystalline phases such as halite (NaCl), quartz, plagioclase and muscovite minerals, which represent the minor phases in the DE. The chemical analysis (Table 1) shows the presence of 5.7 wt% Na_2O that is related with halite (NaCl) mineral which is about 9 wt% as a result from chemical analysis of Cl^-. The Al content is associated to the plagioclase and muscovite minerals.

The X-ray diffractogram of the BNS ash (Fig. 1b) reveals the presence of at least seven different phases such as $CaCO_3$, K_2CO_3 and fairchildite ($K_2Ca(CO_3)_2$), quartz, arcanite (K_2SO_4), magnesium oxide (MgO) and calcium phosphate ($Ca_3(PO_4)_2$), i.e. an impressive array of minerals unlike other ashes originating from organic products used for similar purposes [14]. The presence of these minerals in the BNS ash is corroborated by its chemical composition shown in Table 1. It should be noticed, however, that the phase composition of the ashes may vary depending on the burning conditions like temperature and time.

The morphology and microstructure of the raw materials are shown in Fig. 2. The DE raw material has a characteristic skeletal structure of four different types of fossilized diatom shells (Fig. 2a) corresponding to the examined DE powder from "Murmutani" sediments, located in the region of Potosi-Bolivia. It can also be seen that the particles of diatom shells have highly regular features on a microscale. The shapes of the diatom shells are well defined as elliptical, centric and elongated patterns of open micropores (less than 1 μm) which are characteristic for a typical microstructure of silica-based cell walls. The particle size distribution is in the range of 5–30 μm. The particle diameter at 50% (d_{50})