Elaboration of cellular lightweight firebricks from Algerian kaolin

Boumchedda Khaled* [1], Ghernouti Bahia [1]
[1]Faculty of Engineer Science, Department of Engineering Materials, University "M'hamed Bougara" of Boumerdes, Unity of Research "Materials, Proceeding and environment", 35000 Boumerdes Algeria,

Abstract

Cellular lightweight firebricks were developed starting from foamed slurry of water, formed of an Algerian kaolin, foaming agent, and mineral binder. The consolidation is performed at room temperature, by adding refractory cement which allows the transformation of foamed slurry into a rigid and stiff body that can be machined in the green state after drying.
The sintering of samples which are rectified in form of a regular geometry gives interconnected cellular materials with a good pore size distribution.
The volume of voids and the size of cells of porous refractories are controlled by their processing.
Various porosities were obtained with this novel method of elaboration of porous materials from 60 % to 85 % of vacuum, and various sizes of cells, from 1/100 mm to 1 mm.
Their mechanical resistance is appreciable, and their temperature of melting is > 1650 °C. The expansion as a function of temperature is regular on a large interval of temperature from 25 to 1000 ° C, and the coefficient of thermal expansion is lower than 6 x 10^{-6} $°C^{-1}$.
The structure in form of foam of cellular refractories having voids filled of air confers an high heat insulation to this porous refractory material, and offers a large industrial applications, such thermal insulators, filters.

Key words: Elaboration, cellular material, lightweight firebricks

1. Introduction

The cellular lightweight firebricks are generally porous bodies having a high porosity, which starts of 50 % to over 80 % of vacuum [1,2], they are used in thermal insulation applications such, Intermittent furnaces, tunnel kilns, flues, regenerators, gas generators, reactors and other equipments functioning at high temperature. The cells of these materials are interconnected in all directions and filled of air; indeed air constitutes the best thermal insulating materials. The cellular solids constitute a new generation of materials called "emerging materials" [3]. They are applied in several fields such as the thermal isolation, the catalysis, the purification and the filtration of fluids and of gas, and also in the biotechnology [4]. Porous volume and the size of cell depend of the function which is attributed to cellular material.
The main industrial application is the tunnel kilns, where the cellular firebricks are used to minimize the thickness of walls. These furnaces are usually built with several walls of refractory bricks, one beside the other. Every wall is built with bricks having same structures and same refractory properties. Bricks which are directly exposed to the source of heat are dense and

*Boumchedda Khaled
Address: Faculty of Engineer Science, Department of Engineering Materials, University "M'hamed Bougara" of Boumerdes, Unity of Research "Materials, Proceeding and environment, 35000 Boumerdes ALGERIA E-mail address: boumchedda_k@yahoo.fr, Phone: +213554171751 Fax: +21324911658

impermeable to avoid corrosion whereas the intermediate walls are built by porous refractory bricks to minimize heat exchange with external areas. The external wall is built with classical solid brick, it is far from the heat source, and it must ensure the cohesion and the solidity of the structure. Sometimes one adds an intern partition constituted by chamotte (calcined fireclay). This way of construction of the tunnel kilns makes it possible to strongly decrease the thickness of the walls. Cellular lightweight firebricks have low thermal conductivity and a large capacity thermal isolating which allows the realization of built walls low thickness. Their weak thermal accumulation which is the consequence of their low density and their low thermal conductivity can to reduce the energy consumption of the furnaces.

The methods of elaboration of porous materials are often based on the introduction of organic substances in the mixture of raw materials, which during the heating they burn and thus cause pores in the ceramic [5,6,7]. With these methods it's difficult to master the reproduction of porous materials having the same volume of voids and the same size of cells, because it's impossible to intervene for corrections at high temperature. Often these methods are not reproducible and the samples must undergo dimensional new corrections by machining and can destroy the ceramics products. Then that the method we develop implies the creation of porosity to room temperature, which largely facilitates the reproducibility of the manufacture of porous materials having same porous volume and an only size of cell. This way of elaboration of porous materials makes possible to envisage the final density with an acceptable precision. Indeed it is easy to calculate the density by knowing volume of the mould of casting, the content of dry matter, and the loss on the ignition, and the voluminal withdrawal which the sample undergoes during drying, and the heating.

This study presents a novel method of elaboration of cellular lightweight firebricks, developed according to the principle of elaboration of cellular material at green state; the heat treatment of sintering consolidates this green state to give the final porous product.

2. Materials and Method

2.1. Starting materials

The main raw material is an Algerian kaolin named "DD kaolin", DD for the origin of its deposit which is located in the region of “Djebel Debbagh” in the wilaya of Guelma (eastern Algeria), and 3 for three and last variety. This kaolin is produced and marketed by the Algerian company of kaolin SOALKA for the ceramics industries, and refractory products. The DD kaolin has a hydrothermal geological formation; its representative argillaceous mineral is mainly of kaolinite/halloysite. The first variety (DD1) is whitish and translucent; it was characterized by Boulmokh [8] however its clay reserves are almost exhausted. The others variety (DD2 and DD3) are blackish because they are polluted by a high tenor of manganese oxide which gives a blackish colouring to this clay, and which limit their uses in paper industry, luxury porcelain and sanitary ware, where brightness and whiteness are required [9,10]. However DD kaolin is very rich in alumina (>38 %), the main bearers of alumina are kaolinite and gibbsite. DTA-TG analysis shows the peak which is attributed to gibbsite (DDTA peak at 436.2 °C) (Figure 1). The estimated tenor of kaolinite and gibbsite calculated from weight losses given by TG curve, are respectively 82 % of kaolinite and 8 % of gibbsite. The silica tenor does not exceed 42 wt. %,

which shows a weak tenor of free silica. The flux elements such K_2O, Na_2O, and Fe_2O_3 does not exceed 0.40 wt. %, and the amount of earth-alkaline oxides (CaO and MgO) is very low (<1 %). Its melting point is more than 1700 °C. These characteristics show that DD kaolin constitutes an excellent refractory raw material [10].

2.2. Description of method

The fabrications of cellular lightweight firebricks are carried out by following 4 steps: (1) Preparation of foam of water by mechanical agitation, by mixing water and foaming agent. The characteristics and origin of the air entraining agent are listed in table 1. (2) Mixing of refractory materials with foam for obtaining foamed slurry. These solid ingredients *(SI)* are sieved under 250 microns; they are composed of DD kaolin, of calcined DD kaolin (chamotte), and of mineral binder. The use of chamotte allows decreasing the large contraction which undergoes DD kaolin during firing. Indeed that kaolin undergoes a linear firing shrinkage which goes beyond 25 % (Figure 1) from its initial length which is corresponding to over 50 % of voluminal withdrawal, which can cause deformations to the samples at high temperature. Optimal mixes which avoids the deformations and guarantee a minimal solidity at dry state is composed of the 70 % of kaolin and 30 % of chamotte. This mix undergoes an average voluminal withdrawal of 15 % (Figure 3). The last ingredient which is an refractory cement allows fast drying of the cast foamed slurry without collapse, and it provides solidity to the dry sample. The optimal quantity of cement which avoids collapse is ~20 % in weight based on the weight of crude kaolin and calcined kaolin. (3) Casting the slurry into parallelepipedic metallic moulds to drying at room temperature during 24- 48 hours, followed by total drying in oven at 80 °C. (4) Sintering of specimens is carried out by heating up to 1350 °C according a heating rate of 10 °C/min, followed by soaking during 2 hours.

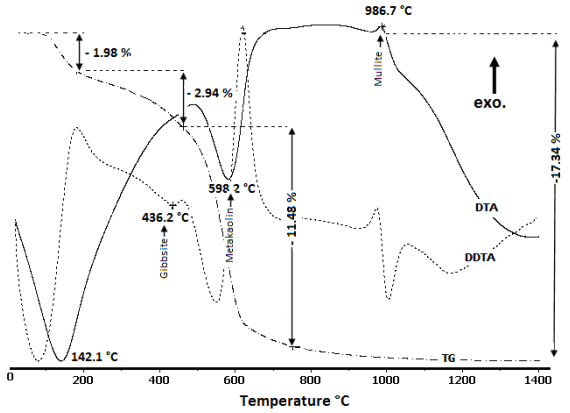

Figure 1. DTA, DDTA, and TG of DD3 kaolin

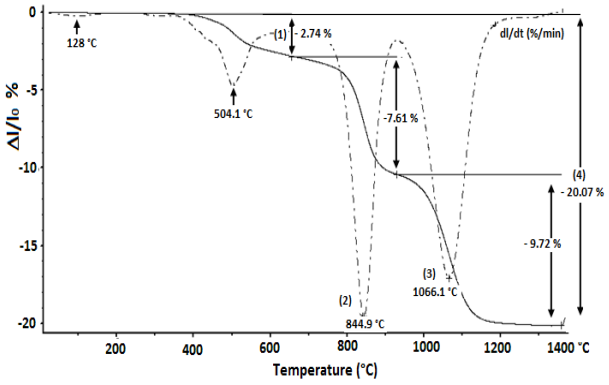

Figure 2. Dilatometer curve of DD3 kaolin

To investigate the reproducibility and the accuracy of this method, five formulations of foamed slurries are prepared with a same weight of solids ingredients (120 g). For increasing or decreasing the bulk densities of cellular specimens, we increase only the volume of the added

water which involves the increasing or decreasing of volume of foamed slurry. The tenors of kaolin, of chamotte, and of binder are 30 g, 70 g and 20 g respectively, which gives 120 g. The quantity of air-entraining (20 cm^3) remains also unchanged. The details of their formulations are given in table 1. The optimal fluidity of foamed slurry which allows easily the filling of moulds is obtained with a ratio «Solids Ingredients/water" (SI/W) which varies from 2.00 to 3.00. This ratio can vary according to the content of argillaceous matter and of its plastic nature; a high content of kaolin decreases this ration and gives a large viscosity of foamed slurry which prevents the filling of the moulds.

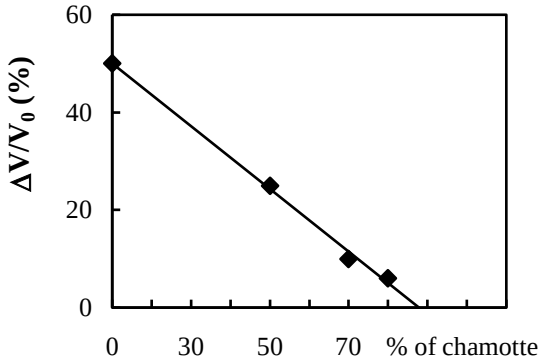

Figure 3. Voluminal shrinkage of fired DD3 kaolin as a function of tenor of chamotte

The predicted densities *(d_p)* are calculated starting from the total weight of foamed slurry without the loss on the ignition of the kaolin + the weight of water and foamed agent, which is divided by the calculated volume of cellular specimens. Total voluminal withdrawal which the specimen undergoes during drying and firing is evaluated at 15 % of the initial volume.

Table 1. Composition of foamed slurries and predicted bulk densities

		Formulation				
Designation	Units	F1	F2	F3	F4	F5
Solids Ingredients (kaolin, chamotte, binder)	g	100				
Foamed agent	cm^3	20				
Water	cm^3	35	40	45	50	55
SI/W ratio	g/g	2.18	2.44	2.73	2.87	2.96
Volume of foamed slurry	cm^3	150	170	200	250	325
Calculated weight of cellular specimen	g	115	115	115	115	115
Calculated volume of cellular specimen	cm^3	127.5	144.5	170	212.5	276.25
Predicted density of cellular specimen	g/cm^3	0.901	0.798	0.68	0.54	0.416

The experimental bulk density *(d_{ex})* of the cellular refractory specimen is evaluated starting from their weight and their geometrical measurements. This experimental value of bulk density is

compared to predicted value (d_{pr}) to evaluate the precision and reproducibility of this method of elaboration of cellular materials. The specific density (d_{sp}) is determined starting from the weight-volume ratio of water moved of cellular specimen powder (<60 μ), then total porosity (P_t) is estimated starting from the ratio (d_{ex}/d_{sp}). Water absorption (W_{ab}) is determined from the weight difference between the dry specimen and the water saturated sample (immersed for 2 h in boiling water). NETZSCH 402 PC dilatometer is used to investigate the thermal behaviour of DD kaolin, and to determine the thermal linear coefficient of expansion (TEC) of cellular lightweight refractory. DTA-TG (STA Netzsch 409 PC) (NETSCZH 409 PC) is also used to characterize the DD kaolin. The appreciation of mechanical resistance at rupture is carried out by the test of flexural strength in three points loading, at room temperature, by using an universal testing machine (Zwick/Roell). Observation of microstructure and cells are carried out by optical microscope.

3. Results

3.1. Physical characteristics

Table 2 summarizes the main characteristics of cellular specimens which are elaborated; bulk density decreases from 0.90 to over 0.40 which corresponds to total porosity of 68 % to 85 % respectively. The accuracy ratio (d_{ex}/d_{pr}) indicates that method gives a good prediction of bulk densities of cellular specimens.
However, one observes that the experimental densities are lower than the calculated densities especially for low densities. That observation can be explained by the matter loss that undergoes the samples during machining before sintering. However, this result shows those values of densities are well comparable, that which confirms the possibility to predict the density of porous specimen of this method.The mechanical resistance is appreciated by the flexural strength, which is respectable for all formulations.

Table 2. Characteristics of cellular specimens

		Formulation				
Designation	Units	F1	F2	F3	F4	F5
Experimental bulk density (d_{ex})	g/ cm^3	0.880	0.743	0.627	0.495	0.392
(d_{ex}/d_{pr}) Ratio	-	0.98	0.93	0.92	0.92	0.92
Specific density (d_{sp})	g/ cm^3	2.75				
Total porosity (P_t)	%	68.00	72.98	77.20	82.00	85.75
Water absorption (W_{ab})	%	65.23	71.00	76.80	81.50	84.00
Flexural strength	kg/cm²	23.00	8.50	6.60	5.50	3.20

Figure 3 which shows the bulk density obtained and a the volume of slurry generated as a function of the volume of water, allows to predict the porosity of cellular specimens by changing

only the volume of water.

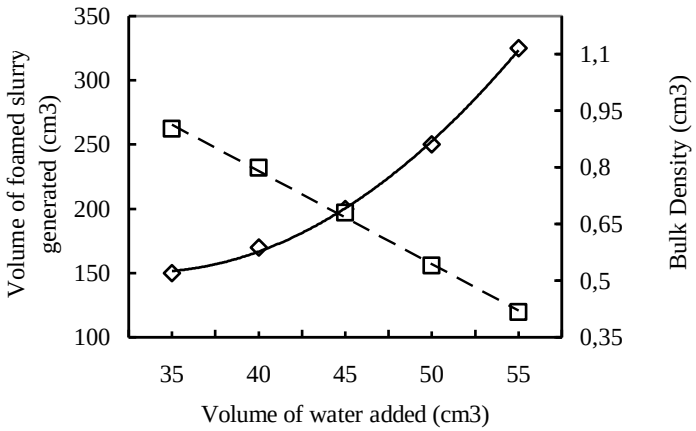

Figure 4. Bulk density and volume of slurry generated as a function of volume of water added

The dilatometric behaviour shows an regular expansion of cellular lightweight refractory from room temperature to 1100 °C (Figure 4). The average thermal coefficient of expansion (TEC) is about 6.1 $x10^{-6}$ $^{-}$°C.

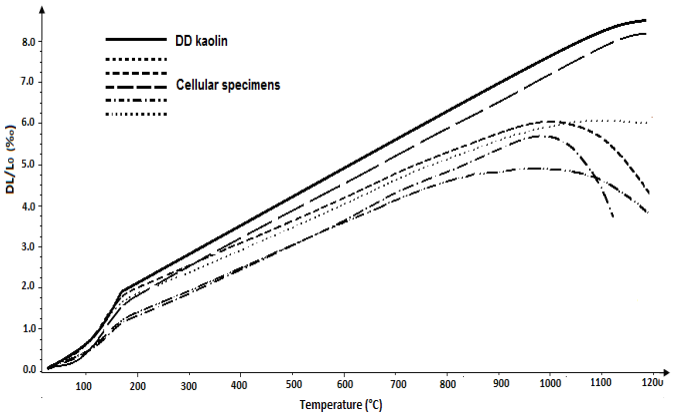

Figure 5. Dilatometric behavior of cellular refractory specimens which are compared to DD kaolin

3.2. Structure

The observation of structure shows that cellular specimens have an interconnected cellular structure with a good pore size distribution. The size of cells varies from 1/100 mm to 1 mm.

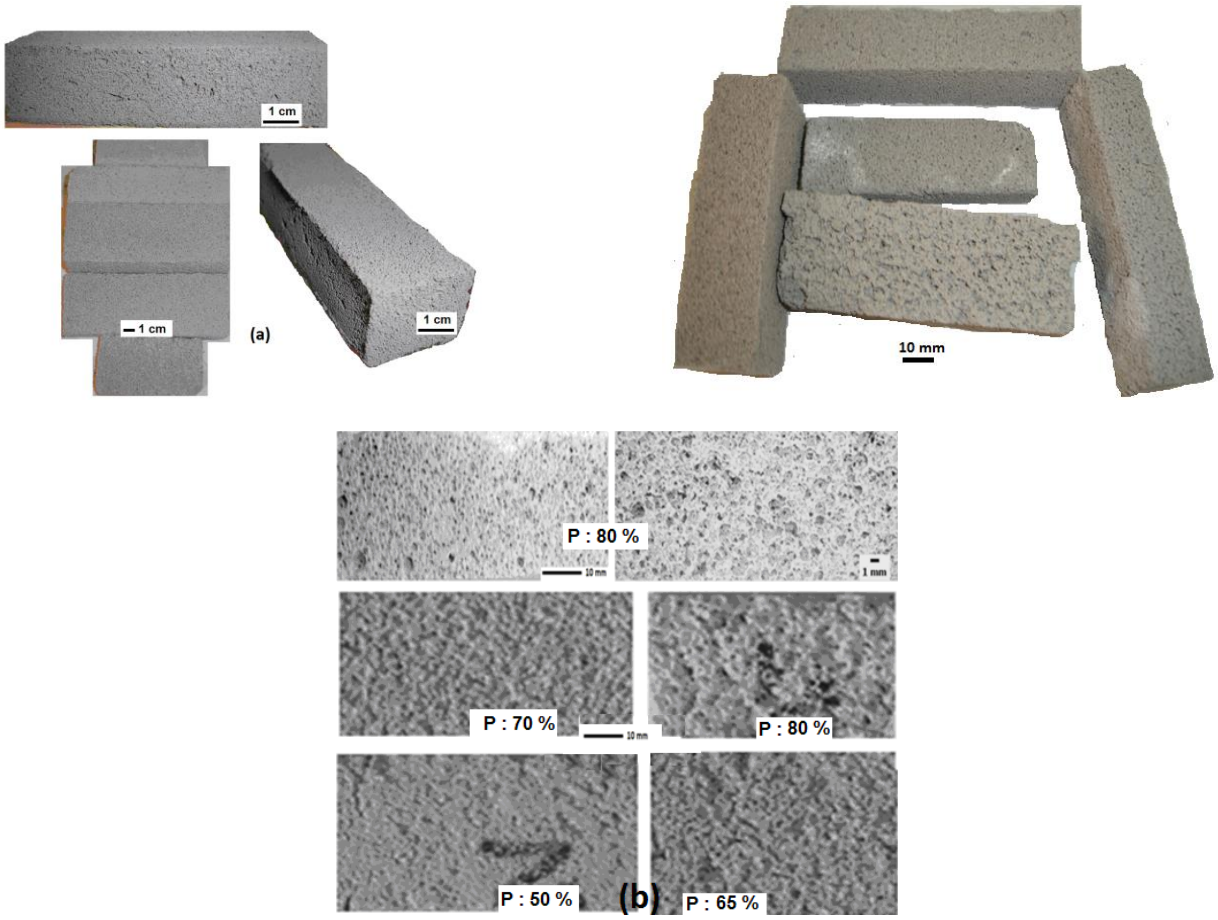

Figure 4. (a) Cellular lightweight firebricks and (b) structure of cellular specimens

4. Conclusion

Cellular lightweight refractory materials were elaborated starting from foamed slurry of water, formed of an Algerian fire-clay, and foaming agent. The consolidation is performed at room temperature, by adding refractory cement which allows the transformation of foamed slurry into a rigid and stiff body that can be machined in the green state after drying. The sintering of samples gives interconnected cellular materials with a good pore size distribution. This way of elaboration confirms well, the possibility to predict the density, the volume of voids, and the size of cells of porous specimen.

Various porosities were obtained with this novel method of elaboration of cellular materials from 60 % to 85 % of vacuum, and various sizes of cells, from 1/100 mm to 1 mm.

Their mechanical resistance of rupture is appreciable, and their temperature of melting is more than 1650 °C. The expansion as a function of temperature is regular on a large interval of temperature from 25 to 1100 ° C, and the thermal coefficient of expansion is about 6 x 10^{-6} $°C^{-1}$.

References

[1] Cellular solids structure and properties, Lorna J. Gibson, and Michael F. Asbhy – second edition Cambridge solid state, science series
[2] Ceramics, Refractories, and Glasses, Materials handbook, 2008, pp 593-689
[3] Matériaux émergeants, Christian Janot, et Bernhard llschner, Presses polytechniques et universitaires Romandes
[4] Nettleship, I., Applications of porous ceramics. Key Engineering Materials 122, 305–324., 1996.
[5] Johnson Kwame Efavi , Lucas Damoah , Delali Yaw Bensah , David Dodoo Arhin , David Tetteh, Development of porous ceramic bodies from kaolin deposits for industrial applications, Applied Clay Science 65–66 (2012) 31–36
[6] Jiahai Bai, Fabrication and properties of porous mullite ceramics from calcined carbonaceous kaolin and a-Al2O3, Ceramics International 36 (2010) 673–678.
[7] Luyten J, Cooymans J, De Wilde A, Thijis I. "Porous materials, synthesis and characterization, in Euro ceramics", Vii (Pt 1–3). Zurich-Uetikon: Trans Tech Publications Ltd; 2002. p. 1937-1940
[8] Boulmokh, A., Berredjem,Y., Guerfi, K., Gheid, A., "kaolin from Djebel debbagh mine, Guelma", Algeria; research Journal of Applied Sciences 2 (4): 435-440, 2007.
[9] Prasad M.S., Reid K.J. and Murray H.H. "Kaolin: processing, properties and applications", Applied Clay Sciences", 6 (1991) 87-119
[10] Jiri Konta J.; Properties of ceramic raw materials, Ceramic Monograph-handbook of Ceramics; 1981 Verlag Schmidt Gmbh freiburg i.Brg.
[11] M. Kolli, M. Hamidouche, G. Fantozzi , J. Chevalier ; Elaboration and characterization of a refractory based on Algerian kaolin; Ceramics International 33 (2007) 1435–1443