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Mullite ($3Al_2O_3{\cdot}2SiO_2$) ceramics obtained by reaction sintering of rice husk ash and alumina, phase evolution, sintering and microstructure

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ABSTRACT

The use of industrial waste (by-products) as raw materials in the ceramic industry has been under study for decades due to the economical, energy, tax and environmental advantages. The specificity of the waste requires a basic characterization and technology thereof.

The applicability of rice husk ash (RHA), as silica (SiO_2) source, in refractory and porous materials with potential structural, insulating and/or filtering applications was carried out by characterizing the ceramic behavior of stoichiometric mixtures of calcined alumina (Al_2O_3) and RHA. A reaction-sintering framework can be defined in the (Al_2O_3–SiO_2) system. The sinterability and conversion during the reaction sintering processes were studied in order to obtain mullite ($3Al_2O_3{\cdot}2SiO_2$) ceramics. Also some microstructural features of the developed materials were studied in the 1100–1600 °C range. The mullitization was studied quantitatively.

Partial densification was achieved (30%) and highly converted materials were obtained. The developed microstructure consisted in a dense ceramic matrix with homogenous interconnected porosity, with a narrow pore size distribution below 20 μm. The developed material gives enough information for designing mullite ceramics materials with either porous or dense microstructures with structural, insulating or filtering applications employing RHA as silica source and calcined alumina as the only other raw material.

1. Introduction

Attributable to the economical, energy, tax and environmental advantages the use of industrial waste (by-products) as raw materials in the ceramic industry has been under study for decades. The specificity of the waste requires a basic characterization [1–7].

The rice processing companies use the husk as fuel for drying, parboiling the cereal and heating for of storage silos. These are generally small businesses and these companies neither have processes for the use nor proper disposal of ash generated by burning. They either deposit these ashes in vacant lots or simply dump those into rivers and streams, causing pollution and aggression to the health of the population, by contaminating the air and eventually causing silicosis [8–13]. On the other hand, recycling of rice husk ash is an excellent alternative to minimize the environmental impact caused by the improper disposal of this material and reducing disposal costs in controlled landfills. The ashes have shown to be great raw material silica, creating a fine material and high reactivity. Accordingly, the silica obtained from rice bran has been used with great success as an alternative raw material for the production of various ceramic materials. Mainly in cement industry [14–16], as inorganic filler of polymer composite [17] and for processing silicon carbide ceramics [18], showing its high technological potential. The combustion of rice husk results in the production of rice husk ash (RHA), around 20% of ash is produced. This RHA presents a significant amount of silicon oxide (SiO_2) (≥90%) and is different from the ash resulting from the combustion of rice straw. In particular, as observed in the results that rice hull ash has a high SiO_2 content even higher than reported from other species of rice. Particularly the predominant polymorph observed in this RHA is cristobalite. This might be related to the relatively high temperature of the particular combustion chamber.

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Mullite ($3Al_2O_3{\cdot}2SiO_2$), the only stable crystalline phase in the Al_2O_3–SiO_2 system presents a great importance in both traditional and advanced ceramics. This is mainly due to its properties such as high thermal stability and favorable properties as conductivity and low thermal expansion, high resistance to creep and corrosion stability, along with good mechanical strength and fracture toughness [19–25]. Mullite is the crystalline phase resulting from the thermal treatment of clay minerals [19].

Mullite and mullite ceramics display a large variety of appearances, reaching from Czochralski-grown single crystals to polycrystalline and polyphase ceramics, and from very large refractory products to very tiny engineering components with high purity and homogeneity [20].

Porous mullite ceramic meet insulating material functions at high temperatures, also as materials for membrane filters of gases and liquids at elevated temperatures, as catalyst supports insulating or as insulating material. Porous mullite materials can be elaborated by different routes and different alumina and silica sources [26–35]. Moreover other by-products have been proposed as starting powders for mullite based materials [36–41].

The adequacy of this material (RHA) for this reutilization strategy was evidenced in a recent work [36] that presents the possibility of processing mullite and mullite-based ceramics and ceramic composites from Al metal wastes and waste by-products of mining tin ores with high kaolinite content. Another recent work [42] obtained porous mullite ceramic employing RHA as silica source. In that case, another alumina source (aluminum acetate) was proposed. This source resulted more expensive and difficult for large scale ceramic production. The resulting ceramics presented higher porosities with filtering applications [42].

In another recent work, mullite based materials with a filter application were proposed: obtaining a porous ceramic material processed from a kaolinitic clay and Rice husk (hull) (not directly the ash like in this work). The calorific power of the husk is a noticeable advantage besides the particular pore distribution that was correlated with the husk properties [43]. The cost effectiveness was assessed [44].

Finally a waste alumina source (with some calcination pretreatment) was employed in a similar work that proposed the elaboration of a mullite based ceramic with alumina and RHA starting powders [45]. In that work the RHA employed was also chemically pre-treated (HCl) to increase the SiO_2 content. They obtained dense materials at 1700 °C thermal cycles. The conversion was studied qualitatively. No acid pre-treatment is assessed in this study.

The principal objective of this article is to study the applicability of this by-product in refractory and porous ceramic materials with potential structural, insulating and/or filtering applications by characterizing the ceramic behavior of stoichiometric mixtures of calcined alumina and RHA without any chemical pretreatment. Particularly to establish sinterability and conversion during the reaction sintering processes in order to obtain mullite ceramic ($3Al_2O_3{\cdot}2SiO_2$). Previously a characterization of the non-conventional silica source was carried out. Finally we intend to describe some microstructural features of the developed materials.

2. Experimental procedure

2.1. Starting materials

The employed alumina source is a typical fine grained calcined alumina usually used for the refractory manufacture ((α-Al_2O_3), A-2G, Alcoa Inc., USA). The rice husk ash employed (RHA) is produced in grill type biomass boiler only fed by rice husk. This produces 10 tons/day of ash. After firing this alternative fuel produces approximately 20% of ashes [8,13]. The XRD pattern is shown in Fig. 1, the results of the Rietveld–LeBail quantification as well [46–49].

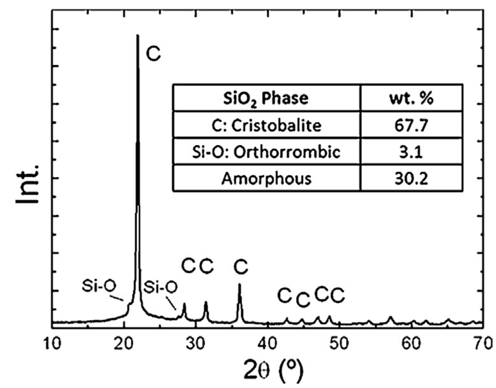

Fig. 1. XRD pattern of the rice husk ash (RHA).

Table 1
Chemical composition of the as received rice husk ash.

Oxide	(% wt.)
SiO_2	94.72
SO_3	0.17
CO_2	0.23
P_2O_5	0.51
CaO	0.21
MgO	0.18
Na_2O	0.05
K_2O	0.46
Al_2O_3	<0.05
Fe_2O_3	0.05
Cl^-	0.02
C	3.39
Melting point (°C)	>1200
Calorific power (cal/g)	351
Specific surface (BET: m^2/g)	11.22

The main properties are shown in Table 1. It can be seen that the important amount of cristobalite is remarkable, probably a consequence of the high firing temperature. The expected amount of amorphous phase was also detected and pondered. The particle size distribution can be observed in Table 2, is coarse and makes necessary the proposed milling pre-treatment: in this case we propose a simple ball milling.

2.2. Processing

Ceramic materials were processed in lab scale employing the RHA as SiO_2 source. Stoichiometric ($3Al_2O_3$:$2SiO_2$) mixtures were wet ball milled (1000 cm^3 for 24 h) in order to make those adequate the particle size and at the same time to achieve an adequate mixture. The resulting powder was then dry pressed with a

Table 2
Particle size distribution of the studied RHA.

Sieve number (ASTM)	Aperture (mm)	Retained (% wt.)
18	1.00	0.78
30	0.595	5.61
50	0.297	42.05
80	0.177	24.32
100	0.149	5.43
120	0.125	6.81
150	0.105	2.33
≤150		12.62