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Short communication

Development of mullite/zirconia composites from a mixture of aluminum dross and zircon

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Abstract

In order to obtain mullite/zirconia composites, mixtures of aluminum dross and zircon were sintered. Aluminum dross was collected and purified by a milling, sieving and washing process. Stoichiometric mixtures of aluminum dross and zircon were sintered at several temperatures (1400, 1450 and 1500 °C) for several periods of time (2, 4 and 6 h). After the purifying treatment the dross contained mainly Al_2O_3, AlN, $MgAl_2O_4$, SiO_2 and metallic Al. A homogeneous mullite matrix with small zirconia particles was obtained by sintering the aluminum dross–zircon samples at 1500 °C for 6 h.

Keywords: B. Composites; Aluminum dross; Mullite/Zirconia

1. Introduction

Aluminum scrap recycling generates important amounts of dross, which forms on the surface of the aluminum liquid bath [1]. The dross composition varies as there are different purification procedures. In general, the dross may contain Al_2O_3, AlN, Al_4C_3, SiO_2, MgO, Al and minor quantities of Si, Fe, and Mg [2]. There are several methods for dross treatment and most of them are focused on the recovering of metallic aluminum [3–5]. There is a great amount of small aluminum recycling companies that do not treat the generated aluminum dross. This is usually confined in landscapes in incandescent state, allowing the metallic aluminum to oxidize, therefore this kind of aluminum dross has a high alumina content. Aluminum dross has been used in several applications, such as reinforcing material in aluminum-dross composites [6], synthesis of spinel [7] by solid-state reaction with MgO and in the synthesis of SiAlON [8] by the carbothermal reduction and nitridation process. In these works the treatment of the dross used as raw materials was not specified and only one source of dross was used, which makes the results only valid for that specific dross. Since aluminum dross contains alumina and silica, this material is suitable for the synthesis of mullite, which is characterized by low-thermal expansion, high strength at elevated temperatures, chemical stability [9], high refractoriness and good thermal shock resistance [10]. On the other hand, mullite has low-fracture toughness (K_{IC}) and, in order to improve K_{IC}, zirconia particles are added to form a composite with a matrix of mullite and zirconia particles homogeneously distributed [11,12]. In this work, aluminum dross samples from different sources were treated and characterized in order to obtain the raw material to fabricate mullite–zirconia composites from mixtures of aluminum dross and zircon.

2. Experimental

Dross samples from three different aluminum processing companies were collected and labeled as dross samples A, B and C. Every sample was ground in a jaw mill to a size below 3.81 cm (1.5 in.). The material was screened and aluminum particles above 1 cm (2/5 in.) were separated. The samples were dry-ball-milled for 40 min and the samples were screened

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using a #12 ASTM mesh. The iron was magnetically separated. Dross samples were then ball-milled for 3 h and screened using a #150 ASTM mesh. In order to eliminate soluble salts (NaCl, KCl), the dross samples were washed in boiling deionized water for 2 h. Due to the presence of calcite, the samples were washed in a solution of 1:1 deionized water:acid acetic for 30 min at 80 °C.

The amount of alumina available for the reaction was determined as follows: pellets of dross were conformed by uniaxial pressing at 100 MPa and sintered at 1400 °C for 2 h and then analyzed quantitatively by XRD using the Rietveld technique. A mixture of the three dross samples in equal amounts was prepared. The composition of this mixture was calculated from the XRD results. Powder mixtures of aluminum dross and zircon (−45 μm) in stoichiometric amounts were homogeneized in a ball mill in acetone for 4 h. The mixture was dried at 80 °C for 4 h and ground in a mortar to eliminate agglomerates. Pellets of 1.5 cm in diameter were conformed by uniaxial pressing at 100 MPa. The pellets were sintered at several temperatures (1400, 1450 and 1500 °C) for different periods of time (2, 4 and 6 h). Additionally, a sample of dross mixture was high-energy milled until the particles reached a size below 45 μm. The zircon was also high-energy milled for 7 h. A mixture of milled zircon and aluminum dross were prepared following the procedures described above. Pellets of 1.5 cm in diameter were conformed by uniaxial pressing at 100 MPa and sintered at 1500 °C for 6 h. The sintered samples were analyzed by XRD (Phillips, X'pert PW3040), with copper anode (λ_{Cu} = 1.54056 Å) at 0.025 step each 1.25 s from 10 to 80 2θ. The pellets were analyzed on the surface in order to identify any tetragonal zirconia retained during the sintering processing. Some of the pellets were cross-sectioned and ground using SiC papers 320, 500, 800, 1200 and #2400 and a final polishing with diamond paste of 3 μm. Samples were carbon-coated and analyzed by SEM.

3. Results and discussion

Table 1 shows the results of the quantitative analysis of the sintered dross samples. The main phases detected were alumina (Al_2O_3), mullite ($Al_6Si_2O_{13}$) and spinel ($MgAl_2O_4$), sample C contains also rutile (TiO_2). These results showed that the dross samples from different process can be treated following the same purification procedure to obtain a raw material for the fabrication of ceramics. Fig. 1 shows the XRD patterns of the sintered pellets of mixtures of aluminum dross and zircon. The formation of mullite was observed at the lowest temperature used (1400 °C) for 2 h. The presence of zirconia indicates that the decomposition of zircon started at low temperature. The XRD pattern corresponding to the sample

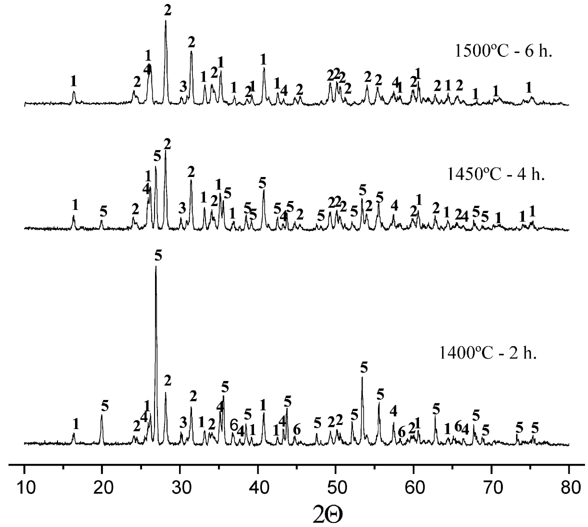

Fig. 1. XRD patterns of mixtures of aluminum dross–zircon sintered at several temperatures for different periods of time: (1) $Al_6Si_2O_{13}$, (2) ZrO_2 monoclinic, (3) ZrO_2 tetragonal, (4) Al_2O_3, (5) $ZrSiO_4$ and (6) $MgAl_2O_4$.

Table 1
Quantification of phases using the Rietveld technique for the sintered dross samples

Compound	wt.%		
	A	B	C
Al_2O_3	46.62	48.96	61.9
$MgAl_2O_4$	20.14	15.16	16.7
$3Al_2O_3$ $2SiO_2$	33.24	35.87	19.25
TiO_2	0	0	2.08

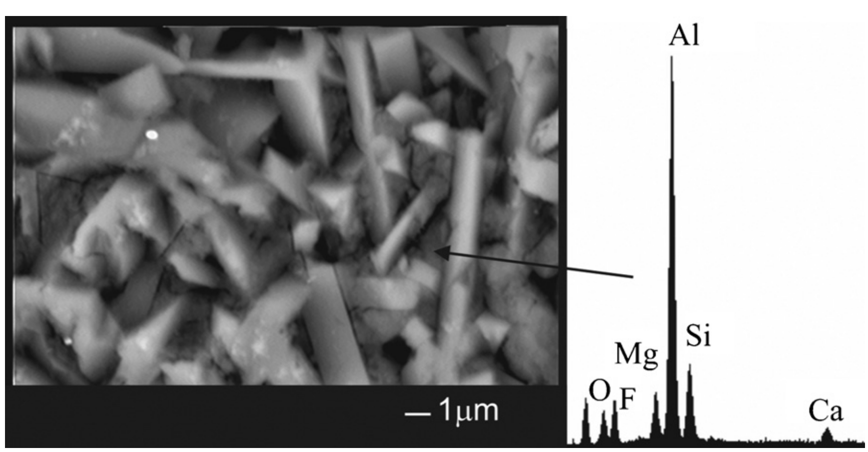

Fig. 2. EDS analysis at the interface of two mullite needles.

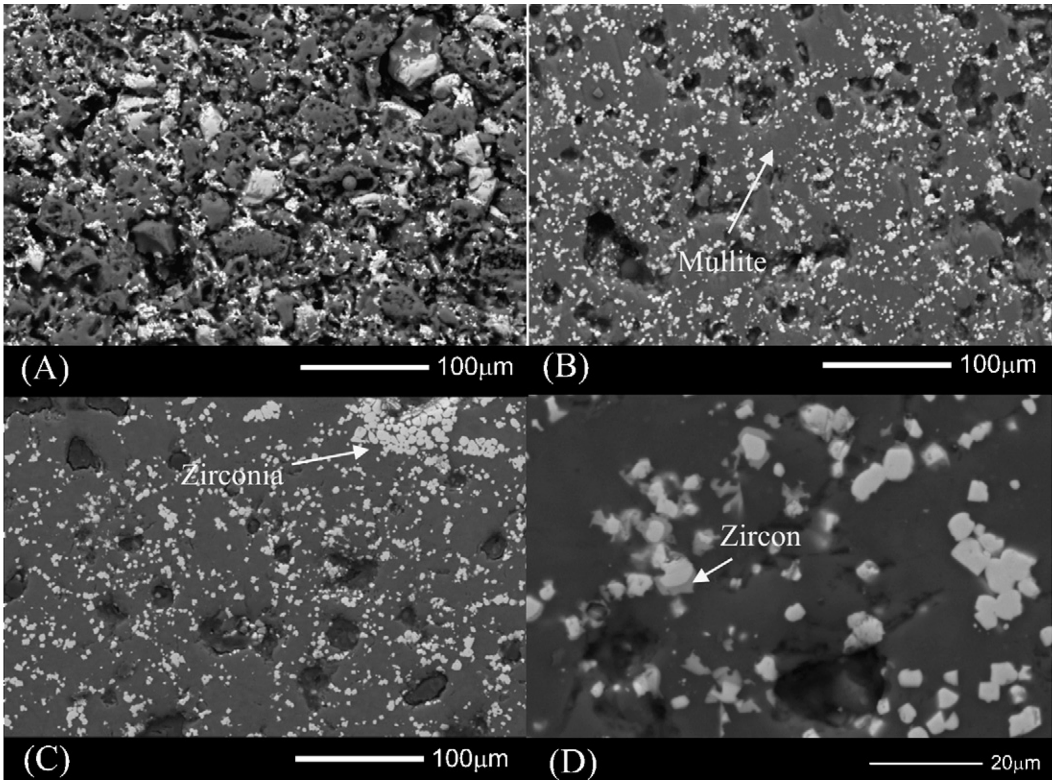

Fig. 3. SEM micrographs of the samples of mixtures of aluminum dross–zircon at 300×: (A) 1400 °C-2 h, (B) 1450 °C-4 h, (C and D) 1500 °C-6 h.

sintered at 1450 °C for 4 h shows an increase in the intensity of the zircon and mullite peaks. However, at this stage the reaction was not complete since peaks corresponding to alumina and zircon were still detected. The XRD pattern of the sample sintered at 1500 °C for 6 h shows that the main components were mullite and zirconia, some alumina was also detected. The XRD pattern of the sample sintered at 1450 °C for 4 h shows the presence of spinel ($MgAl_2O_4$), this spinel was present in the dross sample as received.

Spinel was not detected by XRD diffraction in the sample sintered at 1500 °C for 6 h. Fig. 2 shows an EDS analysis of an interface between two mullite needles, the peaks corresponding to Ca and Mg may indicate reaction of spinel with silica and some impurities. Fig. 3 presents SEM micrographs corresponding to the samples shown in Fig. 1. The sample sintered at 1400 °C for 2 h (Fig. 3A) showed a high amount of porosity and angular zircon particles. As the temperature and time increased (1450 °C for 4 h, Fig. 3B) porosity coalescence and a rearrangement of the zirconia particles in a mullite matrix were observed; additionally, the particles of zircon are not visually identified. The micrograph corresponding to the sample sintered at 1500 °C for 6 h, Fig. 3C, showed a more homogeneous microstructure with a continuous mullite matrix and dispersed zirconia particles. However, porosity and some zirconia agglomerates remain in the microstructure, which indicates that the densification process was not complete. A micrograph corresponding to the sample sintered at 1500 °C for 6 h at higher magnification, Fig. 3D, shows dispersed zirconia particles of about 5–10 μm and some zircon particles that were not completely dissociated, undetectable by X-ray analysis because of its very low content. The presence of alumina and zircon in the sample sintered at 1500 °C for 6 h indicates that the reaction was not complete and a longer time or temperature is needed. Fig. 4 shows the microstructure of a sample obtained with a dross particle size of 45 μm and zircon particle size less that 10 μm, sintered at 1500 °C for 6 h. The porosity was reduced in comparison to the samples with a larger particle size (Fig. 3C). The sample presented a homogeneous structure consisting of a continuous mullite matrix and zirconia particles homogeneously distributed. Particle size has a strong influence on the final microstructure of the composites.

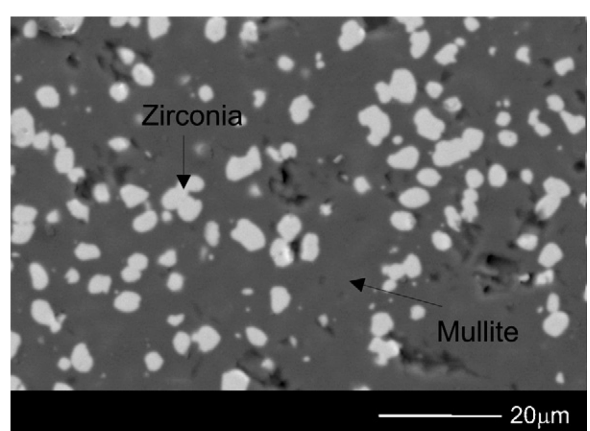

Fig. 4. SEM micrograph of a sample of aluminum dross (45 μm) and zircon (10 μm) sintered at 1500 °C for 6 h.

4. Conclusions

A simple dross treatment process consisting of grinding, sieving and washing was effective to obtain a material suitable for ceramics fabrication. Mullite–zirconia composites were obtained from mixtures of purified aluminum dross and zircon. Samples sintered at 1500 °C for 6 h showed a continuous mullite matrix with dispersed zirconia particles. Residual alumina and zircon indicated incomplete reaction. A more homogeneous mullite matrix with small zirconia particles was obtained in samples prepared by high-energy milling.

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