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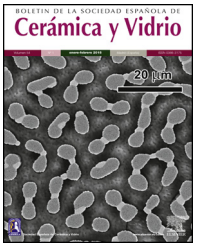

Mullite fabrication from natural kaolin and aluminium slag

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ABSTRACT

The structural transformations of kaolin–aluminium slag mixtures during heating were studied using differential thermal analysis (DTA), thermal gravimetric analysis (TGA), Fourier transform infrared (FTIR) spectroscopy, X-ray diffraction analysis (XRD) and scanning electron microscopy (SEM). The amount of formed mullite increases with the firing temperature. At 1500 °C, the mullitization of the mixture is almost complete. The morphology of the formed mullite is bimodal (primary and secondary phases). The primary mullite, formed from processing of kaolin by the gradual collapse of metakaolin from 990 °C, has a shape of elongated crystals. The other hand, the secondary mullite formed by solution-precipitation from the glass phase in the presence of alumina particles has a shape of acicular grains.

Producción de mullita a partir de caolín natural y escoria de aluminio

Palabras clave:
Mullita
Caolín
Escoria de aluminio
Transformación de fase

RESUMEN

Se estudiaron las transformaciones estructurales de las mezclas de caolín y escoria de aluminio durante el calentamiento mediante análisis térmico diferencial (ATD), análisis termogravimétrico (ATG), espectroscopía infrarroja por transformada de Fourier (EITF), análisis por difracción de rayos X (XRD, por sus siglas en inglés) y microscopia electrónica de barrido (MEB). La cantidad de mullita creada aumenta con la temperatura de cocción. A 1.500 °C, la mullitización de la mezcla es casi completa. La morfología de la mullita creada es bimodal (fases primaria y secundaria). La mullita primaria, creada a partir del

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procesamiento del caolín por la paulatina desintegración del metacaolín a partir de 990 °C, tiene una forma de cristales alargados. Además, la mullita secundaria creada por disolución-precipitación a partir de la fase de vidrio en presencia de partículas de alúmina tiene una forma de granos aciculares.

Introduction

Mullite, whose chemical composition is found between $3Al_2O_3$-$2SiO_2$ and $2Al_2O_3$-SiO_2, is one of the most studied and used ceramics. Its applications are very diverse and cover from refractory field to technical applications [1–3]. In air and under atmospheric pressure, it is thermally and chemically stable from room temperature to melting one [4]. It has very good thermo-mechanical properties, which allows us to use it as structural elements such as thermal engines. Indeed, sensitivity to creep is very limited [5]. Its thermal expansion coefficient is relatively low leading to a good thermal shock resistance [6].

We use different precursors and methods to synthesize mullite. We manufacture it through several reactive processes at different temperatures [7]. Each fabrication method has specific features depending on the wanted application and product performance. Solid phase reaction oxides alumina-silica mixtures at high temperature, generally above 1400 °C [8], can lead to mullite formation. By sol–gel methods [9], or co-precipitation of mixtures of salts in solution based on silicon and aluminium, we synthesize low-temperature mullite [11]. On the contrary, when the precursors are natural alumino-silicate clays, we form the mullite by thermal processes lower than 1300 °C [12].

Kaolin, whose main constituent is kaolinite ($Al_2Si_2O_5(OH)_4$), undergoes successive structural and microstructural transformations during its firing [13]. The decomposition of kaolinite in metakaolinite takes place between 400 °C and 630 °C. The gradual collapse of metakaolin promotes the formation of nanoscale crystals of mullite, in addition to formation of free amorphous silica. At 1200 °C, the crystallization of cristobalite is triggered from amorphous silica, and a considerable amount of mullite crystals is developed [14]. The last transformation step is the verification of cristobalite, which occurs at a temperature generally above 1400 °C. The presence of some impurities, such as CaO, Na_2O and K_2O, in the initial kaolin favours vitrification of cristobalite at lower temperatures. To obtain a secondary mullite, this silica glass can be 'mullitized' by adding alumina, aluminium hydroxides or aluminium slag [15]. Secondary mullite is different to the primary one by the morphology and grain size [16]. Primary mullite forms needles, due to the presence of the vitreous phase [17]. While secondary mullite is in the form of aggregates of needle-like small grains [18], we form it by a solution-precipitation of the glass phase in contact with the particles of alumina. According to Pascual et al., addition of 1–3 wt% of MgO to the kaolin-alumina mixture promotes grain growth of primary and secondary mullite, and the addition of 1–5% Y_2O_3 inhibits grain growth of the secondary mullite [19]. We detect no difference in the X-ray diffraction spectra of the two mullites. However, we observe differences between infrared absorption spectra.

The aim of the present work is the mullitization of two types of Algerian kaolin by adding aluminium slag. We have sintered stoichiometric mixtures at temperatures between 1000 °C and 1500 °C for 2 h. In order to understand microstructural transformations and chemical reactions, we performed differential thermal analysis coupled with thermal gravimetric analysis (DTA/TGA). We have analysed, by X-ray diffraction (XRD), the phase transformations during the various thermal treatments. We determined the morphology of mullite by scanning electron microscopy (SEM). We studied the chemical bonds of the phases formed during the firing of aluminium kaolin-slag mixtures at different temperatures by Fourier transform infrared (FTIR) spectroscopy.

Experimental procedure

We have used two natural kaolin extracted from two various sites of Djebel Debbagh near Guelma (North-East of Algeria). The amount of alumina in their composition and colour differentiate the two kaolin. The first composition noted DD1 is white coloured where as the second one (DD3) in greyish coloured. The difference in their colours is due to the kind of their containing impurities.

Their absolute densities, measured with a helium pycnometer apparatus are respectively 2.63 g/cm^3 for DD1 and 2.61 g/cm^3 for DD3. The used aluminium slag, waste of aluminium industry, is provided by ALGAL company (Algeria). It is white coloured and composed by 87% (in mass) of alumina, its absolute density is about 4.03 g/cm^3.

Tabla 1 represents the chemical compositions of respectively both kaolin and aluminium slag.

To obtain mullite we prepared, with a stoichiometric composition, two mixtures of DD1 and DD3 with aluminium slag, they are respectively noted MDD1 and MDD3.

We milled the mixtures with a planetary ball milling apparatus during 5 h; with a report powder/balls equal to (50/200) g in 80 ml water.

We dried the mixtures at 110 °C and then crushed and sieved to 45 μm meshes. The samples were uniaxial pressed at 100 MPa for obtaining bars shaped (40 mm × 10 mm × 10 mm), after that they were heated up to 600 °C at a rate of 1 °C/min for 1 h to avoid cracking. Then we sintered the samples between 1000 °C and 1500 °C for 2 h, with a heating rate of 5 °C/min. We subjected powders mixtures MDD1 and MDD3 to differential thermal analysis (DTA) and thermo gravimetric analysis (TGA) using Setaram Setsys 16/18 simultaneous TG/DTA analyzer with α-alumina as the reference material. We conducted the test between the room temperature and

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