Ceramics International 42 (2016) 11104–11117

Contents lists available at ScienceDirect

Ceramics International

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

Influence of zircon particle size on conventional and microwave assisted reaction sintering of in-situ mullite–zirconia composites

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ARTICLE INFO

Article history:
Received 24 February 2016
Accepted 5 April 2016
Available online 6 April 2016

Keywords:
Microwave processing
Mullite
Sintering
ZrO_2

ABSTRACT

Mullite–zirconia composites were fabricated by reaction sintering of $ZrSiO_4$ and α-Al_2O_3 using conventional heating and microwave processing. The powder mixtures were prepared from sub-micron zircon powders with three different particle sizes and CIPed as coin shaped samples. The samples sintered both in a muffle furnace and microwave furnace. The open porosities, bulk and true densities were measured. Phase transformations were characterized by X-ray diffraction and microstructures were evaluated by scanning electron microscopy. The effects of zircon particle size on the in-situ transformation system and mullitization was evaluated for both methods. As a result, decreasing zircon particle size decreases the in-situ transformation temperature for 25 °C (1575 °C) in conventional heating. Microwave assisted sintering (MAS) lowers the transformation temperature at least 50 °C by lowering the activation energy more efficiently and gives better densification than conventional sintering. Furthermore, milling also produces structures having finer mullite grains.

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1. Introduction

Alumina-based ceramics show high values of refractoriness, hardness, strength and resistance to chemical attack, being these properties suitable for a broad range of industrial applications. Nevertheless, owing to their brittleness, these ceramics are unsuitable for applications in conditions of severe thermal shock or in structural applications requiring high toughness. In order to overcome the inherent brittleness of alumina and of a great number of ceramic materials whose other properties may be useful in an industrial context (mullite for instance), some mechanisms such as particulate reinforcement can be incorporated in the matrix to make it tougher. The choice of reinforcing phase is crucial in the fabrication and properties of composites. Chemical compatibility and thermal expansion match between reinforcing and matrix phases and the morphology (platelet, whisker or particulates) of the reinforcing phases could affect the processing and final properties of the composites. Recent studies showed that the size of the reinforcing phases also played an important role in the properties of ceramic composites. With nanometer-sized SiC dispersoids in alumina or silicon nitride, the strength and toughness of the composites were greatly improved. The improvement has been attributed to changes in the microstructure (refinement of microstructure; flaw size population), grain boundary/interface strengthening and/or fracture mechanisms in nanocomposites [1].

One of the widely used particulate reinforcement in mullite matrix is *zirconia*. Fine zirconia particles toughen the material mainly as a consequence of the *tetragonal-monoclinic phase transformation* that can occur during the fracture process or during sintering cooling. In the former case, the stress induced phase transformation toughening mechanism is activated, while in the latter, the microcracks produced in the matrix as a result of the phase transformation that occurs during the sintering cooling are responsible for the increase of crack propagation energy [2]. Therefore, zirconia needs dopants like *yttria or ceria* in order to stabilize tetragonal phase at room temperature, which could later turn to monoclinic, and provide volume expansion along with compressive strain in the structure [3].

Mullite–ZrO_2 in the ZrO_2–SiO_2–$Al2O_3$ ternary phase system is one of the composites that has attracted the attention of manufacturers and scientists. This is because of the unique properties of mullite, such as high melting point, chemical stability and excellent creep resistance, low thermal expansion and thermal conductivity, attractive bending strength at high temperature and good thermal shock resistance. However, mullite has low fracture toughness and relatively low strength at room temperature [4–7]

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http://dx.doi.org/10.1016/j.ceramint.2016.04.016
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compared with other ceramics such as ZrO_2, SiC and Si_3N_4 [8] and the difficulties in obtaining fully dense objects [8,9] because of the high activation energy for ion diffusion through the mullite lattice [8] hinder its wide-scale engineering applications [8,10,11]. On the other hand, at high temperature, ZrO_2 has high mechanical strength and is slightly wetted by siliceous and metallic melts compared with mullite. Therefore, Mullite–ZrO_2 composites find extensive applications especially in refractories thanks to their excellent resistance to creep, spalling and high mechanical strength [2,4,12]. In addition, mullite–ZrO_2–Al_2O_3 multicomposites are widely used in forehead feeders and glass melting furnaces as glass contact material such as plungers, spouts, tubes, orifice rings, etc. [13]. The widespread use of this class of materials is due to their high corrosion resistance attributed to the microstructure and to zirconia being slightly wetted by siliceous and metallic melts. Since the solubility potential of zirconia in silica is low, the chemical attack of the composite is comparatively low [12].

Mullite–ZrO_2 composites can be obtained through various processing techniques, including sol–gel mixing of SiO_2, Al_2O_3 and ZrO_2; reaction sintering of zircon and alumina ($ZrSiO_4$–Al_2O_3) by processes such as *plasma arc spraying* [8], *hot-pressing* [9], *sol–gel processing* [14,15], *reaction sintering* [2]; or mechanical mixing of mullite and ZrO_2. Reaction sintering process is more attractive one because it is cheaper compared to the starting raw materials, and because it can easily be implemented industrially (in view of conventional equipment and processing techniques) [16–18]. Reaction sintering of zircon and Al_2O_3 may lead to excessive undesired glassy phases because of the presence of additives that are arbitrarily added to promote the reaction sintering processes by enabling reaction and densification simultaneously [19]. The disadvantage of using reaction sintering is that the dissociation reaction of zircon causes porosity that may lead to degradation of the mechanical properties [2,4]. Reaction sintering process is difficult to control because of occurring chemical reactions, such as dissociation of $ZrSiO_4$ and formation of mullite and SiO_2 free zirconia, as well as densification during sintering at various temperatures [18,20]. On the other hand crystal structure of zirconia in the mullite matrix during reaction sintering has a significant effect to toughening mechanism. Also, in order to favor both chemical reaction and densification during sintering, various additives such as magnesia are commonly used [18].

Recently, several studies on the systems alumina/zirconia, mullite/zirconia and cordierite/zirconia have shown that fine ZrO_2 dispersions in a ceramic matrix can affect the sinterability and considerably improve the mechanical properties. Mullite exhibit significant grain growth, which inhibit sintering kinetics. Micrometric zirconia powder additions to mullite can overcome all the disadvantages resulting from individual mullite [10]. In particular, the addition of ZrO_2 promotes densification and retards grain growth of mullite phase in mullite–zirconia composites [19]. The retention of the zirconia tetragonal phase in the mullite matrix at room temperature is essential for stress induced transformation of the tetragonal phase to the monoclinic one [11]. This toughening of mullite by ZrO_2 is understood to be a result of the transformation of tetragonal-ZrO_2 (t-ZrO_2) to monoclinic-ZrO_2 (m-ZrO_2) which is called transformation toughening and microcrack formation around m-ZrO_2 (microcrack toughening) during crack propagation [12]. A well-known processing route to improve the fracture toughness of mullite is to disperse or precipitate t-ZrO_2 particles in a mullite matrix, this resulting in transformed m-ZrO_2 under an applied stress. The stress-induced phase transformation accompanied by volume expansion and shear deformation may cause beneficial toughening mechanisms to become operative [12].

Besides increased heating rate, uniform heating and energy saving, microwave sintering enables higher density and mechanical properties of ceramics. Stabilized tetragonal zirconia is a transformation toughening agent and also a highly efficient microwave absorber. It can be preferentially heated in a microwave field [10,21]. Bodhak et al. have found that microwave sintering results in better densification and thus better mechanical properties in pure mullite and mullite–zirconia composites [21]. Ebadzadeh and Valefi have found that microwave heating favors the dissociation of zircon compared to conventional heating. However, it is found that microwave assisted heating is more advantageous than susceptorless heating since microwave absorption capability of zircon is quite low at low temperatures [22]. Moreover, microwave assisted heating of zircon/Al_2O_3 mixture was previously studied by our group and positive effect of using microwave energy on transformation kinetics such as lowering zircon–mullite transformation temperature by the help of good microwave interaction of zircon particles was found [23].

The purpose of the present work is to examine the effect of zircon particle size on the transformation kinetics of in-situ mullite–zirconia composites produced by conventional and microwave assisted sintering methods. Furthermore, the advantage of microwave energy was used in order to decrease the transformation temperature and improve the physical and mechanical properties of composites.

2. Experimental work

The stoichiometric ratio of mullite–zirconia composites according to Eq. (1) was used to calculate the composition of the raw material mixtures for the experimental samples. Only the three main oxides (ZrO_2, Al_2O_3 and SiO_2) were considered in these calculations; the presence of all other minor oxides was ignored. Based on the mullitization ($3Al_2O_3 \cdot 2SiO_2$) and dissociation of zircon to ZrO_2 given in Eq. (1), the chemical composition of the starting powders (as shown in Table 1), a mixture of zircon and alumina in proportions equal to the stoichiometric mullite–zirconia composition were prepared. Alumina (having an average particle size (d_{50}) of 7.8 μm) and zircon (d_{50}=498 nm) were used as the starting materials to form mullite–zirconia phase. 1 wt% MgO (d_{50}=10,76 μm) was added as sintering aid. As starting materials, 54.5 wt% Zircon and 45.5 wt% Alumina were used for in-situ mullite–zirconia formation.

Afterwards, zircon powder was milled for 20 and 80 min using a planetary ball mill (Fritsch “Pulverisette 7”) in tungsten carbide (WC) vials with WC balls (10 mm in diameter) at room temperature. To inhibit agglomeration, zinc stearate was added to system as a process control agent. The ball to powder weight ratio was chosen 5:1 and the rotational speed was controlled at 400 rpm. Samples containing various zircon particle sizes depend on milling time were encoded as “Z0” (as received), “Z20” (20 min milling) and “Z80” (80 min milling). The particle size distribution of zircon in various milling times are given in Fig. 1. Average particle size (d_{50}) of Z20 was 367 nm, however for Z80 45% of the average particle size was 128 nm, while 54% was 368 nm. In mixing step, alumina and zircon powders were put in a glass jar. The mixture

Table 1
Chemical composition of the starting powders.

Oxides	Zircon	Alumina
ZrO_2	66	–
Al_2O_3	0.50	99.9
SiO_2	33	–
CaO	0.40	–
Fe_2O	≤0.10	–

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