



Dense mullite–zirconia–zirconium titanate ceramic composites by reaction sintering

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Received 1 September 2015; received in revised form 15 September 2015; accepted 16 September 2015

Available online 26 September 2015

Abstract

Materials from the $\text{Al}_2\text{O}_3\text{--SiO}_2\text{--ZrO}_2$ and the $\text{Al}_2\text{O}_3\text{--SiO}_2\text{--ZrO}_2\text{--TiO}_2$ systems have several high temperature applications because they present the good refractoriness, chemical inertness, adequate mechanical and thermo-mechanical behaviors with a relatively good cost: performance ratio. In this work stoichiometric (3:2:1) molar incompatible mixtures of alumina (Al_2O_3), zircon (ZrSiO_4) and titania (TiO_2) were slip casted and sintered in a 1300–1500 °C temperature range in order to obtain mullite ($3\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2$), zirconia (ZrO_2) and zirconium titanate (ZrTiO_4) dense triple ceramic composite.

Both sintering and reaction occurred after the thermal treatments. Reaction progress and densification evolutions were established. Dense Triplex composite materials were achieved after 1500 °C treatments. The reaction-sintering was followed by XRD, TG–DTA, and dilatometry. Densification started at 1100 °C and the chemical reactions only started above 1300 °C. Aluminum titanate (Al_2TiO_5) was found to be an intermediate of the reaction after 1400 °C treatments. Materials treated below 1300 °C presented a partial densification of the unreacted starting powders. Resulting ceramic materials were characterized. The crystalline phases were evaluated, as well as the texture properties. The achieved microstructure consisted in interlocked multiphase ceramic with zirconia (monoclinic) grains. The achieved Hv and K_{IC} reached 9 GPa and 4.3 MPa m^{1/2} respectively. The dense and interlocked ceramic microstructure and relative high mechanical properties of the developed material encourages several high temperature applications. Finally it can be pointed out that after 1500 °C treatments some detrimental grain growth was observed.

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Keywords: A. Processing; C. Properties; D. Mullite; Zirconia; Zirconium titanate

1. Introduction

Composite materials have an important industrial and technological role. The designing capability of the manufacturer in properties and behaviors is enhanced by combining two or more different materials. However the final properties will not always be between the pure material ones, in fact in several cases the properties are considerably improved. Once the constituent phases and the processing conditions are

chosen the final phase content in the material becomes one of the most important processing variables [1,2].

The mullite ceramics have had and will continue to have a significant role in the development of traditional and advanced ceramics [3–6]. Mullite is the only stable crystalline phase in the aluminosilicate system, under normal atmospheric pressure at room through elevated temperatures [3]. Its chemical composition ranges from $3\text{Al}_2\text{O}_3\text{--}2\text{SiO}_2$ to approximately $2\text{Al}_2\text{O}_3\text{--SiO}_2$. It has received significant attention during the last decades as a potential structural material for high temperature applications [3–6].

Particularly mullite–zirconia composites are materials with important technological applications due to their good properties such as toughness, chemical stability, and high-creep

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resistance [7–12]. In practice they are employed in the glass industry and where high chemical and corrosion resistance are required. Zircon and alumina are largely employed as raw materials in their manufacture [13–15]. The reaction sintering processes is a suitable strategy for obtaining zirconia composites at comparatively lower costs than introducing zirconia as starting powder [16–19].

The addition of TiO₂ to mullite–zirconia composites prepared from different alumina sources leads to change of reaction sintering, densification and microstructure which can alternately change the formation temperature and retention of t-ZrO₂ phase in these composites [20–23].

The mullite–zirconia composites containing different amounts of micro- and nano-TiO₂ particles were prepared by reaction-sintering of alumina and zircon powder by other authors using slip casting as shaping method [23]. Then, the physical properties, phase composition, flexural strength and microstructure of these composites were finally evaluated. This study confirmed the strong influence that nano-TiO₂ particles exert on the microstructure and flexural strength of mullite–zirconia composites.

Zirconium titanate (ZT–ZrTiO₄) is commonly used as a dielectric in microwave devices due to its high permittivity at microwave frequencies [24–26]. ZT has been proposed for structural and thermomechanical severe applications [27,28]. Zirconium titanate (ZrTiO₄) is a well-known compound in the field of electro-ceramics, as constituent of dielectric resonators and components for telecommunications, [29–34]. There is also a family of zirconium titanate ceramic pigments [35].

The traditional preparation of ZT ceramics is based on the solid-state reaction between TiO₂ and ZrO₂ powders at high temperatures. In order to improve the functional properties of the ceramics, expensive and energy consuming post-reaction treatments are generally needed. Chemical methods based on the co-precipitation of reactive precursors were developed to prepare high purity powders and to lower the cost of the post-reaction treatments. The main features of the preparation methods of ZT powders were described by Navio et al. [33]. Controlled hydrolysis of alkoxides and sol–gel syntheses were also investigated to obtain fine particles. Spark plasma sintering of ZT materials has been recently reported [34].

Some materials from the ZrO₂–Al₂O₃–SiO₂–TiO₂ quaternary system have been studied by other authors: The phase diagram has been reported and discussed [36], the projection through the ZrO₂ corner showing secondary phases crystallizing during cooling from ZrO₂–Al₂O₃–SiO₂–TiO₂ mixtures (Z–A–S–T) containing 60 wt% ZrO₂.

The zircon dissociation (Eq. (1)) and mullite formation (Eq. (2)) behavior was described for several compositions [37]. They showed that an interesting family of dense tough ceramics can be processed from inexpensive starting powders in the 1350–1500 °C range. Due to low cost and availability the starting powders chosen in this work are alumina, zircon and titania. The consequent thermal reactions were established; particularly a reaction sintering framework was described. The zircon dissociation and the formation of ZrTiO₄ and 3Al₂O₃·2SiO₂ can be ensured from the phase diagram presented in the mentioned article (Eqs. (1)–(3)). The zircon dissociation [37] occurs at high

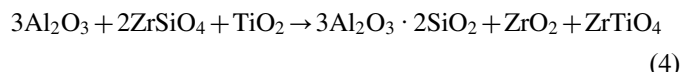
temperatures also. Finally it should be emphasized that TiO₂ forms stable solid solutions with ZrO₂ and mullite. And according to Melo et. al. the solubility of TiO₂ into ZrO₂ and (mullite) is near 4 wt% [36].



The composition studied in the present work differs from the range presented in [36]. In fact it is a simpler one, with less alumina content and an integer molar relation. The concluded sintering mechanism was enhanced by the formation of a transient liquid phase at high temperatures. And probably this mechanism involves a decrease in the mechanical behavior at higher temperatures.

1.1. Objectives

The objective of the present work is to obtain a family of dense polyphasic ceramics, with mullite as continuous matrix, from inexpensive starting powders, and to establish the effect of the key processing variables on the material properties. Another objective is to evaluate the phase composition, the developed microstructure and other technological properties. The actually studied composition corresponds to the stoichiometric molar 3:2:1 which corresponds to the global reaction (Eq. (4)), to form a triple composite from three inexpensive raw ceramic powders.



Eq. (4) is equivalent to twice Eq. (1) plus Eqs. (2) and (3). Table 1 shows the label, formula and properties of the starting and final phases.

2. Experimental procedure

2.1. Starting powders

The zircon starting powder was zirconium silicate with ZrO₂: 64–65.5 wt%, SiO₂: 33–34 wt%, Fe₂O₃: 0.10 wt% and TiO₂: 0.15 wt%, mean diameter (*D*₅₀) of 2.0 μm, specific gravity of 4.6 g/cm³, melting point of 2200 °C and hardness (Mohs) of 7.5 (KreutzonitSuper, MahlwerkeKreutz, Germany). The second starting powder was Alumina (ALMATIS A-16SG) α-alumina (*D*₅₀: 0.5 μm) (Al₂O₃: 99.8%, SiO₂: 0.025 wt%) and the third was an industrial titania powder, (TiO₂: 99.7 wt%), with a crystalline Anatase–Rutile proportions of 95–5 wt% and a mean diameter (*D*₅₀) of around 1.0 μm (Mallinckrodt Chemical Works).

2.2. Materials processing

The processing route was chosen considering processing strategies reported for similar materials belonging to the

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