Characterization of in-situ zirconia/mullite composites prepared by sol-gel technique

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Abstract

The main objective of this study was to investigate the role of zirconia addition to mullite through an in-situ reaction aimed at improving both the mechanical properties and the sinterability behavior. In this work, mullite–zirconia composites were produced using a sol-gel technique. Different amounts of zirconia (0, 10, 15, and 20 wt.%) were added to the mullite, and the calcined gels were sintered at 1550–1700°C for 1 h. The apparent porosity and bulk density of the blank and zirconia/mullite composites were estimated in accordance with ASTM C-20. The phase composition and sample morphology were evaluated via X-ray diffraction (XRD) and scanning electron microscopy analysis (SEM), respectively. Furthermore, the mechanical properties and thermal expansion coefficient (TEC) were also evaluated. The results revealed that the apparent porosity decreased and the density of the zirconia/mullite composites increased when the sintering temperature was increased from 1550 to 1700°C. However, the mechanical properties improved with increasing zirconia content and MZ20 sintered at 1700°C exhibited the maximum bending strength. The TEC results reflected the influence of the composition on the sample TEC. Samples with higher ZrO₂ content yielded higher TEC figures than those with lower content.

1. Introduction

Mullite (3Al₂O₃·2SiO₂) exhibits excellent properties such as chemical and thermal stability, efficient utilization at high temperature, high melting point, and high creep resistance [1–3]. However, this material suffers from drawbacks (e.g. low fracture toughness and poor sinterability) that have limited its application as a functional material [4,5]. Forming composites with this material has provided a solution for such problems. Many materials have been added to mullite to form composites. Addition of zirconia (ZrO₂) to mullite for composite formation enhances both the mechanical properties and the sinterability behavior and hence has gained significant attention [6]. The dispersion of fine ZrO₂ in the ceramic matrix improves the sinterability and the mechanical properties of the obtained composites [7]. In addition, improving the toughening strength of mullite by disseminating tetragonal zirconia (t-ZrO₂) in the mullite matrix has been extensively investigated [8,9]. The toughening mechanism is based on the t-ZrO₂ to m-ZrO₂ phase transformation [8,10,11]. As it is well known that t-ZrO₂ is the best for toughening of the composites. Mullite/zirconia composites have been manufactured using diverse production techniques and starting materials. A low-cost traditional sintering method has been widely used. In such a method, zircon, kaolinite, and alumina are used as starting materials [12,13]. Some authors have used halloysite, boehmite, and zirconia to prepare mullite/zirconia composites [14].

A study considering the effect of different starting materials on the properties of mullite/zirconia composites indicated that under high temperature sintering conditions, changing mullite and ZrO₂ sources had a slight influence on the sintering process and the produced phases. Moreover, the results revealed that the reaction sintered and in-situ formed composites exhibited low mechanical figures compared with the composites formed from the reaction involving ZrO₂, SiO₂, and Al₂O₃ [15].

Many authors have evaluated the effect of mechanical activation on the mullite/zirconia formation temperature [16–18]. Some of these studies indicated that 60 h of activation enhanced the formation of a composite that was sintered at 1400°C for 4 h [16,17]. Another study [18] claimed that 100 h of activation reduced the sintering temperature of 40-h milled composites to 1420°C. Nevertheless, Sistani et al. [19] stated that increasing the milling time to 72 h enhanced uniformity of the phases and restrained the t to m zirconia transformation at 1550°C. Both the tensile strength and Vickers micro-hardness of the sintered samples were enhanced with increasing milling time. The same
authors [20] studied the coinciding impact of the mechanical activation procedure and the addition of TiO₂ and zinc oxide (ZnO) to the starting materials. The impact on the diametral tensile strength, Weibull modulus, and surface roughness of the produced sintered samples was considered. They found that prolonging the mechanical activation to 72 h enhanced the formation of the t-ZrO₂ phase. However, calcination at 1550°C diminished the effect of the activation on the produced phases. They also reported that high sintering temperatures (up to 1550°C) are required to produce mullite/zirconia composites from batches containing ZnO. The results indicated that, despite the high temperatures required, the added ZnO had the most positive effect on the mechanical properties.

Various advanced sintering techniques such as spark plasma sintering, hot-pressing sintering, and microwave-assisted sintering [21–23], have recently been used to improve the sinterability of mullite composites. However, chemical methods such as hydrolytic precipitation and sol-gel techniques have been suggested as a more effective route than mechanical mixing methods [24]. Compared with the mechanical methods, the sol-gel technique shows higher homogeneity, finer grain size accompanied by high particle efficiency, and a reduction in the sintering temperature.

To improve the mechanical strength, specimens with a modified grain orientation were fabricated. Grain orientation modification is achieved by means of templated grain growth. The introduction of templated grains in ceramic bodies has led to the formation of a textured microstructure on sintering at high temperatures [25]. Tape casting is one method that can be used to generate ordered templates, and accordingly, may be employed for controlling the growth orientation of the needlelike mullite grains in the template. Tür et al. [26] found that a textured mullite/zirconia composite fabricated from mixing alumina, zircon, and aluminum borate produced textured mullite. They reported that the mechanical properties of the produced composite improved significantly due to the development of mullite grains in directions perpendicular to and parallel to the longitudinal direction.

The current study focuses on the production of zirconia/mullite ceramic composites using a sol-gel technique. To improve the sinterability and homogeneity, various concentrations (0, 10, 15, and 20 mass%) of ZrO₂ were added in-situ to the mullite sol. The influence of ZrO₂ addition to the mullite matrix on the sinterability, phase composition, and microstructure was investigated. In addition, mechanical and thermal properties for the produced pure and the ZrO₂/mullite composites were evaluated.

2. Experimental procedure

2.1. Starting materials

Chemically pure grade aluminum nitrate nonahydrate (ANN; Al(NO₃)₃·9H₂O, LOBA Cheme. India), tetraethyl orthosilicate TEOS (Si(OC₂H₅)₄, >99%, Chem. Lab. NV. Belgium), monoclinic ZrO₂ powder (99.9%, D50 = 0.4 μm, Zir-Pro, Saint Gobain, France), and nitric acid (HNO₃, >68.5%, BDH Laboratory Supplies, England) were used in this study.

2.2. Sample preparation

As illustrated in Figure 1, pure mullite (M20) was prepared using a stoichiometric composition of Al₂O₃/SiO₂ with a molar ratio of (3:2). Firstly, 0.6 M Al(NO₃)₃·9H₂O was dissolved in a double-distilled water in a ratio of ANN:H₂O = 1:28. 0.4 M Si(OC₂H₅)₄ was separately hydrolyzed with double-distilled water and a few drops of nitric acid to accelerate the hydrolysis process under continuous stirring at 80°C for nearly 2 h to obtain a stable sol. Subsequently, Si(OH)₄ sol was added in a dropwise manner to the Al(NO₃)₃·9H₂O solution. For fabrication of the zirconia/mullite composite samples, the molar ratio of Al₂O₃ and SiO₂ was kept constant at (3:2), and various amounts (10, 15, and 20 mass%) of ZrO₂ were added during the sol formation stage of the mullite. Mixing with a gentle stirring at 80°C was continued until gelation occurred. The formed gel was dried at 110°C for 24 h, and then calcined at 800°C, 1000°C, and 1200°C for formation of the pure mullite phase, with 1 h of

![Figure 1. Processing scheme for mullite and zirconia preparation of mullite composites.](image-url)
soaking time in static air in an electrically heated furnace (heating and cooling rate is 5°C/min) (see Figure 1). The composite batches with 10, 15, and 20 mass% of ZrO₂ were denoted as MZ10, MZ15, and MZ20, respectively.

The calcined powders were subsequently ground (speed: 300 rpm) for 1 h in a planetary ball mill (PM100, Retch GmbH), using a ZrO₂ jar and ZrO₂ balls as a grinding medium (2 cm diameter). To determine the physical properties and chemical composition, the batches were dry pressed (force: 30 kN) into disks (diameter: 10 mm and thickness: ≈3 mm) using a stainless mold. In addition, the mechanical properties were estimated from 5 × 5 × 60 mm rectangular bars. The pellets and rods were sintered at temperatures ranging from 1550°C to 1700°C with 1 h of soaking time in static air in an electrically heated furnace (heating and cooling rate: 5°C/min).

2.3. Characterization

2.3.1. Mullite powder

Mullite phase formation at different calcination temperatures (800, 1000, and 1200°C) was evaluated via X-ray diffraction (XRD) analysis using a Philips X-ray diffractometer with a Cu target and Ni filter. The measurements were performed at a scanning speed of 0.02°/s over a 2θ range of 5–60°. The mullite grain size and morphology were determined by means of transmission electron microscopy (TEM; JEOL JEM-2100 Electron Microscope, HRTEM, Japan) analysis, operated at 300 kV. The sample was prepared by dispersing a small amount of the powder in acetone using ultrasonic bath for 30 min. A drop of the well-dispersed suspension was deposited on a carbon coated copper grid followed by drying the grid for evaporating the solvent before TEM examination.

2.4. Zirconia/mullite composite bodies

The apparent porosity and bulk density of both mullite and zirconia/mullite sintered samples were estimated using the Archimedes method performed in accordance with ASTM C-20 [27]. Moreover, the phase composition of the samples sintered at the optimum sintering temperature (1700°C) was determined by means of XRD analysis with a Philips X-ray diffractometer (model PW1730). In preparation for microstructural characterization, each sintered sample was polished and thermally etched for 15 min in air at a temperature 100°C lower than the respective sintering temperatures. The samples were then coated with gold via sputtering coating, thereby ensuring the electrical conductivity of each sample. Scanning electron microscopy (Quanta FEG 250, Holland) coupled with energy-dispersive X-ray spectroscopy (EDS) were employed for microstructural and elemental evaluations. Furthermore, the thermal expansion coefficient (TEC) was measured using an automatic Netzsch DIL402 PC instrument (Germany). The bending strength was investigated by means of a three-point bending test on a universal testing machine (Model LLOYD LRXS K). The strength was calculated as follows:

\[ \text{Bending strength (}\sigma\text{) (MPa)} = \frac{3FL}{2BD^2} \]  

Where:

\[ F = \text{maximum force before failure}, \ L = \text{span width between the two supports (mm)}, \ B = \text{width of the sample (mm)}, \ D = \text{height of the sample (mm)}.\]

3. Results and discussion

3.1. Characterization of the prepared mullite powder

The phase composition of the mullite gel calcined at various temperatures, (800°, 1000°, 1200°C) is given in Figure 2(a). As shown in the figure, the mullite (JCPDS 83–1881) was present as an amorphous phase at 800°C and started to crystallize at 1000°C. At 1200°C, mullite is present as a well crystalline phase. Some corundum (Al₂O₃) phase peaks (JCPDS 46–1212) were also observed, but silica peaks were absent, owing possibly to the occurrence of silica as an amorphous phase. The particle size of the mullite powder sintered at 1200°C ranged from 6.91 to 20 nm, as shown in the TEM images presented in Figure 2(b). Furthermore, the image showed that the fine mullite particles are present as agglomerated clusters.

3.2. Characterization of mullite and zirconia/mullite composite bodies

The effect of sintering temperature and ZrO₂ content on the bulk density and apparent porosity of the zirconia/mullite composites sintered at temperatures ranging from 1550° to 1700°C for 1 h is demonstrated in Figure 3a and b. As indicated in the figure, increasing both the sintering temperature and the ZrO₂ content led to an enhancement of the densification parameters. Pure mullite bodies were characterized by low bulk density and high porosity levels even for sintering at 1700°C. In contrast, the bodies composed of 20 wt.% ZrO₂ and 80 wt.% mullite sintered at 1700°C were characterized by high bulk density and the lowest apparent porosity. That is, the increase in the ZrO₂ content enhanced the densification process and increased the bulk density figures due to the high theoretical density of the ZrO₂ phase. The increase in density was accompanied by a decrease in the apparent porosity, that is, the density is inversely proportional to the porosity. The studied composites underwent only partial densification and were characterized by low-density figures. This resulted from the fact that on sintering at high temperatures the mullite
grains grew into elongated shapes, which led to the formation of closed pores that diminished the density figures. Figure 4 presents the phase evolution of the zirconia/mullite composites sintered at the optimum sintering temperature. The results revealed that the MZ0 bodies are composed of a pure mullite phase (JCPDS no. 83–1881), whereas the MZ10, MZ15, and MZ20 bodies are composed of mullite and m-ZrO₂ (JCPDS no. 07–0343). No ZrO₂ transformation from m-ZrO₂ to t-ZrO₂ was observed at any of the studied compositions. Figure 5a shows Scanning electron microscopy (SEM) micrographs of the pure mullite bodies densified at 1700°C for 1 h. As shown in the figure, many pores and unwell boundaries mullite particles with sizes ranging from 0.42 to 1.30 µm occurred in each body. In addition, a few needlelike particles and the agglomeration of very fine mullite grains were observed.

An energy-dispersive X-ray analysis (EDS) pattern of the mullite grains is shown in Figure 5b. The pattern shows that mullite phase formation occurred at an Al₂O₃ content of 62.15 wt.%. Data reported for the incongruent formation of mullite [28,29] indicates that mullite with 77.2 wt.% Al₂O₃ (2Al₂O₃·SiO₂, 2/1 mullite) is formed at 1890°C. Furthermore, the formation of 2/1 mullite is affected by many factors, such as the materials used for synthesis and the formation conditions. The decrease in the formation temperature shifts the composition of mullite toward low alumina content and high silica content (3Al₂O₃·2SiO₂, 3/2 mullite). The abovementioned data suggest that the formation of the silica-rich mullite phase resulted from the low sintering temperature employed, that is, 1700°C.

The MZ10 microstructure, Figure 6 (a), consists of randomly distributed secondary mullite acicular grains embedded in a matrix of primary mullite. The porosity increase and the density decrease of the MZ10 sintered
Figure 4. XRD patterns obtained for blank mullite and zirconia/mullite composite bodies. All the bodies were sintered at 1700°C for 1 h.

bodies resulted from the grain size heterogeneity of the mullite matrix [30]. The abovementioned phenomena increased the porosity. However, compared with that of the pure mullite, the porosity of the sintered MZ10 bodies was characterized by coalescence and an arrangement of the ZrO₂ grains (white) in the matrix. Very few round ZrO₂ grains occurred in the intragranular regions, and most of the equiaxed agglomerated ZrO₂ grains occurred in the triple junctions of the mullite grains. Previous studies based on special XRD technology have reported that t-ZrO₂ is always present as intragranular particles, whereas m-ZrO₂ occurs mainly as intergranular particles [31].

When the ZrO₂ content was increased to 15 and 20 wt.% (see Figure 6 (b and c)), the microstructural homogeneity of the sintered bodies increased relative to that of MZ10. The ZrO₂ grains were quite homogeneously distributed in the mullite matrix, which is composed of stick-like randomly oriented secondary mullite grains with almost the same grain size. It was observed that by increasing the zirconia content, both the densification enhancement and the growth and formation of well-crystalline secondary mullite are taking place. That it could be because of the formation of the zircon (ZrSiO₄) phase that formed as a result of the interaction between the mullite' silica and the added zirconia. Pena and De Aza [32] have reported that a system containing zircon and mullite formed a transient liquid phase at temperatures as low as 1450 ± 10°C, which contributes to the enhancement of the rate of reaction and the sintering process. Also, at high temperature, about 1675°C, zircon (ZrSiO₄) is decomposed to zirconia (ZrO₂) and silica (SiO₂) [33]. Such decomposition is mostly associated with the formation of a slight quantity of glassy phase. With the appearance of the glassy phase, the mullite grains start to lose their deficient boundaries (Figure 5) and manifest as rectangular prisms as shown in the SEM micrographs (Figure 6) [7]. The quantity of the formed glassy phase is too small to be detected by either the XRD analysis or the SEM observations.

Figure 5. SEM image of the blank mullite MZ0 (a) and EDS analysis results of the sample (b).

Figure 6. SEM images of the zirconia/mullite composites MZ10 (a), MZ15 (b), and MZ20 (c) (mullite: gray color, zirconia: white color).
Several studies have focused on modulating and enhancing the mechanical behavior of ceramic composites. The three-point bending strength results of sintered zirconia/mullite composites sintered at their optimum sintering temperature are given in Table 1. The XRD patterns of the composites were quite similar. However, Table 1 shows that the bending strength increased significantly from 34.01 ± 1.93 MPa for pure mullite samples to 83.30 ± 2.46 MPa for composites containing 20 wt.% ZrO$_2$. This ZrO$_2$-induced noticeable enhancement in the strength is attributed to the reduction in the apparent porosity and the eventual increase in the bulk density observed for the bodies with high ZrO$_2$ content. In addition, the morphologies of both the mullite and ZrO$_2$ and their distribution and interaction played an important role in improving the bending strength [34].

The thermal expansion coefficient TEC (α) of the sintered samples was measured (see Table 1 for the results). The heating and cooling rate of the samples up to 1000°C was 5°C/min. As indicated in the table, the lowest α (3.91 × 10$^{-6}$/°C) at 1000°C was obtained for the pure mullite samples. The α value of each sample increased with the addition of ZrO$_2$ to mullite. Furthermore, the α value of the sample containing 20 mass% ZrO$_2$ was 5.94 × 10$^{-6}$/°C is similar to that of monoclinic ZrO$_2$ (6 × 10$^{-6}$/°C) [35,36]. The abovementioned results concur with the XRD findings, which indicated that the sintered samples are composed of mullite and monoclinic ZrO$_2$.

It is clear from the obtained results that the combination between mullite and zirconia earned the produced composite unique properties such as excellent thermal shock resistance, improved mechanical stability, and the high chemical stability of the zirconia/mullite composites. All of these properties encourage the employment of these composites in many industrial applications such as: glass industry kilns where the need for high chemical and corrosion stability are required, thermal protection materials for combustors, aircraft, and gas turbine engines, in the steel industries, they are used in slide gate valves, which an integral part of the continuous casting processes for steel. They also used for the production of the nozzles and plugs that need materials having both good thermal shock and erosion resistance.

### Table 1. Three-point bending strength (MPa), apparent porosity (%), and coefficient of thermal expansion obtained for various blank zirconia-mullite composite samples at 1700°C.

| Batch composition | Three point bending strength, MPa | Apparent porosity, % | Coefficient of Thermal Expansion, CTE $\times 10^{-6}$/ K$^{-1}$ |
|-------------------|----------------------------------|----------------------|---------------------------------------------------------------|
| MZ0               | 34.01 ± 1.93                     | 34.80 ± 1.28         | 3.91x10^{-6}                                                 |
| MZ10              | 60.66 ± 3.16                     | 17.48 ± 1.32         | 4.91x10^{-6}                                                 |
| MZ15              | 67.71 ± 2.09                     | 15.30 ± 0.36         | 4.91x10^{-6}                                                 |
| MZ20              | 83.30 ± 2.46                     | 5.66 ± 0.156         | 5.94x10^{-6}                                                 |

### 4. Conclusion

In the present study, a zirconia/mullite nanocomposite was fabricated by means of an in-situ sol-gel preparation technique and sintering of the fabricated bodies at 1700°C. The most important findings of this study are summarized as follows:

1- Increasing both the sintering temperature and the ZrO$_2$ content led to an improvement in the densification parameters.

2- For all the compositions considered, no ZrO$_2$ transformation from m-ZrO$_2$ to t-ZrO$_2$ was observed.

3- The microstructural homogeneity of the sintered bodies increased with increasing ZrO$_2$ content.

4- The mechanical strength was enhanced by the addition of ZrO$_2$. This enhancement is attributed to the improvement in the densification parameters observed for the bodies with a high ZrO$_2$ content. Furthermore, the morphologies of both the mullite and ZrO$_2$ and their distribution and interaction played an important role in improving the bending strength.

5- The thermal expansion coefficient increased with the addition of ZrO$_2$ and its value indicated the presence of m-ZrO$_2$.

### Data availability

The authors declare that all data supporting the findings of this study are available within the paper.

### Disclosure of potential conflicts of interest

No potential conflict of interest was reported by the author(s).

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