Forming Mullite-Ceramics Reinforced with ZrO$_2$-t Starting from Mullite-ZrO$_2$-t and Kyanite-Al$_2$O$_3$-ZrO$_2$-t Mixtures

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FORMING MULLITE-CERAMICS REINFORCED WITH ZrO$_2$-t STARTING FROM MULLITE-ZrO$_2$-t AND KYANITE-Al$_2$O$_3$-ZrO$_2$-t MIXTURES

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ABSTRACT

Mullite-ceramics reinforced with 8 and 10 vol. % YZT were prepared starting from two different powder mixtures. The first one was a mixture of pure mullite and YZT, and the other was a mixture of kyanite, alumina and YZT. The production of ceramics consisted of the pressureless-sintering of mixtures powders which were thoroughly mixed under high energy ball-milling. During sintering, kyanite and alumina of the second mixture react between them to form mullite. Measurements of density, microhardness and $K_{IC}$ were carry out in all produced materials, from it was obtained that samples produced with pure mullite reach major densities, microhardness and toughness in comparison with the values displayed for samples prepared with kyanite and alumina. The reason of this has its explanation since during sintering of the kyanite and alumina samples; there is a competition between two phenomena; the reaction to form mullite and the sintering of the product. As the reaction occurs at low temperature and has high energy consumption, the activation energy necessary for diffusion during sintering is not reached; therefore this phenomenon is the controlling agent during the processing.

INTRODUCTION

Besides its traditional uses, mullite has attracted attention in recent years, as a material for high temperature structural applications, mainly because at these temperatures it may retain a significant portion of the mechanical strength that it has at room temperature, also, because it has a low thermal expansion coefficient, low dielectric constant, high melting point, high creep resistance and high chemical stability$^1$. Table 1 shows some of the values of the main properties of the mullite.

<table>
<thead>
<tr>
<th>Property</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Thermal expansión coefficient</td>
<td>$5 \times 10^{-6}$ K$^{-1}$</td>
</tr>
<tr>
<td>Flexural resistance</td>
<td>200 MPa</td>
</tr>
<tr>
<td>Fracture toghness (K$_{IC}$)</td>
<td>2 MPam$^{1/2}$</td>
</tr>
<tr>
<td>Young modulus</td>
<td>231 MPa</td>
</tr>
<tr>
<td>Density</td>
<td>3.16 gcm$^{-2}$</td>
</tr>
</tbody>
</table>
As can be seen in this table, fracture toughness ($K_{IC}$) is a characteristic in which mullite is deficient. Furthermore, to obtain dense bodies of mullite, it is required long sintering treatment at elevated temperatures ($>1700^\circ$C), this is due to the high value of the activation energy necessary for ion diffusion occurs through the network of mullite. Because of these difficulties, over the past 15 years in many countries around the world, they were conducted a series of investigations in the seek for new processing methods by which they could obtain dense mullite bodies. To increase its tenacity, in some of these studies, it has proposed the use of ZrO$_2$ as reinforcing material, thus values of $K_{IC}$ that have been obtained, varying between 3 and 3.2 MPa-m$^{1/2}$ for 10 to 20 wt. % ZrO$_2$ content.

Some researchers have mentioned that large amounts of ZrO$_2$ on mullite-based composites cause thickening of the microstructure and thus the difficulty in the retention of the tetragonal form of ZrO$_2$. This retention of the ZrO$_2$ is important, because it has been suggested that one method by which the mullite matrix is reinforcing by ZrO$_2$ is the transformation of ZrO$_2$-tetragonal to ZrO$_2$-monoclinic. However, other authors have suggested different possible reinforcement mechanisms, such as microcracking induced by the same transformation of the ZrO$_2$, strengthening grain boundaries caused by a metastable solid solution of ZrO$_2$ and deflection of cracks due to the presence of acicular microstructure.

The aim of this work is to study the formation of mullite composites reinforced with ZrO$_2$, starting from mixtures of mullite + ZrO$_2$ and Kyanite + Al$_2$O$_3$ + ZrO$_2$, using conventional methods of sintering without the application of pressure in an electric furnace.

Because pure mullite is a highly expensive mineral, it is suggest the use of kyanite as mullite precursor material. Kyanite mineral is cheaper than the mullite mineral, with chemical formula (Al$_2$O$_3$·SiO$_2$), this mineral when is heated at high temperatures ($\sim 1300^\circ$C) decomposes forming mullite + silica, for this reason in the mixture with kyanite is added certain amount of alumina to compensate silica excess resulting from the decomposition of kyanite in order to form more mullite.

**EXPERIMENTAL PROCEDURE**

Starting materials were: Mullite powder (99%, 1$\mu$m, Virginia Milling Corporation, USA), Kyanite powder (99%, 3-5 $\mu$m, Virginia Milling Corporation, USA), Al$_2$O$_3$ powder (99.9%, 1$\mu$m, Sigma, USA) and YZT powder (99.9%, 20 $\mu$m, Toyo, Japan). Final YZT contents in the produced composites were: 8 or 10 vol. %. Powder blends of 20 g were prepared in a ball mill with ZrO$_2$ media, the rotation speed of the mill was of 300 rpm, and the studied milling time was 12 h. With the milled powder mixture, green cylindrical compacts 2 cm diameter and 0.2 cm thickness were fabricated by uniaxial pressing, using 300 MPa pressure. Then pressureless sintering in an electrical furnace was performed, at 1500 $^\circ$C during 2 h. Characterization of sintered samples was as follows: Densities were measured using the Archimedes' principle. The microstructure was observed by scanning electron microscopy (SEM), equipped with an energy dispersive spectroscopy analyzer (EDS). Phases present in the sintered composites were determined by X-ray diffraction (XRD). Fracture toughness was estimated by the fracture indentation method, in (all cases ten independent measurements per value were carrying out).

To identify study samples, it was used the following code:
- C8Z: Sample prepared with Kyanite + Al$_2$O$_3$ + 8 % vol. ZrO$_2$
- C10Z: Sample prepared with Kyanite + Al$_2$O$_3$ + 10 % vol. ZrO$_2$
- M8Z: Sample prepared with Mullite + 8 % vol. ZrO$_2$
- M10Z: Sample prepared with Mullite + 10 % vol. ZrO$_2$
Table I. Summary of the dimensions of the notches in the specimen under study.

<table>
<thead>
<tr>
<th>Samples</th>
<th>Aperture [mm]</th>
<th>Depth [mm]</th>
<th>Angle [°]</th>
<th>Radius [mm]</th>
</tr>
</thead>
<tbody>
<tr>
<td>PEZI 0a</td>
<td>1.742</td>
<td>1.869</td>
<td>42.45</td>
<td>0.214</td>
</tr>
<tr>
<td>PEZI 10b</td>
<td>3.117</td>
<td>3.587</td>
<td>39.46</td>
<td>0.224</td>
</tr>
<tr>
<td>PEZI 10a</td>
<td>2.575</td>
<td>2.806</td>
<td>45.22</td>
<td>0.209</td>
</tr>
<tr>
<td>PEZI 10b</td>
<td>2.226</td>
<td>2.322</td>
<td>44.31</td>
<td>0.295</td>
</tr>
<tr>
<td>PEZI 10c</td>
<td>2.132</td>
<td>2.359</td>
<td>41.38</td>
<td>0.201</td>
</tr>
<tr>
<td>PEZI 30a</td>
<td>3.127</td>
<td>3.703</td>
<td>43.00</td>
<td>0.240</td>
</tr>
<tr>
<td>PEZI 30b</td>
<td>3.336</td>
<td>3.830</td>
<td>42.39</td>
<td>0.295</td>
</tr>
<tr>
<td>PEZI 30c</td>
<td>2.639</td>
<td>2.742</td>
<td>46.26</td>
<td>0.197</td>
</tr>
</tbody>
</table>

Bending test.
To apply the theoretical models of Peterson, Irwin and Griffith, it was first necessary to test the 8 specimens, previously prepared and tested in three-point bending to find that maximum force could withstand the test pieces before breaking. This test was conducted in the Instron universal machine, mod. 5500R, using a rate of 0.5 mm/min.

Thereafter, defining the encoding of the specimens based on their geometry, which corresponds to a flexure beam as shown in Figure 2.

Where:
W: the height of the sample [m]
P: is the applied force [N]
S: is the spam formed between the support [m]
a: is the depth of the crack [m]
Besides using other values are also useful.
B: thickness of the sample [m]
r: the radius [m]
d = W - a [m]

Thus using this coding will be able to obtain other parameters in order to find the value of $K_{IC}$.

Using the model of Peterson it is possible to get $K_m$, $\sigma_{max}$ and $\sigma_{nom}$.

Where:
$K_m$: The factor of elastic stress concentration
$\sigma_{max}$: The effort at the root hub of efforts
$\sigma_{nom}$: The nominal stress

In the concentration stress charts developed by Peterson it can be found the value of $K_m$, relating r with d and W with d. However, also it can be obtained applying the equations of the curves and formulas mentioned below.

$$K_m = C_1 + C_2(a/W) + C_3(a/W)^2 + C_4(a/W)^3$$  \hspace{1cm} (1)

In the case where $2.0 < a/r < 20$, the following equations are applied.
ray diffraction patterns do not indicate complete reaction, since there are several peaks correspond to Al2O3 and SiO2 in both cases, however these peaks are very low in intensity, therefore reaction is almost complete. The most important observation here is the presence of ZrO2 in its monoclinic form, and this situation could be a problem, because the reinforcement by transformation of zirconia would not operate in this situation.

Figure 3. X-ray diffraction patterns of different prepared composites.

Microhardness
The microhardness evaluated in different prepared samples is reported in Figure 4. In this figure it is seen that those samples prepared with mullite + ZrO2-t exhibit higher hardness values, compared to the samples prepared with the mixture of kyanite + Al2O3. The greater degree of densification and the situation of has a more homogeneous material in chemical composition in samples prepared with ZrO2 and mullite, are the cause responsible for this condition. Since materials made from kyanite besides being less dense, have a more heterogeneous chemical composition, as have been indicated by XRD results, where phases such as mullite, monoclinic and tetragonal ZrO2, Al2O3 and SiO2 are present.
Fracture toughness

Figure 5 shows the values of fracture toughness determined in different samples prepared here. As expected samples prepared with mullite + ZrO$_2$-t exhibit larger values of fracture toughness because therein ZrO$_2$ is present in tetragonal form, giving the feasibility of the reinforcement by the transformation of ZrO$_2$-t to ZrO$_2$-m exist. On the contrary, the fracture toughness is less in the samples prepared with kyanite + Al$_2$O$_3$ + ZrO$_2$-t, because it was not possible the ZrO$_2$-t retention at the end of processing. The reason of these behaviors have its explanation since during sintering of the kyanite and alumina samples; there is a competition between two phenomena; the reaction to form mullite and the sintering of the product. As the reaction occurs at low temperature and has high energy consumption, the activation energy necessary for ion diffusion through the network of mullite during sintering is not reached; as a result this phenomenon is the controlling agent during the processing.

Figure 5. Fracture toughness of samples sintered at 1500°C, during 2 h.
Microstructure

Typical microstructures of the samples prepared with 10% vol. ZrO₂-t are shown in Figure 6. Figure 6a which corresponds to sample prepared with mullite + ZrO₂-t, clearly shows a good distribution of fine particles of a second phase (ZrO₂-t) in the matrix (mullite). While figure 6b corresponds to a sample prepared with the kyanite + Al₂O₃ and ZrO₂-t mixture, the ZrO₂ distribution is not homogeneous because it is appreciate colony of ZrO₂ agglomerates. ZrO₂ was added to mullite for the purpose of serve as reinforcing material of the same, situation unsuccessful, due to the transformation of ZrO₂-t to ZrO₂-m during any processing stage. The size of the ZrO₂-t in figure 6a is about 1 to 3 µm and the location thereof is in intergranular regions of the mullite matrix. The reinforcing mechanism in these kinds of samples is the transformation of ZrO₂-t to ZrO₂-m, mechanisms before widely documented²,⁴,⁶,⁸,¹¹.

![Microstructure Images](image)

Figure 6. Microstructure observed in scanning electron microscope of samples sintered at 1500°C, during 2 h. (a) mullite + ZrO₂-t, (b) kyanite + Al₂O₃ + ZrO₂-t.

CONCLUSIONS

Samples produced with pure mullite reach major values of density, microhardness and toughness. During sintering of kyanite-alumina mixtures; there is a competition between two phenomena; the reaction to form mullite and the sintering of the product. As the reaction occurs at low temperature and has high energy consumption, the activation energy necessary for diffusion during sintering is not reached; therefore this phenomenon is the controlling agent during the processing. The reinforcing mechanism in these kinds of samples is the transformation of ZrO₂-t to ZrO₂-m.

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