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Improving Fracture Toughness of Mullite Ceramics with Metal Reinforcements

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ABSTRACT: *Mullite/Metal composite ceramics were fabricated by the use of mechanical milling and pressureless sintering. Al2O3 + 10 vol.% of Co, Ni, or Ti were mixed and milled during 12 h at 300 rpm in a horizontal mill, then with the powder mixture it was conformed cylindrical samples by uniaxial pressing using 300 MPa. Pressed samples were sintered during 2 h in an electrical furnace at 1500°C. During sintered it was used an argon atmosphere inside the furnace in order to inhibit metal oxidation. XRD results indicate that alumina and metals retain its crystalline structure. Measurements of densities indicate that they were achieved relative densities between 90% and 98% in the manufacture composites. Optical microscopy observations show mullite's microstructure with very fine and homogeneous distributions of metal particles. Used metals (Co, Ni and Ti) to reinforce mullite, have yielded favorable results improving the fracture toughness of the mullite. However, it should be made more dense materials with them to better explore this potential.*
Keywords *– Fracture toughness, Metals, Reinforcement, Mullite, Ceramics*

I. INTRODUCTION

The ceramic materials to which the mullite belongs, are valued mainly by its thermal stability and corrosion resistance, as well as by its high hardness [1-2]. These virtues are due to the nature of their chemical structure, principally to the strong bonds that hold the constituent atoms in their equilibrium positions. However this nature provides a critical drawback that is the fragility, causing that ceramic are particularly sensitive to slight imperfections in their microstructure, which serve as crack initiation points when they are subjected to mechanical stress. For these reasons, many efforts have gone into research of mullite to develop new processes and materials that minimize these defects at microscopic scales [2-7]. Hence has arisen the idea of strengthening the mullite with other ceramic such as ZrO_2 and SiC, as well as some metals [8-10]. Conventionally, mullite is produced by high temperature calcination of mixtures of SiO_2 and Al_2O_3 [11]. The activation of energy for ion diffusion that takes place through the network of energy requires high temperatures; therefore, high sintering temperatures are necessary (>1700°C) to obtain dense bodies of mullite [12]. This situation makes difficult its processing at industrial level. The aim of this paper is to study the formation of mullite composites reinforced with cobalt, nickel or titanium, using sintering conventional methods without the application of pressure.

II. EXPERIMENTAL

Starting materials were: Mullite ($3Al_2O_3{\cdot}2SiO_2$) powder (99.9 %, 1μm, Sigma-Aldrich, USA) and the following powder metals (Co, Ni and Ti all of them with 99.9% purity, and sizes between 1-2μm, Sigma-Aldrich, USA). The used amount of powders is that one that allows at the end of the processing to obtain the composite Mullite-10 vol.% of the corresponding metal. The powders were dry milled and mixed in a horizontal mill, using a rotation speed of 300 rpm, during 12 h, in ceramic jars and using YSZ's balls as grinding elements, the relation; weight of powder/weight of balls was 1:25. Cylindrical samples were fabricated with the powders mixture, by uniaxial pressing using 200 MPa. Sample's dimensions were: 20 mm in diameter x 3 mm in thickness. Afterward, the pressed samples were pressureless sintered at 1500°C during two hours in an electrical furnace with gas argon atmosphere. The speeds of warming and cooling were kept constant and equal to 10°C/min. The characterization of the synthesized products was of the following way: Densities were evaluated by the Arquimedes' method. Crystalline phases present en each fabricated composite was analyzed by X-ray diffraction. The microstructure of the composites was observed with an optical microscope. The elastic modulus was determined by the ultrasonic method. Microhardness measurements were evaluated with the help of a Vickers indented; finally, fracture toughness was determined by the fracture indentation method, using Evan´s equation [13].

III. RESULTS

3.1 Relative density

The calculation results of the relative density achieved by each sample after sintering is shown in Fig. 1. To construct this figure it was first determined the theoretical density of each compound using the rule of mixtures; subsequently sintered density of each sample was evaluated by the method of Archimedes. With these two values, it could be calculated the relative density attained for each sample after its sintering. The figure 1 shows that the density of the control sample was approximately 90 %, while densities reached by the samples with Ni and Co additions, reached values of 88 and 89 % respectively. These density values are not very high, indicating that the diffusion process during the sintering step does not occurred satisfactorily. This could be due to sintering occurs close in the presence of a liquid phase, formed by the fusion of the used metal in each case (melting point is 1495 and 1453 °C for cobalt and nickel respectively), and surely the liquid formed not wet the surface of the mullite, situation that difficult sintering of the composite. On the other hand, Ti reinforced sample has achieved a relative density of 98%, density considerably better than those of the other samples. In this case, because the melting point of titanium is 1580 °C, there is not the presence of a liquid phase which favored having good diffusion during the sintering of the sample, obtaining the reported density value.

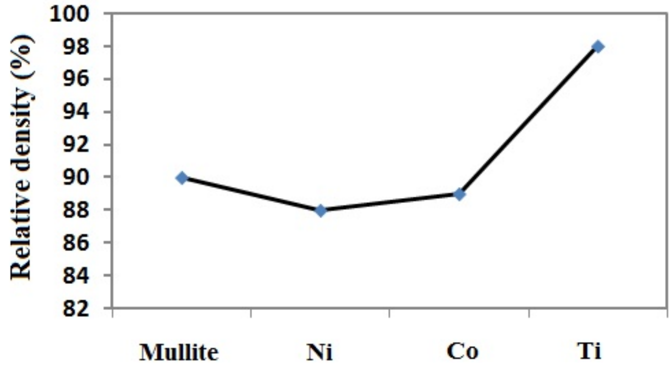

figure 1. relative density of different fabricated composites.

3.2 X-ray Diffraction

X-ray diffraction patterns of the three sintered composites are shown in Fig. 2. As seen in the corresponding pattern to each system, the diffraction spectrum shows just the presence of the mullite and the metal-added. The presence of any oxide, or other element or compound outside the initial composition of the composite is not observed, indicating that there was no contamination in any step of the process.

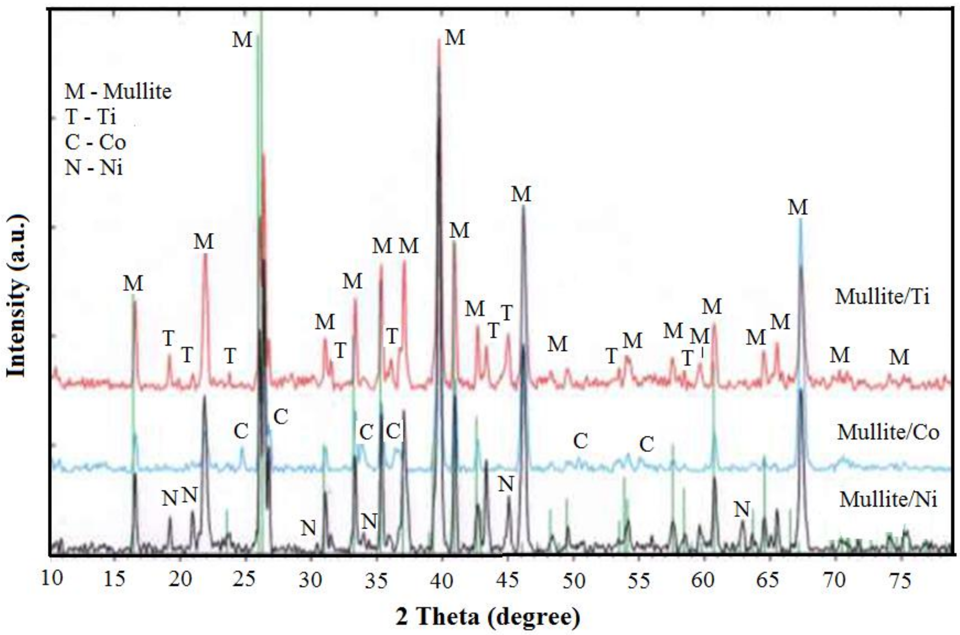

figure 2. x-ray diffraction patterns of the sintered composites.

3.3 Microstructure obsrved with optical microscope

Microstructure photographs of each manufactured composite taken with an optical microscope are shown in Fig. 3. In the cobalt sampled it is observed larger grain size than in the other two, also it has high porosity present in the sample. With respect to the sample with titanium and nickel there is the presence of very fine and homogeneous microstructure, although in both cases alike appreciate some pores. In all three photomicrographs it can be seen that the metallic phase (small and bright particles) are distributed homogeneously in the matrix.

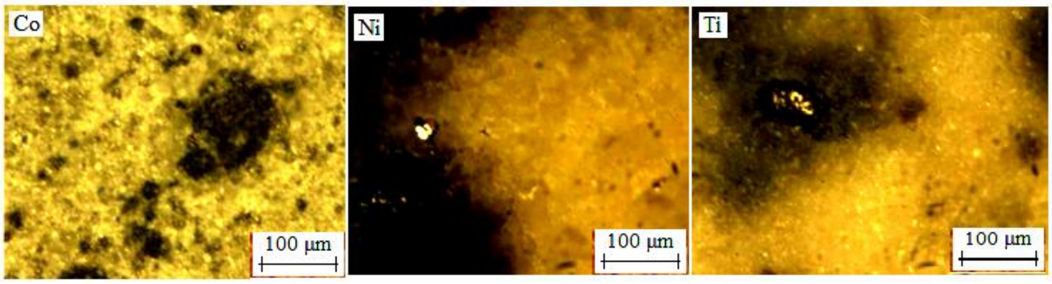

figure 3. optical microstructure of different manufactured composite.

3.4 Mechanical Properties

3.4.1 Young's Modulus

The measurement results of Young's modulus for each composite are presented in Fig. 4. This figure shows that in all cases the value of Young's modulus observed for the control sample (monolithic mullite) is well above of the modules achieved by other samples. What is because adding a ductile material (metal) in a rigid matrix (mullite), the stiffness decreases due to the ductility that gives the metal to the new compound.

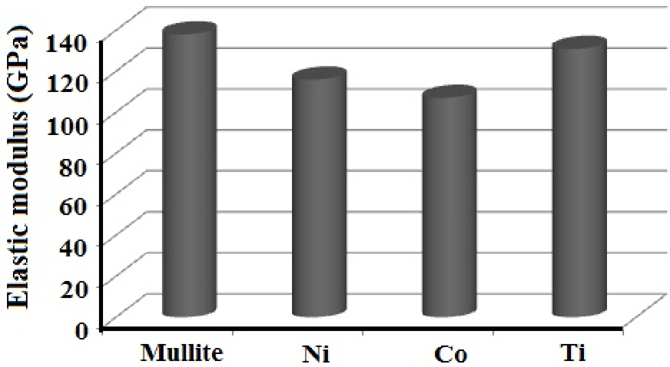

figure 4. young's modulus of sintered composites.

3.4.2 Microhardness

The results of the measurement of microhardness of each sintered sample are shown in Fig. 5. Again in this figure it is apparent that hardness of the pure mullite is well above than hardness of different composites. This result is logical and has two explanations, The first one (for the case of cobalt and nickel systems) is due to the greater degree of densification achieved by purely ceramic sample, the second is that when it is added a ductile metal, much less hard than the ceramic matrix, clearly, the final hardness of the resulting composite will be less. On the other hand, the sample with Ti which is best densified, has a high value of microhardness.

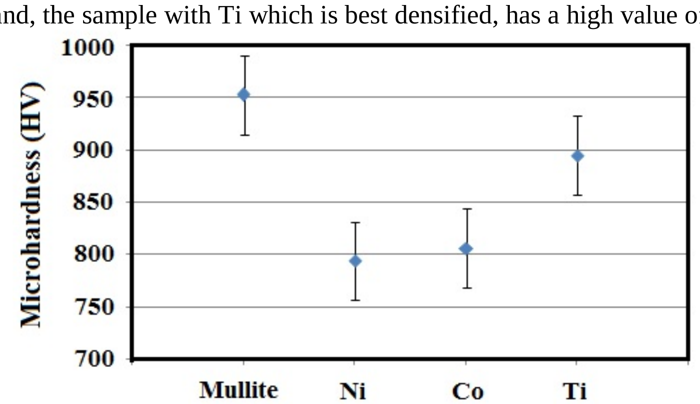

figure 5. microhardness of each sintered sample.

3.5.3 Fracture Toughness

Values of fracture toughness for each sintered composite are shown in Fig. 6. From this figure is that the values of fracture toughness of monolithic mullite, are below of fracture toughness for the composites reinforced with cobalt and nickel, despite the low density values provided by these. On the other hand, the value of this property when is used titanium as reinforcing is improved considerably. The fracture toughness of mullite is improved in 26.3%, 31.5% and 68.4% when added cobalt, nickel and titanium respectively as a reinforcing material. These values would suggest that this type of metal may be promising in the reinforcement of mullite. If it could be adjusted processing conditions to achieve higher densification in the fabricated composites when these metals are used, probably also the fracture toughness could increases significantly. Different authors have reported that the mechanism of enhancement of ceramics, by metals, is due to the closure of cracks through the metal tap when they tend to grow [1-2].

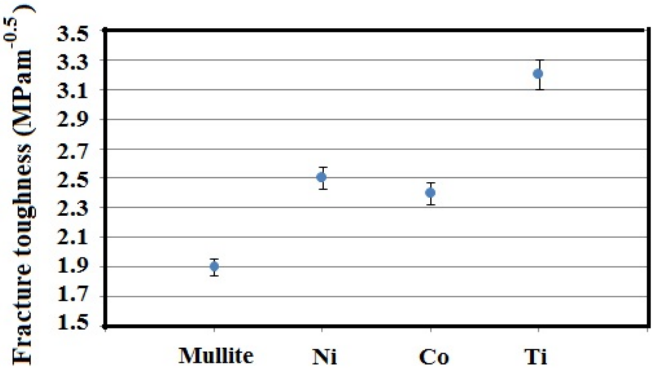

Figure 6. Fracture toughness of each fabricated composite.

IV. CONCLUSIONS

- Through the proposed methodology it could be fabricated mullite-based ceramics with additions of Co and Ni with 90% of densification. On the other hand when mullite is reinforced with Ti, its densification rise to 98%.

- Used metals (Co, Ni and Ti) to reinforce mullite, have yielded favorable results in improving the fracture toughness of mullite. However, it should be made more dense materials with them to better explore this potential.

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