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The Comparison of MgO and TiO_2 Additives Role on Sintering Behavior and Microstructures of Reaction-Sintered Alumina-Zirconia-Mullite Composite

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ABSTRACT

Alumina-Mullite-Zirconia ceramic composites were prepared by reaction sintering of alumina, zircon mixture and the sintering aids of magnesia and titania at temperatures of 1400 and 1600 ºC for 2 hours. The development of different phases and microstructures of the composites were analysed to assess the relative influence of the additives on the formation of alumina-zirconia-mullite composite. Bulk density, phase combination, and microstructure of the sintered samples were examined by Archimedes method, X-ray diffraction (XRD) and scanning electron microscope (SEM), respectively. The results of the study revealed the successful formation of the alumina-mullite-zirconia composite by the reaction sintering method at temperature of 1600ºC. By the same token, the sample containing 6 wt.% of sintering aid shows improvement in density.

1. INTRODUCTION

Alumina-mullite-zirconia (AMZ) composites are important groups of refractory materials which are widely used in glass technology as a refractory lining in different zones like walls of furnaces, feeder sections, spouts, tubes, stirrers, ducts, rotor segments, mantle blocks and orifice rings [1].AMZ refractory composites have better corrosion resistance, mechanical properties and thermal shock resistance compare to pure alumina or mullite ceramics as well as alumina/mullite or zirconia/mullite composites.

In AMZ composites, corundum (alumina), mullite, zirconia (tetragonal and/or monoclinic phase) and minor amount of glassy phase are tailored in a way that improve thermo-mechanical and corrosion resistance properties of composites. Corundum with high refractoriness and hardness, mullite with high creep resistance good thermal shock resistance, and zirconia with high toughness and good corrosion resistance form excellent combined properties to AMZ products [2,3,4].

AMZ refractory composites are produced by different routes such as fused cast and reaction sintering of different raw materials such as zircon ($ZrSiO_4$) and alumina (Al_2O_3). To respond to sintering methods, during firing at high temperature, zircon was decomposed to tetragonal ZrO_2 and amorphous SiO_2 and then silica reacted with alumina and established mullite which can play an important role in AMZ composites sintering [3,4].

During the process of cooling sintering with the temperature, around 1050°C, tetragonal ZrO_2 is transferred to monoclinic form and induced micro cracks in the composites. In the presence of additives like TiO_2 in AMZ composition, some of the tetragonal ZrO_2 remained as a semi-stabilized phase in composites at room temperature. During the application of these composites in the environment with high thermal or mechanical stresses, the transformation of remained tetragonal to monoclinic has been occurred and it toughened the composites inhibiting crack propagation [5, 6].

The effect of TiO_2 or MgO on two component ceramic-ceramic composites such as mullite-ZrO_2 in some areas such as sintering, properties and stability of tetragonal ZrO_2 were investigated by some researchers [7,8]. However in this study, the effects of TiO_2 or/and MgO on the microstructure, mechanical properties and sintering behavior of Al_2O_3-mullite-zirconia composites are investigated.

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2. EXPERIMENTAL PROCEDURES

To prepare the alumina-mullite-zirconia composite, α-Al_2O_3 (d_{50}=63 μm and purity>99.5%), zircon (d_{50}=5 μm and purity>99%) were used as the main raw materials, and $MgCO_3$ (d_{50}=3 μm and purity>99.5%) and TiO_2 (d_{50}=2 μm and purity>99% and rutile form) were employed as sintering aids.

Point A in Figure 1. indicates the location of additives without sintering aid tested in this research. As it is observed, this point is located within the composition triangle of alumina, zirconia and mullite; therefore, after firing the final phases would be alumina-zirconia-mullite as well as equilibrium state. The arrow in the figure indicates the cooling path for the selected composition. After reaching the common boundary of Al_2O_3 and mullite, the mentioned cooling path moves on this line toward Point E (eutectic transformation point), which will constitute the end point for cooling of this compound.

Considering the phase diagram of Al_2O_3-ZrO_2-SiO_2 and equation (1), the original materials were portioned in such a way that the final composite contain, by weight, 60% Al_2O_3, 25% mullite and 15% ZrO_2 phases. The previous studies on alumina-zirconia-mullite composites and the reaction sintering of alumina and zirconia had reported better sintering ability and properties of this composite in the above range [9,10,11].

$$2ZrSiO_4 + (3+x)Al_2O_3 \rightarrow xAl_2O_3 + 2ZrO_2 + 3Al_2O_3.2SiO_2 \quad (1)$$

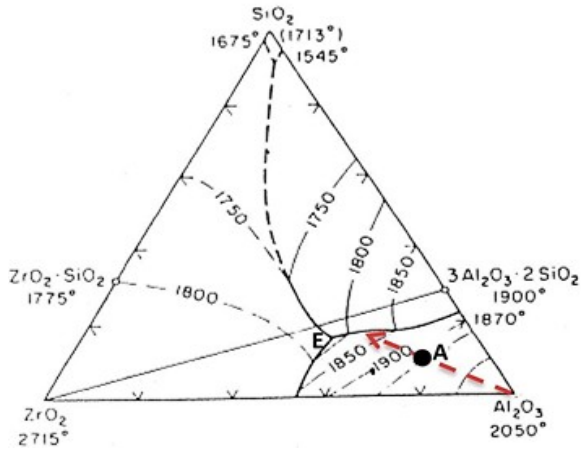

Figure 1. Chemical composition and cooling path of the selected compound in Al_2O_3-ZrO_2-SiO_2 phase diagram.

To prepare the composite, first, the powders of the original raw materials, including alumina and zircon and 5 wt.% of PVA solution along with different amounts of sintering aids, (2, 4 and 6 wt.%) were completely homogenized by means of a fast, one- hour mill . After drying and pressing the samples under a uniaxial pressure of 100 MPa, they were sintered at temperatures of 1400 and 1600 ºC for 2 hours and at a heating rate of 10ºC/min.

In order to identify the samples, they were labeled by the code AMZ-nX, where A, M and Z denote the alumina, mullite and zirconia, respectively, X indicates sintering aid and 'n' is its weight percentage in the mixture. Table 1. shows the different labels of the composite samples.

TABLE 1. Batch design of the investigated samples

Sample code	Sintering aid	%weight
AMZ	without sintering aid	0
AMZ-2T	TiO_2	2
AMZ-4T	TiO_2	4
AMZ-6T	TiO_2	6
AMZ-2M	$MgCO_3$	2
AMZ-4M	$MgCO_3$	4
AMZ-6M	$MgCO_3$	6
AMZ-2TM	TiO_2-$MgCO_3$	2
AMZ-4TM	TiO_2-$MgCO_3$	4
AMZ-6TM	TiO_2-$MgCO_3$	6

To determine the densities of samples, the Archimedes method was used based on the ASTM-C20 standard. The X-ray diffraction (XRD) method was employed to identify the formed phases. XRD patterns were acquired by means of D-500 X-ray diffraction-meter device, which used CuK_α beam with a wavelength of 0.1542 nm. The microscopic examinations of samples were carried out by a Scanning Electron Microscope (SEM Cambridge Stereoscan model).

3. RESULT AND DISCUSSION

Bulk density of the samples sintered at 1600ºC is presented in Figure 2. According to this figure, in AMZ-M and AMZ-T samples with 2% of sintering aid, the bulk densities showed an ascending trend; however, the bulk density of the AMZ-TM sample decreased a little. Two points should be mentioned regarding the bulk density and porosity of the samples; first, the sintering process, which leads to the increasing in bulk density and the reduction of porosity and second, the decomposition of zircon and formation of mullite which can cause expansion and reduction in density. Therefore, by adding 2% sintering aid to the mixture, sintering process becomes dominant in the AMZ-M and AMZ-T composites and the increase of bulk density is mostly due to the reduction of porosity. However, in the AMZ-TM sample, the formation of mullite, which has a lower density in comparison to the original constituent materials, is dominant. Thus, a reduction of density is observed. Subsequently, by increasing the amount of sintering aid in the mixture, the densities of all three samples (AMZ-M, AMZ-T and AMZ-TM) have increased.

It is also observed that the rising curves for different samples don't have the same slope. It can be concluded that the combined sintering aids of magnesia and titania (AMZ-TM) and the sintering aid of magnesia (AMZ-M) in the weight range of 2-4 wt. % have a greater effect on the sintering process which leads to more porosity

reduction whereas for the sample that contains that titania additive, the whole weight range has the same effect on sintering.

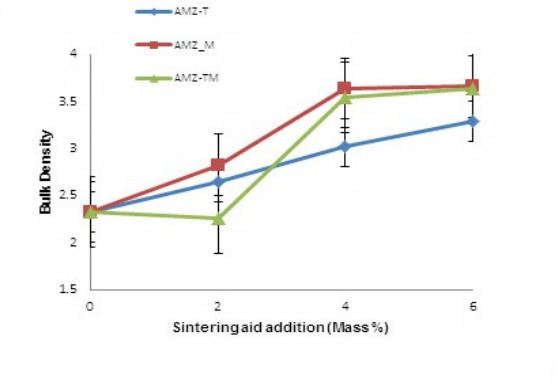

Figure 2. Changes of the bulk density versus sintering aid with the amount and after firing at 1600°C/2h.

Figure 3. shows the X-ray diffraction patterns of the AMZ-2M samples sintered at temperatures of 1400 and 1600ºC. As it is observed, at 1400ºC, mullite has not been established yet and the sample only contains alumina and zircon. Numerous weak peaks of zirconia indicate that zircon starts to decompose. Of course, decomposition is not yet complete, and zircon peaks can still be seen clearly. The decomposition temperature of zircon is above 1600ºC, but we can see that the presence of Al_2O_3 has stimulated the decomposition of zircon at temperature of 1400ºC [5]. Also, the other products of zircon decomposition (SiO_2) was not observed at this temperature, which could indicate the formation of the amorphous SiO_2 phase [12, 13] or the onset of its reaction with alumina to form mullite [14]. Young et al. reported the start of mullite formation at temperature 1450ºC (this temperature is higher than the temperature obtained by thermodynamic estimations) [9]. The XRD pattern of the sample obtained at 1600 ºC lacks the characteristic peak of zircon (2θ = 20º); thus, it can be stated that the zircon phase has decomposed to a large extent so that it cannot be detected by the X-ray and as a result of SiO_2 reaction with alumina, mullite has formed [15]. The major crystalline phases at this temperature are alumina, mullite and zirconia.

ZrO_2 initially forms as a tetragonal phase (t-ZrO_2), after decomposition of the zircon ($ZrSiO_4$) to ZrO_2 and amorphous SiO_2. At high temperature amorphous SiO_2 reacts with alumina and establishes mullite. During cooling, a large quantity of tetragonal zirconia is converted to monoclinic type. It has become well established that the ZrO_2 phase conversion is affected by the size of particles and that the larger particles promote the conversion of t-ZrO_2→ m-ZrO_2 during cooling. In sintering reaction, ZrO_2 grains grow as the temperature rises, and during cooling at room temperature the larger ZrO_2 grains convert to the m-ZrO_2 phase more easily [9].

The absence of mullite concurrent with the formation of ZrO_2 in the sample sintered at 1400ºC shows that the nucleation rate of ZrO_2 is higher than that of mullite; although, the amount and viscosity of the glassy phase can influence the nucleation and crystallization of mullite and ZrO_2. By increasing the temperature to 1600°C and decreasing the viscosity of SiO_2, the nucleation and growth of mullite accelerate [16]. According to the reports of Aksel et al. [17], the radial decomposition of zircon starts from its surface and continues to the grain center, which speeds up the crystallization of zirconia and the formation of amorphous silica.

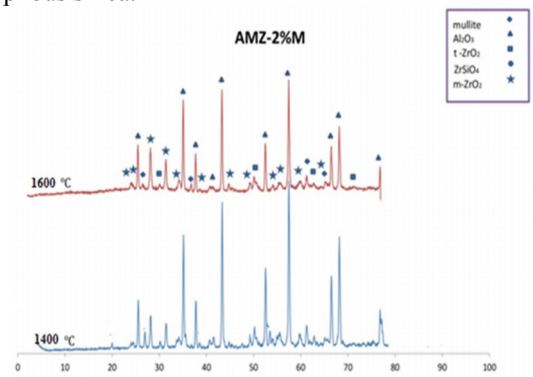

Figure 3. X-Ray diffraction patterns of the AMZ-2M sample sintered 1400 and 1600 ºC

Figure 4. illustrates the X-ray diffraction patterns of the AMZ-M samples with 2 wt. % and 6 wt. % Magnesia sintering aid after firing at 1600ºC for 2 hours. Alumina has formed as the main phase. Mullite, and monoclinic and tetragonal zirconia can be similarly seen in both samples. Considering the reports of Benedetti et al [18], which claim that the existing peaks at 2θ = 28.4º and 2θ = 30.19º specify monoclinic zirconia and tetragonal zirconia, respectively, it can be stated that all the samples have these two phases; of course, the number of tetragonal zirconia peaks and their intensities are less than those of the monoclinic zirconia peaks [18]. Many research works have recently concluded that the presence of MgO additive in the zirconia/mullite ceramic composites not only stabilizes the tetragonal zirconia phase, but, as a sintering aid, it also helps the formation of mullite grains [16].

The XRD pattern of the AMZ-6T sample in Figure 5. shows a similar pattern to that of the AMZ-2T. The lack of zircon and silica (crystobalite and/or tridimite) peaks in both samples is indicative of the complete decomposition of zircon and the reaction of SiO_2 with alumina and thus the formation of mullite, with different peaks of mullite in the AMZ-6T composite are intensive.

According to the X-ray diffraction pattern of the AMZ-TM sample in Figure 6. the peaks are almost similar to each other and the phases of alumina-mullite-monoclinic /tetragonal zirconia are the main phases of

two samples. As it is observed, the TiO_2 sintering aid, like MgO, has been able to stabilize the zirconia [19].

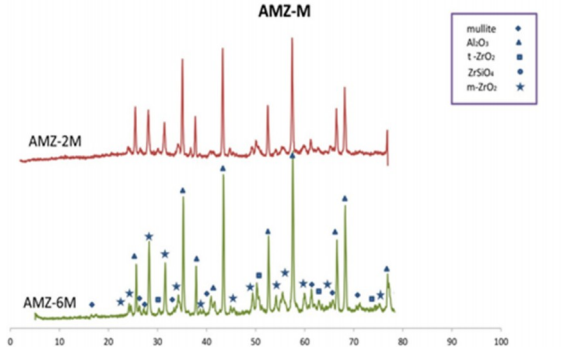

Figure 4. X-Ray diffraction patterns of the AMZ-M samples after sintering at 1600 ºC for 2 hours.

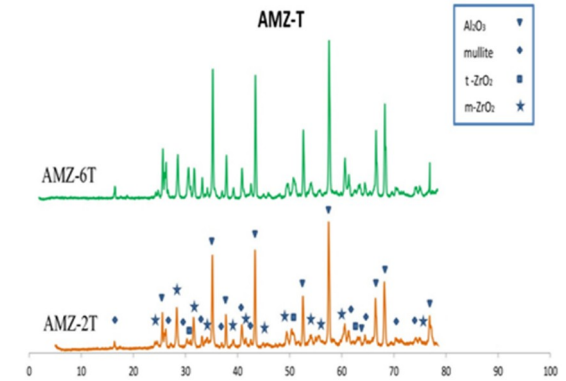

Figure 5. X-Ray diffraction pattern of AMZ-T sample after sintering at 1600ºC for 2 hours.

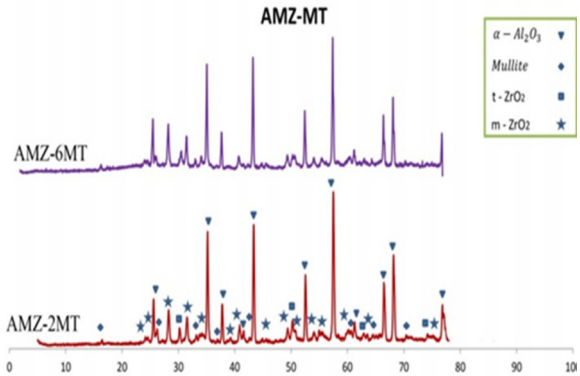

Figure 6. X-Ray diffraction pattern of the AMZ-TM sample after sintering at 1600ºC for 2 hours.

To examine the formation of the mullite phase more accurately, a relativistic and comparative method was employed. In this approach, by measuring the areas under the main peaks of the mullite phase (210) and alumina phase (113), by using formula (2) [20 and 21] the relative concentration of the mullite phase was measured. In this formula, C_M and C_A denote the estimated concentrations of mullite and alumina, respectively, and parameter β indicates the degree of the mullite reaction and formation progress.

$$\beta = \frac{C_M}{C_M + C_A} \times 100 \qquad (2)$$

The obtained results are illustrated in Figure 7. It can be seen that the combined effect of magnesia and titania sintering aids is greater than the sole effect of magnesia or titania by itself. At 2 and 6 wt.% adding sintering aids, the AMZ-T and AMZ-TM samples, respectively, have much greater mullite concentrations than the other samples.

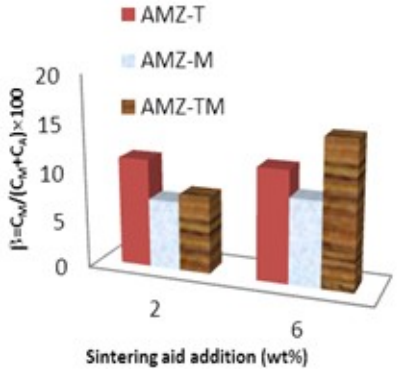

Figure 7. Relative changes of mullite concentration with different types and amounts of sintering aids.

The higher portion of mullite in these samples is probably due to the fact that, in addition to the issues of solid solution and increased diffusion, the additives of MgO and TiO_2 together produce more melt in the sample (considering the phase diagram of MgO-TiO_2 in Figure 8.) in which more alumina is dissolved and thus more mullite is precipitated.

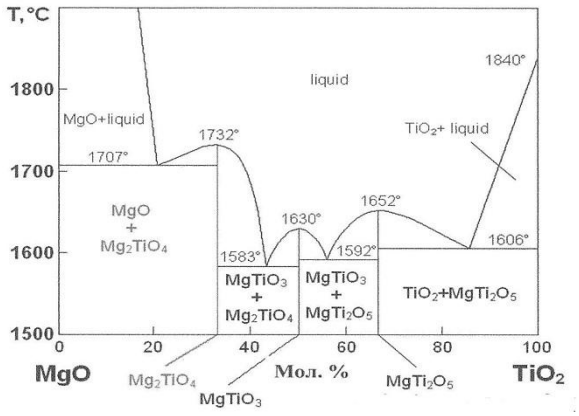

Figure 8. Phase diagram of MgO-TiO_2[22]

The XRD patterns of the samples show, to some extent, the increase in tetragonal zirconia stabilized in the composite by increasing the sintering aid from 2 to 6 wt. %. To verify these observations, the polymorph method of Garvie-Nicholson, in view of Eq. 3, was used [23]. In this equation, I_t (111) is the peak intensity of plane (111) of the tetragonal zirconia, I_m (111) is the peak intensity of plane (111) of the monoclinic zirconia and I_t $(11\bar{1})$ is the peak intensity of plane $(11\bar{1})$ of the tetragonal zirconia.

$$X_t = \frac{I_t(111)}{I_t(111) + I_m(111) + I_m(11\bar{1})} \qquad (3)$$

The obtained results have been illustrated in Figure 9. As it is observed, all the samples display an increase in tetragonal zirconia by the increase in the amount of

sintering aids; and the highest increase can be seen in the composite that contains the mixed sintering aids. Of course, this trend is reversed at lower amounts of sintering aids, and the composite made by adding the mixed sintering aids produced the lowest amount of tetragonal zirconia.

It can be established that the ZrO_2 particles, released from the decomposition of zircon, immediately crystallize as tetragonal zirconia phase, through nucleation and growth, and this phase converts to a monoclinic structure at low temperatures [24]. One of the characteristics of the reaction sintering method used in the preparation of these composites is the semi-stable phase of tetragonal zirconia that exists at ambient temperature [25]. The stability of this phase at low temperature may be justified as such that some zirconia particles are entrapped inside the mullite grains during the crystallization of mullite and, due to the low diffusion coefficient of mullite, these particles are not able to grow; in other words, their dimensions remain smaller than the critical limit necessary for converting them to monoclinic zirconia particles [25-26].

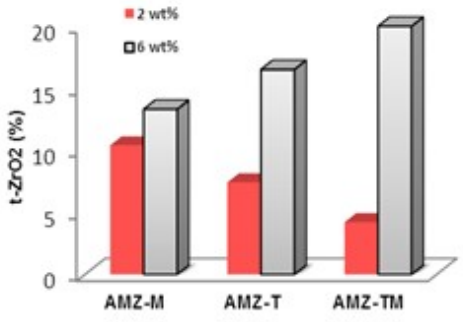

Figure.9. Amounts of t-ZrO_2 estimated by the polymorph method in different composites.

Bochet al. [27] calculated the amounts of mullite and alumina in the alumina-mullite composite by the ratio of peak intensities, according to the relation $I_A(113)/[I_A(113)+I_M(210)]$; where I_A was the peak intensity of plane (113) of alumina located at 2θ = 43.36º and I_M was the peak intensity of plane (210) of mullite situated at 2θ = 26.27º. Similar work was done by Mitra et al. [11,19]on the alumina-zirconia-mullite composite to obtain the share of each phase of alumina, zirconia (monoclinic + tetragonal) and mullite. They also used the mentioned characteristic peak intensities for this purpose. On this basis, the percentages of mullite and alumina phases, different composite samples have been determined and sketched in Figure 10. As it is observed, the increase in the amount of sintering aids in all the samples facilitated the formation of mullite. In this regard, the AMZ-6TM sample has the highest percentage of mullite and the AMZ-2TM sample has the highest percentage of alumina.

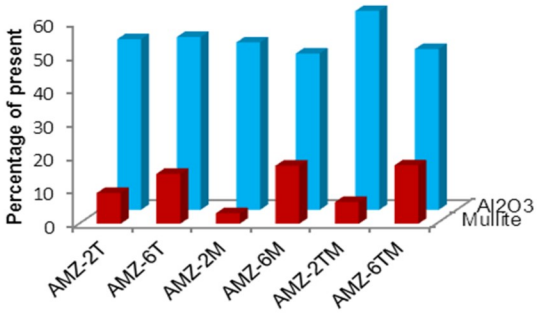

Figure 10. Percentages of mullite and alumina phases present in different composites.

The sizes of the AMZ composite crystallites are listed in Table 2. As it is observed, despite the high sintering temperature and due to the presence of the sintering aids, all the crystallites that form the composite have nanometer sizes. If the sintering aids can be present at grain boundaries, they can retard the size and growth rate of the crystallites by reducing the movement of the grain boundaries.

TABLE 2. Sizes of crystallites in composites with different types and amounts of sintering aids

Sample	mullite crystallite (nm)	alumina crystallite (nm)	Monoclinic phase	
			Tetragonal phase	zirconia crystallite (nm)
AMZ-2M	22.49	84.43	34.75	51.02
AMZ-6M	34.07	97.09	53.03	74.17
AMZ-2T	22.34	70.35	37.98	54.86
AMZ-6T	40.99	84.42	42.53	80.10
AMZ-2TM	33.08	70.35	36.62	45.26
AMZ-6TM	34.08	84.42	48.32	53.40

Figure 11. shows the SEM images of the fracture cross sections of different AMZ samples taken after the completion of polishing operation. Some Pores could be observed in these samples, especially in samples containing 2 wt. % sintering aid; which of course, by increasing the amount of sintering aid and completing the decomposition of zircon, the amount of porosity is reduced and more uniform microstructures are obtained. As the above figure illustrates, the samples containing 2 wt. % sintering aids display nonuniformity in microstructure aid above present of porosity due to low sintering potential of compositions.

While the images of samples containing 6 wt. % sintering aids show a rigid structure with engaged grains for alumina and mullite phases, and the zirconia grains that have dispersed uniformly throughout the matrix. Almost in all samples, the zirconia grains have irregular polygon shapes, but in the AMZ-6M composite, as it shown in Figure 12. they have become more square-like.

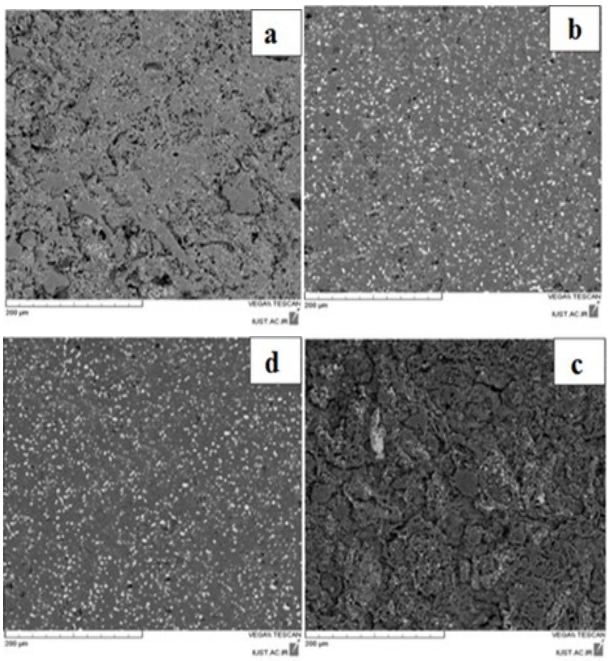

Figure 11. SEM (BSE) images of (a) AMZ-2T, (b) AMZ-6T, (c) AMZ-2TM and (d) AMZ-6TM

In the structure of the AMZ-6M sample, like in the other composites with 6 wt. % weight sintering aid portions, a smooth surface is not observed; this could be for the type and amount of the formed amorphous phase. Also, various types of zirconia particles in these composites can be classified as follows: inter-grain zirconia particles (Figure 12.(2)); and intra-grain zirconia particles (Figure 12.(1)) which can be divided into two groups of zirconia particles within Alumina grains (Figure 12.(1a)) and zirconia particles within mullite grains (Figure 12.(1b)). Also, in terms of shape, these particles can be placed in two classes of almost-round zirconia (Figure 12.(3)) and more square like zirconia particles with cleavage planes (Figure 12.(4)).

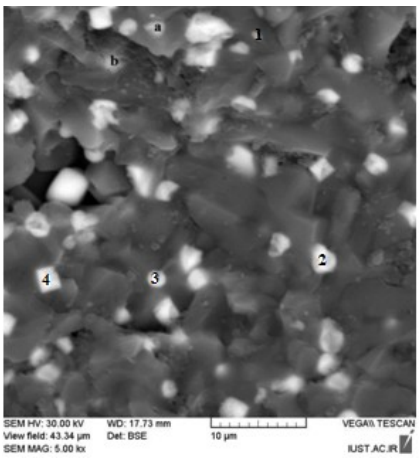

Figure 12. Zirconia classification in the AMZ-6M composite; (1) Intra-grain zirconia particles ((a) inside alumina grains, (b) inside mullite grains), (2) Inter-grain zirconia particles, (3) Round-shape zirconia, (4) Square-shape zirconia.

That part of the zirconia existed in the composite specimen is in tetragonal form. These particles have more round shapes and smaller sizes, and probably the compressive force exerted by the matrix has caused them to remain in tetragonal shape. The square shapes and larger sizes of these particles could be due to their conversion to the monoclinic phase [26].

In view of the SEM images, the average diameter of zirconia grains and also the diameter range of these particles are listed in Table 3. It is observed that in all the examined samples, the increment in the amount of sintering aids has led to the reduction of the average diameter of zirconia grains.

TABLE 3. Range and average size of zirconia particles

Sample	Mean particle size of zirconia (µm)	Range particle size of zirconia (µm)
AMZ-2M	2.22 ±0.75	0.71-4.31
AMZ-6M	2.14±0.48	0.8-3.93
AMZ-2T	3.48±1.12	0.54-6.81
AMZ-6T	2.82±1.07	0.43-5.20
AMZ-2TM	2.87±1.14	0.71-5.36
AMZ-6TM	2.07±0.87	0.54-4.29

4. CONCLUSIONS

This investigation has led to the following conclusions:

1) By increasing the amount of all three sintering aid additives (MgO, TiO_2, MgO+TiO_2) in the samples from 2 to 6 wt. %, the reactions were improved and the density of the samples and the final quantity of mullite in them were increased.

2) The tests showed that, as expected, the decomposition of zircon is facilitated in the presence of alumina and it occurs at lower temperature.

3) Due to the mechanism involved in the reaction sintering method, a part of the zirconia phase remains in a semi-stable tetragonal form.

4) By increasing the amount of sintering aids, the average diameters of zirconia grains are decreased.

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