American Journal of Modern Physics
2013; 2(5): 270-275
Published online September 20, 2013 (http://www.sciencepublishinggroup.com/j/ajmp)
doi: 10.11648/j.ajmp.20130205.16

Effect of MgO addition on the mechanical and thermal properties of mullite synthesised through reaction sintering of Al_2O_3 and Algerian kaolin

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To cite this article:
A. Ouali, M. Heraiz, F. Sahnoune, H. Belhouchet, M. Fatmi, N. Saheb. Effect of MgO Addition on the Mechanical and Thermal Properties of Mullite Synthesised through Reaction Sintering of Al_2O_3 and Algerian Kaolin. *American Journal of Modern Physics*. Vol. 2, No. 5, 2013, pp. 270-275. doi: 10.11648/j.ajmp.20130205.16

Abstract: The influence of MgO addition on the structure and properties of mullite prepared through reaction sintering of Algerian kaolin and Al_2O_3 was investigated. The raw powders were wet ball milled, dried and cold compacted using a uniaxial press. The green compacts were sintered 8 hours at 1600 and 1650°C. The microstructure of samples was characterized using a scanning electron microscope. Mechanical and thermal properties were characterized using Vicker's hardness tester, a universal testing machine and a dilatometer. It was found that the increase of MgO content from 0 to 3 wt-% increased the hardness of samples sintered 8 hours at 1600°C from 1039 to 1316.57 HV. Also, the increase of MgO content in samples sintered 8 hours at 1600 and 1650°C increased the compressive strength up to a maximum then decreased it. For a sintering temperature of 1600°C, the increase of MgO content up to 2 wt-% increased the flexural strength, but a further increase of MgO to 3 wt-% decreased it again, while for a sintering temperature of 1650°C, the increase of MgO content from 0 to 3 wt-% increased the flexural strength from 103.45 to 472.25 MPa. Amongst MgO containing samples, the increase of MgO content increased the coefficient of thermal expansion; however, it remained lower than the coefficient of thermal expansion of the sample without MgO addition.

Keywords: Kaolin, Mullite, Alumina, Reaction Sintering, Mechanical Properties, Thermal Properties

1. Introduction

Synthesis of mullite from clay remains an economical way to prepare mullite and mullite based composites [1-12] ; because mullite powder compacts have poor solid state sinterability [13] and the production of dense mullite compacts requires relatively high sintering temperatures, many studies have been dedicated to investigate the effect of adding sintering aids such as MgO [14-20], SrO [21], B_2O [22], TiO_2 [23], CeO_2[24], V_2O_5[25]and Y_2O_3 [26] on the densification and sinterability of mullite.

Although MgO is one of the most commonly used sintering aids in the processing of mullite, the amount of MgO to be used is still a matter of controversy [26]. It was reported that the addition of MgO above 0.5 wt.% has no effect in controlling mullite grain growth [14]. The efficiency of adding small amounts of MgO (below 0.5 wt.%) on the sintering of industrial mullite was evaluated by Souto and co-workers[26]. They found that the use of 0.1 and 0.5 wt.% of MgO increased the final density of the sintered samples, with the doped samples reaching densities of 99% and the nondoped samples reaching densities of 95%. Also, the use of 0.5% of MgO decreased the sintering temperature by approximately 100^0C. However, many authors have used larger amounts of MgO, usually ranging from 1 to 5 wt.%. Viswabaskaran *et al*. [5] reported that the addition of MgO increased the grain growth and thereby increased the density to maximum value. Also, they found that MgO enhanced the

liquid phase sintering and they obtained a density value of 2.91 g.cm^{-3} for the sample containing 3 wt.% MgO. Also, they concluded that the amount of MgO beyond 3 wt.% had induced bloating and thereby decreased the density. They achieved a strength of 126 MPa for 3 wt.% MgO added samples. In another investigation [18] they found that samples with 3.0 wt.% MgO showed the presence of secondary phases such as α-alumina and spinel; and mullite crystals formed were needle-shaped with rectangular faces; and the MgO (3.0 wt.%) addition slightly decreased the thermal expansion. Doni Jayaseelan and co-workers [17] prepared industrial mullite compacts having near theoretical density using pulse electric current sintering at 1500°C for 2 min. They found that the dopants influenced the morphology of mullite grains and hence the fracture mode. The fracture surface of MgO enriched mullite was highly crystalline in nature and the grains showed extensive fine cleavage.

In previous works we synthesised mullite through reaction sintering Algerian kaolin and Al_2O_3 [12] and investigated the kinetics of mullite formation from Algerian kaolin [27] and analysed its thermal dehydroxylation[28]. Also, we reported the effect of MgO addition on the structure and sintering behaviour of mullite [29] and found that in samples containing 0, 1 and 2 wt-%MgO only mullite was formed while, in addition to mullite, Al_2O_3 was present in samples containing 3 wt-%MgO. At higher MgO content (4, 5 and 6 wt-%), three phases, i.e. mullite, Al_2O_3 and spinel, were formed. Addition of 1 wt-%MgO increased the density of all samples for all sintering times and higher densities corresponded to higher sintering temperatures. At higher MgO content, higher temperatures led to lower densities and lower temperatures led to higher densities for almost all sintering times. The objective of the present work is to investigate the effect of MgO addition on the mechanical and thermal properties of mullite synthesised through reaction sintering of Algerian kaolin and Al_2O_3.

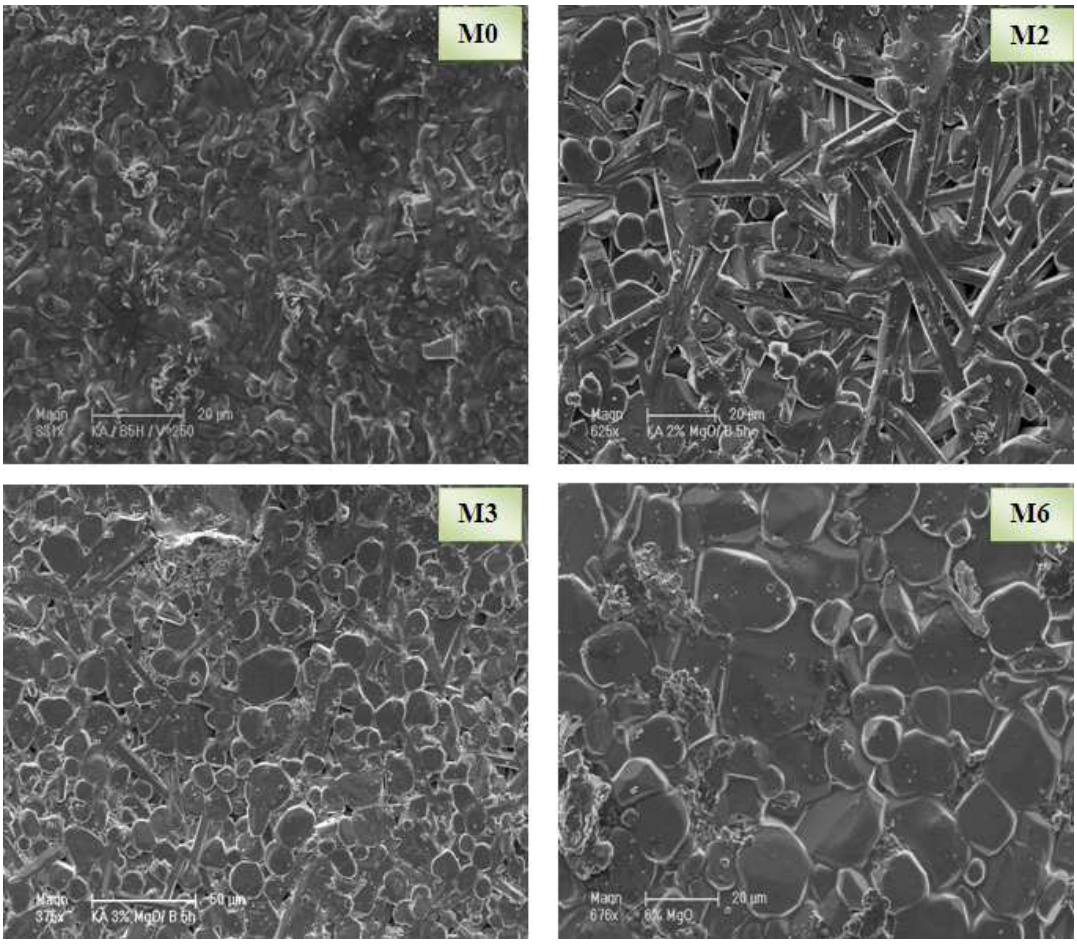

Figure 1. *SEM micrographs of samples sintered at 1650°C for 8 hours.*

2. Materials and Experimental Procedures

Algerian raw kaolin (from Djabal Debagh) was added to Al_2O_3 (99.98 % purity, with average particle size of 0.5 μm) supplied by Fluka according to the stoichiometry that leads to the formation of mullite ($3Al_2O_3.2SiO_2$) only. Samples containing MgO (named M0 to M6) were prepared by addition of 0, 1, 2, 3, 4, 5 and 6 wt-% of MgO to the kaolin-alumina mixture. More information on the chemical composition of the kaolin and samples as well as the

theoretical density of the samples is reported elsewhere [12,27,29]. The mixture of the powders was charged into zirconia vials (250 ml in volume) together with 15 zirconia balls (10 mm in diameter). The ball-to-powder weight ratio was maintained as 10:1 and water was added at a ratio of 2:1. The ball-milling experiments were performed in a high-energy planetary ball mill (Fritsch P6) and were carried out at room temperature at a rotation speed of 250 rpm for 5 hours. The milled mixture was dried at 100 °C for 6 hours then cold compacted at 75 MPa using uniaxial press. Cylindrical specimens of 13 mm diameter were produced. The green compacts were sintered in a furnace at 1600 and 1650°C for 8 hours. The heating rate was 10°C/min. The microstructure of samples was characterized using a JEOL scanning electron microscope (SEM) model JSM 5600. Vicker's hardness was measured by applying a load of 300 g on the polished surface of the specimens. The flexural strength was evaluated through three-point bending test using an INSTRON universal testing machine. Specimens having 8 mm×8 mm×50 mm and a crosshead speed of 0.5 mm/min were used. Each reported value was an average of 5 measurements. The compressive strength of samples was measured using an INSTRON universal testing machine. The linear coefficient of thermal dilation and the relative variation length were measured using a dilatometer (DIL 402C).

3. Results and Discussion

Figure 1 shows SEM micrographs of samples containing 0, 2, 3 and 6 wt-%MgO sintered at 1650°C for 8 hours. It can be clearly seen that the addition of MgO promoted grain growth, this is in agreement with results reported by Viswabaskaran et al [5]. More details on the effect of MgO addition and sintering parameters on the microstructure, phases present and the densification behaviour were reported elsewhere [29].

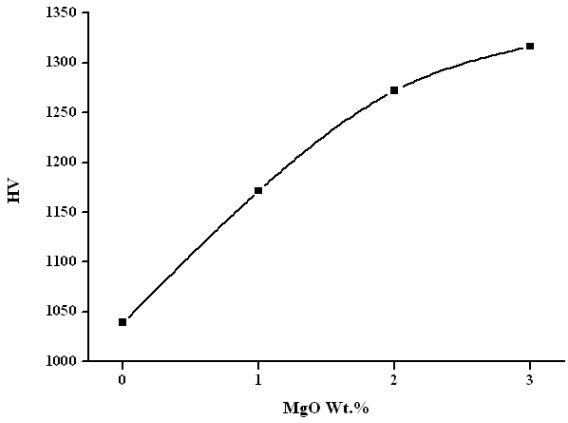

Figure 2. *Vicker's hardness of samples sintered 8 hours at 1600°C.*

Figure 2 shows Vicker's hardness of samples containing 0, 1, 2, 3 wt-%MgO sintered 8 hours at 1600°C. The increase of MgO content from 0 to 3 wt-% increased the hardness from 1039 to 1316.57 HV. The hardness value of 1039 HV obtained for mullite having a relative density of 93.26% without addition of MgO is comparable to that obtained by Hirata *et al*. [30], who obtained a hardness ranging from 12 to 13 GPa for mullite having relative density between 95.8 and 98 % prepared through sintering pure Al_2O_3 and SiO_2 for 3 hours at 1550 °C. The increase of the hardness is due to the increase of the relative density of samples with the increase of MgO content; this trend was also observed by other researchers [5]. It is worth mentioning here that for samples containing 0, 1 and 2 wt-%MgO only mullite was formed while, in addition to mullite, Al_2O_3 was present in samples containing 3 wt-%MgO.

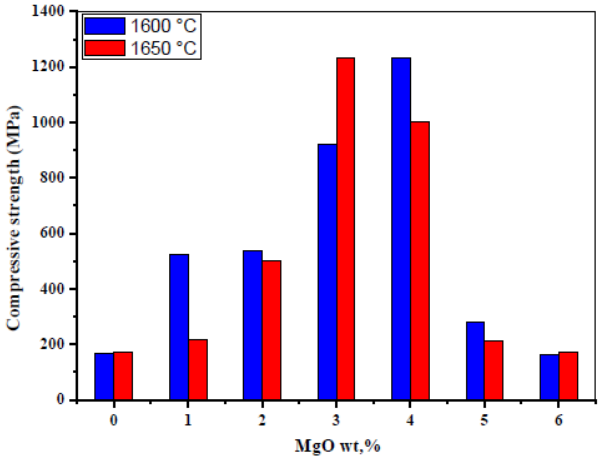

Figure 3. *Compressive strength of samples sintered 8 hours at 1600 °C and 1650 °C.*

Figure 3 shows the compressive strength as a function of MgO content for samples sintered 8 hours at 1600 and 1650°C. It can be clearly seen that at both temperatures the increase in MgO content increased the compressive strength up to a maximum then decreased it. A maximum strength of 1234.17 MPa was achieved at a sintering temperature of 1650°C for the sample containing 3 wt-% MgO; and a maximum strength of 1231.31 MPa was achieved at a sintering temperature of 1600°C for the sample containing 4 wt-% MgO. The increase of sintering temperature from 1600 to 1650°C increased the compressive strength of sample M0 from 169.59 to 172.64 MPa. In MgO containing samples the increase of sintering temperature increased the compressive strength in samples M3 and M6 and decreased it in samples M1, M2, M4 and M5. This trend may be due to the presence of deferent phases in the samples. It was shown in previous work [29] that for samples containing 0, 1 and 2 wt-%MgO only mullite was formed; while, in addition to mullite, Al_2O_3 was present in sample containing 3 wt-%MgO. At higher MgO content (4, 5 and 6 wt-%), three phases, i.e. mullite, Al_2O_3 and spinel, were formed. Also, the relative density, open porosity and closed porosity do affect the strength of samples. The relative density, open porosity, and closed porosity of samples sintered 8 hours at 1600 and 1650°C are presented in figure 4.

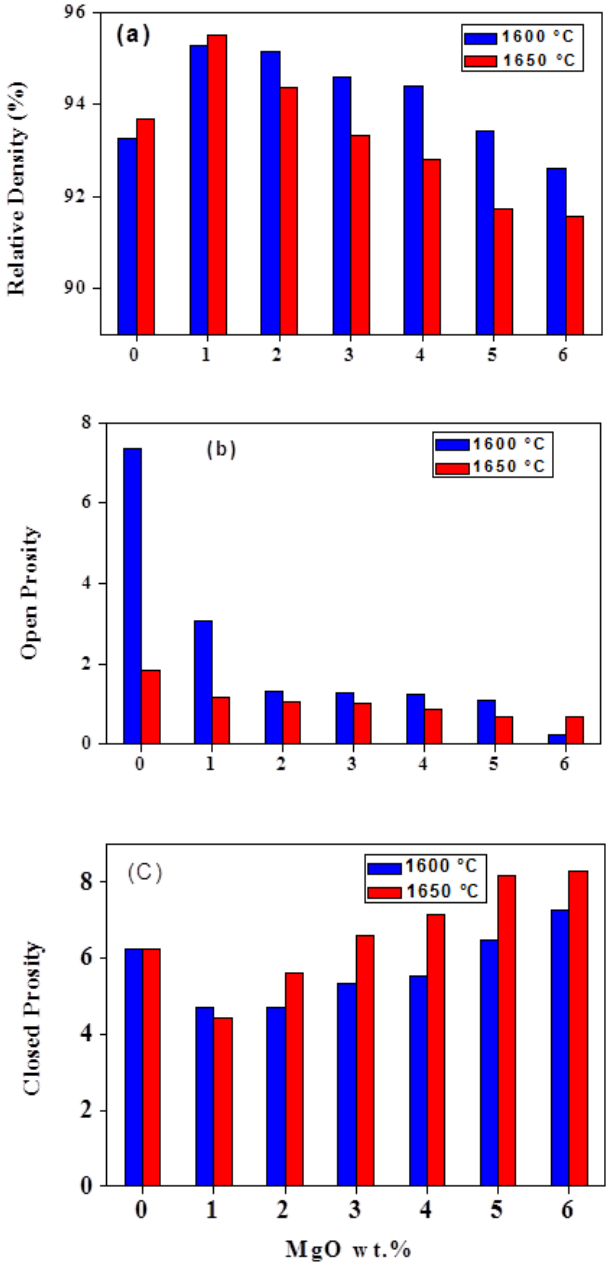

Figure 4. *Relative density (a) open porosity (b) and closed porosity (C) of samples sintered 8 hours at 1600 °C and 1650 °C.*

The increase of sintering temperature from 1600 to 1650°C slightly increased the relative density of samples M0 and M1 while decreasing the relative density of samples M2, M3, M4, M5 and M6 as can be seen in figure 4(a). The open porosity decreased in all samples with the increase of sintering temperature, as shown in figure 4(b). However, the closed porosity remained unchanged in sample M0, slightly decreased in sample M1, and increased in samples M2, M3, M4, M5 and M6 as can be clearly seen in figure 4(c).

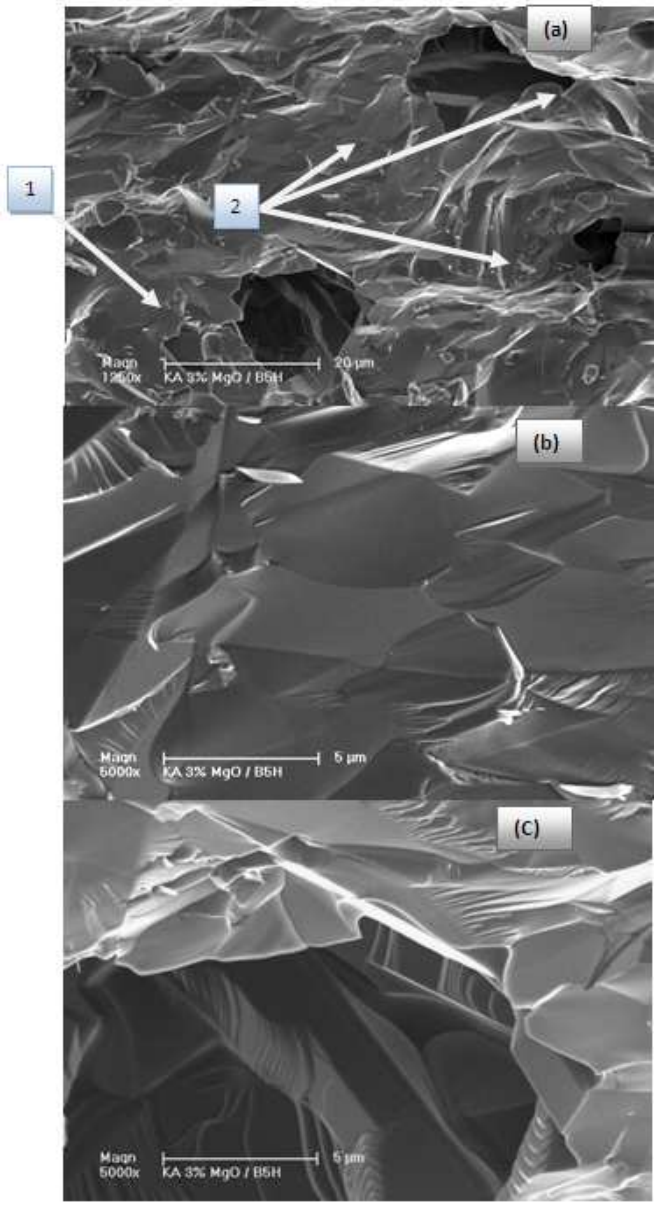

Figure 5. *Surfaces of fractured sample containing 3 wt-%MgO (a) pores and surface markings, (b) higher magnification showing cleavage steps and (c) a river pattern.*

Surfaces of fractured sample M3 containing 3 wt-%MgO are shown in figure 5. It is clear that the mode of fracture in this sample is transgranular. Pores of deferent sizes and shapes, regions 1 and 2 in figure 5(a), were present on the surface. Also, surface markings and cleavages were observed.

Figure 5(b) shows a magnified view of these surface markings believed to be the cleavage planes. It was reported by Torrecillas and co-workers [31] that the presence of sharp-edged pores at grain boundaries and grain boundary junctions favor the cleavage, with the pores acting as stress concentrators. Figure 5(c) shows typical 'river patterns' which form at the passage of grain boundaries. Because adjacent grains have different orientations, cleavage cracks change directions at grain boundaries to continue propagation [32].

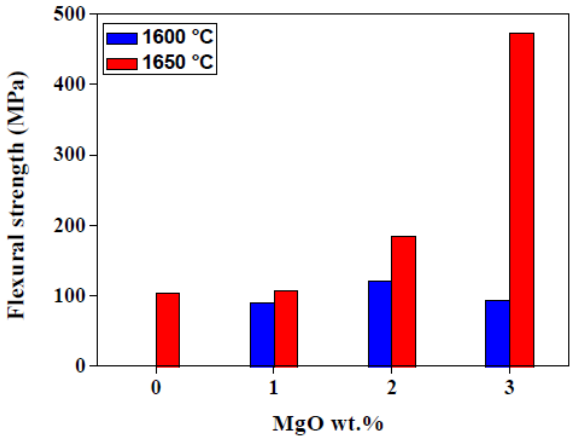

Figure 6. *Flexural strength of samples sintered 8 hours at 1600 °C and 1650 °C.*

Figure 6 shows the flexural strength of samples containing 0, 1, 2, 3 wt-% MgO sintered 8 hours at 1600 and 1650°C. For a sintering temperature of 1600 °C, the increase of MgO content up to 2 wt-% increased the strength, a further increase up to 3 wt-% decreased it again. However, for a sintering temperature of 1650°C, the increase of MgO content from 0 to 3 wt-% increased the flexural strength from 103.45 to 472.25 MPa. This result is in agreement with that reported by Doni *el al*. [17] who obtained a maximum flexural strength of 441 MPa for MgO containing samples.

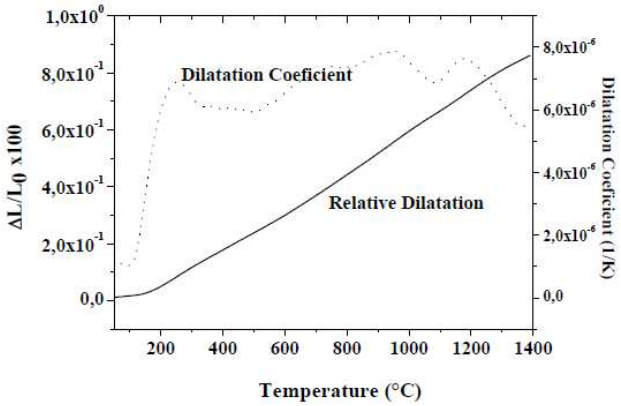

Figure 7. *Thermal expansion of sample M0 sintered 8 hours at 1600°C*

Figure 7 shows typical thermal expansion curve of sample M0 sintered 8 hours at 1600 °C. The coefficient of thermal expansion of samples containing 0, 1, 2 and 3 wt-% MgO between room temperature and 1400°C is presented in figure 8. It is clear that for samples containing MgO, the increase of MgO from 1 to 3 wt-% increased the coefficient of thermal expansion from 4.25 x 10^{-6} to 4.75 x 10^{-6} K^{-1}. However, the coefficient of thermal expansion of samples containing MgO remained lower than the coefficient of thermal expansion of samples without MgO addition i.e. 5.54 x 10^{-6} K^{-1}. The lower MgO content, the lower the coefficient of thermal expansion, this is in agreement with results reported by Somiya and co-workers [33].

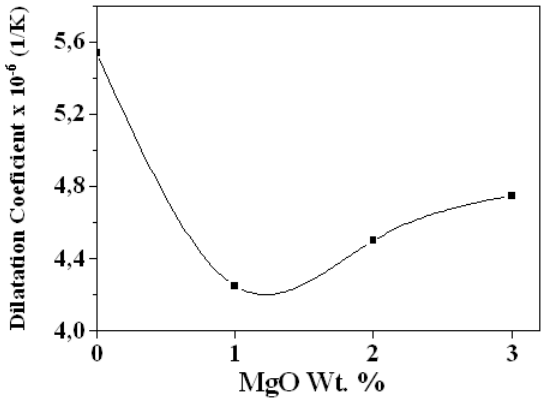

Figure 8. *Coefficient of thermal expansion of samples sintered 8 hours at 1600°C*

4. Conclusion

In this study the mechanical and thermal properties of mullite prepared through reaction sintering of Algerian kaolin and Al_2O_3 were characterized. It was found that the increase of MgO content from 0 to 3 wt-% increased the hardness of samples sintered 8 hours at 1600°C from 1039 to 1316.57 HV. Also, the increase of MgO content in samples sintered 8 hours at 1600 and 1650°C increased the compressive strength up to a maximum then decreased it. For a sintering temperature of 1600°C, the increase of MgO content up to 2 wt-% increased the flexural strength, but a further increase of MgO to 3 wt-% decreased it again; while for a sintering temperature of 1650°C, the increase of MgO content from 0 to 3 wt-% increased the flexural strength from 103.45 to 472.25 MPa. Amongst MgO containing samples, the increase of MgO content increased the coefficient of thermal expansion; however, it remained lower than the coefficient of thermal expansion of samples without MgO addition.

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