**Effect of Y$_2$O$_3$ Additions on Microstructure and Properties of Alumina–Magnesia Ceramics**

**Aurawan Rittidech, Pimpun Wisuwan and Thitima Pinkhunthod**

*Department of Physics, Faculty of Science, Mahasarakham University, Mahasarakham, Thailand, 44150, Thailand*

**Abstract:** The purposes of this research are to prepare and investigate the characterization of 0.7MgO-xY$_2$O$_3$-(99.3-x)Al$_2$O$_3$ by varies x at 0.0, 2.0, 4.0, 6.0, 8.0 and 10.0 wt% (AMY). AMY samples were synthesized by solid state reaction under two different sintering methods conventional sintering method (NS) and two stage sintering method (TS). Ceramic samples were sintered by conventional sintering at 1600°C for 5 h and using two stage sintering with the first sintering temperature (T$_1$) at 1600°C for 30 min and cooling down to the second sintering temperature (T$_2$) at 1450°C for 10 h. The phase composition of the samples were characterized using XRD technique. XRD patterns from two method sintering revealed phase combination of Al$_2$O$_3$, MgAl$_2$O$_4$, MgO and Y$_2$O$_3$. It was found that it is not different XRD patterns from two method sintering. SEM micrographs form NM and TS showed that the shape of the ceramic grains were polyhedron and ellipsoid, while the grain sizes form NS were in the range of 0.91-1.00 µm and the grain sizes from TS were in the range of 0.34-0.48 µm. The samples from NS and TS is showed optimum mechanical properties by AMY with Y$_2$O$_3$ additions between 4-6 wt%.

**Keywords:** Al$_2$O$_3$, MgO, The Solid State Reaction, Two-Stage Sintering

**Introduction**

Alumina (Al$_2$O$_3$) ceramics have high hardness, good wear resistance and high temperature stability. Rao *et al.* (2003) revealed that Al$_2$O$_3$ ceramics has been widely used for structural ceramics application similar results were reported recently (Rejab *et al.*, 2014). In spite of the variety of useful physical properties of sintered oxide ceramics based on chemically and thermally stable alpha modification of alumina (α-Al$_2$O$_3$) their application as cutting tool inserts working under mechanical loads and thermal shock conditions is limited due to their brittleness and low strength. Among dopants in Al$_2$O$_3$, Magnesium Oxide (MgO) has an important effect on mechanical and electrical properties of bulk Al$_2$O$_3$. Lu *et al.* (2005) has shown that MgO is a traditional additive to Al$_2$O$_3$ since it can reduce the sintering temperature and grain size with the results are supported good mechanical properties agreement with other works (Rittidech *et al.*, 2006). Moreover, The work by Azhar *et al.* (2010) confirmed that small amounts of magnesia (<0.70 wt%), when added to zirconia-toughened alumina, enable it to a promising material for machining applications. Another group of dopants is represented by metal oxides, which strongly segregate at alumina-alumina interfaces, such as yttria and zirconia. Due to its limited solubility in alumina crystal lattice (~10 atomic ppm) yttrium segregates to α-Al$_2$O$_3$ surfaces and improves the creep resistance at high temperatures. This makes yttria a common dopant in many applications. Yttria doping was found to inhibit both densification and grain growth of alumina but the effect is much reduced with increasing temperature. Galusek *et al.* (2012) confirmed doping with Mg, Y and Zr resulted in suppression of grain growth in the final stage of sintering. Ceramic material is made by high temperature sintering process from the raw powder and there is a close link between the microstructure mechanical properties of ceramic material. In the load process of raw powders, there are a number of pores between powders. The reduction process of pore is the major process in ceramic material sintering densification process that revealed by Min *et al.* (2014). The mechanical properties of the Al$_2$O$_3$-based ceramics depend strongly on the microstructure as well as composition that reported by Rittidech *et al.* (2013). The microstructure of Al$_2$O$_3$ can be controlled by two ways...
i.e., either by using additives to prohibit the grain growth for obtaining highly dense ceramics or by using the novel processing technique to modify the microstructure. Two-stage sintering process is one of the ways of eliminating grain growth in the final stage of sintering reported by Chen and Wang (2000). This was originally successfully applied to the densification of a nanometer-sized yttria powder without the final stage of grain growth. This work will study the effect of two stage sintering technique on phase formation, microstructure, densities and mechanical properties of alumina-magnesia-yttria ceramic with various Y$_2$O$_3$ additives.

**Materials and Methods**

Powders with 0.7MgO-$x$Y$_2$O$_3$-(99.3-$x$)Al$_2$O$_3$ where $x$ = 0.0, 2.0, 4.0, 6.0, 8.0 and 10.0 were prepared from MgO, Y$_2$O$_3$ and Al$_2$O$_3$ as precursors and isopropyl alcohol as solvent. All the five different batches were then ball milled for 24 h. After ball-milling, drying in alcohol as solvent. All the five different batches were then ball milled for 24 h. After ball-milling, drying in alcohol as solvent.

The crystalline size of the samples calculated from the XRD patterns is summarized in Table 1. The crystalline size of the samples calculated from the X-ray line broadening using the Scherrer equation; $D = \frac{0.9 \lambda}{B \cos \theta}$ (reported by Jenkins and Snyder, 1996). Microstructural analysis was examined by using Scanning Electron Microscopy (SEM) and Energy-Dispersive X-ray spectrometry (EDX) (JEOL JSM-840A) on a polished surface of sintered samples. The micro hardness of the bulk ceramics was measured using a micro scan from Vickers and Knoops (FM-700e type D, Future Tech., Japan).

**Results and Discussion**

Densities of the sintered samples were determined by using Archimedes principle. Figure 1 and 2 show the data on the densities and shrinkage of the 0.7MgO-$x$Y$_2$O$_3$-(99.3-$x$)Al$_2$O$_3$ where $x$ = 0.0, 2.0, 4.0, 6.0, 8.0 and 10.0 under TS sintering and NS sintering. It is observed that a density of between 3.31 and 3.73 g/cm$^3$. The maximum density, under 2 type sintering, were obtained in the samples of 0.7MgO-$x$Y$_2$O$_3$-(99.3-$x$)Al$_2$O$_3$ ceramics with 6 wt% Y$_2$O$_3$ added. It can be found that TS samples show higher densification than NS samples. Densities tend to increase with increasing concentrations of Y$_2$O$_3$. The promotion of densification by the addition of yttria in TS is reflected by the lower temperature required to achieve a high final density than when using an undoped method. Moreover, linear shrinkage was showed that increasing with Y$_2$O$_3$ contents, corresponding to densities.

| Content of $x$ | Average grain size (µm) | Crystal size (nm) | Vickers hardness (MPa) | Knoop Hardness (MPa) | Fracture toughness (MPa.m$^{1/2}$) |
|----------------|-------------------------|-------------------|------------------------|----------------------|---------------------------------|
| 0.0            | NS 1.344                | TS 0.566          | NS 79.6                | TS 63.3              | NS 7.14                        | TS 8.11                        | TS 2.16                        | TS 2.27                        |
| 2.0            | NS 1.089                | TS 0.532          | NS 68.4                | TS 61.7              | NS 10.28                       | TS 11.84                       | TS 10.81                       | TS 11.22                       | TS 2.48                        | TS 2.55                        |
| 4.0            | NS 1.004                | TS 0.488          | NS 67.5                | TS 53.8              | NS 13.47                       | TS 11.57                       | TS 10.12                       | TS 12.18                       | TS 2.84                        | TS 2.79                        |
| 6.0            | NS 0.760                | TS 0.466          | NS 61.2                | TS 50.4              | NS 11.85                       | TS 12.76                       | TS 12.09                       | TS 14.29                       | TS 2.77                        | TS 3.24                        |
| 8.0            | NS 0.825                | TS 0.477          | NS 62.4                | TS 50.1              | NS 11.11                       | TS 10.80                       | TS 10.79                       | TS 11.05                       | TS 2.56                        | TS 2.68                        |
| 10.0           | NS 0.910                | TS 0.342          | NS 61.5                | TS 48.5              | NS 10.08                       | TS 9.86                        | TS 9.58                        | TS 10.21                       | TS 2.42                        | TS 2.52                        |
Fig. 1: Densification of 0.7MgO-$x$Y$_2$O$_3$-(99.3-$x$)Al$_2$O$_3$ ceramics with variation of Y$_2$O$_3$ using NS and TS sintering

Fig. 2: Linear shrinkage of 0.7MgO-$x$Y$_2$O$_3$-(99.3-$x$)Al$_2$O$_3$ ceramics with variation of Y$_2$O$_3$ between NS and TS sintering
Fig. 3: X-ray diffraction patterns of 0.7MgO-Y2O3-(99.3-x)Al2O3 ceramics with variation of Y2O3 using NS sintering at 1600°C (T1) for 5 h.

Fig. 4: X-ray diffraction patterns of 0.7MgO-xY2O3-(99.3-x)Al2O3 ceramics with variation of Y2O3 using TS sintering at 1600°C (T1) for 30 min and T2 at 1450°C with a hold time of 10
The microstructure comparing between the two type sintered condition of 0.7MgO-\(x\)Y\(_2\)O\(_3\)-(99.3-\(x\))Al\(_2\)O\(_3\) with 6 wt% Y\(_2\)O\(_3\) added are shown in Fig. 5. Microstructural evaluation were observed, i.e., uniformly sized grains with well-packed and continuous grain structure in TS sintered ceramic and abnormal grain growth were appeared in NS sintered ceramic. The average grain size values using SEM technique were obtained in Table 1. It
can be seen that NS samples exhibited average grain size range of 0.760-1.344 μm and TS samples exhibited average grain size range of 0.342-0.566 μm. Figure 6 exhibits grain growth rather larger than the other the 0.7MgO-0.3Y2O3-(99.3-χ)Al2O3 ceramics with content of Y2O3 under TS sintering. This result indicates that the role of MgO and Y2O3 is to inhibit grain growth of Al2O3 ceramics, in agreement with other studies as Azhar et al. (2010; Galusek et al., 2012; Lukianova and Lukianova (2018). After performing a comparison of the grain sizes in 0.7MgO-xY2O3-(99.3-χ)Al2O3 ceramics after conventional sintering and grain sizes in 0.7MgO-xY2O3-(99.3-χ)Al2O3 ceramics after two stage sintering, it was found that the sizes of the grains in 0.7MgO-xY2O3-(99.3-χ)Al2O3 ceramics after two stage sintering were smaller than in ceramics from conventional sintering. This result agreed with previously published works (Wang et al., 2009; Galusek et al., 2012). The main reason for the application of two stage sintering is suppression of grain growth in the final stage sintering by application of the second, low temperature, heating step. Thus, the optimal content of Y2O3 is an important parameter for the development of ceramic microstructures.

Corresponding EDX analysis and chemical compositions for some of these 0.7MgO-xY2O3-(99.3-χ)Al2O3 ceramics under TS sintering are shown Table 2. It is seen that the Y concentration increases with increasing Y2O3. The mechanical properties of 0.7MgO-xY2O3-(99.3-χ)Al2O3 ceramics as a function of different Y2O3 concentrations with two sintering condition (NS and TS) is shows in Table 1. The micro hardness of all the compositions is higher than that of the Al2O3-MgO ceramics undoped with Y2O3. The optimal Y2O3 addition (4-6 wt%) inhibited grain growth in ceramics and gave rise to homogeneous and dense ceramics. TS samples were obtained higher micro hardness and toughness than NS samples. This observation is in agreement with the result of XRD, which are high percentage of MgAl2O4 phases in NS. This result indicate that the effect of MgAl2O4 phases on mechanical properties. Previous studies (Lu et al., 2005) showed that occurrence of MgAl2O4 phases caused a decrease in fracture toughness.

The highest value for fracture toughness was obtained from 0.7MgO-xY2O3-(99.3-χ)Al2O3 ceramics with 6 wt% Y2O3 added from TS samples, which corresponds to the high dense ceramics and optimal microstructure.

**Conclusion**

A two stage sintering process was performed to evaluate the influence of the addition of different ratios of Y2O3 on densification, phase analysis and mechanical properties of 0.7MgO-xY2O3-(99.3-χ)Al2O3 ceramics as a function of different Y2O3 additions. The key parameter that controlled the grain growth here would be attributed to the addition of Y2O3. 0.7MgO-0.6Y2O3-93.3Al2O3 ceramics with average grain size of 0.46 μm shows the highest fracture toughness of 3.24 MPa.m1/2. The relationship between the microstructure and mechanical property is discussed.

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**Author’s Contributions**

Aurawan Rittidech: Designed the work, analysis and interpretation of data, contributed to the writing of manuscript.

Pimpan Wisuwan: Participated in experiments, data correction.

Thitima Pinkhunthod: Collected and interpolation XRD data.

**Ethics**

This article is original and contains unpublished material. The corresponding author confirms that all of the other authors have read and approved the manuscript.

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