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Effect of Y_2O_3 Additions on Microstructure and Properties of Alumina–Magnesia Ceramics

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Abstract: The purposes of this research are to prepare and investigate the characterization of $0.7MgO\text{-}xY_2O_3\text{-}(99.3\text{-}x)Al_2O_3$ 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_2O_3, $MgAl_2O_4$, MgO and Y_2O_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_2O_3 additions between 4-6 wt%.

Keywords: Al_2O_3, MgO, The Solid State Reaction, Two-Stage Sintering

Introduction

Alumina (Al_2O_3) ceramics have high hardness, good wear resistance and high temperature stability. Rao *et al*. (2003) revealed that Al_2O_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_2O_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_2O_3, Magnesium Oxide (MgO) has an important effect on mechanical and electrical properties of bulk Al_2O_3. Lu *et al*. (2005) has shown that MgO is a traditional additive to Al_2O_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_2O_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_2O_3-based ceramics depend strongly on the microstructure as well as composition that reported by Rittidech *et al*. (2013). The microstructure of Al_2O_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_2O_3 additives.

Materials and Methods

Powders with 0.7MgO-$x$$Y_2O_3$-(99.3-$x$)$Al_2O_3$ where x = 0.0, 2.0, 4.0, 6.0, 8.0 and 10.0 were prepared from MgO, Y_2O_3 and Al_2O_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 electronic furnaces and sieving with 120 mesh, the resulting powders were calcined at 1200°C, with dwell times 2 h and heating/cooling rates of 10°C/min. The powders were then pressed at 3MPa into form pellets having 1.5 cm diameter using hydraulic press and were sintered in alumina crucible at a temperature of 1600°C for 5 h with heating rate of 10°C/min (NS sintering). The sintering experiments were carried out in an electrical furnace (Nabertherm, Germany). For the two stage sintering, samples are sintered in furnace with heating rate of 10°C/min to desired temperature (T_1) at 1600°C with a hold time of 30 min and then cooling down to T_2 at 1450°C with a hold time of 10 h (TS sintering). The bulk densities of sintered sample were calculated using Archimedes' method. The crystallized phase of the ceramic samples was measured by X-Ray Diffraction (XRD) using Cuk$_\alpha$ radiation (Philips PW 1729 diffractometer, Netherlands). The crystalline size of the ceramics was determined 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$$Y_2O_3$-(99.3-$x$)$Al_2O_3$ where x = 0.0, 2.0, 4.0, 6.0, 8.0 and 10.0 under TS sintering and NS sintering. It is observed that a density of between 3.31 and 3.73 g/cm^3. The maximum density, under 2 type sintering, were obtained in the samples of 0.7MgO-$x$$Y_2O_3$-(99.3-$x$)$Al_2O_3$ ceramics with 6 wt% Y_2O_3 added. It can be found that TS samples show higher densification than NS samples. Densities tend to increase with increasing concentrations of Y_2O_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_2O_3 contents, corresponding to densities.

The XRD patterns of 0.7MgO-$x$$Y_2O_3$-(99.3-$x$)$Al_2O_3$ ceramics with added 0.0-10.0 wt% Y_2O_3 after sintering with NS sintering and TS sintering condition are shown in Fig. 3 and 4, which presents the difference of content phases. Apart from alpha (α)-Al_2O_3, MgO, $MgAl_2O_4$ and Y_2O_3 are identified. The XRD patterns for CS samples showed a similar trend compared to TS samples. However, the addition of MgO to Al_2O_3 can reduce grain size but more MgO additive led to the formation of $MgAl_2O_4$ phase confirmed with Rejab *et al*. (2014). The XRD patterns in NS samples and TS samples were observed $MgAl_2O_4$ phases tend to increase with increasing Y_2O_3 contents.

It was observed that higher $MgAl_2O_4$ phases content in NS samples than TS samples caused from the high sintering temperature for long dwell time. In addition, the crystalline size of the samples calculated from the XRD patterns is summarized in Table 1. The crystalline sizes of NS samples are in the range 61.2-79.6 nm, while TS samples are in the range 48.3-63.3 nm.

Table 1: Average grain size and mechanical properties of 0.7MgO-$x$$Y_2O_3$-(99.3-$x$)$Al_2O_3$ ceramics with variation of Y_2O_3 using different sintering between NS sintering and TS sintering

Content of x	Average grain size (μm)		Crystal size (nm)		Vickers hardness (MPa)		Knoop Hardness (MPa)		Fracture toughness (MPa.m$^{1/2}$)	
	NS	TS	NS	TS	NS	TS	NS	TS	NS	TS
0.0	1.344	0.566	79.6	63.3	7.14	8.11	7.14	8.92	2.16	2.27
2.0	1.089	0.532	68.4	61.7	10.28	11.84	10.81	11.22	2.48	2.55
4.0	1.004	0.488	67.5	53.8	13.47	11.57	10.12	12.18	2.84	2.79
6.0	0.760	0.466	61.2	50.4	11.85	12.76	12.09	14.29	2.77	3.24
8.0	0.825	0.477	62.4	50.1	11.11	10.80	10.79	11.05	2.56	2.68
10.0	0.910	0.342	61.5	48.5	10.08	9.86	9.58	10.21	2.42	2.52

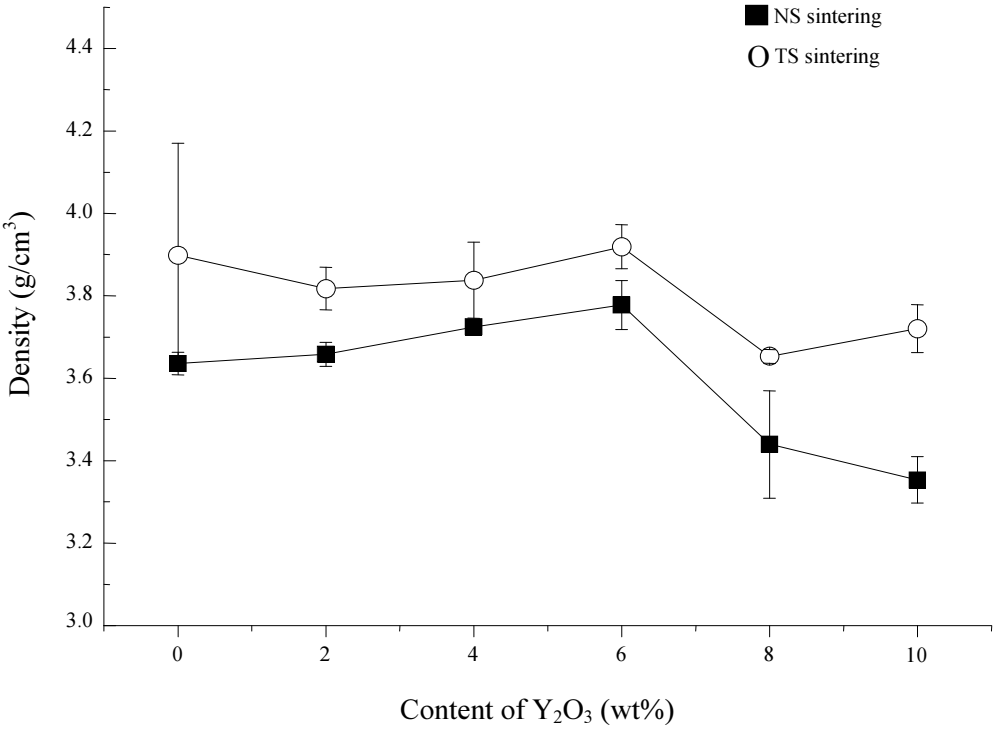

Fig. 1: Densification of 0.7MgO-$x$$Y_2O_3$-(99.3-$x$)$Al_2O_3$ ceramics with variation of Y_2O_3 using NS and TS sintering

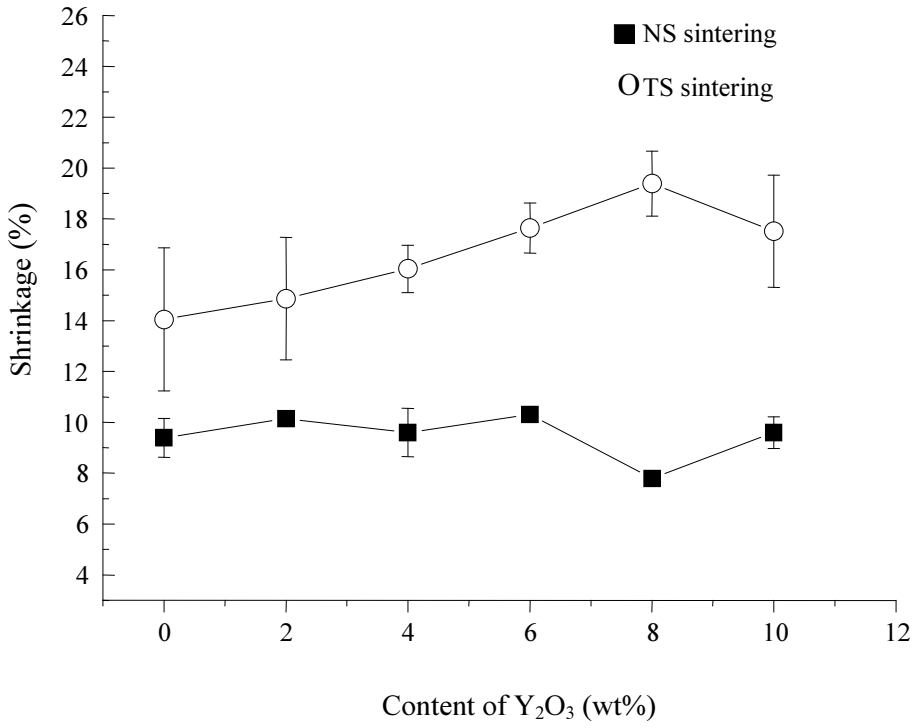

Fig. 2: Linear shrinkage of 0.7MgO-$x$$Y_2O_3$-(99.3-$x$)$Al_2O_3$ ceramics with variation of Y_2O_3 between NS and TS sintering

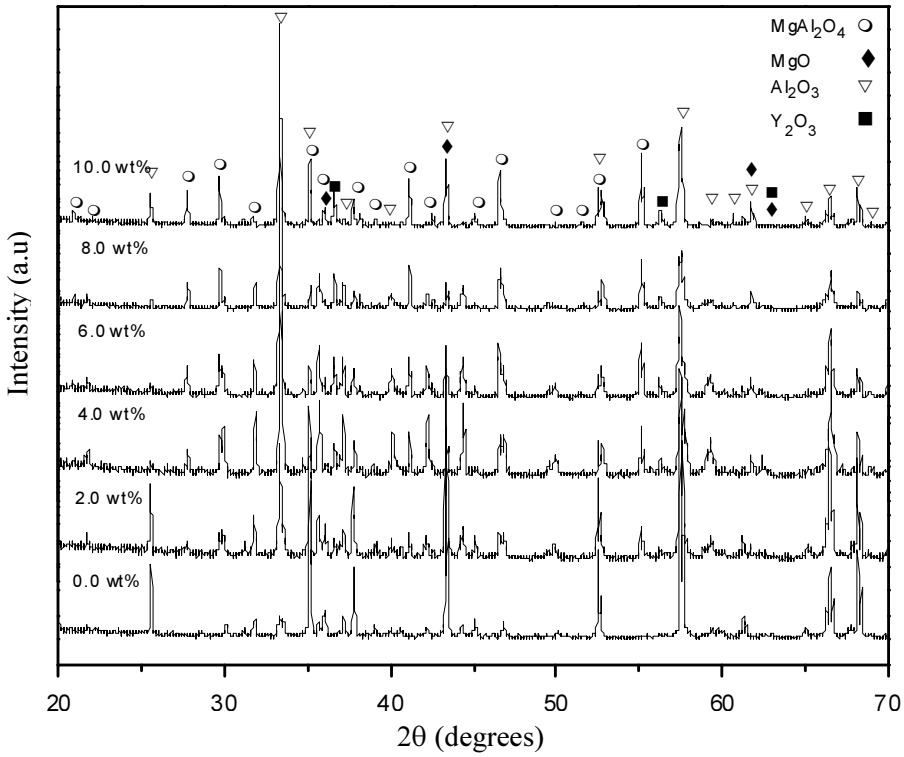

Fig. 3: X-ray diffraction patterns of 0.7MgO-$x$$Y_2O_3$-(99.3-$x$)$Al_2O_3$ ceramics with variation of Y_2O_3 using NS sintering at 1600°C (T_1) for 5 h

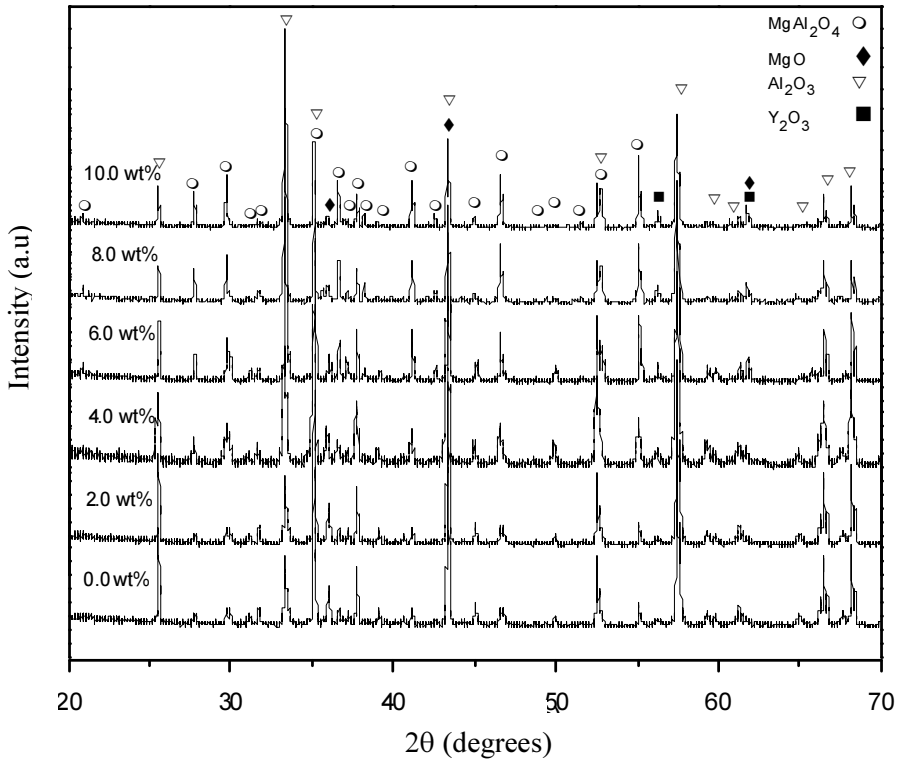

Fig. 4: X-ray diffraction patterns of 0.7MgO-$x$$Y_2O_3$-(99.3-$x$)$Al_2O_3$ eramics with variation of Y_2O_3 using TS sintering at 1600°C (T_1) for 30 min and T_2 at 1450°C with a hold time of 10

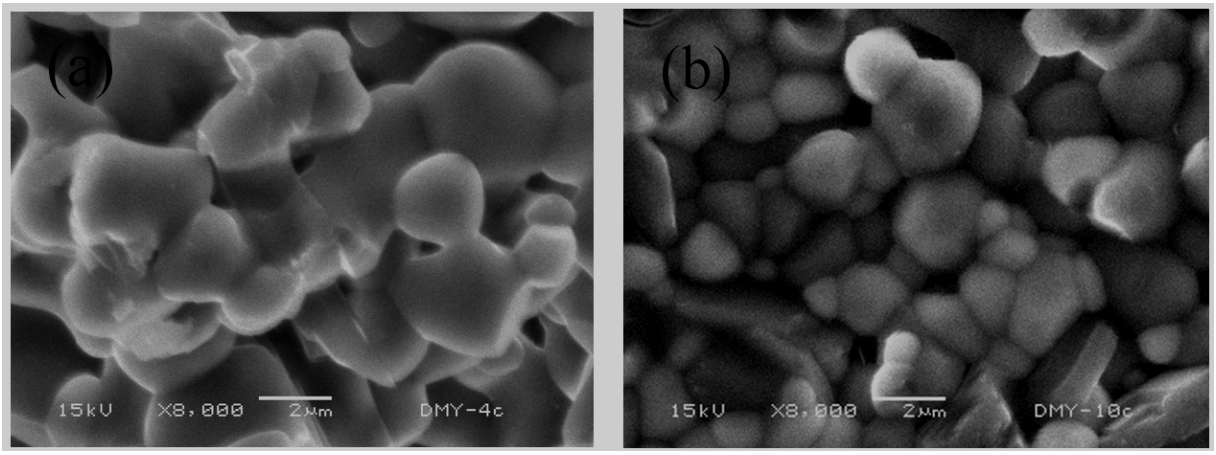

Fig. 5: SEM micrograph of (0.7MgO-$x$$Y_2O_3$-(99.3-$x$)$Al_2O_3$ ceramics with x= 6 wt% using different sintering (a) NS sintering (b) TS sintering

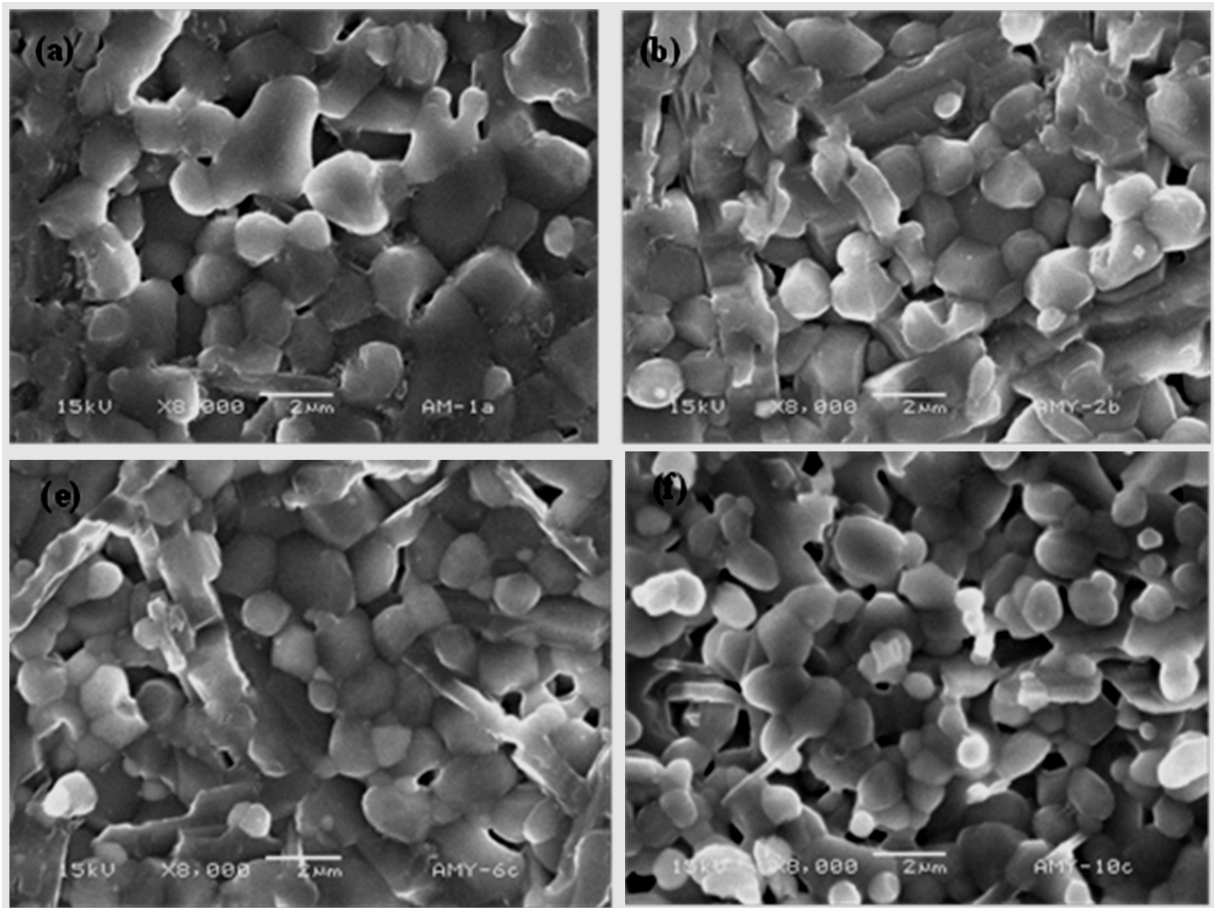

Fig. 6: SEM micrograph of 0.7MgO-$x$$Y_2O_3$-(99.3-$x$)$Al_2O_3$ ceramics with variation of Y_2O_3 using TS sintering (a) 0.0 wt% Y_2O_3 (b) 2.0 wt% Y_2O_3 (e) 8.0 wt% Y_2O_3 (f) 10.0 wt% Y_2O_3

The microstructure comparing between the two type sintered condition of 0.7MgO-$x$$Y_2O_3$-(99.3-$x$)$Al_2O_3$ with 6 wt% Y_2O_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 $0.7MgO\text{-}xY_2O_3\text{-}(99.3\text{-}x)Al_2O_3$ ceramics with content of Y_2O_3 under TS sintering. This result indicates that the role of MgO and Y_2O_3 is to inhibit grain growth of Al_2O_3 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 $0.7MgO\text{-}xY_2O_3\text{-}(99.3\text{-}x)Al_2O_3$ ceramics after conventional sintering and grain sizes in $0.7MgO\text{-}xY_2O_3\text{-}(99.3\text{-}x)Al_2O_3$ ceramics after two stage sintering, it was found that the sizes of the grains in $0.7MgO\text{-}xY_2O_3\text{-}(99.3\text{-}x)Al_2O_3$ 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 Y_2O_3 is an important parameter for the development of ceramic microstructures.

Corresponding EDX analysis and chemical compositions for some of these $0.7MgO\text{-}xY_2O_3\text{-}(99.3\text{-}x)Al_2O_3$ ceramics under TS sintering are shown Table 2. It is seen that the Y concentration increases with increasing Y_2O_3. The mechanical properties of $0.7MgO\text{-}xY_2O_3\text{-}(99.3\text{-}x)Al_2O_3$ ceramics as a function of different Y_2O_3 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 Al_2O_3-MgO ceramics undoped with Y_2O_3. The optimal Y_2O_3 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 $MgAl_2O_4$ phases in NS. This result indicate that the effect of $MgAl_2O_4$ phases on mechanical properties. Previous studies (Lu *et al*., 2005) showed that occurrence of $MgAl_2O_4$ phases caused a decrease in fracture toughness.

Table 2: Chemical compositions of $0.7MgO\text{-}xY_2O_3\text{-}(99.3\text{-}x)Al_2O_3$

Contents of Y_2O_3 (wt%)	Composition (at.%)			
	Al(K)	Mg(K)	Y(K)	O(K)
2.0	36.02	6.55	2.48	55.44
4.0	38.58	7.01	3.62	52.69
6.0	31.15	6.87	5.58	54.86
8.0	31.81	7.54	6.15	56.50
10.0	32.50	7.21	8.97	51.88

The highest value for fracture toughness was obtained from $0.7MgO\text{-}xY_2O_3\text{-}(99.3\text{-}x)Al_2O_3$ ceramics with 6 wt% Y_2O_3 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 Y_2O_3 on densification, phase analysis and mechanical properties of $0.7MgO\text{-}xY_2O_3\text{-}(99.3\text{-}x)Al_2O_3$ ceramics as a function of different Y_2O_3 additions. The key parameter that controlled the grain growth here would be attributed to the addition of Y_2O_3. $0.7MgO\text{-}6.0Y_2O_3\text{-}93.3Al_2O_3$ ceramics with average grain size of 0.46 μm shows the highest fracture toughness of 3.24 $MPa.m^{1/2}$. 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 interpitation 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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