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Influence of Pentavalent Niobium on Microstructural and Mechanical Properties of ZTA Ceramic Composite via Cold Isostatic Pressing

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ABSTRACT. The influence of pentavalent niobium (Nb_2O_5) on microstructural and mechanical properties of ZTA ceramic composite via cold isostatic pressing (CIP) method was investigated. The microstructural analysis and phase transformation was determined by scanning electron microscopy (SEM) and X-ray diffraction (XRD). XRD analysis revealed the presence of t-ZrO_2, m-ZrO_2 and $Nb_2Zr_6O_{17}$ phase. The maximum value of hardness, fracture toughness and flexural strength was found at 1 wt.% of Nb_2O_5 addition with the value of 1840 HV, 8.6 MPa.√m and 351 MPa, respectively. Thus, the addition of Nb_2O_5 results in an enhancement for the mechanical properties of ZTA ceramic.

Keywords: *Hardness, Fracture toughness, Flexural strength, Cold isostatic pressing (CIP);*

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1. INTRODUCTION

Zirconia toughened alumina (ZTA) is a zirconia-alumina system where the toughness of alumina is enhanced by adding zirconia [1]. Zirconia undergo transformation toughening which lead to an improvement on mechanical properties of the composites [2]. Pentavalent niobium (Nb_2O_5) is found as one potential additive that can improve the mechanical properties of ceramics [3-5]. Previous work done by Hassan et al. [3] found that Nb_2O_5 improved the mechanical properties of ZTA ceramic by sol gel method using uniaxial press as compaction method. For this work, cold isostatic pressing (CIP) method was chosen as compaction method in producing the homogeneity of powder compact [6,7]. CIP can improved the mechanical properties through lowering the porosity of ceramic compared to uniaxial press method [8-10]. Therefore, this study focuses on the role of CIP as compaction method on the microstructural and mechanical properties such as hardness, fracture toughness and flexural strength behaviour for ZTA added Nb_2O_5 ceramics.

2. MATERIALS AND METHODS

For this work, 80 wt.% of Al_2O_3 (Alcoa, A16SG,99.0% purity) and 20 wt.% YSZ (Goodfellow, 94.6% purity) were used as starting materials. The amount of Nb_2O_5 (Merck, 99.5% purity) in weight percent was varied from 0 wt.% to 7 wt.% into the initial compositions by wet milling method. Then, the mixtures were dried to

100 ºC and grinded to form powders. The mixed powders were compacted by uniaxial press at 15 MPa and followed by cold isostatic pressing (CIP) at 150 MPa for 2 min to form a cylindrical shapes with dimensions of 13 mm in diameter and 4 mm in height (for physical and microstructural characterization) and rectangular bars of dimensions of 40 × 10 × 10 mm (for mechanical evaluation). The green body was sintered in atmosphere at 1600 ºC for 4 hours. The phase transformations of sintered body were analyzed by X-ray diffraction (XRD). Scanning electron microscopy (SEM) was used to observe the microstructure of each sample. The values of hardness and fracture toughness were measured by a Vickers indentation technique tester using a 30 kgf load. The fracture toughness values were calculated for all samples using the Palmqvist crack formula proposed by Niihara (1983)[11]:

$$3K_{Ic} = 0.035\,(H^* a^{1/2})\,(3E/H)^{0.4}\,(c/a)^{-0.5} \quad (1)$$

K_{Ic} is the fracture toughness, H is Vickers hardness, a is the half length of Vickers diagonal (μm), E is equal to the Young modulus of the samples and c is the length of the radial crack size (μm). Flexural strength was measured in a three-point bending test on a universal testing machine at a crosshead speed of 1 mm/min, and support distance of 20 mm.

3. RESULTS AND DISCUSSION

Table 1 shows the XRD analysis of the samples for different composition of Nb_2O_5 additions (0 - 7 wt.%). The phase existed in ZTA-Nb_2O_5 consist of alumina (Al_2O_3), tetragonal zirconia (t-ZrO_2), monoclinic zirconia (m-ZrO_2) and $Nb_2Zr_6O_{17}$ phase. Al_2O_3 are present as corundum matching ICSD pattern No. 98-005-7635 with hexagonal structure. Tetragonal zirconia and monoclinic zirconia was observed for all composition matching ICSD pattern No. 98-005-7635 and No. 98-007-1860. The XRD analysis also revealed the formation of secondary phase, $Nb_2Zr_6O_{17}$ (ICSD pattern No. 98-000-5068) starting at 3.0 wt.% Nb_2O_5 with orthorhombic structure. These identification phase's percentages correspond to the addition of Nb_2O_5. The lower addition of Nb_2O_5 (0 - 1 wt.%) related to the effect of t-ZrO_2 and m-ZrO_2 phase, which is attributed to the toughening mechanism (t → m). These mechanism are one of the factors to improve the toughness in ZTA-Nb_2O_5 composite [12]. Meanwhile, the higher Nb_2O_5 addition (3 - 7 wt.%) revealed the formation of $Nb_2Zr_6O_{17}$ phase. It was recorded that the highest formation of $Nb_2Zr_6O_{17}$ is 0.9 % at 7 wt.% Nb_2O_5 addition. This was proved by Energy dispersive X-ray analysis (EDX) in Fig. 2 with the presence of Nb element at YSZ grains.

Table 1 XRD analysis on phase composition for ZTA-Nb_2O_5

Nb_2O_5 addition (wt.%)	Phase Composition (%)			
	Al_2O_3	t-ZrO_2	m-ZrO_2	$Nb_2Zr_6O_{17}$
0	80.4	18.4	1.1	0
0.1	80.7	17.7	1.6	0
0.3	77.0	17.1	5.9	0
0.5	77.2	16.4	6.3	0
0.7	75.3	13.1	11.6	0
1	68.6	13.9	17.5	0
3	69.8	15.8	14.3	0.1
5	77.2	16.9	5.1	0.7
7	78.5	17.5	3.0	0.9

SEM micrographs for sintered samples of 0 wt.% and 7 wt.% of ZTA-Nb_2O_5 are shown in Fig. 1. Based on the microstructures, the grains for YSZ and Al_2O_3 are well distributed among each other. EDX analysis in Fig. 2 shows that the dark grains are Al_2O_3 and light grains are yttria stabilized zirconia (YSZ). The presence of Nb element was detected in YSZ grains, confirmed by the EDX analysis (Fig. 2). Meanwhile, $Nb_2Zr_6O_{17}$ phase found to be appeared at the YSZ grains when its amount exceeds the solubility limit.

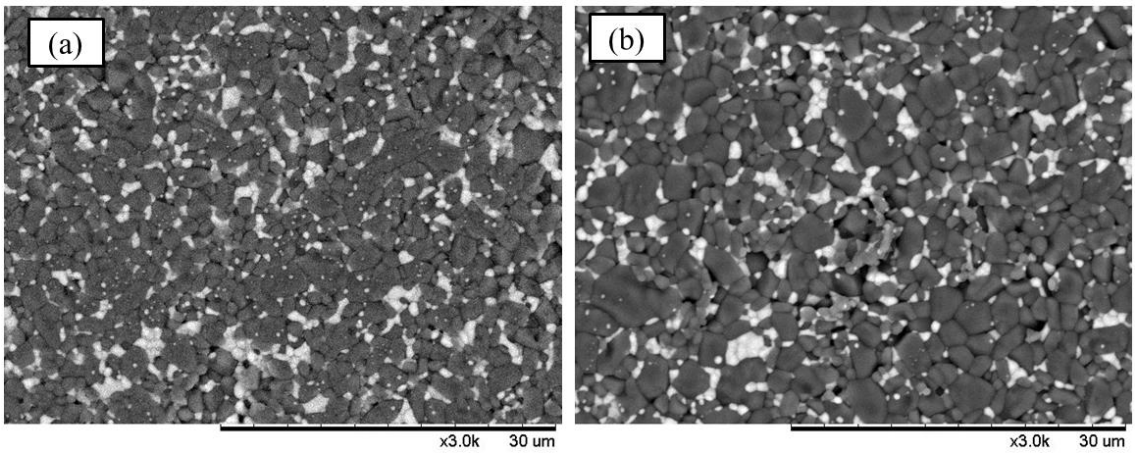

Fig. 1 SEM micrograph of ZTA- Nb_2O_5 composition (a) 0 wt.% and (b) 7.0 wt.%, at 3.0 K magnification

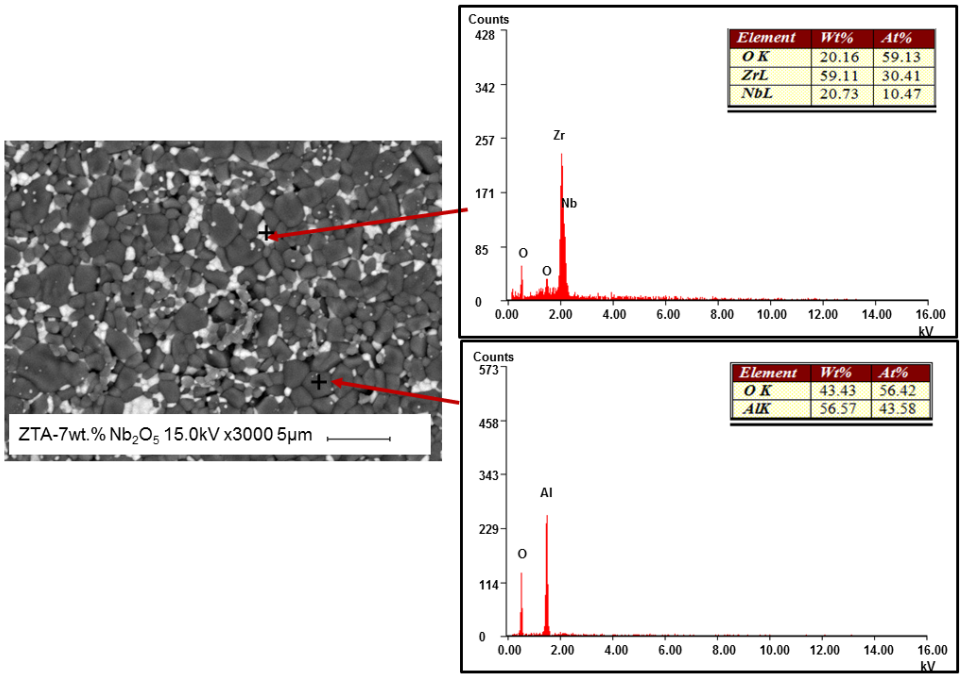

Fig. 2 Microstructure and EDX analysis for ZTA with 7 wt.% of Nb_2O_5 addition

Fig. 3 presents the results of hardness, fracture toughness and flexural strength of ZTA-Nb_2O_5 composites as a function of Nb_2O_5 content. As seen, the hardness, fracture toughness and flexural strength increased with increasing Nb_2O_5 content. The addition of 1 wt.% Nb_2O_5 gives the maximum value for hardness (1840 HV), fracture toughness (8.6 MPa.√m) and flexural strength (351 MPa). The dense compact through CIP enhanced the mechanical properties of this system, as the relative density achieved 97.4%-99.8% to the theoretical

density, where sample for 1 wt.% Nb_2O_5 gives the highest relative density (99.8%). CIP improved the particle packing and reorganized the microstructure, resulting in higher densification for sintered body [13]. Furthermore, the improvement of fracture toughness also can be related to the transformation toughening (t → m) due to crack deflection and crack bridging that occurs at the crack path after indentation. However, the properties gradually decreased when reached at 3 wt.% of Nb_2O_5 addition. This phenomenon was related with the presence of secondary phase, $Nb_2Zr_6O_{17}$. A secondary phase can promotes pore formation which might weaken the ceramics by reducing some of the mechanical properties. Pores were described as a discontinuity in the material, resulting in an undesirable effect [14]. This was confirmed by the results of porosity in Table 2, where the porosity found to increase at 3 wt.% of Nb_2O_5 addition.

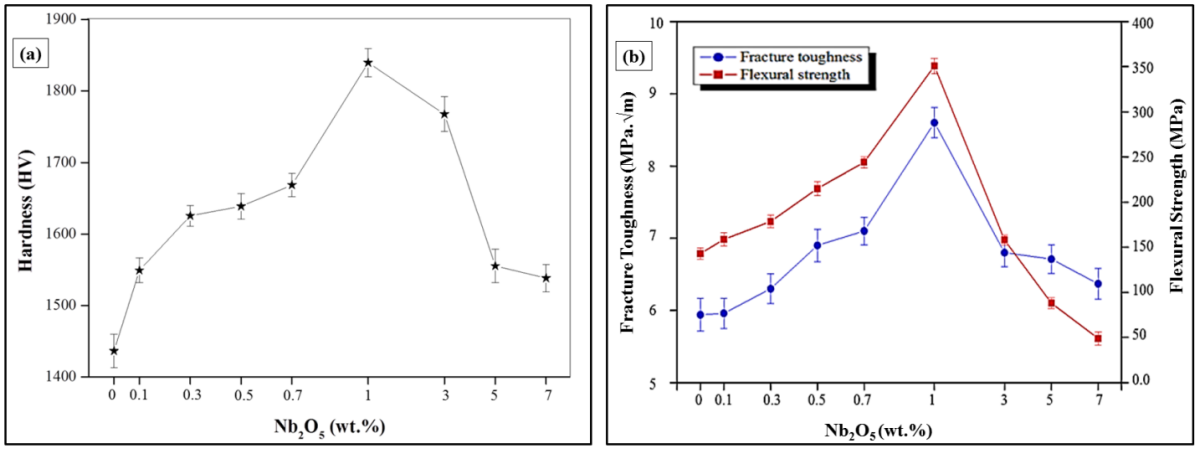

Fig. 3 Results of (a) hardness and (b) fracture toughness and flexural strength of ZTA-Nb_2O_5

Fig. 4 indicates the results of bulk density and theoretical density of ZTA samples with different addition of Nb_2O_5. The result of bulk density follows the pattern of theoretical density. This result clearly indicates that Nb_2O_5 enhances sinterability and the density of ceramics. However, at 3 wt.% the densities decreased due to excess of Nb_2O_5 beyond the solubility limit, which results in the presence of $Nb_2Zr_6O_{17}$ phase (Table 1). The results show that by increasing the Nb_2O_5 addition, the bulk density increased and the percentage of porosity decreased as illustrated in Table 2. Lower percentage of porosity enhances the mechanical properties of the ZTA ceramic. The highest density (4.6 g/cm^3) with low percentage of porosity (0.08%), was found at 1 wt.% of Nb_2O_5 addition. Hassan et al. [3] stated that the ionic radius of Nb^{5+} (0.68 Å) is smaller compared to Zr^{4+} (0.79 Å) and Y^{3+} (1.019 Å). When Nb^{5+} enter into the Zr^{4+}, it will caused a large amount of oxygen vacancies formed in the samples and accelerate the lattice diffusion, thus enhance the densification rate [15]. Furthermore, it also led to the formation of the observed new phase, confirmed by XRD result, which indicated in Table 1.

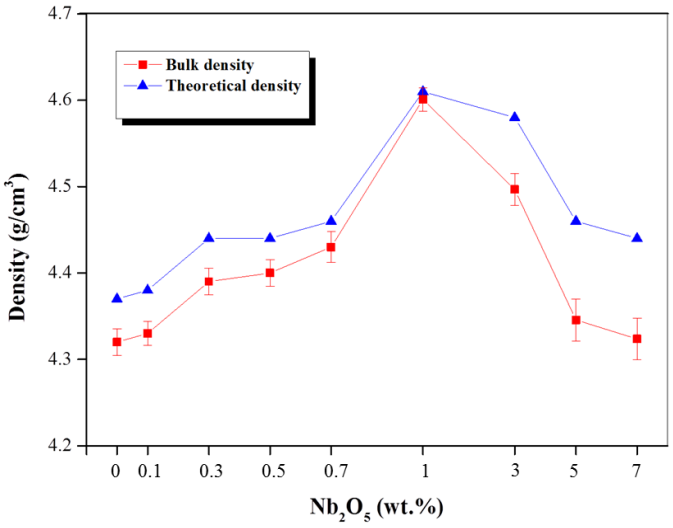

Fig. 4 Result of bulk density and theoretical density of ZTA-Nb_2O_5

Table 2 Result for porosity of ZTA-Nb_2O_5

Nb_2O_5 addition (wt.%)	Porosity (%)
0	0.79
0.1	0.23
0.3	0.24
0.5	0.24
0.7	0.09
1	0.08
3	0.33
5	0.71
7	2.23

4. SUMMARY

The influence of pentavalent niobium (Nb_2O_5) on microstructural and mechanical properties of ZTA via CIP method was successfully investigated. The optimum composition are 1 wt.% Nb_2O_5 addition. The addition of Nb_2O_5 enhanced the hardness, fracture toughness and flexural strength compared to ZTA without Nb_2O_5 addition. The enhancements reach a limit at 1 wt.% addition, which is attribute to the transformation of t → m phase. These proved that the toughening mechanism occurs for 1 wt.% addition. However, the effectiveness of these toughening mechanism deteriorate the mechanical properties with further additions (3 - 7 wt.%) which is corresponds to an existence of $Nb_2Zr_6O_{17}$ phase.

REFERENCES

[1] N.A. Rejab, A.Z.A. Azhar, M.M. Ratnam, Z.A. Ahmad, The effects of CeO_2 addition on the physical, microstructural and mechanical properties of yttria stabilized zirconia toughened alumina (ZTA), Int. J. Refract. Met. Hard Mater., 36 (2013) 162-166.

[2] B. Basu, Toughening of Yttria-stabilised tetragonal zirconia ceramics, Int. Mater. Rev., 50 (2005) 239-256.

[3] A.M. Hassan, S.M. Naga, M.Awaad, Toughening and strengthening of Nb_2O_5 doped zirconia / alumina (ZTA) composites., Int. J. Refract. Met. Hard Mater., 48 (2015) 338-345.

[4] Y.F. Hsu, Influence of Nb_2O_5 additive on the densification and microstructural evolution of fine alumina powders, Mater. Sci. Eng. A, 399 (2005) 232-237.

[5] J.E.M. Lopes, M.M. Oliveira, G.S. Costa, G.P. De Figueredo, J.S. Vasconcelos, J.H.G. Rangel, Chemical synthesis and characterization of Nb-ZrO_2, Ceram. Int., 42 (2016) 861-873.

[6] A. Shui, A. Makiya, S. Tanaka, N. Uchida, K. Uematsu, Effect of cold isostatic pressing on microstructure and shrinkge anisotropy during sintering of uniaxially pressed alumina compacts, J. Ceram. Soc. Jpn., 110 (2002) 264-269.

[7] E. Djurado, F. Boulc'h, A. Pivkina, Y. Frolov, N. van Landschoot, J. Schoonman, Cold isostatic and explosive isodynamic compaction of Y-TZP nanoparticles, Solid State Ionics, 154-155 (2002) 375-380.

[8] G. Bajpai, R. Purohit, R.S. Rana, Development of Al-Nano composites through powder metallurgy process using a newly designed cold isostatic compaction chamber, Mater. Today Proc., 2 (2015) 2737-2746.

[9] G.Y. Akimov, I.Y. Prokhorov, I. V. Gorelik, V.M. Timchenko, V.G. Vereshchak, A.D. Vasil'ev, Role of cold isostatic pressing in the formation of the properties of zro_2-base ceramics obtained from ultradisperse powders, Refractories, 36 (1995) 47-53.

[10] N.K. Su, M. Fadhlullah, A.A. Razak, B. Johar, M.A. Idris, Z.A. Ahmad, The role of $Nb_2Zr_6O_{17}$ phase on the hardness and fracture toughness of ZTA/Nb_2O_5 by cold isostatic pressing, Mater. Sci. Forum, 888 (2017) 126-130.

[11] K. Niihara, A fracture mechanics analysis of indentation-induced Palmqvist crack in ceramics, J. Mater. Sci. Lett., 2 (1983) 221-223.

[12] F. Zhang, L. Li, E. Wang, Effect of micro-alumina content on mechanical properties of Al_2O_3/3Y-TZP composites, Ceram. Int., 41 (2015) 12417-12425.

[13] D. Galusek, P. Znášik, J. Majling, Influence of cold isostatic pressing on compaction and properties of Mg-PSZ ceramics, J. Mater. Sci. Lett., 18 (1999) 1347-1351.

[14] Dr.G.Viola, Mechanical Properties of Ceramics, Springer Science + Business Media, LLC, Switzerland, (2011).

[15] L. Hamzioui, F. Kahoul, A. Boutarfaia, The effect of Nb_2O_5 addition on the structural, dielectric and piezoelectric properties of $Pb_{0,98}$ $Ba_{0,02}$ $[(Zr_{0.52}Ti_{0.48})_{0,98}(Cr^{3+}{}_{0.5}, Ta^{5+}{}_{0.5})_{0,02}]$ ceramics, Energy Procedia. 74 (2015) 198-204.