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Thermal Properties and Microstructural Characterization of Aluminium Titanate (Al_2TiO_5) / La_2O_3 -Stabilized Zirconia (ZrO_2) Ceramics

Tahsin BOYRAZ

Cumhuriyet University, Department of Metallurgical and Materials Engineering, Sivas, TURKEY

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Abstract: In this paper defined the productions and densification behaviour of 8 mole% Lanthana (Lanthanum oxide, La_2O_3) -stabilized Zirconia (Zirconium Oxide, ZrO_2) based composites fabricated by the conventional ceramic production process. La_2O_3 stabilized zirconia (LSZ, with mole 8% La_2O_3) has excellent high temperature properties like low thermal conductivity, mechanical properties, elevated melting point, high toleration for thermal shock, superior oxidation resistance and well phase stability. Aluminium titanate (Al_2TiO_5) low thermal conductivity coupled with well chemical resistance in melted metals and exhibit extremely well thermal shock resistance. In this study, Aluminium titanate / LSZ ceramic with different proportions of Al_2TiO_5 was prepared by powder metallurgy techniques. The mechanical, thermal and microstructural properties were characterized by XRD, SEM, hardness and dilatometer. The thermal shock resistance performance under water quenching of the as-prepared ceramics was also estimated. As a results shown that the adding of aluminium titanate to LSZ matrix improves the properties of the aluminium titanate / LSZ ceramics.

***Keywords*:** Lanthanum oxide, stabilized zirconia, Aluminium Titanate, Characterization, Hardness, Thermal Properties, Sintering

Alüminyum Titanat/La_2O_3 -Stabilize Edilmiş Zirkonya Seramiklerin Termal Özellikleri ve Mikroyapısal Karakterizasyonu

Özet: Bu yazıda, geleneksel seramik üretim yöntemleri ile imal edilen% 8 mol Lanthana (Lantanyum oksit, La_2O_3) ile stabilize edilmiş Zirkonya (Zirkonyum Oksit, ZrO_2) esaslı kompozitlerin üretim ve yoğunlaşma davranışları anlatılmaktadır. , La_2O_3 ile Stabilize edilmiş (dengelenmiş) zirkonyum dioksit (LSZ, mol% 8 La_2O_3 ile) termal darbeye karşı yüksek tolerans, düşük termal iletkenlik, mekanik özellikler, yüksek erime noktası, iyi bir faz dengesi ve mükemmel oksidasyon direnci gibi yüksek üstün sıcaklık özelliklerine sahiptir. Alüminyum titanat (Al_2Ti0_5), erimiş metallerde iyi kimyasal dirençle birlikte, termal şok direnci ve düşük termal iletkenliğe sahiptir. Bu çalışmada, farklı yüzdelerdeki Al_2TiO_5 ile alüminyum titanat / LSZ seramikleri toz metalurjisi teknikleri kullanılarak hazırlandı. Mikroyapısal, mekanik ve termal özellikler XRD, SEM, dilatometre ve sertlik ile karakterize edildi. Hazırlanan seramiklerin suda soğutulmasıyla termal şok direnci davranışı da değerlendirildi. Sonuçlar, alüminyum titanatın LSZ matrisine eklenmesinin alüminyum titanat / LSZ seramiklerinin özelliklerini geliştirdiğini ortaya koydu.

***Anahtar Kelimeler*:** Lantan oksit, stabilize zirkonya, alüminyum titanat, karakterizasyon, sertlik, termal özellikler, sinterleme

* Corresponding author. *Email address: tahsinboyraz@cumhuriyet.edu.tr*

1. INTRODUCTION

Pristine zirconia (ZrO_2) occurs in three forms: Monoclinic, tetragonal, and cubic. The monoclinic phase exists at the room temperature and the temperatures greater than 1170 ˚C it transforms to the tetragonal form. The tetragonal phase is constant at temperatures of 1170 - 2370 ºC, and the cubic phase occurs at over 2370 °C [1-7]. The transition from tetragonal form to monoclinic phase (about 9%) undergoes large volume change in microstructure which makes difficult to obtain stable sintered zirconia ceramic products. Slightly higher ions e.g., those of La^{3+} (ionic radius of 1,15 Å) replace the lesser ions like some of the Zr^{4+} ions in the crystal matrix provides a stabilization of polymorphic tetragonal zirconia over a varied range of temperatures. Obtained the doped zirconia powders are identified as stabilized zirconia [3]. In recent years, great interest has been concentrated understanding of the superior electrical, chemical and mechanical properties such as thermal, fracture toughness and strength like high tolerance for thermal shock and low thermal conductivity of Lanthana stabilized zirconia [8-14]. Thermal expansion coefficients of ZrO_2 and La_2O_3 are 10.5 x 10^{-6} and 8.6 × 10−6 K^{-1}, respectively [9]. LaSZ ceramics have very high mechanical resistance [13].

As a synthetic ceramic, Aluminum titanate (Al_2TiO_5) is the most widely used as structural materials due to their low thermal conductivity, good thermal shock resistance and great melting point. Even though aluminium titanate is a popular material, it has specific disadvantages such as inter granular micro cracking, which exhibits due to the great thermal anisotropy of singular particles, that greatly limits the mechanical properties of polycrystalline Al_2TiO_5 [3]. Aluminium titanate (Al_2TiO_5) is usually prepared by several procedures. It is obtained by the solid-state reaction between Al_2O_3 and TiO_2, above the eutectoid temperature 1280°C and exhibits low thermal expansion coefficient (from 0.2 x 10^{-6} to 1.0x10^{-6}) low thermal conductivity (0.9 x 1.5 Wm^{-1} K^{-1}), and an perfect thermal shock resistance. These properties make Al_2TiO_5 a good choice for high-temperature applications such as exhaust port liners, valves, turbochargers, piston buttons, manifold insulations, swirl chambers and other internal combustion engines components. Moreover, aluminium titanate ceramics are suitable materials in many other crucial applications such as in burner nozzles, thermocouple sleeves and refractory crucibles for metal casting and flow regulations [15-25].

In the present study, an introduction of different percentages of Al_2TiO_5 to LSZ ceramics were undertaken. Physical, microstructural, thermal and mechanical characterization investigations on resultant ceramics were carried out.

2. MATERIALS and METHODS

Appropriate amount of the two singular oxides (La_2O_3 which is an additive material for monoclinic ZrO_2 (Merck, Germany) and submicron-sized ZrO_2 (Serp, France)) formulated in this study were prepared by solid state sintering method. Lanthana stabilized zirconia (LSZ, with mol 8% La_2O_3) and aluminium titanate (Al_2TiO_5) were prepared from ZrO_2 (Serp, France) and La_2O_3, Al_2O_3, TiO_2 (Merck, Germany) by the conventional ceramic powder processing technique. Lanthana and zirconia were estimated in relations of mole percentage of LSZ, with mole 8% La_2O_3 mixtures were scattered in acetone**.** After zirconia oxide balls of 10 mm were utilized for wet-milling of the precursor ceramics used in acetone for 2h, the powders were grounded. Then, Al_2O_3 and TiO_2 were also brought together using the similar wet-milling route to achieve Al_2TiO_5 ceramic materials. Al_2TiO_5 ceramics were manufactured using reaction sintering of an equimolar combination of Al_2O_3 and TiO_2 powders. Then powders were standardized using wet-ball-milling. The resulting mixture powders were sintered at 1550 ºC for lanthana-stabilized zirconia (LSZ) and 1600 ºC for AT (aluminium titanate (Al_2TiO_5)) in air for 2 hours. The process completed with the heating speed of 5 ºC /min longed for two consecutive hours.

Additions of AT (Aluminium Titanate) in mass amounts of 0, 10, 20 wt. % were prepared to the LSZAT powders (these mixtures are symbolized LSZAT0, LSZAT10 and LSZAT20). Each party was again wet-ball-milling mixed with ZrO_2 balls of 10 mm Ø for 2 h. Then, mixtures we pressed at 100 MPa into 23 mm dimension specimens. The ceramic powder compacts were sintered to 1600 °C with a soaking time of 120 min and a heating rate of 5 °C/min in conventional sintering processes. The densities of sintered samples were determined by the water-immersion technique using the Archimedes method. Density, water absorption and porosity of the sintered samples were measured. X-ray diffraction analysis was performed using CuKα radiation (40 kV and 40mA, Rigaku, Dmax, IIIC) to identify the presence of LSZ or Al_2TiO_5 related phases after the sintering process. Thermal expansion analysis was measurement by using dilatometer (USA, Anter). SEM (Scanning electron microscopy, Leo440) was used to investigate the microstructural characterization of the sintered ceramic samples. The morphological parameters of the different phases were characterized by using EDX (semiautomatic image analyser). The Vickers micro hardness measurements (Shimadzu Micro Hardness Tester HMV-2) were measured on the polished surface of the examples at room temperature. Minimum six particular tests with a peak load of 2000 g and a loading period of 15 s. were made for each set of ceramics.

3. RESULT and DISCUSSION

The physical analysis of different mixtures of the sintered LSZAT ceramics obtained in this study is presented in Table 1. In the literature [4,5], the theoretical density of the LSZ ceramics is 5.75 g.cm^{-3}, and that of the Al_2TiO_5 ceramics is 3.20 g.cm^{-3}, respectively (see Table 1). Moreover, the values of porosity and water absorption for the samples were presented in Table 1 which clearly indicate that an increase in Al_2TiO_5 content between 0 and 20 wt % as well as sintering temperature increases porosity and water absorption of the resultant samples.

Table 1. Physical properties of sintered samples.

	ρ_{bulk}, gr/cm^3	ρ_{true}, gr/cm^3	$\rho_{relative}$, %	Porosity %	w.abs. %
		1600 ºC			
LSZAT0	5,55	5,75	96,6	4,118	0,742
LSZAT10	4,83	5,56	87,02	6,505	1,346
LSZAT20	4,59	5,36	85,62	8,415	1,832

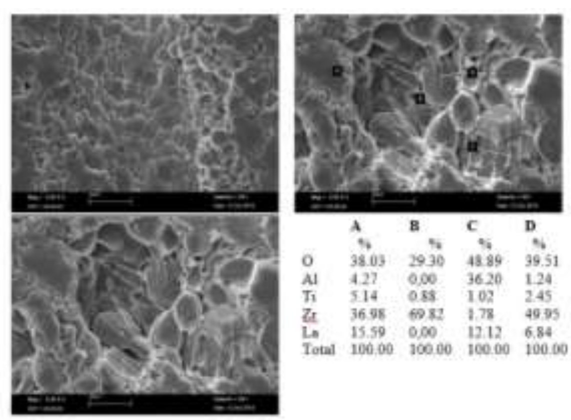

Figure 1. SEM images of LSZAT0, LSZAT20 and EDX images analyses of LSZAT20 samples.

SEM surface view images of the sintered LSZAT0 and LSZAT20 ceramic samples were depicted in Figure 1. It can be obviously seen that LSZAT0 appears denser and fewer pore structures than LSZAT20 samples. Therefore, Aluminium titanate will be a uniformly dispersed phase in the matrix structure. The resultant dense form was perceived with an increasing Aluminum titanate (Al_2TiO_5) content. Besides, an addition of Aluminium titanate (Al_2TiO_5) influences the particle morphology as is observed in the microstructural view of the composites obtained and eventually, an increase in grain size will enlarge an aluminium titanate volume. Representative EDX result indicated the microstructural view of the LSZAT10 samples used which enables to analyse phases. It can be clearly seen that due to the presence of Zr, O, Al and Ti elements in Region A, two main phase formation (LSZ and Al_2TiO_5) was identified. Also, other regions were determined as: region B (includes Zr and O elements), region C (includes Zr, O, Al, Ti, and La elements), and region D (includes Zr, O, Al, Ti, and La elements), respectively. Moreover, the content of Titanium element in the samples obtained was measured as 5.14 % (region A), 2.38 % in within the grain structure (region B) and 0,88 % at the grain boundary (region C). Hence, it appears that TiO2 is more denser at the grain boundaries and the average grain size indicated as 4,2 µm for LSZAT0 and 10,8 µm for LSZAT20 respectively.

X-ray diffraction patterns of the sintered samples shown in Figure 2. Accordingly, X-ray diffraction analysis of the as-sintered samples explained that the highest phases are LSZ and aluminum titanate (Al_2TiO_5). The results show that monoclinic and tetragonal phases occur and observed that lanthana does not stabilize t-phase in samples added to zirconia. At last, a pyrochlore-type cubic phase $La_2Zr_2O_7$, is formed [13].

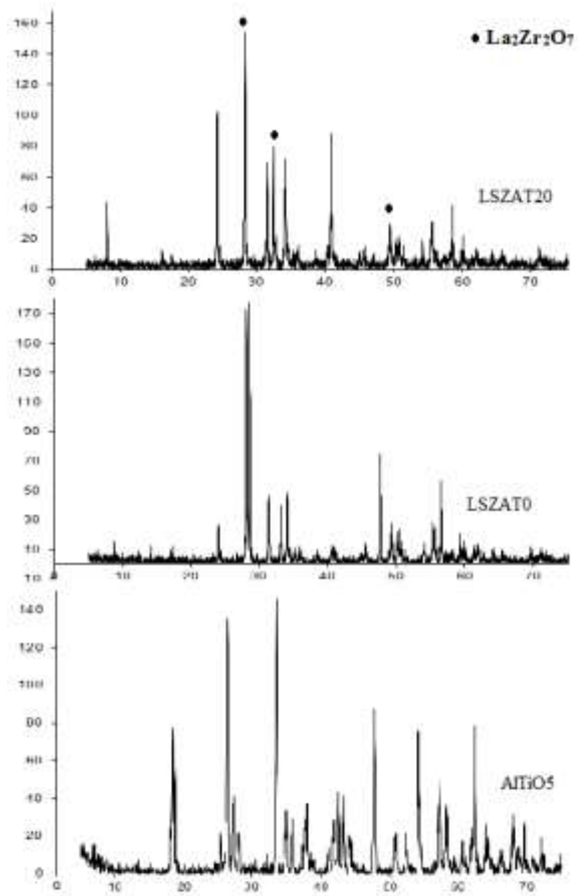

Figure 2. X-ray diffraction patterns of the samples LSZAT20, LSZAT0 and aluminium titanate.

Figure 3 shows that the thermal expansion behaviour of LSZAT20 ceramics. According to this, LSZAT0 has coefficient of thermal expansion (CTE) of 10.35×10^{-6} K^{-1} for 8 mole % LSZ in the range 298 and 1273 K [16] and Al_2TiO_5 has coefficient of thermal expansion (CTE) of $0,2\times10^{-6}$ K^{-1} [17]. According to the results, addition of AT is observed to decrease the coefficient of thermal expansion of LSZAT0. Samples LSZAT0, LSZAT10 and LSZAT20 in the range of 298-1273 K. have coefficient of thermal expansion of $10.35x10^{-6}$ K^{-1}, 9,33 $x10^{-6}$ K^{-1} and 8,32 $x10^{-6}$ K^{-1}, respectively. It is well known that a low coefficient of thermal expansion is one of the basic requirements for development thermal shock resistance at high temperatures.

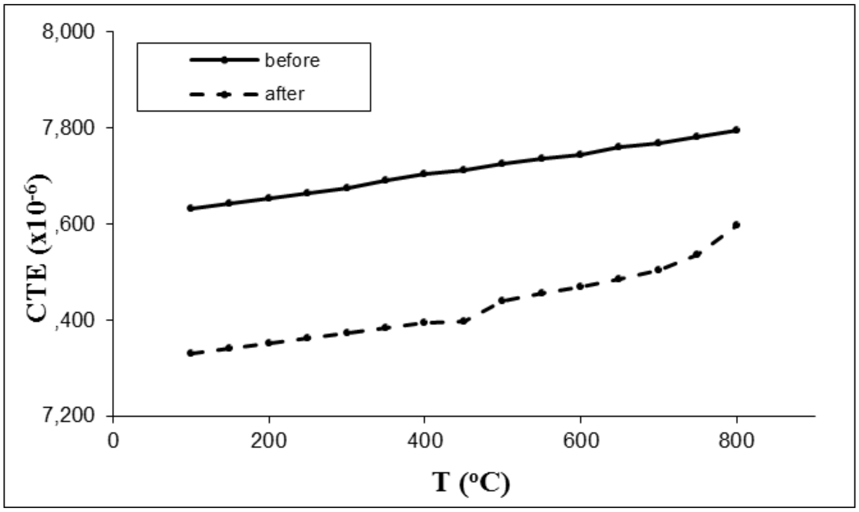

Figure 3. Average coefficient of thermal expansion (CTE) designs of LSZAT20 ceramics (After and before from thermal shock testing).

Figure 4 shown micro hardness of LSZAT0, LSZAT10 and LSZAT20 examples before and after thermal shock tests. Micro hardness (Shimadzu, HMV) was measured on the polished surface of the samples at room temperature. At least six singular tests with a peak load of 2000 g and a loading time of 20 s were applied for each set of composites. After thermally etching at 1500 ºC for 4 hour in air, micro hardness was measured again. The micro hardness rises with increasing AT content. In the case of LSZAT20, the hardness received the biggest value (1452 Hv). Hence, growing AT content can be related to the mechanical properties like hardness.

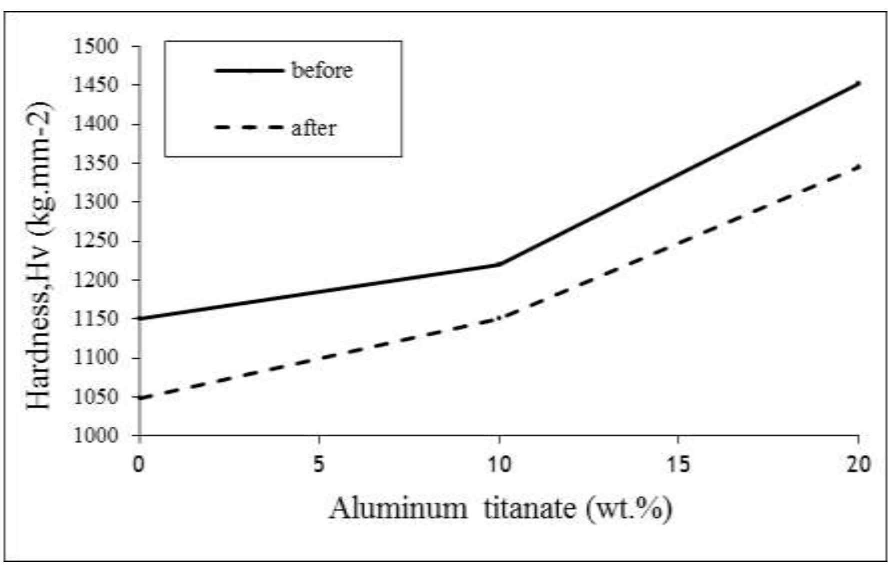

Figure 4. Vickers micro hardness LSZAT0, LSZAT10 and LSZAT20 samples before and after thermal shock testing.

4. CONCLUSION

Aluminium Titanate / LSZ ceramics with different percentages of Al_2Ti0_5 was prepared, thermal properties and microstructural were characterized. The increase in the content of aluminum titanate was observed to be denser and less porous. The addition of Aluminium titanate also actions the grain morphology as is observed in the microstructures of the composites. The grain size was increased when the amount of aluminium titanate was increased. LSZ-AT ceramic composites demonstrate increased hardness depending upon the AT content. As seen in the example LSZAT0, it has a hardness of 1150 Hv which increases to 1452 Hv by addition of 20% by weight of AT. Consequently, it was observed that as the content of aluminum titanate was increased the hardness was increased. XRD analysis of the sintered samples displayed that main phases are LSZ and aluminium titanate (Al_2TiO_5). The results showed that the tetragonal phase of the samples increased as the content of Aluminum titanate increased The thermal expansion coefficient of the LSZ decreased when AT was added. Samples LSZAT0, LSZAT10 and LSZAT20 in the range 298-1273 K have the coefficient of thermal expansion of $10.35x10^{-6}$ K^{-1}, 9,33 $x10^{-6}$ K^{-1} and $8,32x10^{-6}$ K^{-1}, respectively. It should be well known that the low coefficient of thermal expansion is necessary to increase the thermal shock resistance at high temperatures.

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