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Fabrication and Characterization of Mullite Reinforced CeO_2 Added ZrO_2 Ceramics

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ABSTRACT

We examined in this study whether there are phase differences in the CeO_2-ZrO_2 mixture depending on sintering temperatures and times. In addition, the effect of mullite was investigated on the mechanical, physical and microstructural properties of this mixture. Mullite ($3Al_2O_3.2SiO_2$) and 10 mol % cerium oxide doped zirconium dioxide (10 mol % CeO_2 - 90 mol % ZrO_2) powders were synthesized by conventional ceramic production technique. The mixtures were prepared by mechanical alloying method using zirconia ball mill in acetone environment. To synthesize mullite, Al_2O_3 and SiO_2 powders mixture was prepared with stoichiometric proportions and fired it in the air at 1600 ^oC for 3 h. And the ceria added zirconia composites were fired at 1300 ^oC for 2 h. Thus, ceria - zirconia and mullite composite phases were obtained and milling and sieving processes were carried out. Then, mullite-free and 10% by weight mullite reinforced cerium oxide added zirconia mixtures were prepared by powder metallurgy method. For 24 h, the mixtures were milled with zirconia ball mill in acetone environment and sieved by 75 µm sieve. The powders were pressed by uniaxial pressing after drying. The formed samples were sintered in a high temperature furnace in air conditions for 1 and 5 h at 1500 and 1600 ^oC sintering temperatures. Finally, microstructure examinations of the composites with SEM, phase analysis with XRD, hardness, three-point bending and wear tests were performed. In addition, the results of water absorption, porosity and density from physical properties were investigated.

Mullit Takviyeli CeO_2 Katkılı ZrO_2 Seramiklerinin İmalatı ve Karakterizasyonu

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ÖZET

Bu çalışmada CeO_2-ZrO_2 karışımında sinterleme sıcaklıkları ve sürelerine bağlı olarak faz farklılıkları olup olmadığını inceledik. Ayrıca mullitin bu karışımın mekanik, fiziksel ve mikroyapısal özelliklerine etkisi araştırılmıştır. Mullit ($3Al_2O_3.2SiO_2$) ve %10 mol seryum oksit katkılı zirkonyum dioksit (%10 mol CeO_2 - %90 mol ZrO_2) tozları geleneksel seramik üretim tekniği ile sentezlendi. Karışımlar, aseton ortamında zirkonya bilyalı değirmen kullanılarak mekanik alaşımlama yöntemiyle hazırlanmıştır. Mullit sentezi için, Al_2O_3 ve SiO_2 toz karışımı stokiyometrik oranlarda hazırlanmış ve 1600 ^oC'de 3 saat hava atmosferinde fırınlanmıştır. Serya katkılı zirkonya kompozitleri 1300 ^oC'de 2 saat fırınlanmıştır. Böylece serya - zirkonya ve mullit kompozit fazlar elde edilmiş, öğütme ve eleme işlemleri gerçekleştirilmiştir. Daha sonra toz metalurjisi yöntemiyle mullitsiz ve ağırlıkça %10 mullit takviyeli seryum oksit katkılı zirkonya karışımları hazırlanmıştır. Karışımlar 24 saat aseton ortamında zirkonya bilyalı değirmende öğütülmüş ve 75 µm elekten elenmiştir. Tozlar kurutulduktan sonra tek eksenli presleme ile preslenmiştir. Oluşan numuneler hava atmosferinde bir yüksek sıcaklık fırınında 1500 ve 1600 ^{o}C sinterleme sıcaklıklarında 1 ve 5 saat sinterlenmiştir. Son olarak kompozitlerin SEM ile mikroyapı incelemeleri, XRD ile faz analizi, sertlik, üç nokta eğilme ve aşınma testleri yapılmıştır. Ayrıca fiziksel özelliklerden su emme, gözeneklilik ve yoğunluk sonuçları araştırılmıştır.

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Introduction

Among ceramics, zirconia (ZrO_2) and its composites have become very popular for technological and many scientific studies because of their good mechanical properties, corrosion resistance, low thermal conductivities, higher temperature stabilities and higher chemical stabilities (Ceylan, 2006; Boyraz, 2008; Pekdemir, 2018). They are preferred as significant materials for refractory materials, high temperature furnaces, components that are resistant to wear, various cutting tools, dental studies and a lot of fields (Ceylan, 2006; Boyraz, 2008; Abi, 2009; Pekdemir, 2018). High-purity zirconia (ZrO_2) exhibits three polymorphs depending on temperature: monoclinic zirconia phase is stable at temperatures up to 1170 °C. After this temperature, the conversion from the monoclinic phase to the tetragonal phase begins and the tetragonal phase is stable at temperatures up to 2370 °C. From this temperature to the melting temperature of 2680 °C, it is in the cubic zirconia phase. (Boyraz, 2008; Abi, 2009). Depending on the cooling processes, conversion from the t-ZrO_2 phase to the m-ZrO_2 phase takes place. Transformation is very important as it causes volumetric changes of around 3% to 5% and thus cracks. Due to preventing this transformation and stabilizing the zirconia, it is common to use stabilizers. Addition of stabilizers to zirconia, lowers temperature of the transformations, reduces volumetric growth or shrinking and blocks the polymorphic transformations. By using stabilizers, it is possible to make stable the high-temperature phases at low temperatures too (Abi, 2009). Different stabilizers, such as, Al_2O_3, CaO, CeO_2, MgO, SiO_2, TiO_2, Y_2O_3 and even a combination of them, stabilize and hold stable the zirconia in the t-ZrO_2 and/or c-ZrO_2 forms at low temperatures (Hafızoğlu, 2021; Hafızoğlu, Akkuş and Boyraz, 2021; Hafızoğlu, Boyraz and Akkuş, 2021). It is possible to produce materials including only t-ZrO_2 or c-ZrO_2 or a mixture of these with m-ZrO_2 phases by adding different quantities of stabilizers. If less than sufficient stabilizing oxide is added, partially stabilized zirconia (PSZ) is obtained instead of fully stabilized zirconia. PSZ usually consists of two or more closely mixed phases. As a result of using stabilizers and obtaining fully or partially stabilized zirconia, could be achieved superb mechanical properties for example bending strength, hardness, fracture toughness (Cutler et al., 1992).

Zirconia exhibits better mechanical properties than other ceramics. However, like all other ceramics, it is fragile and cannot be formed at room temperature. Therefore, it is desirable to increase the toughness of these materials. So, some energy absorbing mechanisms such as transformation toughening and fiber reinforcement are used in ceramic matrices (Boyraz, 2008).

Increasing fracture toughness with transformation toughness, works on the principle of a phase transformation caused by tension, by reducing the driving force that propagates existing cracks in the material structure (Boyraz, 2008). The best example for this can be indicated zirconia. Zirconia which is added to the ceramic main phase, keeps important role in increasing the fracture toughness and thus the bending strength with its tetragonal - monoclinic transformation toughening feature. With the discovery of the transformation toughening that occurs in zirconia, this material has found wide use. Because the main factor limiting the use of ZrO_2 in advanced engineering applications was the low toughness of this material. Increasing the toughness property with transformation toughness has also increased the interest in zirconia. The volume increase of 3 - 5%, which occurs with the tetragonal - monoclinic phase transformation in zirconia, prevents crack propagation, so increases the toughness of the material and its resistance to fracture (Boyacıoğlu, 2007). The t-m transformation in zirconia can occur in different ways. For example: t-ZrO_2 can be free as a single particle or crystal; It can also be found in a compacted form in a matrix as a precipitate phase. Tetragonal zirconia particle size and the matrix in which the particles are compacted are the most important factors. A critical t-ZrO_2 grain size is determined for ZrO_2 matrix phase. If the grain size is below this critical size, there will be no transformation; above, the t-m transformation occurs either continuously or as a result of applying a stress (Boyraz, 1998).

In the ceramic - ceramic mixed structure formation process, which is the other method of increasing the fracture toughness, the strength and toughness are increased by adding ceramic whiskers, fibers or particles to the main phase. This method is based on creating a physical barrier to the progressive crack. With higher tensile strength than polycrystalline material, whiskers are a good barrier to propagation cracking. So, the fracture toughness of zirconia can be increased more by adding some secondary phases for example nano particles, nano sized fibers or nano sheets into zirconia matrix (Liu et al., 2018). For the last decades, improvement of nanomaterials offers new alternatives to reinforce ceramics. Carbon nanotubes have attracted important attention as reinforcement materials because of their superior properties. However, at high temperatures, they are prone to react with oxide matrixes leading to degradation in some mechanical properties of carbon nanotubes and limited the reinforcing effect on the ceramics (Sun et al., 2005; Eichler et al., 2007). So, it is stated that incorporation of mullite which is another type of ceramic toughening method with its high temperature oxidation and corrosion resistance and the other superior properties, might be preferred (Liu et al., 2018). In the literature, mullite ($3\ Al_2O_3.2SiO_2$) is described as the matchless stable middle crystalline phase for Al_2O_3-SiO_2 binary system, cost-friendly and exhibiting good refractory ability (El Ouatib et al., 2005; Kucuk and Boyraz, 2019). Mullite has received significant attention for technological applications because of its well properties like low coefficient of thermal expansion, high melting temperature, good resistance to creep, good chemical stability and satisfactory hardness (Kumar et al., 2015; Roy et al., 2015). In short, fracture toughness of zirconia can be advanced with mullite reinforcement as the secondary phase into the ZrO_2 matrix and so, the other mechanical properties can be improved too (Liu et al., 2018). In addition, the temperature of sintering is also important, because of affecting some properties of ceramics through changing of the crystalline phases and microstructure (Denry and Kelly, 2008).

In this study, mullite ($3Al_2O_3.2SiO_2$) and 10 mol % cerium oxide doped zirconium dioxide powders were synthesized by conventional ceramic production technique. It was examined whether there are phase differences in the CeO_2-ZrO_2 mixture depending on sintering temperatures and times and the effect of mullite on the mechanical, physical and microstructural properties of this mixture.

Material and Methods

Mullite and ceria doped zirconia powders were produced by conventional ceramic production method in this study. All precursory powder materials were obtained from Company Eczacıbası (Al_2O_3 and SiO_2 powders), Merck (CeO_2) and Chemicals of Handan Yaxiang Trading Co. (ZrO_2). The powders were mixed in acetone environment by mechanical alloying method. The powders were heated for 24 hours in oven at 110 ºC before and after mixing. Mullite ($3Al_2O_3.2SiO_2$) and 10 mol% ceria doped zirconia (CeO_2-ZrO_2) powders were synthetized by sintering from the powders prepared with stoichiometric ratios of Al_2O_3, SiO_2, CeO_2 and ZrO_2 powders after homogenized in ball mill. Mullite ($3Al_2O_3.2SiO_2$) was synthetized for 3h at 1600 ^{o}C and 10 mol% ceria doped zirconia (CeO_2-ZrO_2) composite powders were synthetized for 2 h at 1300 ^{o}C. Thus, ceria - zirconia and mullite composite phases were obtained and milling and sieving processes were carried out. Then, mullite-free and 10% by weight mullite reinforced cerium oxide added zirconium oxide composites were prepared by powder metallurgy technique (hereinafter these mixtures will be named CeZ00M and CeZ10M respectively). The sample was coded as CeZ10M16005 (CeZ: Ceria doped zirconia; 10M: 10% Mullite addition and 16005: 1600 °C sintering temperature and 5 hours sintering time). After the composite powders were milled for 24 h in acetone environment with zirconia ball mill, sieved by 75 µm sieve and dried. Then, the composite mixtures were pressed to 56x12 mm sizes mold gap by uniaxial pressing machine at 200 MPa load. The pressed samples were sintered in a high temperature furnace (Protherm™ Furnace) and in air conditions for 1-5 h sintering times and 1500-1600 ^{o}C temperatures. The heating rate was 5 ^{o}C/min. Then, microstructure investigations with SEM, phase analysis with XRD, the hardness, 3-point bending and wear tests and physical properties that are water absorption, porosity, shrinkage and density results were examined on the composites.

The three-point flexural strength tests were executed with crosshead speed of 0.5 mm/min in a Shimadzu brand tensile-compression device. For each sample, measurements were taken five times and their average were taken as the bending strength results of the samples.

The strength calculations were made with the formula (1):

$$\sigma = 3/2*P*L / (b*h^2) \quad (1)$$

(In (1) the letters mean that, P: maximum load, L: the distance of between supports, h: height, b: width).

After 180, 320, 600, 1200 and 2500 grit sanding process, polishing is done for each sample. With Vickers hardness tester that was Mitutoyo brand, the measurements of hardness were executed by 1 kg load for 10 seconds. For each sample, measurements were taken five times and their average were taken as the hardness results of the samples (Önen and Boyraz, 2014; Akkuş and Boyraz, 2018; Boyraz and Akkuş, 2021). The wear tests of samples were executed with Plint brand wear tester. For wear tests, steel discs were used. For each sample, wear tests were executed at 400 rpm rate, 5, 10 and 15 min wear durations and 100 N force. The samples were measured with a precision scale of 10^{-4} g. After the assigned wear times, the samples were scaled again and the wear amounts were calculated (Akkuş and Boyraz, 2018; Boyraz and Akkuş, 2021). To determine the phases, XRD with Cu Kα radiation (Bruker AXS D8 Advance; 20kV-60kV, 6mA-80mA and $\theta = 10^{o}$-90^{o}, 0.002^{o}) was used. The phases of the samples seen in XRD patterns were defined with the Panalytical X'Pert program. The microstructural characterization of the samples was done with the Mira3XMU FE-SEM (Tescan, Czech Republic) brand scanning electron microscope machine and energy dispersion spectrum. The results were presented in various graphics and tables and some comments on these results were made.

Results and Discussion

Physical (shrinkage, water absorption, density and porosity tests) and mechanical (hardness, 3-point bending and wear) tests, SEM, EDS and XRD analysis results were included in this section. Calculations and measurements were repeated 5 times and arithmetic averages were taken. The shrinkage, porosity, water absorption, relative density and bulk density results are shown in Table 1, and also Fig. 1. In Fig. 1, the relative density values were taken at the rate of 1/3 of the actual values for the graph.

Table 1. Physical test results of CeZ00M and CeZ10M samples

Samples	Bulk density (g/cm^3)	Relative density (%)	Water absorption (%)	Porosity (%)	Shrinkage (%)
CeZ00M15001	5,21	88,80	2,08	11,20	12,35
CeZ00M15005	5,63	96,03	0,20	3,97	14,32
CeZ00M16001	5,80	98,87	0,18	1,13	15,17
CeZ00M16005	5,82	99,22	0,17	0,78	15,96
CeZ10M15001	4,84	88,22	1,39	11,78	12,57
CeZ10M15005	4,89	89,14	1,31	10,86	12,72
CeZ10M16001	4,92	89,68	1,25	10,32	13,14
CeZ10M16005	4,91	89,50	1,26	10,50	13,15

Ming-qing et al. (2002), produced the samples stabilized with 12 mol% CeO_2 by sintering at 1500 – 1600 °C temperatures for 3 hours. They stated that the highest density value was obtained from 16003 samples similarly our study.

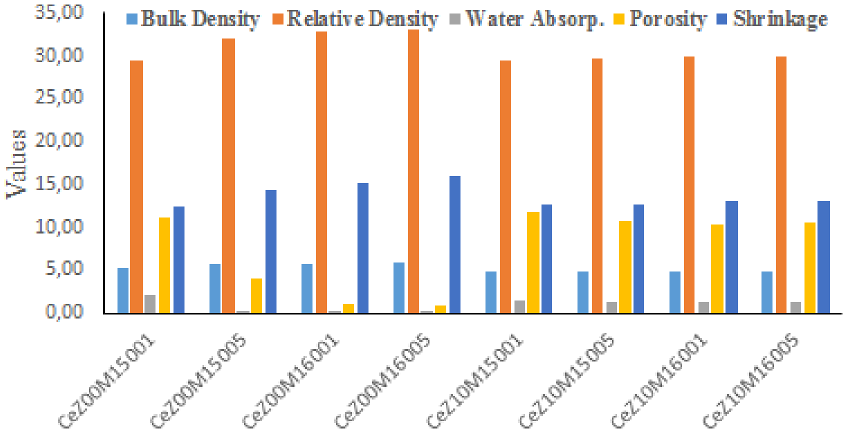


Fig. 1. Physical test results graph of CeZ00M and CeZ10M samples

When the results were examined, it was found that with increasing sintering temperature and time, the shrinkage values increased in general, and accordingly the water absorption and porosity values decreased; It is seen that experimental density and relative density values increase. In addition, it is seen that the shrinkage, experimental density and relative density values are lower and the water absorption and porosity values are higher in mullite added samples.

Table 2 and Fig. 2 indicated the hardness and three-point flexural or bending strength results of CeZ00M and CeZ10M samples.

Table 2. Hardness and three-point bending strength results of CeZ00M and CeZ10M samples

Samples	Hardness (HV)	3-Point bending strength (MPa)
CeZ00M15001	400,80	127,52
CeZ00M15005	511,30	140,95
CeZ00M16001	592,25	147,40
CeZ00M16005	595,20	77,28
CeZ10M15001	395,20	15,60
CeZ10M15005	400,55	16,54
CeZ10M16001	418,28	18,24
CeZ10M16005	376,70	15,06

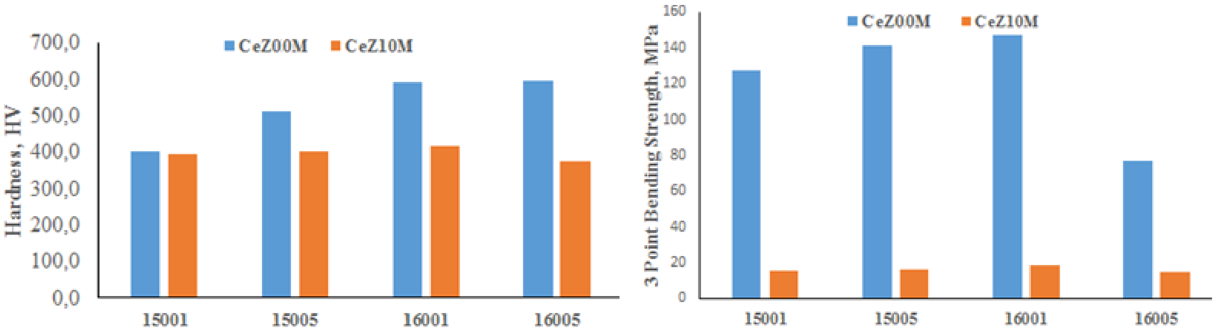


Fig. 2. Hardness and three-point bending strength graphics of samples

It is seen that the hardness values generally increase with increasing sintering temperature and time. The hardness values of the mullite added samples are lower and the CeZ10M16005 sample has the lowest hardness value despite the high sintering temperature and time. This is related to the pores that occur in the microstructure.

According to the 3-point bending test results, it is seen that the three-point flexural strength values increase with increasing sintering temperature and time in the samples, and the flexural strength values of the 16005 samples with and without mullite additives have the lowest values. In addition, it is understood that the three-point bending strength values of the mullite added samples are very low. It is thought that this situation is a result of the defects in the microstructure and possible phase changes depending on the sintering temperature, sintering time and mullite additive.

The wear tests of samples were executed with Plint brand wear tester. For wear tests, steel discs were used. For each sample, wear tests were executed at 400 rpm rate, 5, 10 and 15 min wear durations and 100 N force. The samples were measured with a precision scale of 10^{-4} g. After the assigned wear times, the samples were scaled again and the wear amounts were calculated. Wear results are seen in Table 3 and Fig. 3.

Table 3. Wear results of CeZ00M and CeZ10M samples

Samples	**Wear Volume (mm^3), 100 N**		
	5 min.	**10 min.**	**15 min.**
CeZ00M15001	0,995	1,847	2,874
CeZ00M15005	0,755	1,211	1,751
CeZ00M16001	0,318	0,404	0,756
CeZ00M16005	0,287	0,601	0,946
CeZ10M15001	1,650	2,781	4,754
CeZ10M15005	1,099	1,783	3,105
CeZ10M16001	0,451	1,065	1,553
CeZ10M16005	0,605	1,258	2,148

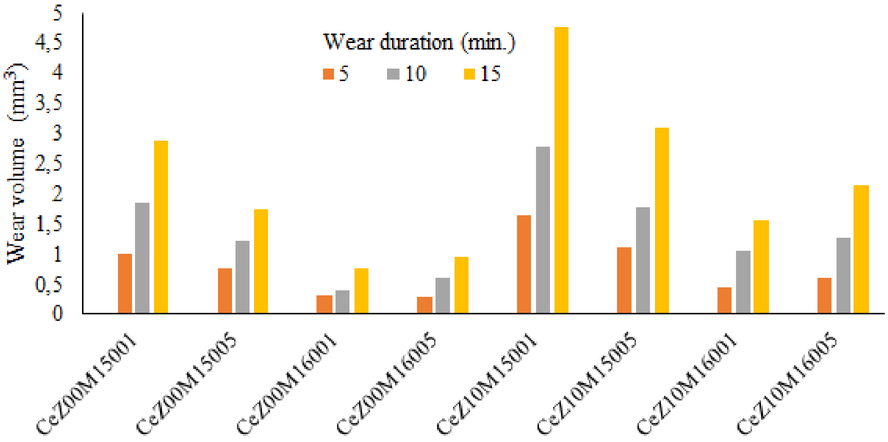


Fig. 3. Wear test results graph of samples

When the data obtained are examined, it is seen that the wear results vary depending on the bending strength, hardness and wear time of the specimens. In general, as the wear time increases, the amount of wear of the samples also increases; It is understood that the wear resistance of the samples with high hardness and bending strength is higher and the mullite additive negatively affects these samples and decreases the wear resistance. Although adhesive type wear is observed in the samples in general, it has been observed that with the increase in the wear time in some samples, cracks occur on the worn surface and very small pieces break off and cause abrasive wear.

The phase changes in the sample structure related with the temperature and time of CeZ00M and CeZ10M samples were analyzed and the basic phases that emerged in the structure were shown in Fig. 4.

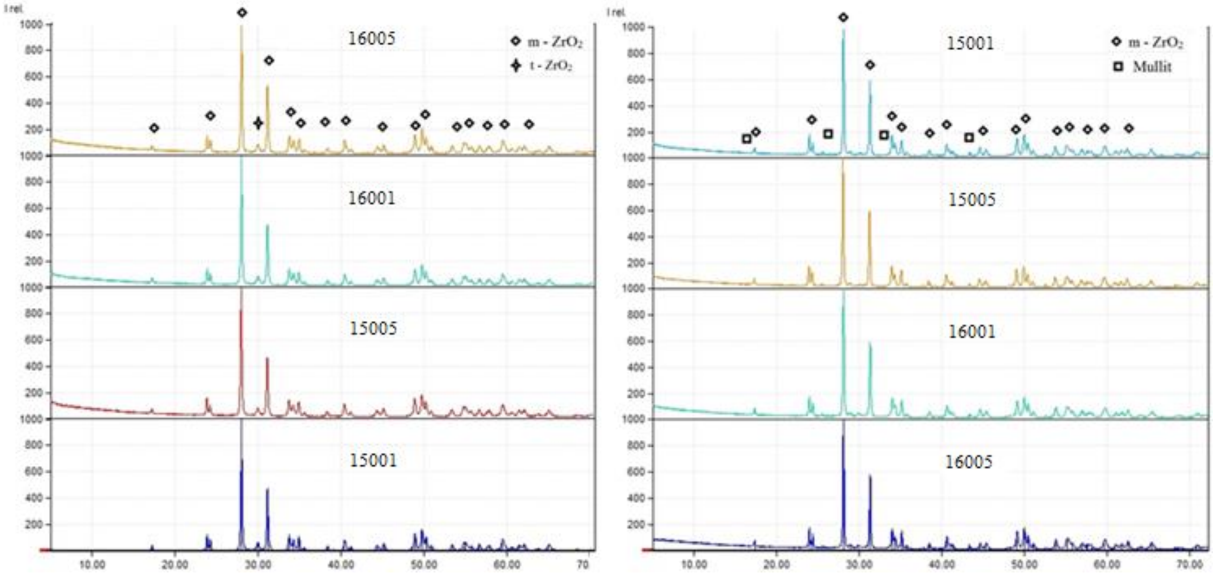


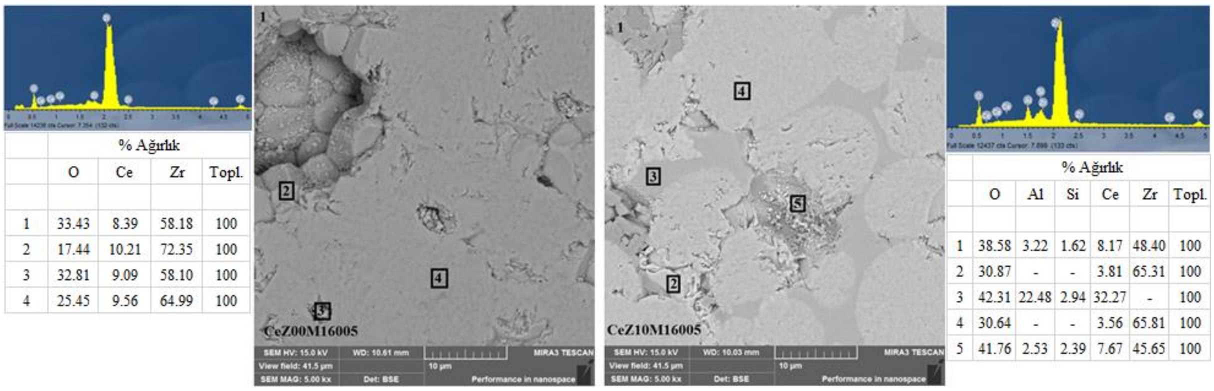
Fig. 4. XRD patterns of CeZ00M (left) and CeZ10M (right) samples

From XRD patterns of the samples, m-ZrO_2, t-ZrO_2 and mullite phases were detected. The m-ZrO_2 (JCPDS 37-1484), t-ZrO_2 (JCPDS 81-1544) and mullite (JCPDS 83-1881) phases were verified by comparison with JCPDS standard cards.

In our mullite-free (CeZ00M) samples, while the m-ZrO_2 phase decreased from 15001 to 16005 in the sample XRD patterns given in Figure 4; In the 16005 sample, the m-ZrO_2 phase increased compared to the other samples. The t-ZrO_2 phase is seen in the samples, albeit slightly. The fact that the 16005 sample has the lowest bending strength is because of the increase in the monoclinic phase and grain coarsening in this direction.

In our mullite added (CeZ10M) samples given in Figure 4, XRD patterns are the almost same and there is no t-ZrO_2 or c-ZrO_2 phase in the structure. In addition, macro cracks were formed in the mullite added samples during the production stage. Therefore, mullite additive affected these samples negatively and also reduced the mechanical properties. The decrease in flexural strength in CeZ10M16005 sample is thought to be due to excessive grain growth.

The microstructure image and elemental analyzes of CeZ00M16005 and CeZ10M16005 samples with EDS are given in Figure 5. From the SEM image given in Figure 5, we see that the grain size is significantly larger in the mullite added sample, which will adversely affect the microstructure. It can be predicted that this situation will have a negative effect on the hardness results of the specimens and decrease the hardness of the mullite added sample. As a matter of fact, in the experimental results, the hardness value of the CeZ10M16005 sample was found to be lower than the hardness value of the CeZ00M16005 sample.



	% Ağırlık			
	O	Ce	Zr	Topl.
1	33.43	8.39	58.18	100
2	17.44	10.21	72.35	100
3	32.81	9.09	58.10	100
4	25.45	9.56	64.99	100

	% Ağırlık					
	O	Al	Si	Ce	Zr	Topl.
1	38.58	3.22	1.62	8.17	48.40	100
2	30.87	-	-	3.81	65.31	100
3	42.31	22.48	2.94	32.27	-	100
4	30.64	-	-	3.56	65.81	100
5	41.76	2.53	2.39	7.67	45.65	100

Fig. 5. SEM images and elemental analyzes of CeZ00M16005 and CeZ10M16005 samples

According to the EDS analyzes given in Fig. 5, the evaluation of the EDS analysis results on the CeZ00M16005 and CeZ10M16005 samples was made from general (1) and parts 2, 3, 4, 5. It has been observed that the results of the EDS elemental analysis made from the general field survey (1) and other parts are compatible with the contribution rates and XRD results made to the samples.

Conclusion

In our study, utilization of mullite in the production of ceria doped zirconia was explored. The effect of CeO_2 and mullite contribution to ZrO_2 main matrix was evaluated.

In all samples except CeZ10M16005, shrinkage, experimental and relative density and hardness values increased with increasing sintering temperature and time; It was observed that the porosity and water absorption values decreased. This situation in the CeZ10M16005 sample is thought to be a result of the pores formed in the microstructure and possible phase changes related to the decrease in the density value. In addition, with increasing sintering temperature and time, the three-point flexural strength values and wear resistance of the samples generally increased; On the other hand, it was observed that the wear resistance decreased and the flexural strength values got the lowest values in the 16005 samples with and without mullite additives.

From the XRD patterns, while m-ZrO_2 phase decreases from CeZ00M15001 to CeZ00M16005 samples; In the CeZ00M16005 sample, the m-ZrO_2 phase increased compared to the other samples. A small amount of t-ZrO_2 phase is seen in the samples. The fact that the CeZ00M16005 samples have the lowest bending strength is due to the increasing the monoclinic phase and grain coarsening in this direction. There is no significant difference in the XRD patterns of the mullite-doped samples and no t-ZrO_2 or c-ZrO_2 phase was detected in the structure. Therefore, we think that the decrease in flexural strength of mullite-added CeZ10M16005 sample is due to excessive grain growth.

The density value of mullite additive is lower than ZrO_2 - CeO_2 composite and it is more stable about shrinkage value at high temperatures compared to this composite. Therefore, in mullite added samples, shrinkage, experimental density, relative density and hardness values are lower, whereas water absorption and porosity values are higher. The SEM images show that the grain size is significantly larger in the mullite added sample. This adversely affect the microstructure. It can be said that this situation has a negative effect on the hardness results of the specimens and decrease the hardness of the mullite added samples. Also, from the XRD patterns of mullite-added samples, it was seen that there is no t-ZrO_2 or c-ZrO_2 phase in the structure. This is a reason of lower bending strength values. In addition, macro cracks were formed in the mullite added samples during the production stage. Therefore, mullite additive affected these samples negatively and also reduced the mechanical properties which are hardness, bending strength and wear values.

In samples without mullite additives, m-ZrO_2 and t-ZrO_2 phases were detected, whereas m-ZrO_2 and mullite phases were found in samples with mullite additives. In these samples, no significant phase change was observed depending on temperature and time.

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Statement of Conflict of Interest

No conflict of interest was declared by the authors.

Author’s Contributions

The authors declare that they have contributed equally to the article.

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