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Article

Effect of Sintering and Various Fillers in Zirconia Composite Coating for High Temperature Application

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Abstract. Zirconia is a ceramic material that is relatively cheap and easy to purify from mineral form. Zirconia powder has stable properties under high temperature conditions making it suitable for use as a coating for steel substrates. Ceramic composite coating is one option that can be used to increase its durability by adding filler which has lubricant properties. In this research, hBN, MoS2 and graphite were used as filler coatings. The coating method used is slurry spray, which is a simple method and there is a subsequent sintering process so that the resistance of the coating to the substrate is better. The effect of the coating is seen before and after the sintering process on the surface and thickness. And to see the adhesion of the coating to the substrate, a thermal shock test was carried out. From the test results, it was found that sintering had a significant effect on the coating surface, where the defects on the coating surface became fewer and more even. The optimum temperature for sintering is 600^{o}C where the least porosity is obtained.

Keywords: Zirconia composite coating, lubricant, slurry spray, thermal shock, sintering.

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1. Introduction

Zircon is resistant to thermal shock and chemical attack, abrasion resistant, impact resistant, and has a high melting point so that because of its characteristics, zircon is classified as a refractory material [1]. Based on the Oxford Dictionary of Earth Sciences, refractory minerals can be defined as minerals that are resistant to decomposition by high temperature, pressure, or chemical attack [2]. A refractory material is expected to be able to maintain its bond strength and structural shape at high temperatures and various destructive forces. The world's need for zircon is supported by zircon production spread across various countries. For many years, Australia has led the way as the largest zircon producing country, followed by South Africa. Indonesia is also one of the countries that contributes to zircon supplies to meet global zircon needs [3]. Data obtained by the Geological Resources Center in 2006 stated that Indonesia's zircon potential was more focused on the island of Kalimantan. Zircon has been widely applied in various industrial fields, including the ceramic, refractory, molding sand and abrasive industries [4].

The addition of a coating layer can be done as a way to improve the surface properties of a material, including steel [5]. Research on coatings that utilize ZrO_2 has been carried out for various purposes, for example as a coating mixture that has anti-fouling properties, increases resistance to corrosion, and increases coating resistance to high temperatures [6-8]. This is a supporting reason that the use of zircon as a coating material could be an effort to increase the added value of local Indonesian minerals. According to research conducted by Gurusamy et al [9], the composition of the coating material mixture will influence the properties of the coating formed. This is the reason why in this research ceramic slurry was made with zircon powder.

The slurry coating technique refers to the technique of applying a slurry or making a paste first, followed by drying and sintering of the coating on the substrate. It has long been used for enamels that produce a layer of glassy substance on the surface of a component for decoration or protection. The coating process using the slurry method is divided into three, namely, mixing, coating and sintering processes. This sintering aims to remove volatiles from the ceramic layer at high temperatures. As porosity increases, the particle contact area will increase and the mechanical strength will increase. Therefore, it is very important to study the sintering phenomenon of ceramic coatings, and the relationship between diffusion and material structure during the process [10].

In this research, fillers was added to the zircon coating matrix. hBN, MoS_2 and graphite function as a lubricant which provides thermal shock resistance properties to coatings if given a certain composition [11-13]. Sintering temperature is also an important parameter that influences coating performance, therefore it is necessary to see its effect on the coating surface before and after the sintering process to see whether it results in many defects appearing on the coating.

2. Materials and Method

In this study, Zircon powder was used as the main component of ceramic coatings, and hBN, MoS_2 and graphite as a filler which will produce a coating with a smooth surface and resistance to high temperatures. The substrate as the specimen material used in this research is low carbon steel AISI 1005 in the form of a tube. The specimen was cut using a wire cutting tools machine to a size of 20x15x7 mm.. Sand blasting is an important step for surface cleaning and increasing the roughness of the steel surface which can strengthen the bond between the substrate surface and the ceramic coating [14].

The slurry mixture was mixed with a stirrer for 30 minutes until mixed evenly. After that, the slurry mixture is subjected to a wet milling process with a ball mill for 2 hours then sieved with a certain mesh size and the slurry resulting from the sieve is ready for the next process, namely spraying on a substrate that has been treated with surface preparation [15]. The process of making slurry coating is by mixing ingredients whose composition is shown in Table 1. The spraying process is carried out by shooting the slurry at a spray distance of 30 cm and a time of 15 seconds so that the coating powder is evenly distributed and adheres to the surface of the steel substrate. The coated steel substrate was dried at room temperature for 5 hours then oven at 100oC for 1 hour. After that, the sintering process was carried out in a furnace supplied with argon gas at varying temperatures for 2 hours in various temperatures (400^{o}C, 500^{o}C, 600^{o}C and 700^{o}C). The sintered specimens were cooled in air at room temperature. Then, the coated steel substrate is subjected to several tests in the laboratory [10].

The coating thickness analysis was carried out by the metallographic method according to the ASTM E3-11 standard. In this research, thermal shock testing uses the cooling method with water. The sintered specimens were heated again in a furnace at a temperature of 600^{o}C for 10 minutes. Then the specimen was quickly transferred into a container filled with water and then cooled in air at room temperature. The surface of the specimen was observed for damage using Scanning Electron Microscopy (SEM), if more than 1/3 of the area experienced damage, spallation and cracks then the specimen could not withstand thermal shock [16].

Table 1. Composition of Ceramic Slurry Coating.

Materials	**Composition (%w)**			
	ZB3	**ZM3**	**ZC3**	**ZA3**
ZrO_2 Powder	3	3	3	3
hBN Powder	9	-	-	3
MoS_2 Powder	-	9	-	3
Graphite Powder	-	-	9	3
Waterglass	18	18	18	18
Sodium Lauryl Suphate (SLS)	5	5	5	5
Aquadest	Balanced	Balanced	Balanced	Balanced

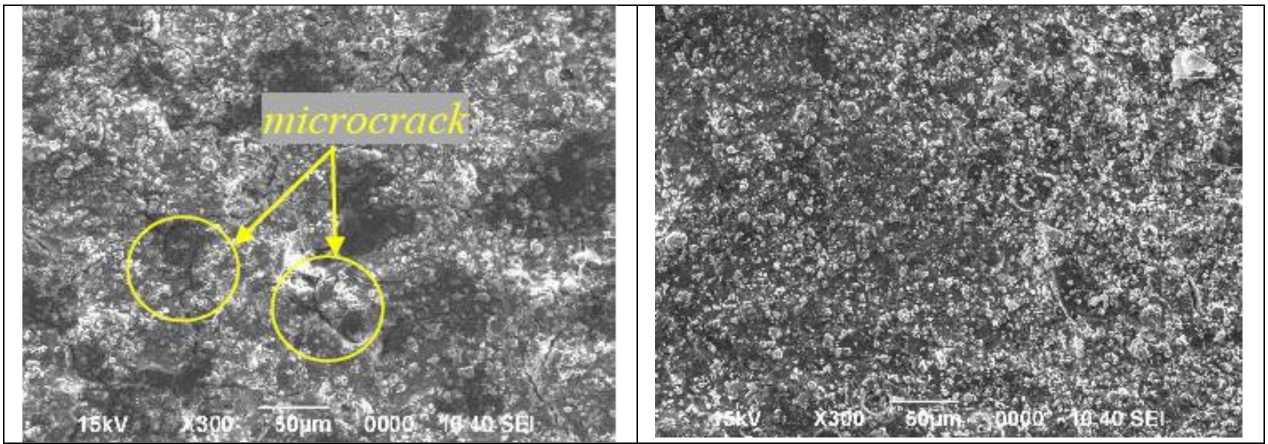
3. Results and Discussion

3.1. Analysis of the Coating Surface Morphology

SEM-EDS testing is carried out on the upper side of the coating with a magnification of 300x so that it shows the microstructure of the surface morphology of the coating on sintered and not sintered. In Fig. 1(a), it can be seen that the coating surface after sintering is denser and more even when compared to Fig. 1(b), which is the coating surface without sintering. It can also be seen from the size of the microcrack which becomes narrower. Figure 1 shows that sintering causes each particle to stick together better and reduces porosity and microcracks that form on the coating surface. This is supported by research which states that sintering causes the merging of particles which is indicated by the expansion of the particle grain boundary area so that a continuous area is formed [17].

3.2. Analysis of Coating Thickness

The change in layer thickness before and after sintering is very important to observe how much mass is lost after the heating process. This affects the amount of air trapped during the coating process as well as the adhesion of the coating bond to the substrate. Figure 6.4 shows the change in thickness of commercial zircon composite coating with variations in lubricant before and after sintering at a temperature of 600°C. For the ZB3 sample which uses hBN as a lubricant, it can be seen that there is no significant change in coating thickness before and after sintering as well as for ZA3 which uses a hybrid lubricant mixed between hBN, MoS_2 and graphite. This indicates that not much air was trapped during the coating process. The ZM3 sample that used MoS_2 lubricant experienced a significant reduction in thickness, while the change in thickness for ZC3 with graphite lubricant was not too large. This is because MoS_2 easily undergoes oxidation at temperatures above 500°C [18].



(a) (b)

Fig. 1. The surface morphology of the coating at 300x magnification on sample (a) not sintered; (b) sintered.

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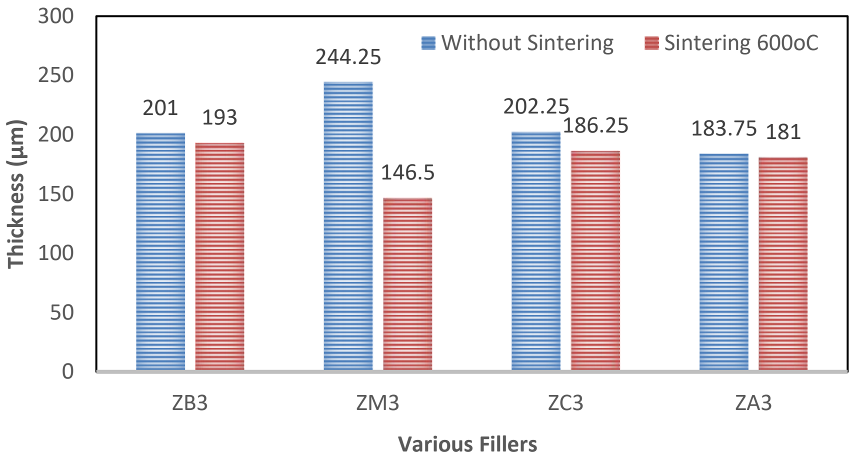

Fig. 2. Effect of Various fillers and Sintering on Layer Thickness.

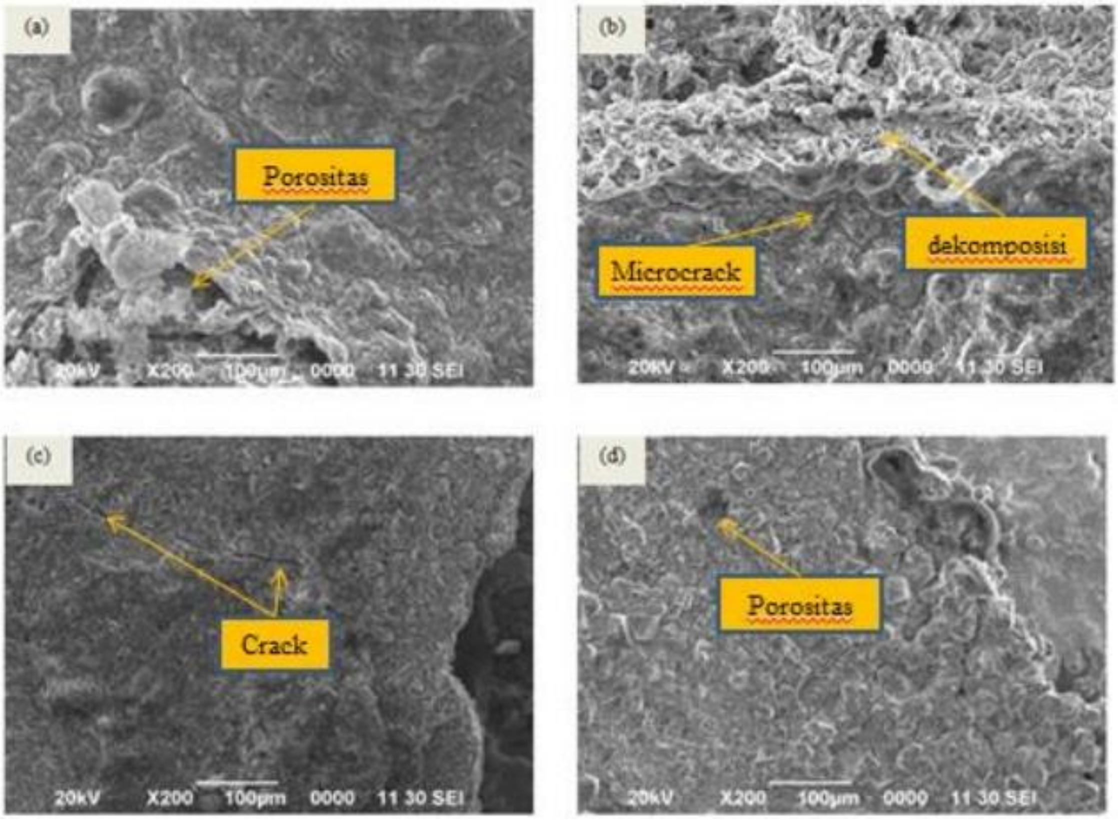

Fig. 3. Morphology of the coating surface after thermal shock test.

3.3. Analysis of Thermal Shock Resistance

Figure 3 is the SEM-EDS result with a magnification of 100x on the surface of the coating after a thermal cycle treatment of 1 cycle at a temperature of 600°C and cooled quickly with water media. Figure 3(a) shows a ZA3 sample with a sintering temperature of 400°C. It can be observed that the surface morphology of the layer is rough and there is porosity. This can be caused by the solidification process being less than perfect and the presence of oxygen trapped during solvent evaporation, resulting in porosity on the surface of the layer. An incomplete solidification process will leave ZrO_2 particles that have not melted, so the particles are still round. The round shape of the particles can produce cavities on the surface of the coating resulting in greater roughness and porosity [19]. It can be seen that the shape of the BN grains is round and lumpy, evenly distributed on the coating surface.

Figure 3(b) shows a ZA3 sample with a sintering temperature of 500°C, it can be observed that there are microcracks in the layer. This is caused by the side stresses of the sintering process. In the sintering process, a shrinkage process will occur due to evaporation of the solvent. When the coating is no longer able to shrink, it will leave residual stress which can cause microcracks to form. Microcracks can also be caused by evaporation of water and decomposition of additives contained in the slurry during sintering or due to differences in the thermal expansion coefficient between the coating and the substrate [20].

Figure 3(c) shows the SEM results for the ZA3 sample with a sintering temperature of 600ºC. It can be seen that the surface morphology of the layer formed has a slight crack on the coating surface. Figure 3(d) shows the SEM results for the ZA3 sample with a sintering temperature of 700ºC. It can be seen that the surface morphology of the layer formed does not have cracks, but has porosity and an uneven surface.

From Fig. 3 as a whole it can be seen that the best surface morphology is found on coating surface 3 (c) with a smooth surface and better than Figs. 3(a) and 3(b). In Fig. 3(d), the sample surface is also rougher and the coating layer has almost completely peeled off due to the sintering temperature being too high.

4. Conclusion

The sintering process of zircon coatings at high temperatures greatly influences the coating surface, namely showing a finer microstructure and fewer boundaries forming. Sintering after the coating process greatly influences the smoothness of the coating surface grains. Comparison of sintering temperature variations can be seen that the coating surface at a sintering temperature of 400^{o}C, and at a sintering temperature of 500^{o}C can be seen that the sample surface is not smooth, there is a lot of porosity and decomposition, this can be caused by the solidification process being less than perfect and the presence of oxygen trapped during solvent evaporation. thus producing porosity on the surface of the layer. An incomplete solidification process will leave ZrO_2 particles that have not melted, so the particles are still round. At a sintering temperature of 600^{o}C the sample surface is smoother and nicer, whereas at a temperature of 700^{o}C the sample surface is rough because the coating layer is almost gone due to the temperature being too high. So it can be concluded that the optimum temperature for sintering zircon coating is 600^{o}C.

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