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Investigation of Ceramic Coating by Thermal Spray with Diffusion of Copper

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Flame thermal spray techniques were used to prepare composites of ceramic powders, alumina and silica ($\text{Al}_2\text{O}_3+\text{SiO}_2$), zirconia with titanium ($\text{TiO}_2+\text{ZrSiO}_4$) and zirconia with alumina ($\text{Al}_2\text{O}_3+\text{ZrSiO}_4$) by weight percentages (5%, 10%, 15%, 20%), to study the performance of composites coating on the diffusion of copper during the sintering process at a temperature (1500°C) for two hours. Result of energy dispersive X-ray test showed that the higher value of the diffusion copper was in alumina brick substrate without coating 46.11%, while ceramic coating specimens of SiO_2 with Al_2O_3 , Al_2O_3 and TiO_2 with ZrSiO_4 made barrier to reduce the diffused atoms of copper especially of SiO_2 with Al_2O_3 and Al_2O_3 with ZrSiO_4 specimens; this led to improving the mechanical properties. Scanning electron microscopy and atomic force microscope test showed that the dense ceramic coating structures are more organized and homogeneous with absence of large pores or voids especially after copper diffusion inside bricks within ceramic coating. Greatest adhesion strength 5.46 MPa of the coating layer with the alumina brick substrate, it was achieved by zircon silicate with 20% $\alpha\text{-Al}_2\text{O}_3$ (with higher alumina ratio) and zircon silicate with 20% TiO_2 which has lower value of adhesive bonding strength (3.352 MPa).

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1. Introduction

Thermal barrier coatings (TBCs) like zirconia, which has a melting point of about 2700°C , low thermal conductivity, wear and oxidation resistance, as well as high hardness and fracture toughness with relatively low density, are mostly sprayed with TBC process [1]. The performance of the flame thermal sprayed pure TiO_2 has been compared with those of $\text{Al}_2\text{O}_3\text{-}40\%\text{TiO}_2$ and pure Al_2O_3 shows the best results [2].

The wetting behavior of copper (and its alloys) on alumina has been widely studied because of its relevance to metallization of alumina substrates in the fabrication of microelectronic devices [3, 4]. The direct reactive joining of alumina with copper, known as the copper direct bonding (CDB) [5], in which the joint is formed through the synthesis of copper oxides with alumina in the presence of the molten $\text{Cu-Cu}_2\text{O}$ eutectic, which well wets the ceramic surface liquid formation, plays an important role in the joining process [6]. The interface consisted of a homogeneous reaction layer produced by chemical interaction, homogeneous interface free of crack and porosity [7].

2. Experimental

Different types of commercial powders were selected: Al_2O_3 (90.0% pure– $150\ \mu\text{m}$ particle size), SiO_2 (99.9% $150\ \mu\text{m}$ particle size), TiO_2 (99.5% $50\ \mu\text{m}$ particle size) and ZrSiO_4 (99.9% $50\ \mu\text{m}$ particle size). Spray gun produces thermal flame which has maximum temperature

of 3000°C by two gases oxygen and acetylene, melting powder is carried in gases mixture. Operating parameters during coating deposition process are the oxygen and acetylene pressure was 4 and 0.7 bar, respectively, distance 20 cm, powder feed rate ($7\ \text{cm}^3/\text{min}$), and substrate temperature (1000°C). The powder was supplied through a special tube in the flame gun; the specimen is fixed on the flange to make 90°C with the powder flame flow. Then, the coating is heated to about 800°C for a suitable time to permit the adhesion for layers.

Infrared thermometer was used to know the temperature of surface before spraying process and the temperature of the fusion after spraying process. Finally, the molten pure copper (Cu–99.9% purity) was overheated about 100°C above the melting point and infiltrated into specimens (coating and alumina substrate) during sintering process of the samples was performed in programmable furnace at temperature (1500°C) for 2 h the rate of heating and cooling was $15^\circ\text{C}/\text{min}$.

3. Results and discussion

3.1. Atomic force microscope for surface roughness results

The topographic structures in 2D and 3D views for the preparation ceramic coating specimen after copper diffusion shows the intermetallic particles and the Cu-rich particles are visible leads to increase in the density in the ceramic coating surface compared with the ceramic coating specimens before copper diffusion as show in Table I.

Figure 1 shows the scanning electron microscopy (SEM) image of the coated specimen with coating; it was noticed that alumina, cristobalite and mullite are present in all the specimens with silica composite coating. The

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structure is heterogeneous and consists of lamellar structure with flattened splats of different in the shape and size. Figures 2 and 3 display the SEM micrographs of all ceramic composite coatings specimens with four different percentages of ceramic powders (5, 10, 15, and 20%) from SiO_2 with Al_2O_3 and Al_2O_3 , TiO_2 with ZrSiO_4 on alumina brick substrate before and after copper diffusion. These figures show that the coated specimens are more homogeneous than the pure alumina specimens with still have un-molten and partially melted particles in this coating with the corollary of hollows in the cross-section.

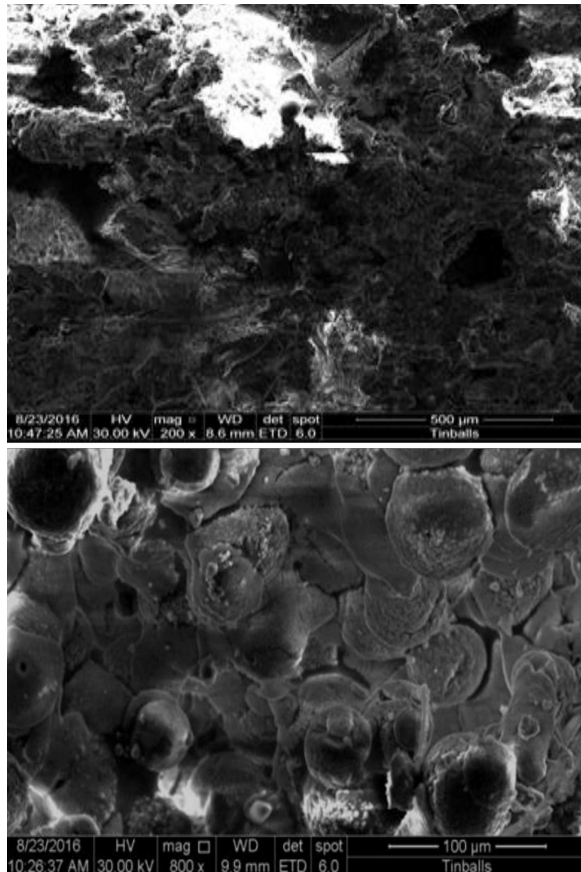


Fig. 1. SEM image of Al_2O_3 -20% SiO_2 thermal spray ceramic coated specimen on alumina brick substrate before and after diffusion of copper.

The microstructure of ZrSiO_4 or TiO_2 composite coating (with Al_2O_3 and TiO_2) in Figs. 2 and 3 showed that the dense ceramic matrix composite coating with ZrSiO_4 splats embedded inside a dense Al_2O_3 matrix. This anisotropic composite coating combined with the large difference in thermal conductivity between ZrSiO_4 and Al_2O_3 will alter the thermal behavior of the coating [8].

The copper particles are concentrated on the figures as bright areas alternating with ceramic component in coating structure. The splat grey phases visible can be assigned to a Cu-rich or intermetallic phases (Al_3Cu ,

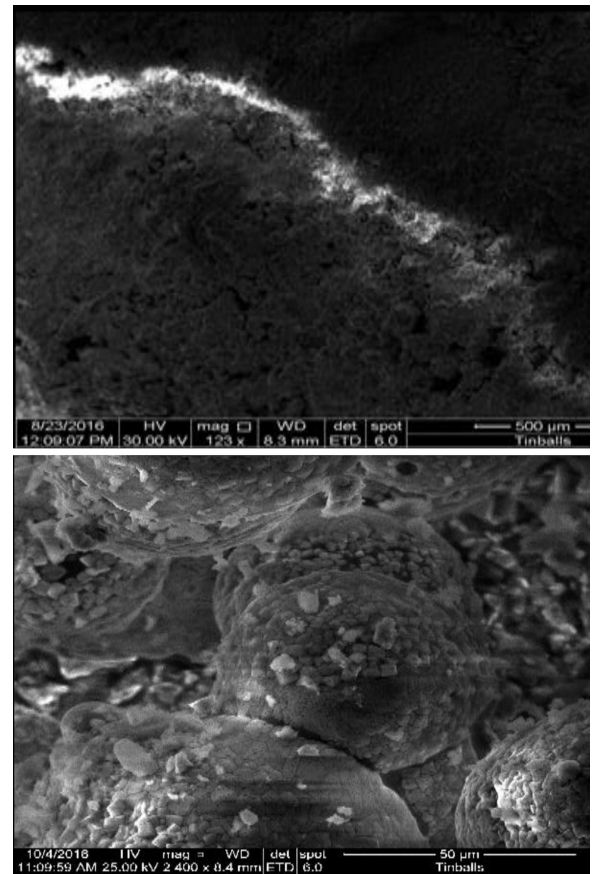


Fig. 2. SEM image of ZrSiO_4 -20% Al_2O_3 thermal spray ceramic coated on alumina brick substrate after and before copper diffusion.

TABLE I

Surface roughness and average diameter by AFM measurement

No. of group	Material [wt%]	Surface roughness [nm]	Average diameter [nm]
group 1	alumina with copper	1.55	119.62
group 2	alumina+20% SiO_2	0.88	158.77
	Al_2O_3 +20% SiO_2 with cu	1.27	80.61
group 3	ZrSiO_4 +20% Al_2O_3	1.12	148.27
	ZrSiO_4 +5% Al_2O_3 with cu	1.38	90.15
group 4	ZrSiO_4 +5% TiO_2	0.774	80.20
	ZrSiO_4 +10% TiO_2 with cu	0.881	71.30

CuAl). The light grey zones are identified with the oxides Al_2O_3 and CuO . The $\text{Cu Al}_2\text{O}_3$ phase could not be found on the surface. Thus, it can be concluded that the microstructure of the composite coating is built of three main phases with different morphologies (metallic/intermetallic phase, oxide phase and spinel phase), to avoid this, which allows us to increase the CuAl_2O_3 phase while at the same time reducing the porosity. The

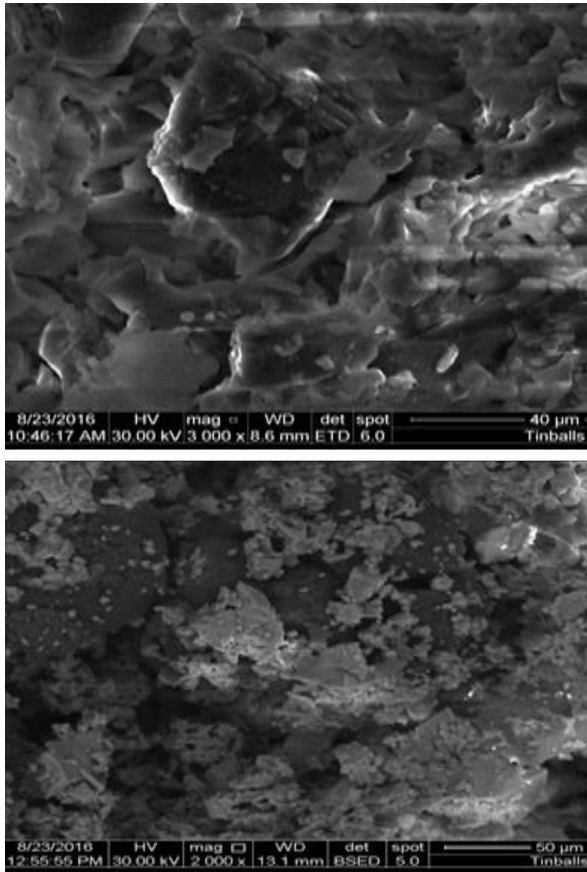


Fig. 3. SEM image of 20% TiO₂-ZrSiO₄ thermal spray ceramic coated on alumina brick substrate before and after copper diffusion.

average coating thickness was measured by a computerized optical microscope and is 40 μm and 173.669 μm, respectively.

Table II shows a comparison between the diffusion of copper values for ceramic coating specimens with the alumina brick substrate without any coating and it can be concluded that the higher value of the diffusion copper was in alumina brick substrate without coating, while ceramic coating specimens of SiO₂ with Al₂O₃, Al₂O₃ and TiO₂ with ZrSiO₄ worked barrier reduced the diffused atoms of copper especially of SiO₂ with Al₂O₃ and Al₂O₃ with ZrSiO₄ specimens.

TABLE II

EDX analysis copper counts in ceramic coating on alumina substrate.

Type of coating	EDX analysis of copper
pure alumina	46.11%
α - Al ₂ O ₃ -20% SiO ₂	1.89%
ZrSiO ₄ -20% TiO ₂	1.79%
ZrSiO ₄ -20% α - Al ₂ O ₃	0.32%

3.2. Mechanical properties

Figures 4 and 5 reveal the Vickers micro-hardness and the splitting tensile strength value of ceramic composite coating specimens with different percentages of ceramic powders (5, 10, 15, and 20%) from SiO₂ with Al₂O₃, Al₂O₃ and TiO₂ with ZrSiO₄ on alumina brick substrate compared with of pure alumina after diffusion process by copper molten, the hardness has a maximum value of at 20 wt%Al₂O₃-ZrSiO₄ content after diffusion of copper during sintering process. An increased micro-hardness value when compared to that of pure alumina will result in a material being more resistant to indentation at a given load, which will signify that the material will be able to plastically deform more so than the monolithic ceramic. The strength increases from 5.709 to 7.986 MPa with diffusion of copper for Al₂O₃ with ZrSiO₄ ceramic composite coating sintered at 1500 °C to reach the maximum value. For TiO₂ with ZrSiO₄ the strength increases from 4.41 to 5.42 MPa. This may be due to the increase in compressive strength with copper content in ceramic coating matrix. The copper diffusion into ceramic coating increase the dislocation density at the coating interface and difference in coefficient of thermal expansion.

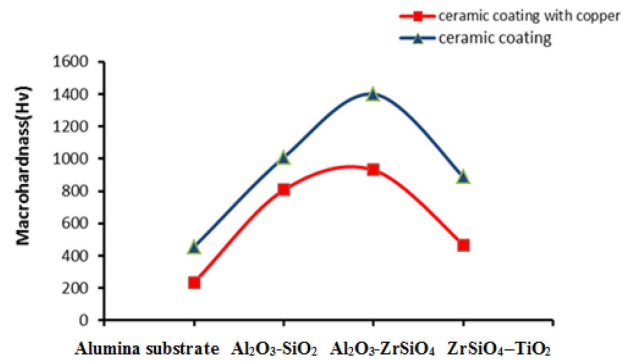


Fig. 4. Variation of Vickers hardness for ceramic strength coating on alumina brick substrate after copper diffusion.

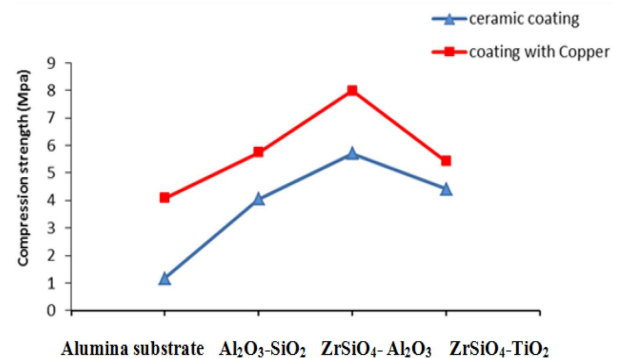


Fig. 5. Variation of the splitting tensile with different percentages of ceramic powders.

sion between the hard metal and brittle ceramic matrix within ceramic composite coating which leads to interaction stress and uniform distribution with better interfacial strength of copper ceramic coating.

3.3. Adhesion strength of ceramic sprayed coating

Table III shows the adhesion strength for the all type coated specimens. Therefore the strength of peeling is much higher than these numbers and that may be due to good adhesion between the coat and substrate which has a good roughness before coating. The higher hardness, higher adhesion strength, and lower porosity can be obtained at a lower coating thickness. Greatest adhesion (5.46 MPa) was achieved by zircon silicate with 20% Al_2O_3 (with higher alumina ratio) and alumina with 20% SiO_2 as compared to zircon silicate with 20% TiO_2 which has lower value of adhesive bonding strength (3.352 MPa).

TABLE III

Adhesion strength of ceramic coating specimens.

Type of coating	Adhesion strength [MPa]
pure alumina	1.66
ZrSiO_4 - TiO_2	3.352
Al_2O_3 - SiO_2	5.309
ZrSiO_4 - Al_2O_3	5.46

4. Conclusion

In this study the mechanical properties of coating made of reference powder increase with percentage of ceramic particles (5, 10, 15, and 20%) from Al_2O_3 with SiO_2 , ZrSiO_4 with Al_2O_3 , and TiO_2 . The hardness values for alumina- ZrSiO_4 coating are higher.

Hardness values of ceramic coated specimens with different percentages of ceramic materials are higher than that of alumina brick substrate, while hardness value for TiO_2 - ZrSiO_4 decrease with diffusion copper content. Higher value of the copper diffusion was in alumina brick substrate without coating, while composite ceramic coating specimens formed barrier reducing the content of diffused atoms copper especially at SiO_2 with Al_2O_3 and Al_2O_3 with ZrSiO_4 specimens.

The alumina coating has a high adhesive bonding strength at the interface between coating and substrate. Greatest adhesion was achieved by zircon silicate with 20% Al_2O_3 as compared to zircon silicate with 20% TiO_2 which has lower value of adhesive bonding strength.

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