Fabrication of YSZ coatings on nickel-based alloys by anodic electrophoretic deposition

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In the paper, YSZ coatings were prepared on nickel-based alloy substrates by anodic electrophoretic deposition. The YSZ suspension solution was obtained under stirring and ultrasonic treatment, in which the anhydrous ethanol and acetylacetone were used as the dispersion medium and ammonium polyacrylate was used as the dispersant of the suspension. The effects of different deposition voltage and deposition time on YSZ coating were investigated. Meantime, the microstructure of the coating surface was observed by metallographic microscope. It was found that the high-quality YSZ coating could be obtained by deposition at 60 V for 2–3 min. Finally, the effect of sintering temperature on coating quality was investigated by X-ray diffractometer and scanning electron microscopy. The results showed that the YSZ coating bonded closely with the substrate after sintering at 1200 °C, and the porosity of the YSZ coating increased after sintering

Keywords: YSZ coating; anodic electrophoretic deposition; suspension solution; nickel-based alloys.

INTRODUCTION

Ceramic coatings are widely used in electronics, aerospace, marine shipping, biology and other technical fields due to their excellent physical properties. Zirconia and some related compounds, such as yttrium stabilized zirconia (YSZ), have excellent properties such as high hardness and corrosion resistance, and are commonly used to prepare coatings¹. YSZ coating can be applied to fuel cell electrolyte (a few microns to hundreds of microns), and thermal barrier coating (about 100 μ m–300 μ m)^{2, 3}, etc. To improve the surface properties of materials (such as oxidation resistanceand corrosion resistance), the surface protection of ceramic coatings can be used to solve the problem⁴⁻⁶. Therefore, the research on the coating process has received great attention and has a huge application market.

There are various preparation methods for ceramic coatings, such as air plasma spraying (APS)⁷, electron beam physical vapor deposition (EB-PVD)⁸, sol-gel⁹ and electrophoretic deposition (EPD)¹⁰, etc. Among them, EPD is considered to be an effective technology for preparing thermal barrier coatings, which has good universality for different materials and complex structures with simple equipment and low cost¹¹⁻¹⁴. Among them, the preparation of the suspension is the key to electrophoretic deposition. Khanali et al.15 studied the effects of the solvent, zeta potential, particle size, and pH value of the suspension on the deposition process. Pantoja-Pertegal et al.¹⁶ used YSZ with different morphology and particle size to explore the influence of different suspension media on the deposition process. Borojeni et al.¹⁷ studied the aging behavior of YSZ in different non-aqueous suspensions, and found that the mixture of acetylacetone-ethanol suspension was used as the electrophoretic deposition solvent to obtain high--quality crack-free coatings. The different suspension compositions and deposition parameters have important effects on the anodic electrophoresis process. To obtain a relatively dense YSZ coating, the sintering temperature should be in the range of 1350-1500 °C, and the higher temperature was easy to cause damage to the nickel base

alloy matrix. Therefore, many researchers controlled the sintering temperature at 1100–1300 °C by adding sintering additives sintered^{14, 18–20}.

In this paper, YSZ coating was prepared by anodic electrophoretic deposition, and the effects of deposition voltage and deposition time on anodic electrophoretic deposition were studied. And the effects of sintering temperature on the microstructure of anodic electrophoretic deposition coating were explored.

EXPERIMENTAL

Preparation of YSZ coating

The nickel-based alloy with a size of 20 mm \times 20 mm \times 6 mm was used as the substrate. Firstly, the alloy substrate was ultrasonically cleaned in absolute ethanol and deionized water for 30 minutes, respectively. Then, the YSZ suspension solution was prepared. Commercially available YSZ nanopowder (8 mol% Y₂O₃, cubic,>99%, Hangzhou Jiupeng Co. Ltd, China) with spherical morphology, an average particle size of 200 nm was used as initial core particles. Ammonium polyacrylatewith with an average molecular weight of 30 million (Guangdong Wengjiang Co. Ltd, China) was put into deionized water and stirred to obtain 2.5 g/L dispersant solution. The mixture of absolute ethanol (>99.7%, Macklin, China) and acetyl acetone (ACAC,>99%, Macklin, China) was used as dispersive medium with volume ratio for 1:1, and then different amounts of YSZ powder and 0.5 ml ammonium polyacrylate solution were put into the dispersion medium to obtain suspensions with solid contents of 2.5 g/l, 5 g/L, 10 g/l and 20 g/l, respectively. The stability of suspensions with different solid contents was determined by static sedimentation method. A stable and uniform suspension was obtained after stirring for 1 h with a magnetic stirrer and sonication for 20 min. Finally, the suspension particles were deposited on the nickel-based alloy substrate using a DC regulated power. The electrophoretic deposition device used the nickel--based alloy block as the anode and the graphite as the cathode. The electrophoretic deposition was carried out at room temperature with different electric voltages

and times. The YSZ coating was dried at 120 $^{\circ}$ C for 60 min, and then the dried coating surface was sintered at 900 $^{\circ}$ C, 1200 $^{\circ}$ C and 1300 $^{\circ}$ C in nitrogen atmosphere for 2 h, respectively.

Characterization

The surface microstructure of YSZ was investigated by the optical microscope (Metallurgical Microscope, 9XB-PC, China). X-ray diffraction (XRD) patterns of YSZ samples were recorded on a Rigaku Ultima IV diffractometer operating with Cu K α radiation. The morphologies were investigated using a Scanning Electron Microscope (JEOL JSM-7800F).

RESULTS AND DISCUSSION

Through the sedimentation test of the suspension, it was found that the newly prepared suspension with a solid content of 5 g/L will not appear obvious sedimentation until 7 h, as shown in Fig. 1. When the solid content is greater than 5 g/l, the particles in the suspension will settle rapidly. To ensure the particle content in the suspension, 5 g/l suspension is the best content.



Figure 1. Sedimentation photos of 5 g/L YSZ suspension at different times

Figure 2 showed the metallographic micrograph images of YSZ coating obtained under different voltages and times. When the deposition time was set to 2 min, the change of the deposition layer was observed by changing the deposition voltage. From Fig. 2a, it can be found that the deposited YSZ coating did not completely cover the substrate at 45 V. When the voltage was increased to 60 V, the coating coverage was significantly improved (Fig. 2b). The coating partially peeled off with the voltage continued to increase to 75 V (Fig. 2c), which may be due to the turbulence phenomenon of the suspension caused by the high voltage, resulting in the rapid movement of the charged suspended particles and the rupture of the sediment layer^{21.} Thus, the deposition voltage of 60V was considered the optimal value. When the deposition time was extended to 3 min, it can be seen from Fig. 2d that the surface of YSZ coating had high coverage without cracks. When the deposition time was extended to 4 min, it can be found that the surface appeared defects. Meantime, with the increase in deposition time, the number and size of surface defects had increased significantly (as shown in Fig. 2f). The adhesion of particles on the substrate surface decreased with the increase of coating thickness, which was easy to cause particles to separate from the substrate. Thus, it can be seen that when the deposition voltage and time were 60 V and 2-3 min, respectively, the quality of the YSZ coating was good.

Figure 3 showed the SEM images of the YSZ coating obtained at 60 V for 2 min. When the coating was only dried at 120 °C for 60 min, it can be seen from the SEM image (Fig. 3a, 3b and 3c) that the image was not clear and there was a drift phenomenon during the SEM photography, which may be the bonding force between the coating surface and the substrate was not good. When the coating was continually sintered at 1200 °C for 2 h, the porosity of the coating increased significantly and cracks appeared (Fig. 3d, 3e and 3f). This is due to the cracks caused by the decomposition of ammonium polyacrylate in the coating during sintering. The porosity in Fig. 3a measured by Image J is about 11.08%, and the porosity in Fig. 3d is 23.38%. Meantime, it was found that the coating wasn't ceramized at 900 °C, indicating that the sintering temperature was too low. When the sintering temperature of the coating rose to 1300 °C, a layer of black powder appeared on the surface of the substrate without the coating and the coating on the surface of



Figure 2. Metallographic microscope images under different deposition voltages and times: (a) 45 V, 2 min; (b) 60 V, 2 min; (c) 75 V, 2 min; (d) 60 V, 3 min; (e) 60 V, 4 min; (f) 60 V, 5 min



Figure 3. SEM images of YSZ coating: (a, b, c) dried at 120 °C for 60 min; (d, e, f) sintered at 1200 °C for 2 h

the substrate with the coating felled off, indicating that the surface of the substrate was damaged at 1300 $^{\circ}$ C.

To characterize the thickness of the material, cross-sectional SEM image was conducted. From Fig. 4, it can be seen that the thickness of YSZ coating was about $20 \ \mu\text{m}$ - $30 \ \mu\text{m}$.



Figure 4. Cross-sectional SEM images of the as-sintered YSZ coatings

Figure 5 was the XRD pattern of substrate, coating before sintering and YSZ coating sintered at 1200 °C for 2 h. The subtract had the obvious peaks at $2\theta = 43.450$ °, 50.497 ° and 74.336 °, which corresponded to the (111), (200), and (220) crystal planes of Ni-Cr-Co-Mo (PDF No. 00-035-1489), respectively. The YSZ coating after sinter-



Figure 5. XRD patterns of the samples

ing had obvious peaks at $2\theta = 29.899^{\circ}$, 34.618° , 49.844° , 59.137° , 62.120° and 73° , which corresponded to the (101), (021), (110), (113), (202) and (220) crystal planes of $Zr_{0.8}Y_{0.2}O_{1.91}$ (PDF No. 00-037-1307), respectively.

CONCLUSIONS

In the paper, YSZ coatings were successfully prepared on nickel-based alloys by anodic electrophoretic deposition. Ammonium polyacrylate was used as a dispersant to make the surface of particles negatively charged, and the absolute ethanol and acetyl acetone was used as dispersive medium. The sedimentation experiment showed that the suspension with solid content of 5 g/L had good stability, and the sedimentation phenomenon will occur after 7 h. The metallographic microscope results indicated that high-quality YSZ coating without crack can be obtained at 60V for 2-3 min. The sintering experiment of the YSZ coating at different temperatures showed that the YSZ coating was still powder particles if the sintering temperature was too low, and the nickel-based alloy substrate will be destroyed if sintering temperature was too high. When sintered at 1200 °C, the YSZ coating had a porous structure, and a few cracks appeared on the surface of the coating. The SEM results show that YSZ coating thickness was 20-30 µm. This provides a possibility for anodic electrophoretic deposition of ceramic coatings.

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