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Preparation and characterization of aluminide/zirconia composite coatings by a three-step combined process

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ABSTRACT

A new process involving low-temperature pack cementation, sol-gel and electrodeposition methods was developed to prepare aluminide bond coat/zirconia composite coatings. The morphology and phase structure of the bond coat and ZrO_2 coating were examined by scanning electron microscope equipped with energy dispersive spectroscopy, thermogravimetric/differential thermal analysis and X-ray diffraction. The thermal shock resistance was further studied. The results revealed that the bond coat contained Fe_3Al, FeAl, Fe_2Al_5 and $FeAl_3$ phases and had a surface roughness of about 2.641 μm. Pore structure was observed on the surface of ZrO_2 film after sol-gel process, while dense surface without pores was detected after electrodeposition. The annealing temperature had an important influence on the surface morphology and phase structure of the ZrO_2 coating. In addition, the coating showed a good thermal shock resistance without peeling off after 40 thermal cycles.

1. Introduction

Zirconia (ZrO_2) is widely used for the fabrication of functional coatings such as hydrogen permeation barrier coatings, anticorrosion coatings and thermal barrier coatings due to its excellent physical and chemical properties [1–8]. Various methods have been adopted to prepare the ZrO_2 coatings: physical vapor deposition [9,10], chemical vapor deposition (CVD) [11–13], electrodeposition [2,14–17], plasma spray [5,6,8,18,19], ion beam deposition [20], atomic layer deposition [21], etc. However, the above methods either do not apply to complex components or the equipment required is expensive. The sol-gel method has become a promising method due to its easy control of process parameters, lower crystallization temperature, relatively low cost, and applicability of large and complex geometries [1,3,22]. Many studies have been conducted on ZrO_2 films prepared by sol-gel process [1,3,22–29]. The preparation of sols is roughly as follows: the precursor (zirconium *n*-butoxide, zirconium *n*-propoxide, zirconium acetate hydroxide, etc.), alcohol solvent (ethanol, propanol) and deionized water were mixed and a rapid hydrolysis occurred. At the same time, chelating agent (acetylacetone, acetone, etc.) was added to control the hydrolysis and the formation of zirconium hydroxide. In some studies, acidic catalyst (nitric acid) was introduced to prevent the immediate precipitation of ZrO_2 to form a stable sol. Lee et al. [1] prepared a thin ZrO_2 film on 316L stainless steel by sol-gel method. They found that the annealing temperature had an important influence on the surface morphology and crystalline structure of the ZrO_2 film. The film annealed at 500 °C for 1 h had the best quality with micro-cracks on the surface and 595 μN adhesive force. The film consisted of cubic ZrO_2. Nouri et al. [22] researched the structural evolution of ZrO_2 films at different temperatures for 50 min. A cracked film was obtained by heat treatment at 800 °C. As the temperature decreased, the films showed improved morphologies, and a crack-free and uniform film could be obtained by heat treatment at 500 °C. Meanwhile, the surface of the films became uneven as the heat treatment temperature increased.

The thermal expansion coefficient of ZrO_2 (12×10^{-6}–$14 \times 10^{-6}\,K^{-1}$) is quite different from that of 316L stainless steel (18.76×10^{-6}–$21.94 \times 10^{-6}\,K^{-1}$ between 300 and 700 °C) commonly used as a substrate in the hydrogen permeation barrier coatings [30–31]. This mismatch in thermal expansion can easily cause cracking and peeling of the coating during thermal cycling. In the preparation of thermal barrier coatings, a MCrAlY coating having a suitable thermal expansion coefficient is generally prepared between the substrate and the ZrO_2 coating [32–40]. This idea can also be used in the field of hydrogen permeation barrier coatings. The commonly used preparation methods of bond coat include atmospheric plasma spraying (APS) [32–34], electron beam physical vapor deposition (EB-PVD) [35–37], high velocity oxygen fuel spraying (HVOF) [38,39], low pressure plasma spraying (LPPS) [40], etc. However, the above methods all require expensive investments and are not suitable for complex shaped components. In addition, due to the large surface

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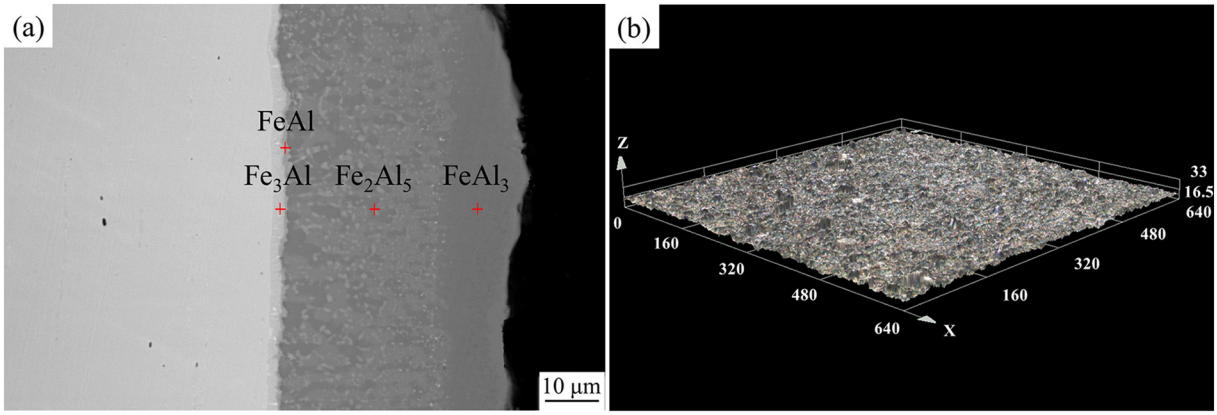

Fig. 1. (a) SEM micrograph of the cross-sectional morphology of the aluminide bond coat (b) The surface three-dimensional topography of the bond coat.

Table 1
EDS results at the positions in Fig. 1(a) in at.%.

Possible phase	Al	Fe	Cr	Ni
Fe_3Al	8.67	66.45	19.34	5.55
FeAl	41.02	37.61	12.44	8.93
Fe_2Al_5	65.94	28.70	3.88	1.48
$FeAl_3$	70.92	21.73	4.53	2.82

roughness, some bond coats require further surface modification treatment to obtain an excellent top coat [32,35,41]. Karaoglanli et al. [32] produced a high porosity bond coat by the APS process. The surface roughness was 7.34 μm, which negatively affected the adhesion of the top coat on it. Shot peening process was adopted to modify the surface layer of the bond coat, and a denser structure was obtained with lower roughness of about 3 μm. The overall quality of the composite coatings was improved. In recent years, some researchers have used pack cementation process to prepare the aluminide diffusion coating as bond coat. Because of the low cost of pack cementation equipment and suitability for complex shapes, this process has great development prospects [42,43].

The sol-gel process must be annealed to remove organic residues and obtain ZrO_2. The three phase structures of ZrO_2 are monoclinic, tetragonal and cubic. Some researchers believe that tetragonal ZrO_2 is stable at low temperatures because of the critical particle size effect of ZrO_2 grains. The tetragonal phase is stable when the grain size is smaller than the critical particle size, while the monoclinic phase can be stably present when the grain size is larger than the critical size [44]. A lot of researchers have researched the impact of annealing temperature on the ZrO_2 coating crystal transformation. Lee et al. [1] found that the ZrO_2 film was cubic phase after annealing at 500 °C, and transformed to tetragonal phase at 700 °C. Nouri et al. [22] revealed that after annealing at 700 °C, ZrO_2 coating was tetragonal phase. As the temperature rose to 900 °C, it changed to monoclinic phase. However, other researchers found that tetragonal phase ZrO_2 formed at lower temperatures. Yen et al. [45] reported that the amorphous ZrO_2 completely transformed into tetragonal phase at 573 °C. Lin and Aguilar et al. [46,47] found that tetragonal ZrO_2 began to form at about 300 °C, and started to transform into monoclinic phase at about 600 °C. In the range of 900–1000 °C, the ZrO_2 coating completely transformed into single monoclinic phase.

Considering that the thermal expansion coefficient of the iron-aluminum intermetallic compound is between those of 316L stainless steel and ZrO_2, it is used as bond coat to replace the traditional ones. Based on this, this study proposes a new composite coating preparation process used for hydrogen permeation barrier: low-temperature pack cementation process is adopted to prepare the aluminide bond coat, and ZrO_2 film is formed on the surface of the bond coat by sol-gel method.

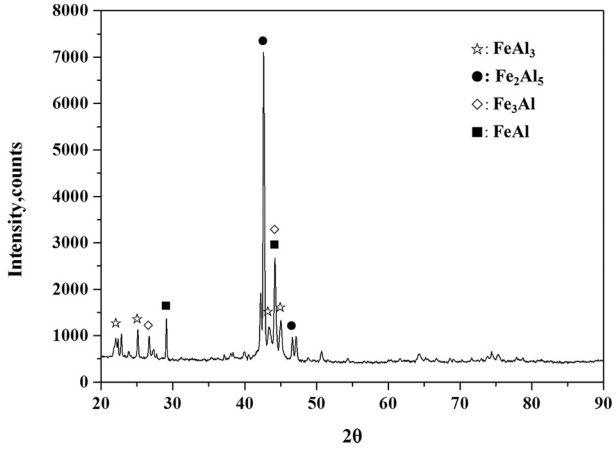

Fig. 2. X-ray diffraction pattern of the aluminide bond coat.

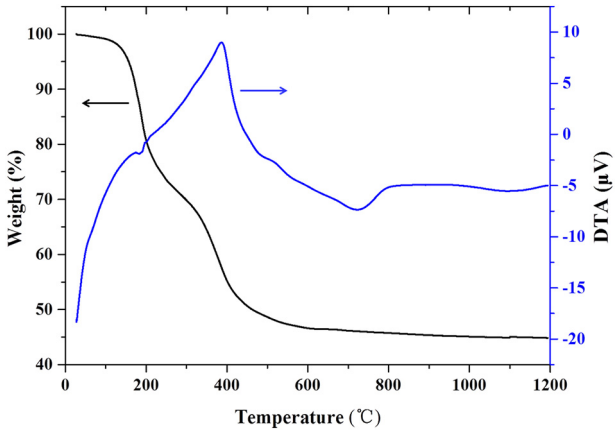

Fig. 3. TG-DTA curves for the as-dried gel.

Electrodeposition is further used to densify the ZrO_2 film since the sol-gel method usually produces pores. Low-temperature pack cementation can avoid the potential impact of high temperature on the substrate. The low-temperature pack cementation, sol-gel and electrodeposition methods are all suitable for treating components with complex shapes. This process innovatively combines the three methods to ensure its wide applicability. Composite coating of certain thickness consisted of aluminide bond coat and ZrO_2 top coating was prepared with this process. The influence of different annealing temperatures on the ZrO_2 coating was studied.