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ARTICLE

Preparation of Alumina Coatings as Tritium Permeation Barrier by a Composite Treatment of Low Temperature Plasma

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Abstract: The alumina coatings as a tritium permeation barrier were prepared on stainless steel bulk by a composite low temperature plasma method of magnetron sputtering, heat treatment and O ion implantation. The phases, surface morphologies, chemical composition and O element distribution of the coatings were characterized by XRD, SEM, EDS and AES individually, and scratch adhesion test, thermal shock resistance and tritium permeability test were also performed. The results show that the Al coatings deposited by magnetron sputtering are well, and Fe-Al alloys interlayer is formed after heat treatment. In O ion implantation process, the oxygen depth is deepened and the oxygen density gradient decreases with higher accelerating voltage at definite doses; meanwhile it can be seen that 8×10^{17} ions/cm^2 is the crucial dose, above which the oxygen density is more homogeneous. After a series of tests, the obtained coatings have the good performance of film-substrate cohesion, thermal shock resistance and the tritium permeation resistance, and the coatings, which were treated by superposition at 8×10^{17} ions/cm^2 doses, have the best tritium permeability resistance, and the trillium permeability decreases 3 orders of magnitude compared with that of the stainless steel bulk at 600 ºC.

Key words: alumina coatings; tritium permeation barrier; magnetron sputtering; O ion implantation; heat treatment

Tritium is the important nuclear material and it is widely used in the nuclear field. Because tritium has high permeability and toxicity, the tritium permeation not only causes nuclear pollution, but also wastes large sums of raw material. Tritium permeation resistance is always an important issue in the nuclear field [1]. The preparation of coatings of ceramic material on structure material with low diffusivity (so-called penetration barriers) seems to be a better practical method to reduce or hinder the permeation of tritium through the substrates. Preparation of alumina coatings as tritium permeation barrier on stainless steel is one of the research focus. During the working process, the thermal stresses were set up in film-substrate interfaces by the mismatch of thermal expansion coefficients and cause the alumina coatings shed. In order to handle this situation, the FeAl/Al_2O_3 coatings is a good way since formed FeAl alloy can decrease the thermal stresses after the Al coatings on stainless steel was heated. There are many preparation methods of FeAl/Al_2O_3 coatings, VPS-heat treatment, HAD- high temperature oxidation, PC-CVD, etc[2]. The coatings which were obtained by VPS-heat treatment crack easily, the coatings which were obtained by HAD- high temperature oxidation has bad film-substrate cohesion, while the coatings which were obtained by PC-CVD cause stress corrosion due to chloride exits in PC process[3,4]. However, the coatings, which were obtained through the way of low temperature plasma, have no disadvantages mentioned above. Meanwhile the low temperature plasma is

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harmless to substrate. It is worthy studying that $FeAl/Al_2O_3$ is obtained through the low temperature plasma on stainless steel.

In the present paper, Al thin films were deposited with magnetron sputtering process on stainless steel first and then the FeAl alloy was produced after heat treating, and finally Al_2O_3 coatings were obtained after oxygen implantation. The parameters of magnetron sputtering and oxygen implantation were investigated. After a series performance tests (such as thermal shock resistance and the tritium permeation resistance), better coatings were obtained.

1 Experiment

The 316L stainless steel samples of Φ10 mm×1 mm were polished with abrasive papers, degreased in the acetone medium and thoroughly dried. The Al coating was deposited with magnetron sputtering on stainless steel under the condition of base vacuum 2×10^{-3} Pa, temperature 100 ºC, Ar pressure 0.30 Pa. And then it was put into a vacuum firing furnace in base vacuum 6×10^{-4} Pa at 600 ºC for 180 min. In the oxygen ion implantation experimental process, base vacuum, O pressure and accelerating current were 3.0×10^{-4} Pa, 2×10^{-2} Pa and 4 mA, respectively, accelerating voltage was in the range from 30 kV to 70 kV, and implantation dose was in the range from 4×10^{17} to 1×10^{18} ions/cm^2. The phases were characterized by X-ray diffraction, which was made on the surface of the treated samples, by X'PertProMPD powder diffract metric with Ni filtered Cu Kα radiation (λ=0.15418 nm) and scintillation detector within 2θ in the range 25^o~100^o. The microstructure was observed by scanning electron microscopy (SEM). The chemical composition was characterized by EDS, which was own equipment of SEM. The film-substrate cohesion test was performed with scratch tester under different loads (50, 60, 70, 80, 90, and 100 N). The thermal shock resistance was tested in the muffle furnace repeatedly, the sample was heated to a fixed temperature (550 ºC)[5], which was the sensitizing temperature of 316L stainless steel, and then was immersed in room temperature water until the sample was cooled down completely. The Al coating thickness was measured by an eddy current method. As the oxygen implantation coatings thickness is difficult to test by conventional methods, it was analyzed by auger electron spectroscopy (AES). The coatings tritium permeability was tested by deuterium permeation measurement equipment.

2 Results and Discussion

2.1 Magnetron sputtering coatings

After magnetron sputtering, the obtained coating thickness is about 5 um with the smooth surface. As can be seen from Fig.1, the diffraction peaks corresponding to the angles 38.5°, 44.8°, 65.1°,78.2°,82.5° show the presence of Al phase in coatings. The pronounced diffraction peaks illustrate the crystalline is well, which is about 94% by the Jade analysis. As can be seen from Fig.2, the grain size is uniform with no obvious defects. From the above, it can be seen that the Al coatings quality is well.

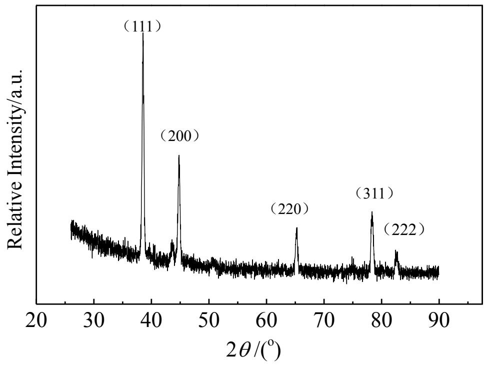

Fig.1 XRD pattern of coatings obtained by magnetron sputtering

2.2 Heat treatment coatings

After heat treatment, grazing incidence X-ray diffraction was performed, the XRD pattern of coatings is shown in Fig.3. The diffraction peaks corresponding to the angles 43.97^o, 63.94^o, 80.84^o show the presence of FeAl phase in heat treated coatings. Under high temperature heat treatment, the Al atoms of the coating diffused into Fe bulk, meanwhile the Fe atoms diffused into Al coating, and the Fe-Al alloys (FeAl, Fe_2Al_5 phase) formed after holding a certain amount of time[6].

2.3 Oxygen ion implantation coatings

2.3.1 XRD analysis

XRD pattern of oxygen ion implantation coatings is given in Fig.4. The diffraction peaks corresponding to the angles 38.5°, 44.8°, 65.2°, 78.2°, 82.5° show the presence of Al phase and the diffraction peaks corresponding to the angles 66.7°, 45.7°, 37.5° show the presence of γ-Al_2O_3 phase.

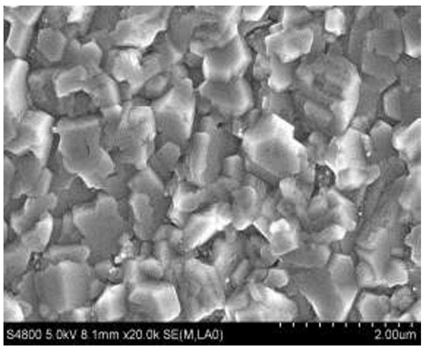

Fig.2 SEM image of coatings obtained by magnetron sputtering