Fusion Engineering and Design xxx (2017) xxx–xxx

Contents lists available at ScienceDirect

Fusion Engineering and Design

journal homepage: www.elsevier.com/locate/fusengdes

A novel low-temperature approach for fabricating α-Al_2O_3-based ceramic coating as tritium permeation barrier

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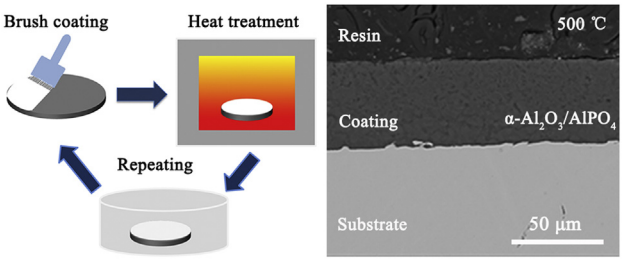
HIGHLIGHTS

- A new low-temperature method was developed for fabricating α-Al_2O_3 based coating.
- α-Al_2O_3 based coating was successfully fabricated on a steel substrate at 500 °C.
- The resulted coating was rather compact, without pores and cracks.
- The coating adhered to the substrate firmly with a considerable binding force.
- This method is cost-effective, facile, and applicable to complex geometry.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 30 October 2016
Received in revised form 21 March 2017
Accepted 23 March 2017
Available online xxx

Keywords:
Al_2O_3
Coating
Low-temperature fabrication
Fusion reactor
Tritium permeation barrier

ABSTRACT

α-Al_2O_3-based coatings have attracted great interest as promising tritium permeation barrier used in the D-T fusion reactor system. However, it is still difficult to fabricate α-Al_2O_3 coatings at a low temperature that can be endured by the steel substrate. Herein, thermal-chemical reaction method was developed for the fabrication of α-Al_2O_3-based coatings. α-Al_2O_3/$AlPO_4$ composite coating was successfully formed on the steel substrate at 500 °C. The resulted coating was uniform and compact without presence of pores and cracks. The binding strength of the coating was tested and the result indicated its good adherence to the substrate. Moreover, the coating thickness could be easily controlled. The low-temperature approach developed is expected to provide a new platform for fabricating α-Al_2O_3-based coatings as tritium permeation barrier. This approach is also applicable to the fabrication of other ceramic coatings.

1. Introduction

During the past few decades, lessening the tritium permeability of structural materials has been a critical issue in the D-T fusion reactor systems. A great many efforts have been dedicated to solve this issue [1]. According to the literatures, fabrication of ceramic coatings on the structural material surface was an effective method for preventing the tritium from permeating, owing to the low tritium permeability, high thermal stability, high electrical resistivity and high corrosion resistance of ceramic materials [2]. A wide variety of ceramic coatings have been synthesized as tritium permeation barrier, such as TiO_2, TiN, Cr_2O_3, Er_2O_3, Y_2O_3 and Al_2O_3 [3–9]. Among them, Al_2O_3 based coatings have attracted increasing attention due to their ultra high permeation reduction factor, which surpasses most of the currently obtained ceramic coatings.

Several techniques have been developed to form Al_2O_3 based composite coatings as tritium barrier, such as sol-gel method [10],

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http://dx.doi.org/10.1016/j.fusengdes.2017.03.136
0920-3796/

metal-organic chemical vapor deposition [11] and hot-dip aluminizing [12]. Al_2O_3, Al_2O_3/Cr_2O_3, and $FeAl/Al_2O_3$ coatings were successfully fabricated [10–13]. However, Al_2O_3 in these coatings was either amorphous or γ-Al_2O_3 phase [14,15]. A desirable α-Al_2O_3 phase was only achieved when the coating was annealed above 1000 °C. It is a high temperature that can not be borne by the steel substrate. Even though α-Al_2O_3/TiC composite coating was successfully fabricated by air plasma spraying after annealing at 600 °C for 30 h, a high level of porosity and cracks were also formed in the coating [16,17]. These pores and cracks can provide extra avenues for the tritium to pass through and thus greatly deteriorate its tritium permeation reduction capability.

So, how to develop a low-temperature platform, which can produce compact α-Al_2O_3-based tritium barriers, is an urgent need for the application of α-Al_2O_3 coating in the fusion reactor.

Al phosphate coatings can be fabricated by thermal-chemical reaction method at a low temperature (about 500 °C). As reported in the previous study [18], the hydrogen permeation rates for the phosphate coated specimens were much smaller than those for the uncoated ones, which concluded that Al phosphate layers could provide strong barrier effects against permeation of hydrogen isotopes through ferritic steel.

Herein, α-Al_2O_3-based coatings were fabricated by thermal-chemical reaction method. Using α-Al_2O_3 powder as the starting material and home-made $Al(H_2PO_4)_3$ as the binder, α-$Al_2O_3/AlPO_4$ composite coating was successfully synthesized at 500 °C, an ultra low temperature in comparison with those reported in other literatures. Additionally, the obtained coating was compact, free of porosity and cracks, and well adhered to the steel substrate. The coating thickness could be easily controlled. The thermal-chemical reaction method is facile, low cost and applicable to complex geometry. With these advantages, it is expected to facilitate the fabrication of α-Al_2O_3-based coatings as tritium permeation barrier.

2. Experiment

At first, a slurry was prepared by using aluminum hydroxide ($Al(OH)_3$), Chemical Reagent Ltd.), phosphoric acid (H_3PO_4, 85%, Chemical Reagent Ltd.), and α-Al_2O_3 powder (30 nm, 99.9%, Aladdin Industrial Corporation) as the starting materials. These materials were all used as obtained. Firstly, H_3PO_4 and deionized water were mixed together with a mole ratio of 1:10, and then the mixture was stirred at 80 °C in water bath for 10 min. Afterwards, $Al(OH)_3$ was added into the solution followed by further stirring for 30 min. After stirring, α-Al_2O_3 powder was mixed with the solution. Continuously stirred for about 12 h, a well-distributed homogeneous slurry was obtained eventually. Using 316L steel as the substrate, the as-prepared slurry was uniformly coated on the surface of the pre-treated substrate by brushing method. The slurry-coated specimen was baked in the oven at 60 °C for 4 h and was then annealed at 500 °C for 2 h. The thickness of the single layer coating was mainly related to the viscosity of the slurry. The coating thickness was easily increased by repeating the brush coating process.

The microstructure of the coatings both before and after annealing was determined by X-ray diffraction (XRD, a PHILIPS X'pert PRO powder diffractometer with CuKα radiation). The surface and cross-section morphology were characterized by scanning electron microscope (SEM, Nova NanoSEM 450 microscope, FEI, Netherlands). Thermal shock resistance test was performed by quickly heating the sample at 600 °C for 10 min and then rapidly quenching the sample into water at ambient environment. The binding force of the coating was tested by a pull-off method with a stretch rate of 0.5 mm/min.

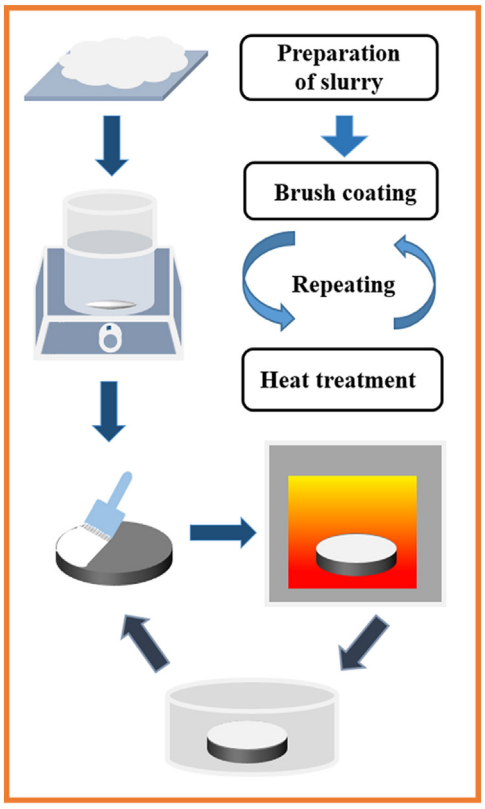

Fig. 1. Schematic diagram of the whole procedure for fabricating α-$Al_2O_3/AlPO_4$ composite coating.

The compactness of the α-$Al_2O_3/AlPO_4$ composite coating was evaluated by potentiodynamic polarization, which is usually used to test the corrosion behaviors of the materials [4,16]. The electrochemical measurements were carried out for specimens coated with epoxy resin while leaving an exposed surface area of 1 cm^2 in a three-electrode cell. The platinum was used as the counter electrode, saturated calomel electrode as the reference electrode, and the coated samples as the working electrode. The electrolyte used was 3.5% NaCl solution open to air.

3. Results and discussion

The whole procedure for fabricating α-$Al_2O_3/AlPO_4$ composite coating is schematically shown in Fig. 1, which contains three steps. First, a slurry was prepared by using $Al(OH)_3$, H_3PO_4, and α-Al_2O_3 powder. $Al(OH)_3$ and H_3PO_4 were chosen as the starting materials because the reaction between them can generate $Al(H_2PO_4)_3$. Usually, $Al(H_2PO_4)_3$ is used as an inorganic binder, which can adhere the ceramic particles together and also make the coating adhere to the substrate firmly [19,20]. Second, the as-prepared slurry was brushed manually on the surface of the 316L steel substrate to form a precursor coating. Finally, the slurry-coated substrate was baked in an oven at 60 °C and was then annealed at 500 °C for 2 h. In the baking process, $Al(OH)_3$ and H_3PO_4 in the precursor coating were effectively reacted, leading to the product of $Al(H_2PO_4)_3$, which made the coating compact and adhered the coating to the substrate. In the following annealing procedure, $Al(H_2PO_4)_3$ was converted into $AlPO_4$, resulting in the formation of α-$Al_2O_3/AlPO_4$ composite coating on the substrate surface. In the present study, the weight percentage of α-Al_2O_3 in the composite coating was about 72 wt.% and the rest consisted of $AlPO_4$. The optimal percentage of α-Al_2O_3 in the coating depended on its particle size. It was found that the weight percentage of α-Al_2O_3 could be increased to 90 wt.% when

Please cite this article in press as: H. Li, et al., A novel low-temperature approach for fabricating α-Al_2O_3-based ceramic coating as tritium permeation barrier, Fusion Eng. Des. (2017), http://dx.doi.org/10.1016/j.fusengdes.2017.03.136