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Preparation of Al_2O_3/Cr_2O_3 tritium permeation barrier with combination of pack cementation and sol–gel methods

Shuaijie Feng, Yan Wang, Cheng Zhang, Chunlan Luo, Juexin Xu, Jinping Suo*

State Key Laboratory of Material Processing and Die & Mould Technology, School of Materials Science and Engineering, Huazhong University of Science and Technology, Wuhan, 430074, PR China

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ABSTRACT

In this paper, a layered Al_2O_3/Cr_2O_3 composite coating was prepared on the surface of super-clean reduced activation martensitic (SCRAM) steel by the combination of pack cementation and sol-gel methods as a tritium permeation barrier. First, the pack cementation processes was conducted at 980 °C for 30 min to form a chromed layer and then the sample was oxidized to form Cr_2O_3 layer at 760 °C for 2 h. Subsequently, the sol-gel method was conducted on the as-prepared Cr_2O_3 coating and Al_2O_3/Cr_2O_3 composite coating was finally prepared with subsequent thermochemical treatment. The results showed the first forming Cr_2O_3 coating was consisted of a 6 μm thick Cr_2O_3 exterior layer and a 2 μm thick interior diffusion layer due to inter-diffusion of Cr and Fe. The Al_2O_3/Cr_2O_3 composite coating was a three-layered structure with a total thickness of 10.5 μm. The outer layer was Al_2O_3, the middle layer was a mixing layer consisting of Al_2O_3 and Cr_2O_3, and the inner layer was a diffusion Cr,Fe layer. Well- crystallized α-Al_2O_3 formed in the coating at 960 °C which is below the formation temperature of α-Al_2O_3 resulting from the templet effect of isostructural Cr_2O_3. The hydrogen permeation test results revealed Al_2O_3/Cr_2O_3 composite coating exhibited a good barrier property against hydrogen penetration, thus the composite coating maybe a good choice to increase the hydrogen permeation resistance of SCRAM steel.

1. Introduction

Efficient tritium permeation treatment is essential for the design of a water-cooled lithium-lead fusion reactor. In recent year, preparing a thin ceramic coating as a tritium permeation barrier (TPB) on structural metallic material has been considered a promising way to solve the problem of permeation and leakage of tritium. Many TPBs have been researched such as Cr_2O_3 [1], Al_2O_3 [2], Y_2O_3 [3] or Al_2O_3/Cr_2O_3 [4] etc.

Alumina coating has been proved a promising choice for the TPB_S application due to its good properties such as high temperature thermal oxidation resistance, good compatibility with liquid Pb-Li and the ability to suppress hydrogen isotope permeation [5]. Several preparation methods have been developed to prepare alumina coating including hot-dip aluminization, pack cementation (PC), chemical vapor deposition (CVD), physical vapor deposition (PVD), thermal spraying and sol-gel [6,7]. Among these methods, sol-gel is a low-cost and flexible method which can offer good adhesion to the substrate via the chemical bonding [8]. However, cracking or delaminating [9] is a common phenomenon in Al_2O_3 coating prepared via sol–gel due to mismatch in thermal expansion coefficient between the metal structural substrate for nuclear fusion and Al_2O_3 coating. To improve the situation, an intermediate transition layer whose thermal expansion coefficient is between Al_2O_3 and metallic matrix is often used to prepare a gradient composite coating. Besides, we often expect crystalline Al_2O_3 phase to supply better property. Among complex and variable phases of Al_2O_3, well-crystallized α-Al_2O_3 shows the best stability and tritium permeation resistance. However, the formation temperature of α-Al_2O_3 is up to 1200 °C, the elevated temperature may damage the property of metal structure substrate and cause failure such as pores and cracks in the coating due to the thermal stress. Many efforts have been made to lower the formation temperature of α-Al_2O_3 [10,11]. Using isostructural Cr_2O_3 as a template has been proposed and proved to be a promising method to stimulate the formation of α-Al_2O_3 at lower temperature.α-Al_2O_3 thin film [12] was deposited on Cr_2O_3 layer at 280–560 °C with radio frequency sputtering and the local epitaxy phenomenon was observed at the interface of α-Al_2O_3 layer and Cr_2O_3 layer. Min Zhang [13] et al. prepared α-Al_2O_3 film by low temperature thermal oxidation of Al_8Cr_5 coating at 720 °C in argon for100 h and Jun Cai [14] et al. observed the interface of α-Al_2O_3 and Cr_2O_3 prepared by oxidation of Al $_{45}Cr_7$ powders at 650–720 °C for 9–500 h in air, and the result showed that α-Al_2O_3 nucleate on the (0 0 1) facet of α-Al_2O_3

* Corresponding author.
E-mail address: jinpingsuo@hust.edu.cn (J. Suo).

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Table 1
Chemical composition (wt%) of SCRAM steel.

Composition wt.%	Cr	W	Mn	V	Ti	Si
	9.26	2.3	0.43	0.097	0.021	0.28
	P	C	S	N/ppm	O/ppm	Fe
	0.0061	0.073	0.005	150	31	Balance

directly. Thus Cr_2O_3 is supposed to be a good transition layer between the Al_2O_3 and metallic matrix.

With the above background, we prepared Al_2O_3/Cr_2O_3 composite coating on the SCRAM steel with the pack cementation and sol-gel methods. The two methods are both cost effective and can be easily applied on substrate with various shapes and sizes in fusion reactors, even for inside pipe walls, which is convenient [15]. Besides, to minimize the influence of thermal treatment on the mechanical properties of metal substrate, coatings were prepared simultaneously with the thermal treatment of the matrix steels: the Cr_2O_3 layer was prepared with pack cementation method while the first quenching was implemented and the outer Al_2O_3 layer was obtained with sol-gel methods while the second quenching was conducted.

2. Experimental

2.1. Samples preparation

The SCRAM steel was employed as the substrates on which multilayered coatings were prepared and the chemical composition is shown in Table 1. In our previous study [16], we found that austenitic grain size and martensite lath width were refined with twice-quenching and tempering processes (980 °C/0.5 h/ W.C (water cool) + 760 °C/2 h/ A.C(air cool) + 960 °C/0.5 h/W.C + 760 °C/2 h/A.C),and therefore the ductile brittle transition temperature (DBTT) of the SCRAM steel was lowered, which is benefit for fusion application. To avoid affecting the property of the substrate, the coatings were prepared simultaneously with the heat treatment of the steel: the pack cementation process was conducted at 980 °C for 0.5 h to form chromed layer, after being oxidized at 760 °C for 2 h, Cr_2O_3 coating was developed. Subsequently, the sol-gel process was conducted and the Al_2O_3/Cr_2O_3 composite coating was prepared with the second quenching and tempering processes. Before experiment, the substrates were cut into 20 mm (diameter) × 5mm(thickness)species which were subsequently polished with sandpaper to 800-grit and then were ultrasonically cleaned for 20 min in acetone and dried in a drying oven.

2.2. Preparation of Cr_2O_3 layer

The Cr_2O_3 coating was prepared via two steps including pack cementation and succedent thermal oxidation. First, the substrate was buried in the middle of an alumina crucible with a well-mixed pack powder mixture containing 48.5 wt% Cr powder as raw material, 1.5 wt % NH_4Cl as an activator and 50 wt% Al_2O_3 powder as inert filler. The particle size of chromium, ammonia chloride and alumina powders were 75 μm, 125 μm and 150 μm respectively. After being sealed with an alumina lid using high-temperature cements, the crucible was placed in air at room temperature for 24 h to dry the cements and then in an electric furnace at 80 °C for an hour to cure the cements. Afterwards, the crucible was placed into an electric muffle furnace and heated to 980 °C for 0.5 h to deposit Cr on the substrate. After the deposition processes, the packs were taken out from the furnace and cooled down to room temperature in the water. Then the chromed sample was taken out and ultrasonically cleaned to remove impurities adhered on the surface. After that, the chromed sample was oxidized at 760 °C for 2 h to obtain the Cr_2O_3 layer.

2.3. Sol-gel coating preparation

The Al_2O_3 sol was prepared by the Yoldas method [7,10]. Al-isopropoxide $Al(OC_3H_7)_3$ was chosen as the precursor (99.5 wt.% purity) and HNO_3(99.7 wt.% purity) as the peptizing agent. To form the solution, $Al(OC_3H_7)_3$ was added into a beaker containing distilled water (H_2O and $Al(OC_3H_7)_3$ in a molar ratio of 100–1) held at a temperature of 85 °C with kept magnetic stirring for about 3 h. A series of hydrolysis and polymerization reactions simultaneously occurred, resulting in an opaque solution. Then the peptizing agent HNO_3 was added to scatter the aggregated AlOOH particles by making the particles loaded the positive charge resulting from the adsorption of H^+, the molar ratio of HNO_3 to $Al(OC_3H_7)_3$ is 0.1–1, keep stirring at 85 °C,until a transparent colloidal suspension was obtained.

In order to exam the effect of Cr_2O_3 interlayer on the formation of Al_2O_3 layer, the dip-coating process was conducted both on the substrate and the as-prepared Cr_2O_3 sample to make a comparison. The samples were dipped into the sol and then withdrew at an evenly speed of 3 mm/s. Dried in air for 24 h and then in an oven at 120 °C for 3 h. According to the previous experiment, the dip-coating process can be repeated to control the thickness of the coating: The coating thickness increased by about 400 nm per dip-coating processes, and the coating exhibited the most compact structure after 5 times of dip-coating process. Thus in the experiment, dip-coating processes was repeated 5 times. Later, the coated samples were heat-treated at 960 °C for 0.5 h in the electric muffle furnace and then water-cooled to room temperature. In order to reduce thermal stress in the coating at thermochemical treatment process, the heating rates before 960 °C varied in different temperature ranges: the rate was controlled 1 °C/min in the temperature ranges of 100–200 °C and 300–500 °C, out which the heating rate was 5 °C/min, Which was decided by the results of Thermogravimetric (TG) and Differential Thermal Analysis (DTA) (Shown in Fig. 2).

2.4. Characterization

Thermogravimetric (TG) and Differential Thermal Analysis (DTA) was used to analyze the change of weight and heat with temperature during sol crystallization to formulate appropriate heating rate for sol-gel coated samples. The surface and cross-section morphologies of coatings were characterized by an environmental scanning electron microscopy(ESEM), the chemical composition and the multilayered structure were identified by an energy dispersive X-ray spectroscopy (EDS) attached to the ESEM instrument. The crystal phases was detected by X-ray diffraction (XRD) with CuKa radiation at 40 kV.

The hydrogen permeation property was tested at 25 °C with the Devanathan &Stachurski [17] two-compartment cell, which has been widely used due to its simplicity, flexibility, and high sensitivity [18]. As described in Fig. 2, the apparatus include a cathode chamber and an anode chamber. The detailed information about electrodes in Fig. 1 are listed in Table 2. Before the test, the samples were cut, grounded and polished to thin slice of Φ20 mm × 0.5 mm and then washed in deionized water, ultrasonically cleaned in acetone, dried quickly in high-pressure air stream. The samples worked as the double working electrodes were fixed between the two compartments. In the cathode chamber, hydrogen atom produce by H^+ reduction through galvanostatic polarization at a constant current density of 10 mA cm^{-2} in 0.2 mol/L KOH + 1 g/LNa_2S solution. In the anode chamber, 0.2 mol/L KOH solution was used and a constant potential of +0.25 V vs was held. The hydrogen atoms that permeate through the sample are oxidized at the substrate surface at the constant potential and the resulting anodic current was recorded using the potentiostat which is connected with a computer to collect and show the current density-time curves. During charging process, the anodic current rose until reached a steady state. The steady-state permeation current density can reflect the hydrogen flow rate permeating through samples, thus it is able to measure the hydrogen permeation resistance of the coatings.

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