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Sol–gel prepared Al_2O_3 coatings for the application as tritium permeation barrier

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ABSTRACT

One of the critical issues in test blanket module (TBM) system is to reduce tritium permeation from Pb–Li into the water/helium coolant. Use of alumina coating is one of the most promising methods to solve this problem. In the present study, well-adhered alumina coatings on ferritic steel substrates were prepared by the sol–gel method. X-ray diffraction (XRD) and field-emission scanning electron microscope (FSEM) were employed to identify the phase and examine the microstructure of the coating. The coating prepared at 500 °C is amorphous, while that prepared at 650 °C is γ-Al_2O_3. When calcination temperature is elevated to 1100 °C, α-Al_2O_3 is formed. The surface of the well-adhered coating was smooth and uniform. No spallations and cracks were observed in the coating or at the scale/alloy interface. The grain size of the coating remains very small even at 1100 °C (in the range 30–40 nm for 1100 °C). The thickness of the coating can be controlled by changing aging time and the number of dipping cycles. The flexibility to coat complex geometries by this method, even inside tubes, is guaranteed.

1. Introduction

One of the essential issues in design of the blanket is the permeation of tritium through the structural material into the secondary circuit. The reduction of tritium permeation from liquid Pb–17Li breeder and plasma into water/helium coolant, in order to reduce radiological hazard and to optimize tritium balance, is of critical importance. Besides, studies on reduced-activation-ferritic–martensitic (RAFM) steels (MANET I, F82H-mod, Optifer and EUROFER 97), indicate that steel elements Fe and Cr are strongly dissolved when exposed to Pb–17Li at 480 °C, and therefore, a corrosion resistant coating is also needed for this purpose [1–3].

Al_2O_3 protective coating has some advantages in the Pb–17Li system, because it is expected to be compatible with Pb–17Li, stable with neutron irradiation and reduce tritium permeation effectively [4,5]. Several fabrication approaches for preparing Al_2O_3 coating have been developed [6], including hot-dip aluminization, chemical vapor deposition (CVD), vacuum plasma spray (VPS), detonation jet, low pressure plasma spray (LPPS) and air plasma spray (APS). And inspiring developments have been achieved in the last few years. The efficiency of the barrier is characterized by the permeation reduction factor (PRF), which is the ratio of permeation fluxes measured without and with the barrier. It was found that the PRF ranges from 10 to 10,000 in experimental tests and some of these coatings have already fulfilled the requirement of PRF ≥1000 in gas phase [7]. However, structural imperfections of the protection layer on the surface, such as cracks and porosity, can decrease the quality of the protection layer as hydrogen/tritium molecules can penetrate through cracks to the substrate surface [8]. Pisarev's model for hydrogen permeation through a membrane covered by a protection layer with cracks predicts that minor cracking (0.001% uncoated surface) can bring the PRF down to 10 in the pure diffusion limited regime (DLR) [9]. Unfortunately, these imperfects are common [8,10,11] in fabricated coatings.

Fe–Cr–Al alloys have been also considered to be a suitable coating for the RAFM steels as a thin layer of dense, self-healing and adherent Al_2O_3 forms on the surface upon oxidation in Pb–17Li alloy [11]. Fe–Cr–Al alloys are extremely oxidation resistant even at high temperature like 1100 °C, benefiting from the formation of Al_2O_3 film [12]. In the present study, a Fe–Cr–Al alloy was selected as the base material of investigation, with which formation of Al_2O_3 protective films by pre-oxidation and sol–gel process were explored as a potential tritium permeation barrier (TPB). The purposes of pre-oxidation of the base alloy are (1) to improve the adherence of Al_2O_3 coating to the substrate; (2) to restraint crack propagation throughout the coating for safety concern; (3) to increase the thickness of the Al_2O_3 protective layer. And sol–gel technique is an effective method for preparing thin Al_2O_3 films [13,14]. The characteristics of the Al_2O_3 scales, both thermally grown and sol–gel coated, on the surface of the Fe–Cr–Al foil were examined. The application potential of such prepared TPB was also discussed.

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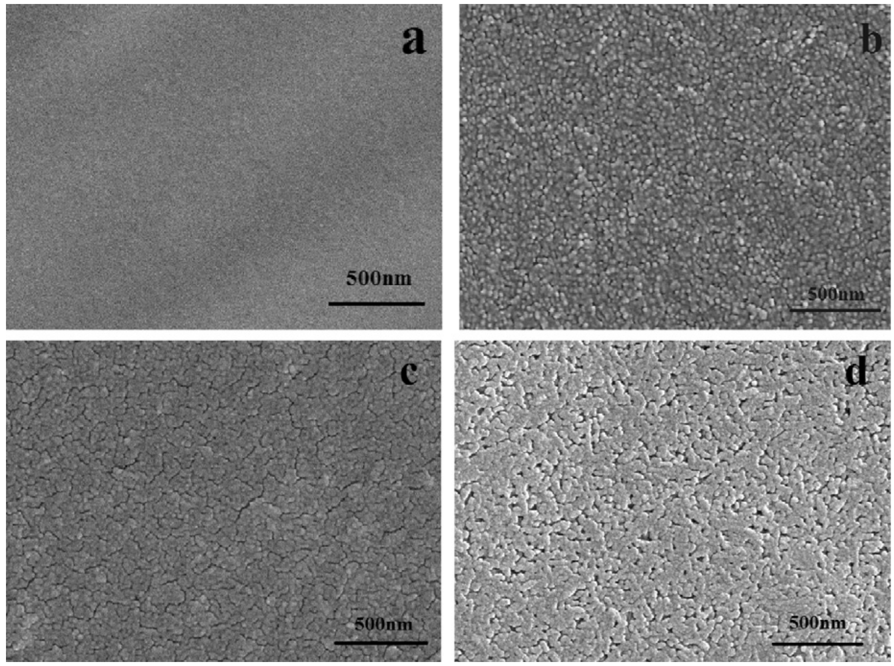

Fig. 1. SEM images of surface morphology of the coatings heat-treated for 1 h at 500 °C (a), 650 °C (b), 800 °C (c), and 1100 °C (d), respectively.

2. Experimental

2.1. Pre-treatment of metal substrates

Fe–Cr–Al alloy foils (JFE, Japan) with a thickness of 50 μm and chemical composition of Fe–20Cr–5.5Al–0.1Mn–0.08La (wt.%) were employed as the substrates on which Al_2O_3 coatings were prepared. Rectangular foil coupons (50 mm × 15 mm) were sized, and ultrasonically cleaned in acetone for 30 min and rinsed in de-ionized water to remove inorganic and organic contaminants. Pre-oxidation of the foils was carried out in air at 1100 °C for 3.5 h after being heated in a box furnace from room temperature at

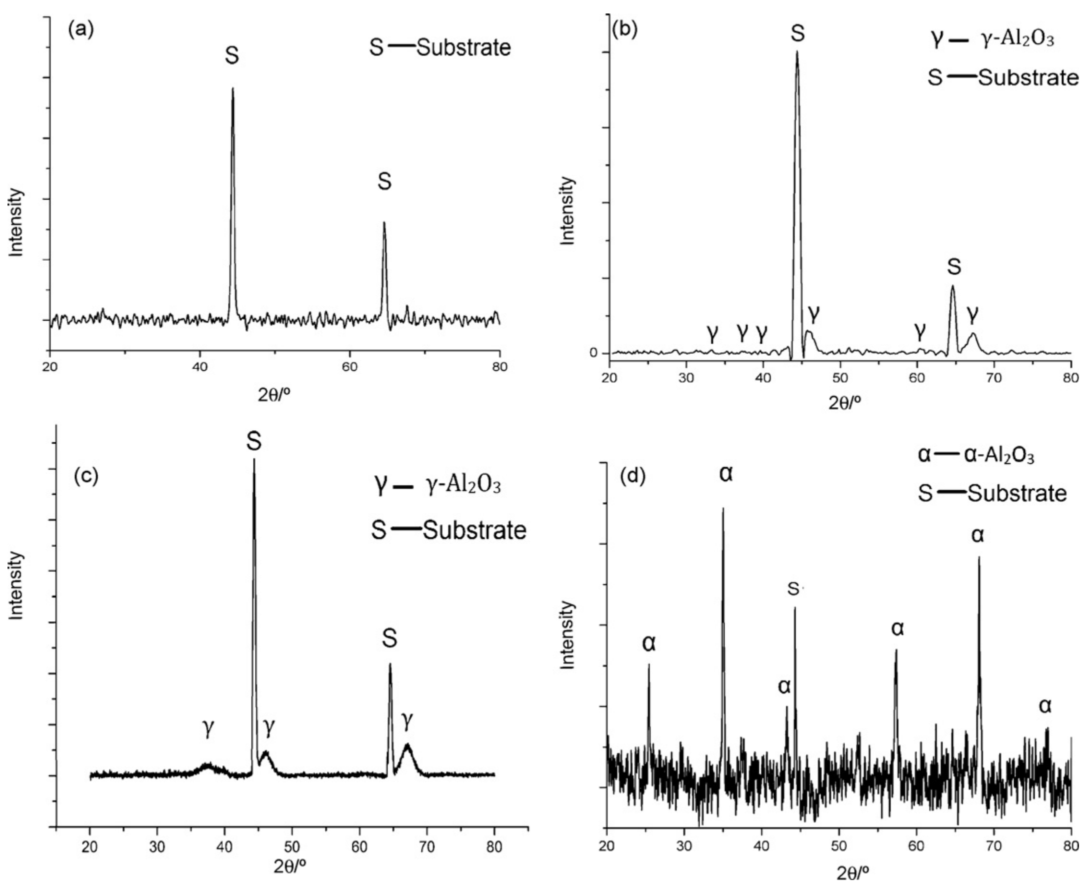

Fig. 2. Thin-film XRD patterns of coatings sintered at 500 °C (a), 650 °C (b), 800 °C (c) and 1100 °C (d); γ for γ-Al_2O_3, α for α-Al_2O_3 and S for substrate.