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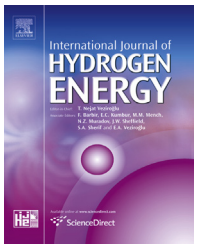

Influence of annealing atmosphere on the deuterium permeation of Y_2O_3 coatings

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ABSTRACT

In this work, Y_2O_3 coatings were deposited on 316L stainless steel substrate by metal organic chemical vapor deposition (MOCVD), followed by annealing at 700 °C in argon or hydrogen atmosphere. The effect of annealing atmosphere on the deuterium permeation inhibition properties of Y_2O_3 coatings was investigated. The coating was characterized by X-ray photoelectron spectroscopy (XPS), X-ray diffraction (XRD) and scanning electron microscope (SEM). The hydrogen permeation inhibition performance of coatings was investigated by deuterium permeation measurement. When post-annealed in hydrogen atmosphere, a $FeCr_2O_4$ layer formed at the interface as a result of element diffusion, leading to obvious increase of the permeation current from the first measurement cycle to the second. The deuterium permeation reduction factor (PRF) values of the coating were in the range of 412–102 at 500°C–700 °C in the first measurement cycle. However, the PRF values decreased to 276–43 at 500°C–700 °C in the second measurement cycle. The coating post-annealed in hydrogen atmosphere exhibited lower permeation inhibition performance than that post-annealed in argon atmosphere, which might be ascribed to the cracks and holes presented in the coating.

Introduction

Hydrogen energy and thermonuclear fusion energy are considered as the most prospective alternative clean energy. Hydrogen diffusion and permeation at high pressure are detrimental to the integrity of structural components for hydrogen storage and distribution [1–4]. In the fields of hydrogen storage devices, vacuum solar receivers and fusion reactors, etc., Type 316L stainless steel is widely used as the material of construction [5]. However, Type 316L stainless steel is susceptible to hydrogen embrittlement due to high hydrogen permeation rates at elevated temperatures. The application of tritium permeation barriers (TPBs) in the form of a coating to control tritium permeation has been widely investigated [6–21]. Among the coating materials that have been considered, Al_2O_3 is the subject of much current research because of its high permeation reduction factor (PRF) and stability at high temperature. However, in order to obtain crystalline Al_2O_3 coating, high deposition or annealing temperatures are necessary, e.g. above 900 °C [22–26]. Recently, yttrium oxide (Y_2O_3) was studied as a candidate for TPB coatings because of its high thermal chemical stability and permeation reduction factor [27–31]. The deuterium permeation inhibition performance of Y_2O_3 coating can be as good as Al-based coatings [32,33].

The hydrogen isotope permeation property may not only relate to material itself, but also depend much on the

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microstructure of coatings [34–36]. Since hydrogen isotope diffusion in materials is often governed by structural imperfections, the preparation of crack-free and dense coating is essential to achieving coating with excellent permeation inhibition performance. Therefore, it is necessary to analyze the microstructure of coating to understand the formation mechanism of imperfections, and then find their effect on the permeation behavior of hydrogen isotopes.

Until now, previous studies has been mainly focused on understanding the detrimental effect of hydrogen isotope containing atmosphere on metal materials such as stainless steel [2,3]. However, less attention has been dedicated to figuring out the effect of microstructure evolution in different atmospheres on the hydrogen isotope permeation inhibition behavior. TPB coatings often work in hydrogen isotopes-containing harsh environments, including high-temperature and high-pressure hydrogen isotopes, which would incite microstructure change. For ensuring the effectiveness of TPB coatings, the performance evolution in working environments should be considered. In this work, Y_2O_3 coatings were deposited on 316L stainless steel by metal organic chemical vapor deposition (MOCVD), followed by annealing at 700 °C in different atmospheres. In order to investigate the microstructure evolution of Y_2O_3 coating in working environment systematically, high-temperature annealing in hydrogen atmosphere was used to simulate the working environment. For comparison, high-temperature annealing in argon atmosphere was used to simulate the ideal environment. Deuterium permeation measurement was used to evaluate the effect of microstructure change on the deuterium permeation properties of coatings.

Experimental

Material synthesis

Deposition of the Y_2O_3 coatings was performed by MOCVD in a horizontal hot wall reactor, which is described elsewhere [37]. Type 316L stainless steel disks of 29 mm in diameter and 0.5 mm in thickness were used as substrates. Yttrium β-diketonates organometallic ($Y(tmhd)_3$, Alfa AESAR) was used as precursor. $Y(tmhd)_3$ was heated at 150 °C to evaporate. H_2 was used as carrier gas with a flow rate of 20 sccm. Oxide coating deposited by using organometallic substances often contain some amount of carbon due to incomplete decomposition of organic precursor [38,39]. It was reported that water vapor could accelerate removal of organic product by forming volatile hydrocarbon complexes, thus facilitating formation of carbon-free coatings [40]. Therefore, the carrier gas H_2 was mixed with water vapor by flowing through a water bubbler before arriving at the precursor evaporation zone. The Y_2O_3 coatings were deposited at 500 °C for 2 h with a chamber pressure of 1.2–1.4 kPa. In order to study the effect of different environment on the microstructure evolution and permeation properties of the coatings, the as-deposited coatings were post-annealed at 700 °C in argon or hydrogen atmosphere. The as-deposited coating, the coating annealed in argon and in hydrogen atmospheres on 316L stainless steel substrates were denoted as S1, S2 and S3, respectively.

Material characterization

The crystal structure of the coating was analyzed by grazing incidence X-ray diffraction (GIXRD, Rigaku-D/max2500). The surface morphology of the coatings was analyzed by scanning electron microscopy (SEM,Hitachi-S4800). The chemical composition and binding states were examined by X-ray photoelectron spectroscopy (XPS, PHI Quantera SXM). Deuterium permeation measurements were conducted by a gas permeation self-made apparatus. Before the deuterium measurement, the uncoated side of 316L stainless steel was polished to eliminate the influence of surface oxide layer formed during thermal processing. The permeation chamber was divided by the coated disk into two parts: the upstream and the downstream chamber. The coated side of the sample was mounted facing the upstream side. Deuterium was used as permeation gas and introduced into the upstream chamber at 40–100 kPa using a needle valve. The pressure of deuterium was monitored by a quartz vacuum gauge (10–100 kPa, DL-10,Beijing Xinhengjiu Tech.). During the permeation measurement, the downstream chamber was continuously pumped to maintain the pressure at 2×10^{-5} Pa. The pressure of the downstream chamber was measured by an ionization gauge (6×10^{-8}–10^{-1} Pa, DL-7, Beijing Xinhengjiu Tech.). The flux of deuterium permeating through the sample to the downstream chamber was measured by a quadrupole mass spectrometer (QMS, Hiden HPR30). The measurement temperatures for deuterium permeation were 500°C–700 °C. The measurement procedure was described in detail elsewhere [37].

Results and discussion

The GIXRD analysis results of the Y_2O_3 coatings were presented in Fig. 1. Here, the incident angle of GIXRD was set at 1°. After post-annealed at 700 °C, the diffraction peaks intensified

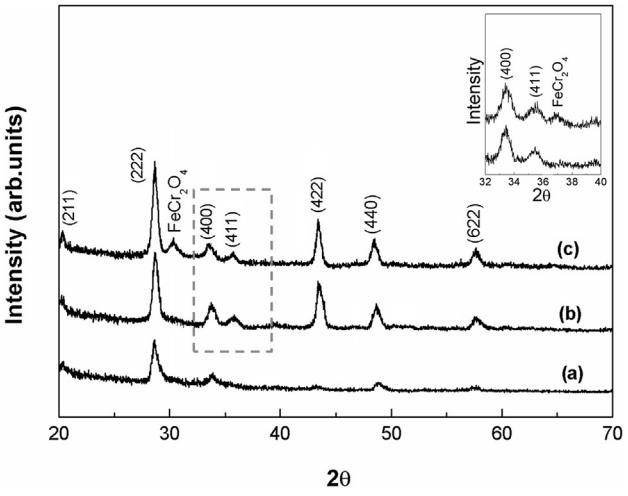

Fig. 1 – X-ray diffraction patterns of the Y_2O_3 coatings: (a) S1, (b) S2 and (c) S3. The inset showed the enlarged XRD pattern with diffraction angle 2θ between 32° and 40° marked by rectangle.