Fusion Engineering and Design 89 (2014) 2755–2758

Contents lists available at ScienceDirect

Fusion Engineering and Design

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

Preparation and characterization of zirconium films for first mirror application in fusion devices

Wei Liu, Chengwei Wen, Xinggui Long*, Xiaohong Zhang, Jinhua Liu

Institute of Nuclear Physics and Chemistry, China Academy of Engineering Physics, Mian Yang 621900, China

HIGHLIGHTS

- The nanostructured Zr films deposited on Al_2O_3 substrate are obtained by pulsed laser deposition (PLD) for the application of first mirror.
- The Zr films showed a very smooth surface and low values of root mean square roughness (RMS).
- The high reflectivities make it one of great candidate of first mirror materials for the applications of nuclear fusion.

ARTICLE INFO

Article history:
Received 20 February 2014
Received in revised form 12 June 2014
Accepted 27 July 2014
Available online 27 August 2014

Keywords:
Nuclear materials
Laser processing
Zr
Thin films
Specular reflectivity

ABSTRACT

The nanostructured zirconium (Zr) films deposited on Al_2O_3 substrate are obtained by pulsed laser deposition (PLD) for the application of first mirror. Structural features, optical properties and surface morphologies of as-grown Zr films are systematically investigated as a function of pulse repetition rate. It is found that the Zr films show a typical hexagonal close packed structure and all deposited films exhibit a very smooth surface. There are no voids and folds on the surface of Zr films. The root mean square roughness (RMS) values increase with increasing pulse repetition rate. The variation of pulse repetition rate has no obvious effects on the reflectivity because of the smooth film surface. Up to the wavelength of 800 nm, the reflectivity is higher than 70%, which is excellent for the application of first mirror.

1. Introduction

In order to control the nuclear fusion process, various metallic films are developed as diagnostic tools in Tokamak apparatus. Diagnostic mirrors are foreseen as necessary optical components in ITER diagnostic systems. Due to the harsh environment in the fusion reactor, mirrors placed on the first wall are subjected to erosion by sputtering, high temperature and neutron flux [1]. Materials with good optical performance (high reflectivity) and low sputtering yield have drawn a great attention for the application of nuclear fusion. Rhodium films [2,3] and molybdenum thin films [4] prepared by PLD have been investigated for first mirror. Specular reflectivity of rhodium films deposited by PLD can reach the value of 75% at 800 nm [2,3].

Compared to other deposition methods, PLD is considered as a versatile technique that is suitable for growing extremely pure thin films [5]. By varying the laser parameters (laser fluence, pulse repetition rate) and experimental conditions (substrate temperature), it is possible to obtain desired crystalline structure and film surface [5–7].

Thanks to its properties such as low thermal neutron cross-section under ambient temperature and atmospheric environment, exceptional corrosion resistance under high temperature, Zr is one of the ideal candidates for the exploration of mirror thin films [8,9]. Some other deposition techniques, such as sputtering or evaporation have been applied to investigate the Zr thin films with various properties, including special ω phase (at room temperature and high pressure (above 2–8 GPa), an ω phase with a three atoms hexagonal structure could be formed [10,11]), outstanding microstructure, and mechanical properties [12–15]. To the best of our knowledge, no study has been reported on the Zr films deposited by PLD. Furthermore, the optical properties of Zr films are also not clear. In this paper, the influences of pulse repetition rates

* Corresponding author. Tel.: +86 186 8169 5416/2496715; fax: +86 15281199125.
E-mail address: liuweizigong@163.com (X. Long).

http://dx.doi.org/10.1016/j.fusengdes.2014.07.023
0920-3796/

on the surface morphologies and optical properties of Zr films are explored. The aim of this study was to find appropriate conditions of PLD in order to obtain a pure film with good optical properties for tokamak exposure.

2. Materials and methods

In our experiments, Zr films approximately 75–100 nm were deposited by PLD on the rolled sapphire (Al_2O_3 (0 0 0 1)) substrates. As a comparison, Si (1 1 1) substrates were introduced to these experiments. A nanosecond laser pulse (KrF excimer laser: $\lambda = 248$ nm, pulse duration 25 ns, laser fluence 6.5 J/cm^2) was focused on the Zr target (purity 99.9%) at an angle of 45°. The pulse repetition rate (2, 6, 10 Hz) was changed to explore the reflectivity, roughness and morphology of as-deposited films. The deposition was carried out at base pressure of about 1×10^{-5} Pa and substrate temperature of 200 °C. To reduce the drilling of target, the Zr target was also rotated continuously at a constant rate of 12 revolutions per minute. Before the deposition process, the target was cleaned by 2000 laser shots to remove the surface contaminations.

The microstructures of the as-deposited Zr films are identified by grazing-incidence X-ray diffraction (GIXRD). The morphologies were characterized by scanning electron microscope (SEM) and atomic force microscopy (AFM). A UV–vis–NIR PerkinElmer Lambda 1050 spectrophotometer was used to obtain the reflectivity of Zr films.

3. Results and discussions

Fig. 1 shows the X-ray diffraction patterns obtained from Zr films deposited at three pulse repetition rates. According to the identified diffraction lines, Zr films exhibit a polycrystalline hexagonal close packed structure, which matched the JCPDS file (where, JCPDS is an abbreviation of Joint Committee on Powder Diffraction Standards). Compared with a standard bulk Zr (JCPDS pattern #05-0665), an apparent preferential orientation in the (1 1 0) and (0 0 2) directions are observed for all the films. The film deposited at 10 Hz exhibited the highest (0 0 2), (1 0 1) and (1 1 0) peak intensities and the minimum values of full width at half maximum (FWHM), indicating a higher crystallinity of the film compared with other films. In the process of deposition, high energy particles produced by laser change the pathways of nucleation and migration of adatoms, leading to the apparent preferential orientations. High energy introduced by higher pulse repetition rate can enhance the migration and diffusion of the deposition atoms on the surface of substrate [16]. With increasing pulse repetition rates, the migration and the diffusion of the atoms in the films will release the strain in the film, and thus decrease the compressive stress.

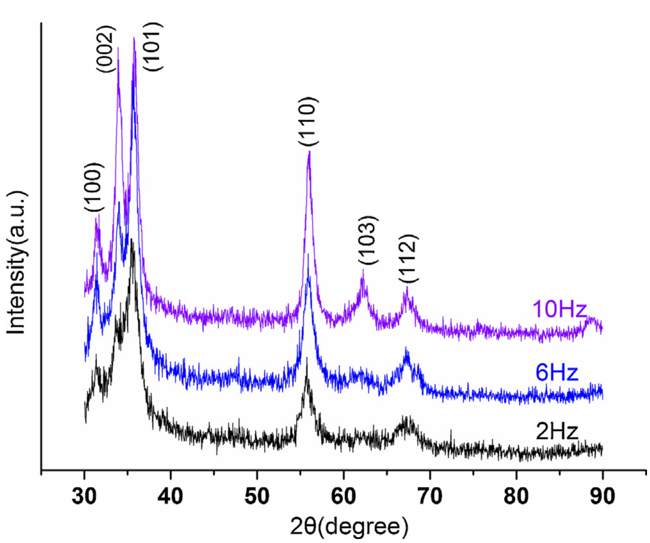

Fig. 1. XRD pattern of Zr films with various pulse repetition rates.

The surface morphologies of Zr films with various pulse repetition rates are depicted in Fig. 2(a)–(f). On the top view, all films show very smooth and compact surfaces, which are consistent with the XRD analysis. Although the presence of some small droplets was observed in Fig. 2(a)–(c), the smooth film surface domains constituting the film are clearly visible. Compared to the Mo films [4] and Rh films [2] deposited by PLD, the Zr films reveal a smoother film surface and less liquid droplets. Due to a complex physical process during the target ablation and plasma plume expansion, the presence of droplets is inevitable. In general, the liquid-phase under the action of vapor and plasma pressure could be the origin of droplets [17]. However, the relationship between laser parameters and the dimensions and quantities of droplets is still not clear.

The AFM images (scan areas of size 1 μm × 1 μm) of Zr films are showed in Fig. 2(d)–(f). The RMS roughness of the films under different pulse repetition rate is depicted on the corresponding AFM image. The RMS roughness increases with increasing pulse repetition rate. In a suitable range of pulse repetition rate, lower pulse repetition rate can cause less island density, increased average island size and the diminished surface roughness [18]. In Fig. 2(d), the film deposited at 2 Hz revealed a lower RMS roughness value. A higher RMS roughness obtained for Zr films grown at other pulse repetition rates can be ascribed to its enhanced pulse repetition rates. The higher energy and deposition rate caused by pulse laser make the film exhibit a smooth morphology [19]. All RMS roughness (2 Hz: 0.57 nm; 6 Hz: 0.6 nm; 10 Hz: 1.2 nm) showed in Fig. 2(d)–(f) reveal that the surface of Zr films is very smooth. From the observations obtained from SEM and AFM images, we predict that the optical properties of as-deposited Zr films can meet the fundamental requirement of first mirror.

As we have already mentioned, the optical properties are an important issue for applications of these films in fusion experiments. We then investigate the reflectivity of Zr films. Specular reflectivity of: (a) Zr films deposited at different pulse repetition rates and (b) films grown on different substrate are depicted in Fig. 3. In Fig. 3(a), the specular reflectivity of these smooth Zr films are shown versus the incident beam wavelength in the range 200–800 nm. It can be seen that the values of specular reflectivity increase with increasing the wavelength and the specular reflectivity is higher than 70% at the wavelength of 800 nm. The results obtained in Fig. 3(a) are in good agreement with Rh films [2,3] but higher than Mo films deposited on SS substrate by PLD [3]. To verify the influence of substrates, Si (1 1 1) is used as a comparison. In Fig. 3(b), the reflectivity values obtained with different substrates are very close. However, the values obtained from Al_2O_3 (0 0 0 1) are higher than the values obtained from Si (1 1 1). It is known that the substrate that has a similar thermal expansion coefficient or same crystal structure with the film materials can produce a smoother surface and higher reflectivity. In above experiments, Al_2O_3 and Zr are hexagonal close packed crystal structure. However, Si is face centered cubic structure. Compared to the thermal expansion coefficient of Si (4.2×10^{-6} K^{-1}), Zr (5.78×10^{-6} K^{-1}) is closer to the thermal expansion coefficient of Al_2O_3 (5.8×10^{-6} K^{-1}). Thus, a smoother surface is obtained with Al_2O_3.

In fact, it is possible to calculate the specular reflectivity with the well-known Bennett's formula [20], which correlates the specular reflectivity at normal incidence with the surface roughness:

$$R_s = R_0 \exp^{-\left(\frac{4\pi \times \mathrm{RMS}}{\lambda}\right)^2}$$