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Deuterium plasma exposure of rhodium films: Role of morphology and crystal structure

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HIGHLIGHTS

- Rh films with different morphologies and crystal structures were deposited.
- Rh films were exposed to a deuterium plasma to investigate erosion properties.
- Rh film morphology and crystal structure affected its performance during exposure.
- Granular Rh film with few nm crystallite size prevents reflectivity degradation.

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ABSTRACT

The behavior of rhodium film mirrors with different crystal structure and morphology toward a deuterium plasma is presented. The specular reflectivity of rhodium films was monitored before, during and after exposure. To understand the reflectivity behavior of the rhodium films during exposure, samples were characterized by scanning electron microscopy, X-ray photoelectron spectroscopy and atomic force microscopy. Crystal structure and morphology of rhodium films strongly affect the change of the specular reflectivity during deuterium plasma exposure. In particular, films with few nm crystallite size and granular-like morphology prevent the reflectivity degradation, probably as a consequence of the inhibition of rhodium deuteride sub-superficial layer formation.

1. Introduction

Metallic mirrors will be crucial components of all optical systems for plasma diagnostics and imaging tools in ITER [1]. They must survive a harsh environment consisting of intense thermal loads, strong radiation fields and high particle fluxes. Two main effects of these extreme conditions are the erosion of the mirror surface by charge exchange neutrals and the re-deposition of the sputtered material transported from the first wall onto its surface. These phenomena could lead to a dramatic decrease of the specular reflectivity of the mirror, posing a serious threat on the operation of the entire diagnostic system.

Various mirror types have been investigated so far, including polycrystalline, monocrystalline and nanostructured thin film mirrors [2]. The former choice appears to be the simplest solution in terms of cost and availability of the production techniques. However polycrystalline mirrors develop a complex step relief pattern when eroded by an ITER-relevant plasma [3,4]. This behavior has been attributed to the difference of the sputtering yield of grains oriented differently with respect to the surface. A monocrystalline mirror or a mirror composed of crystallites with nanometric size should guarantee a more uniform erosion during tokamak operation (the surface relief pattern maintains small with respect to the wavelength of the reflected light, thus ensuring a low diffuse reflectivity) and ultimately a more reliable diagnostic system. Since both high production costs and technical difficulties prevent the manufacture of monocrystalline mirrors with diameters larger than 10 cm, thin film technologies can offer a cheaper and feasible solution for ITER scientists and engineers.

One of the principal materials considered for the production of thin film mirrors is rhodium (Rh), thanks to its high reflectivity in a wide wavelength range and acceptable sputtering yield. Recently, two techniques have been exploited to produce nanostructured Rh film mirrors: magnetron sputtering [5,6] and pulsed laser deposition (PLD) [7,8]. The former permits to obtain planar films on large and complex shaped areas and first mock-ups have already

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been produced [9], whereas the latter guarantees a detailed control on both morphology and nanostructure of the deposited films. Exploiting these techniques, it is possible to obtain films with a great variety of morphologies and crystal structures, in terms of crystallographic orientation and crystallite size (i.e. the sub-grains dimension measured by X-ray diffractometer) from a few nanometers to ~30 nm. A major open issue is the behavior of Rh film mirrors in erosion dominated zones as a function of their morphology and crystal structure.

In this work, four Rh film mirrors with different morphologies and crystal structures were produced by magnetron sputtering and PLD, and exposed to a laboratory deuterium plasma. Such an approach permits to conduct a dedicated *in situ* analysis of the physical problem of mirrors under erosion during tokamak operation. The specular reflectivity of the rhodium films was measured both before and after plasma exposure by a spectrophotometer and was monitored during the exposure by an *in situ* reflectometry system [10]. In order to understand the changes in the reflectivity of the Rh films under deuterium ions bombardment, the samples were characterized by scanning electron microscopy (SEM), atomic force microscopy (AFM) and X-ray photoelectron spectroscopy (XPS).

In Section 2, the preparation methods of the Rh films, the deuterium plasma exposure facility and the characterization techniques are presented. The experimental results, focused on the effect of the deuterium plasma exposure of Rh films on their optical properties, morphology changes, and chemical composition, are shown in Section 3 and discussed in Section 4. Concluding remarks are presented in Section 5.

2. Experimental

One Rh film was produced by the magnetron sputtering technique, while three films by the PLD method. The latter were deposited in different experimental conditions in order to obtain various morphologies and crystallite size. All the samples were deposited on stainless steel-316L (SS) substrates (25 mm diameter, 2 mm thick) which were mechanically polished (first by abrasive SiC paper, then by diamond paste, and finally by an alumina powder of 0.05 μm particle size).

Details about the complete deposition procedure and characterizations of the magnetron sputtered Rh film, henceforth referred to as “MS”, are given in [5,6]. In summary, the average crystallite size was around 10 nm, the crystallites did not show any specific texture or preferential orientation and SEM cross-section analysis revealed dense columnar structure.

The production method of the PLD Rh films is described in [8]. The first PLD Rh coating was deposited in high-vacuum (3×10^{-3} Pa) and is labeled as “columnar” in the rest of the text, because its SEM cross-section reveals a columnar structure [8]. The deposition of the second and the third kind of samples, labeled as “NC1” (“NC” stands for nanocrystalline) and “NC2”, was performed in helium atmosphere at 40 Pa and 55 Pa, respectively. The laser fluence and the distance between the target and the substrate were fixed to 10 J/cm^2 and 70 mm. In order to guarantee a high mechanical stability for NC1 and NC2 coatings, a Rh columnar adhesion layer was deposited directly above the stainless steel substrate [8]. To investigate the crystallite size and orientation of the PLD Rh films, they were characterized by X-ray diffraction (XRD) using a SIEMENS D5000 instrument with monochromatic Cu Kα radiation at a glancing incidence (with different angles of the X-ray source) and using a Panalytical X’Pert Pro X-ray diffractometer with monochromatic Cu Kα radiation in $\theta/2\theta$ configuration. The mean crystallite size, estimated using the Scherrer formula from the grazing XRD spectra, for columnar, NC1 and NC2 Rh films was 12 nm, 8 nm and 6 nm, respectively. The $\theta/2\theta$ XRD spectrum revealed how the columnar sample was highly-orientated along the (111) direction. This feature, although still present, became less evident for NC1 and NC2 specimens. The ratio between the areas of peaks (111) and (200) was 217.1, 31.4 and 2.9 for columnar, NC1 and NC2 Rh films, against 2.1 for the bulk.

To investigate Rh films morphology, the four samples were analysed by a Hitachi S-4800 field emission SEM (acceleration voltage 5 kV). Fig. 1 shows the SEM top-view images for MS (a), columnar (b), NC1 (c) and NC2 (d) Rh films, where compact film morphologies can be observed. In the case of the NC2 film, it is also possible to notice the presence of a slight open structure or “granular-like” morphology. Moreover, a crack network around the scratches of the polished stainless steel substrate can be observed (Fig. 2a). This is the result of the combination between the tensile stress present in the film and its brittle nature [8]. A typical crack width is of some tens of nanometers (Fig. 2b).

The details of the magnetron sputtered and the PLD samples are summarized in Table 1.

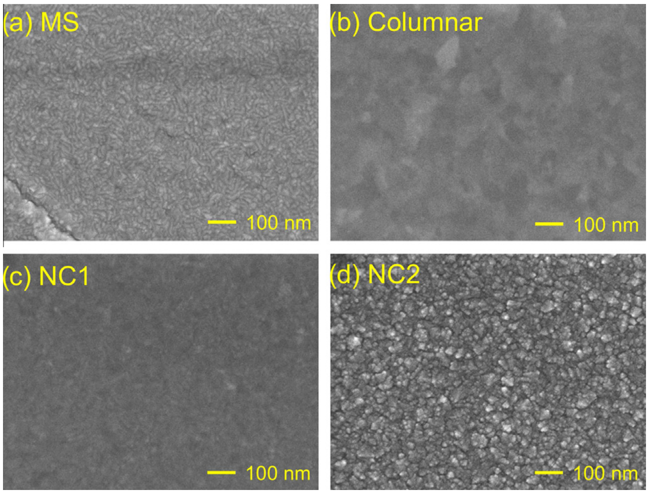

Fig. 1. SEM top-view images for: MS (a), columnar (b), NC1 (c), NC2 (d) Rh films as-deposited.

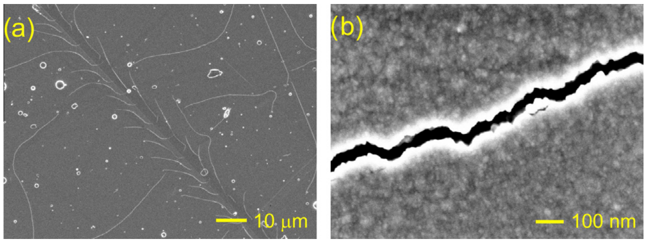

Fig. 2. SEM top-view images of the NC2 Rh film at two different magnifications. (a) Crack network close to a scratch of the polished stainless steel substrate. (b) Magnified image around a crack.

Table 1
Samples and their characteristics: deposition technique, mean crystallite size (nm) and morphology.

Sample name	Deposition technique	Mean crystallite size (nm)	Morphology
Columnar	PLD	12	No features
MS [6]	Magnetron sputtering	10	No features
NC1	PLD	8	No features
NC2	PLD	6	Granular-like