Applied Surface Science 443 (2018) 68–73

Contents lists available at ScienceDirect

Applied Surface Science

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

Full Length Article

Damage evaluation of proton irradiated titanium deuteride thin films to be used as neutron production targets

Manuel Suarez Anzorena [a,b], Alma A. Bertolo [a,b], Leonardo Gagetti [a,c], Pedro A. Gaviola [a,b], Mariela F. del Grosso [a,c,d,*], Andrés J. Kreiner [a,c,e]

[a] Gerencia de Investigación y Aplicaciones, Comisión Nacional de Energía Atómica, Av. Gral Paz 1499, B1650KNA San Martín, Buenos Aires, Argentina
[b] Instituto Sabato, Universidad Nacional de San Martín (UNSAM) – Comisión Nacional de Energía Atómica (CNEA), Buenos Aires, Argentina
[c] Consejo Nacional de Investigaciones Científicas y Técnológicas (CONICET), Rivadavia 1917, C1033AAJ Ciudad de Buenos Aires, Argentina
[d] GRUCAMM, Universidad Tecnológica Nacional Gral. Pacheco, H. Yrigoyen 288, B1617FRP, General Pacheco, Buenos Aires, Argentina
[e] Escuela de Ciencia y Tecnología, Universidad Nacional de San Martín (UNSAM), B1650HMQ San Martín, Buenos Aires, Argentina

ARTICLE INFO

Article history:
Received 3 January 2018
Revised 20 February 2018
Accepted 22 February 2018
Available online 23 February 2018

Keywords:
Titanium deuteride films
Neutron production targets
Irradiation
X-ray diffraction
LIBS
ERDA

ABSTRACT

Titanium deuteride thin films have been manufactured under different conditions specified by deuterium gas pressure, substrate temperature and time. The films were characterized by different techniques to evaluate the deuterium content and the homogeneity of such films. Samples with different concentrations of deuterium, including non deuterated samples, were irradiated with a 150 keV proton beam. Both deposits, pristine and irradiated, were characterized by optical profilometry and scanning electron microscopy.

1. Introduction

Research and development of neutron production targets through nuclear reactions, induced by charged particle beams provided by high power accelerators, can be used as a powerful tool in numerous fields where neutron beams are required. Some of those fields are: accelerator based boron neutron capture therapy (AB-BNCT) [1], homeland security, non-proliferation and in particular the accelerator driven systems (ADS) which are being developed to be used for the incineration of nuclear wastes throughout a transmutation process as well as for advanced application in the power generation [2,3], among many other medical and nuclear applications [4,5].

Neutron production targets are being studied around the world and in particular in Argentina as part of a project underway to develop high current accelerators for medical and nuclear applications [6]. One of the main characteristics of these targets will be the ability to withstand the radiation and hydrogen damage induced by the deuteron beam's high fluence [7]. In particular, when the D(d,n) reaction is considered, the development of TiD_2 targets is an important matter in the nuclear materials development and in neutron production.

Ti and TiD_2 thin films generation and characterization have been extensively studied and reported [8–10]. Irradiation of such films has been carried out using fluences of up to 1.2×10^{17} ions/cm^2 [11,12]. In this work, we show the results of the development of TiD_2 thin films and their behavior under proton irradiation with fluences of up to 3×10^{18} ions/cm^2. To study the irradiation damage, non irradiated and irradiated films were characterized by several techniques, and irradiation simulations were performed.

2. Materials and methods

In order to obtain TiD_2 thin films, Ti deposits were made using a physical vapor deposition (PVD) process on two different substrates: 1050 aluminum alloy and ETPHC copper, in the form of 3 mm thick discs. The substrates were previously polished up to 1 μm grit polishing cloth and cleaned in an ultrasonic bath for 15 min. Ti thin films were made with a substrate temperature of

* Corresponding author at: Gerencia de Investigación y Aplicaciones, Comisión Nacional de Energía Atómica, Av. Gral Paz 1499, B1650KNA San Martín, Buenos Aires, Argentina.
E-mail address: delgross@tandar.cnea.gov.ar (M.F. del Grosso).

https://doi.org/10.1016/j.apsusc.2018.02.222
0169-4332/

423 K and 523 K, for Cu and Al respectively, a pressure between 4 and 8 $\times 10^{-3}$ Pa and a deposition rate between 20–25 Å/s. Once the titanium film is deposited on the substrates, and without loosing vacuum inside the deposition chamber, the samples were impregnated with deuterium under controlled temperature, time and D_2 pressure conditions [10,13,14]. The deuterium pressure (P_{D_2}) varied between 0.1 $\times 10^5$ Pa, 0.5 $\times 10^5$ Pa and 1.0 $\times 10^5$ Pa, the substrate temperature (T_s) was taken within 373 K and 473 K, and the exposure time (t_e) was set among 0.5 h, 1 h and 2 h. Since deuterium concentration may not reach the stoichiometric relation of two deuteriums per each titanium, due to the inherent experimental performance, we will refer to it as TiD_x samples.

TiD_x films with the highest deuterium concentration, were irradiated at the ion injector of the TANDAR accelerator at National Atomic Energy Commission, Argentine, by a proton beam of 150 keV and currents between 3 and 12 μA, depending on ion source conditions. Beam diameters varied between 7 and 20 mm, which corresponded to current density of 9.5×10^{-4} mA/cm^2 and 3.1×10^{-2} mA/cm^2 respectively, reaching fluences up to 3×10^{18} ions/cm^2. In order to evaluate the temperature reached by the system, under the irradiation conditions, finite element simulations were carried out. For all cases, the simulations showed maximum temperatures of the samples below 363 K.

All samples were characterized by different techniques. Physical characterization includes the measurement of TiD_x film thickness with an optical profiler NT1100 (Veeco Instruments Inc.). Adhesion tests were carried out with the tape test technique [8], and surface micrographs were taken with an environmental scanning electron microscopy (ESEM) Quanta 200 (FEI Company), which includes an energy dispersive spectroscopy (EDS) Apollo X detector (EDAX Inc.). Crystallographic characterizations were performed by grazing incidence X-ray diffraction (GIXRD) Empyrean (PANalytical B.V.) with a PIXcel3D detector. Finally to determine the elemental composition of the samples we used two different techniques: laser induced breakdown spectroscopy (LIBS) that was carried out using a LIBS2500plus (Ocean Optics Inc.), the equipment has a 50 mJ Nd:YAG laser, a lens 25x60 NIRI (ACH-NIR) to focus the emitted light and a HR2000 + CCD [15]; and elastic recoil detection analysis (ERDA) [16], using a telescope detector (a solid state detector plus a gaseous type detector) and a TNT-N1728 (CAEN) multichannel analyzer for data acquisition. The use of a telescope detector allowed us to study the depth profile of deuterium and titanium at the same time. Data processing for the ERDA technique was performed using a MATLAB code, written specifically for this purpose.

The SRIM code [17] was used to calculate the beam range and the displacements per atom (DPA) caused by irradiation, using the recommended calculation method from Stoller et al. [18].

3. Results and discussion

3.1. Ti thin films

Well adhered Ti deposits on Cu and Al substrates were obtained, achieving mean roughness values between (11.09 ± 3.50) nm and (19.30 ± 4.00) nm and thicknesses between (1.89 ± 0.01) μm and (2.58 ± 0.01) μm. On Fig. 1 SEM micrographs can be seen: Fig. 1 (a) shows the granular surface morphology of the deposit and in Fig. 1(b) it can be observed a lateral view of the Ti film and a measure of its thickness.

3.2. Characterization of titanium deuteride films

SEM micrographs were made to characterize the surface morphology of the titanium deuteride films. As can be seen in Fig. 2, the titanium deuteride kept the same surface morphology of the titanium thin film, shown in Fig. 1. In order to study deuterium distribution and concentration in Ti thin films, all samples were analyzed with ERDA, LIBS and GIXRD. We were able to evaluate the homogeneity of the deuterium distribution, in the depth of the deposit by means of ERDA and across the surface by means of LIBS. The deuterium concentration and crystalline structure was evaluated by GIXRD. Also, by means of the previously mentioned techniques, we could find that a group of samples retained a higher deuterium concentration, while for other samples, the amount of deuterium retained was lower.

In Fig. 3 can be seen two different TiD_x samples measured with ERDA, (a) with low concentration of deuterium and (b) with a higher concentration. Regarding the relation between the deuterium and titanium counts we can find that the parameters that benefit the deuterium absorption are: $P_{D_2} = 1 \times 10^5$ Pa, T_s = 473 K and t_e = 0.5–2 h. It was observed that deuterium impregnation saturates after such times. The signal level of oxygen is due to the surface layer of TiO_2 generated by being in contact with the atmosphere, and the small signal levels of carbon indicate film contamination, that could either come from impurities present in the deposition chamber or simply from environmental pollution. On the other hand, substrate ions (Al or Cu) lose all their energy and do not reach the solid detector E, therefore they do not present events on these graphics. The concentration depth profile, for

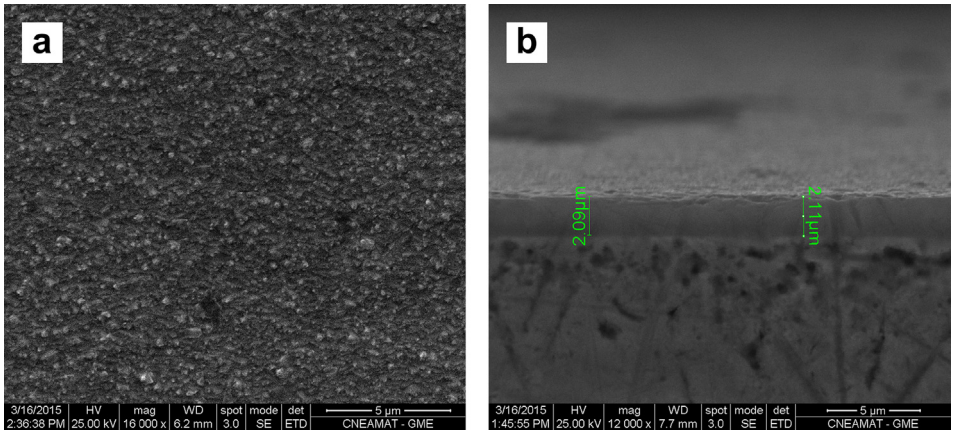

Fig. 1. SEM micrograph of (a) the surface and (b) a lateral view of a Ti deposit on a Cu substrate.