Nuclear Inst, and Methods in Physics Research B 430 (2018) 64–71

Contents lists available at ScienceDirect

Nuclear Inst. and Methods in Physics Research B

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

Depth profiling of titanium nitride thin films deposited on stainless steel utilizing combined EBS and NRA techniques

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ARTICLE INFO

Keywords:
Titanium nitride
Elastic backscattering
Nuclear reaction analysis
Depth profiling
Vacuum arc discharge

ABSTRACT

Composition and depth profiling of titanium nitride thin films deposited on stainless steel using vacuum arc discharge device have been determined. A combination of two ion beam analysis techniques has been applied to allow accurate quantification of nitrogen in the prepared titanium nitride thin films. These include elastic backscattering (EBS) spectrometry together with nuclear reaction analysis (NRA) technique utilizing $^{14}N(\alpha,p_0)^{17}O$ nuclear reaction, both performed at resonance energy for incident alpha beam of 4500 keV. They were applied simultaneously, using two independent charged particle detectors, one is dedicated for combined EBS/NRA measurements and the other for pure NRA one. Experimental conditions have been carefully optimized to avoid overlapping of peaks of the two spectra correspond to each reaction. Novel Multi-SIMNRA software has been utilized to enable simultaneous elemental depth profile determination of both data sets. Experimental setup of the in-vacuum system has been carefully characterized. Independent complement measurements utilizing scanning electron microscopy (SEM) equipped with energy dispersive X-ray analysis (EDX) were also applied to verify the experimental findings obtained by combined EBS and NRA techniques. The results have revealed N/Ti ratio of around 1 for the prepared films, with low levels of contamination elements. Finally, X-ray diffraction (XRD) measurements have demonstrated TiN structure with low oxygen contamination.

1. Introduction

Elemental composition characterization of titanium nitride (TiN_x) thin films deposited on metallic surfaces has been given significant interest by scientists. Such deposition considerably enhances the mechanical and chemical properties of metallic surfaces [1]. Several analytical techniques have been applied for elemental composition characterization of the deposited TiN_x thin films [2,3]. Among them, ion beam analysis (IBA) techniques have been successfully utilized and have shown to be effective tools to reveal elemental depth profile information of TiN_x thin films [4,5]. Elastic backscattering technique using alpha beam was comprehensively applied. It might be directly utilized as conventional Rutherford backscattering (RBS) if TiN_x thin film is deposited on top of substrate made of light elements, like graphite or hydrocarbon polymer materials [6,7]. If substrate comprises heavier elements like silicon or stainless steel, elastic backscattering technique (EBS) using alpha beam may successfully be applied at resonance energies to enhance the signal of elastic backscattering of nitrogen [8–10]. Many researchers have demonstrated an elastic backscattering (EBS) resonance using alpha beam at energy of 3700 keV to detect nitrogen in different systems [9,11–15]. Additionally, proton beam can be used to induce elastic backscattering protons on nitrogen at resonance energies [16]. Finally, both elastic recoil detection analysis (ERDA) and nuclear reaction analysis (NRA) techniques were also widely applied simultaneously with EBS technique [5,17]. Many NRA measurements are performed using deuterium or ^{3}He ions beams, given that relevant induced nuclear reactions have large cross sections, together with positive Q-values, hence reaction can occur at any energy of the incident particle [18]. However, deuterium and ^{3}He ion beams have to be used with special care due to potential unwanted reactions, especially those associated with production of neutrons. Additionally, both deuterium and ^{3}He ion beams are not always available at many accelerator facilities, especially ^{3}He one [19].

We aim, in this work, to perform elemental analysis of titanium nitride thin films deposited on stainless steel substrates by vacuum arc discharge technique that can be operated at industrial level and has been regularly applied at our institution [12,20]. Nitrogen depth profile information was achieved by applying simultaneous EBS and NRA techniques using alpha beam (instead of deuterium or ^{3}He ones). Protons induced from $^{14}N(\alpha,p)^{17}O$ nuclear reaction together with elastic

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https://doi.org/10.1016/j.nimb.2018.06.012
Received 29 October 2017; Accepted 11 June 2018

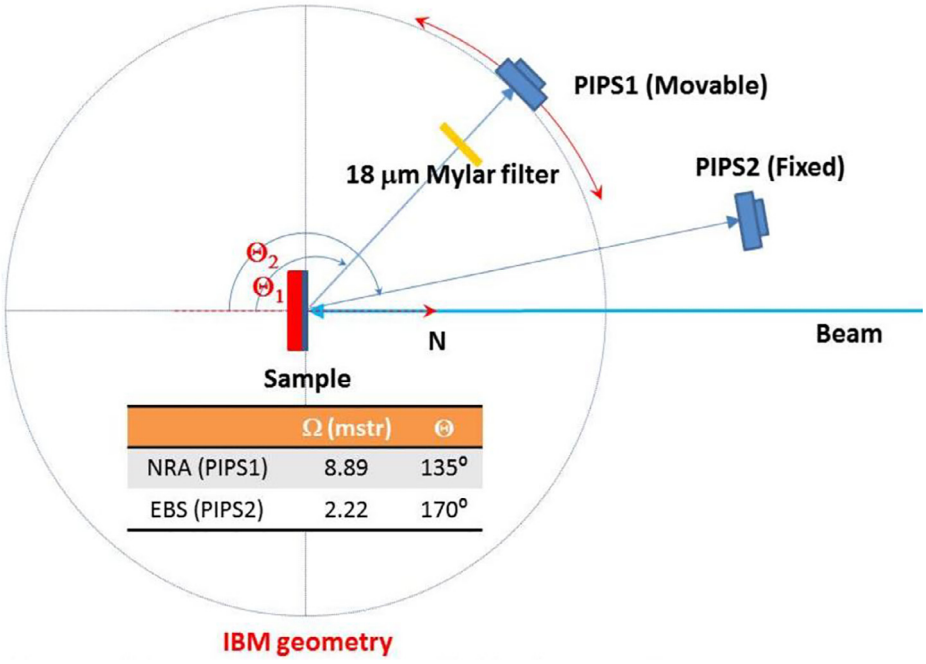

Fig. 1. An outline of upper view of the geometrical arrangements inside the vacuum chamber.

backscattered alphas from $^{14}N(\alpha, \alpha)^{14}N$ reaction, both performed using alpha beam at energy of 4500 keV (instead of conventional 3700 keV) corresponding to resonances on nitrogen, were collected to gain the required depth profile information of nitrogen. Experimental conditions were optimized to resolve potential peak interference of nitrogen in both EBS and NRA spectra. Utilizing of novel Multi-SIMNRA software enables self-consistent analysis of data obtained by different experimental conditions (two different techniques here, EBS and NRA) of a given sample.

2. Experimental

2.1. Sample preparation

Two SS316 stainless steel plates of 1 mm thick were cut into square pieces of 10 mm wide. The substrates were then cleaned using ultrasonic cleaner in ethanol prior to mounting in the vacuum chamber. Finally, the substrates surface was cleaned by argon ion bombardment before thin film deposition. The TiN_x thin films were prepared by vacuum arc deposition using an industrial V-1000 "U" system, (Belarus) from a high pure Ti target (99.99%). Comprehensive characterization and setup of this system is discussed elsewhere [12,21]. High pure titanium target was first cleaned by pure argon bombardment for 10 min. Then, the film was deposited, on the SS316 stainless steel substrate, in nitrogen discharge for 10 min. The residual pressure was lower than 4×10^{-6} mbar. The working pressure in the vacuum chamber was maintained at 1.3×10^{-3} or 2×10^{-3} mbar during the deposition to prepare two samples, labeled TiN-1/SS316 and TiN-2/SS316 respectively.

2.2. Instrumentation

EBS and NRA measurements were performed using 4500 keV alpha beam generated by our 3 MV HVEE™ tandem accelerator [22] at the Atomic Energy Commission of Syria (AECS). A large in-vacuum target chamber, located at the end station of the −60° beam line, is dedicated to enable standard ion beam analysis techniques associated with charged particle detection, including elastic backscattering spectrometry (RBS/EBS), conventional elastic recoil detection analysis (ERDA) and nuclear reaction analysis (NRA). It is equipped with "wheel" target holder that is attached to a four-axis automatic-controlled goniometer to enable different motion possibilities of the samples holder in-vacuum. Samples were irradiated with an incident alpha beam normal to their surface with a beam current of 10 nA. Two passivated implanted planar silicon (PIPS) detectors were used (Canberra's PIPS detector, No. PD50-12-100AM, with nominal resolution of 12 keV and active thickness and area of 100 μm and 50 mm^2, respectively). They are both positioned in IBM scattering geometry (i.e. the incident beam, exit beam and surface normal of the sample are in the same plane). The first detector is movable to enable variation in scattering angles in between −170° to +170° and has a detection solid angle of 8.89 msr. Additionally, it is equipped with an automated rotatable foil holder in front of its window to enable easy selection of variable absorbers within the vacuum chamber. It is mainly dedicated for both NRA and conventional ERDA measurements. For the current investigation, this detector was fixed at a lab scattering angles angle of 135°, and a Mylar™ absorber of 18 μm thick was placed in front of its window so that the backscattered α-particles can totally stop in it, and only the emitted protons induced for (α,p) reaction can pass it with energy loss that can be easily estimated using, for instance, SRIM software [23,24]. Using nuclear reaction kinematics [18] for the $^{14}N(\alpha,p)^{17}O$, incident alpha beam of energy E_α = 4500 keV results in the protons emission of energy of about 1747 keV. Then, they are subjected to energy loss during the passage of the 18 μm Mylar absorber so their energy decreases to about 1300 keV. The second detector was placed at a lab scattering angle of 170°, subtending a solid angle of 2.22 msr without any absorber foil in front of its window to enable detection of the backscattered particles as well as other charged particles products induced form nuclear reactions.

Fig. 1 shows an outline of upper view of the geometrical arrangements inside the vacuum chamber. The accumulated beam charge is measured directly from the sample holder that is connected to a charge integrator; the total collected charge was10 μC. The alpha beam is collimated to dimensions of 2×2 mm^2 using two sets of slits located at the beam line. Additionally, the position, intensity and profile of ion beam can be monitored using a beam profile monitor (BPM) device located just behind the vacuum chamber. Furthermore, we have efficiently applied this device for indirect beam charge measurements [25].