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STRUCTURE AND SURFACE MORPHOLOGY OF SPUTTERED POLYCRYSTALLINE AND NANOCRYSTALLINE Ti FILMS

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We investigate the effect of ion bombardment on the structure and surface morphology of polycrystalline Ti (purity of 99.5 %) and nanocrystalline Ti-SS (stainless steel) films deposited by DC magnetron sputter ion plating (MSIP) process. The development of the structure and surface roughness with increasing negative substrate bias U_s, strongly differs in polycrystalline Ti and in nanocrystalline Ti-SS films. The Ti films sputtered under U_s ranging from 0 to –1500 V are regularly polycrystalline, while the Ti-SS films with a high content of SS elements, are nanocrystalline, even if sputtered at zero bias. The surface roughness of polycrystalline films increases with increasing negative bias U_s and in Ti films sputtered at high U_s (greater than –1000 V) reaches high values up to several micrometers. On the contrary, the surface of Ti-SS films sputtered at U_s from 0 up to –1000 V is smooth and the surface roughness does not depend on the ion bombardment. Moreover, nanocrystalline Ti-SS films have a smoother surface than polycrystalline Ti films sputtered at zero bias. This is due to the large difference in the grain size of nanocrystalline (about 1 nm) and polycrystalline (hundreds of nm) films.

1. *Introduction*

Recent studies have shown that ion bombardment during growth of thin films is an efficient method to control properties of the films. The ion bombardment strongly influences processes on the surface and in a subsurface region of the growing film. It affects the mechanism of growth, hence the film density [1], degree of crystallinity, preferred orientation [2], grain size [3], microstrain and macrostress, i.e. the film microstructure [4].

Thin films are applied with the purpose of achieving special features of the surfaces such as friction, radiation reflection, heat absorption, corrosion resistance, gas or humidity sorption, optical reflection [5], decorative effects etc. Therefore, considerable attention has been devoted to investigations of their surface morphology.

Main factors influencing surface morphology of the films are the substrate roughness and the film microstructure. The film roughness induced by the substrate roughness can be easily eliminated by polishing of substrate. More difficult is to eliminate the roughness caused by the film microstructure, because it depends on many parameters of the deposition process. Therefore, production of a film with defined properties still remains a serious problem.

2. *Experimental*

Both types of the films were deposited by the DC magnetron sputter ion plating (MSIP) process, using a small circular planar magnetron with a Ti (99.5 %) target of a diameter of 60 mm. The magnetron was placed at the bottom flange of a cylindrical stainless steel deposition chamber (l = 310 mm, d = 250 mm). After evacuation to a base pressure ($< 10^{-3}$ Pa), argon (99.99 %) was introduced into the chamber. Substrates (25 mm × 12 mm × 3 mm) made of CSN 15330 steel were fixed onto an electrically isolated, movable and heatable substrate holder. The substrates were prepared by a standard grinding and polishing procedure using a STRUERS PEDEMAX 2 device that guaranteed the same surface roughness of all samples. Prior to deposition, the substrates were sputter cleaned.

The Ti films were deposited under the following conditions: discharge current I_d = 1.34 A, discharge voltage U_d = 480 V, argon pressure p = 0.6 Pa, substrate-to-target distance d_{s-t} = 45 mm, substrate temperature T_s = 200 °C, different values of negative substrate bias U_s ranging from 0 to -1500 V and deposition time t_d = 15 min. The substrate ion current densities, i_s, were relatively low, from 0.17 to 0.49 mA/cm^2.

The Ti-SS (46.2 Fe, 11.8 Cr, 5.7 Ni, 0.2 Si, 1.0 Mn) films were sputtered from a Ti target equipped with two SS rings of diametere 27 mm, located symmetrically around the target axis. The operating conditions were: I_d = 0.85 A, U_d = 620 V, p = 0.6 Pa, T_s = 200 °C, d_{s-t} = 45 mm, different value of U_s ranging from 0 to –1000 V and t_d = 15 min. The elemental composition of the T-SS films was determined by EDAX.

Film thickness of both types of the films was approximately the same, 4.5–5 μm.

The structure of the films was characterized by X-ray diffraction measurements, using XRD spectrometer Micrometa 2. The surface morphology was characterized by surface roughness measurements with a Laser stylus RM 600 device using an infrared laser beam (800 nm), and by SEM.

3. *Results and discussion*

3.1. *Polycrystalline Ti films*

Pure Ti films are polycrystalline with grains of about several hundred nm. The preferred orientation of crystallites changes with increasing negative bias from the (002) orientation at low bias voltages (below 200 V) to the (110) orientation at high bias voltages of about 300 V and higher. At all bias voltages, the reflection lines are strong and very narrow (FWHM< 1°). That indicates that the films have good crystallinity even at a high negative bias (see Fig. 1).

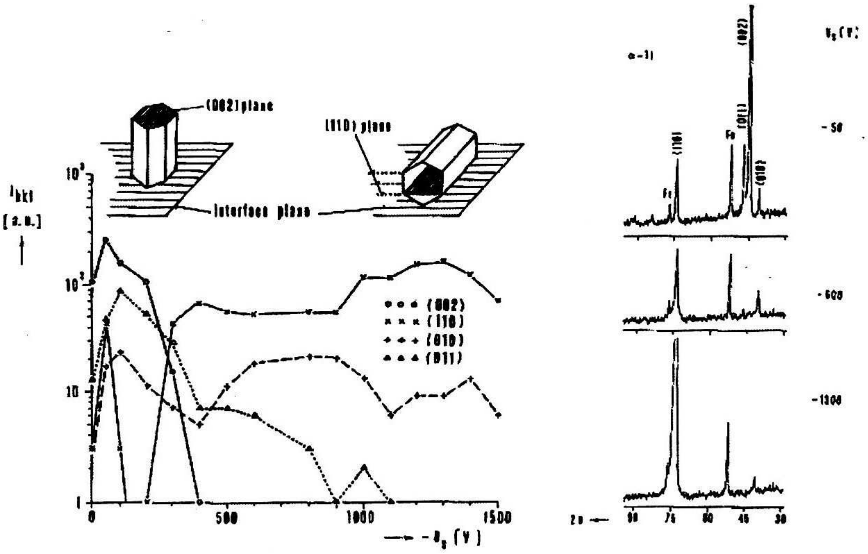

Fig. 1. Intensities of X-ray reflection from pure Ti *(99.5 %) films deposited with different values of the negative substrate bias (left). XRD patterns for three values of negative substrate bias U_s (right).*

The surface roughness of Ti films does not develop continuously with increasing negative bias. Three roughness regions can be distinguished for U_s ranging from 0 to –250 V, from –300 V to about -1000 V and from -1000 V to higher negative voltages, respectively (see Fig.2). In the first region, the effect of the ion bombardment is low and in the whole region the surface roughness is very similar to that of the substrate prior to the film deposition. The transition from the first region to the second one is relatively sharp. Experiments indicate that a change in the magnitude of surface roughness in the second region is due to the change in crystalline orientation from (002) do (110). Also, in this region the ion bombardment practically does not change the surface roughness. This is determined by the crystalline orientation which is almost constant in the whole region. The third region is characterized by an increase of surface roughness with increasing negative U_s. The effect of ion bombardment, resulting in stopping of the growth of crystal grains, probably starts to be compensated and dominated by the crystal growth due to the heat delivered to the growing film by bombarding ions. The surface morphology corresponding to the three typical surface roughness is also displayed in Fig. 2.

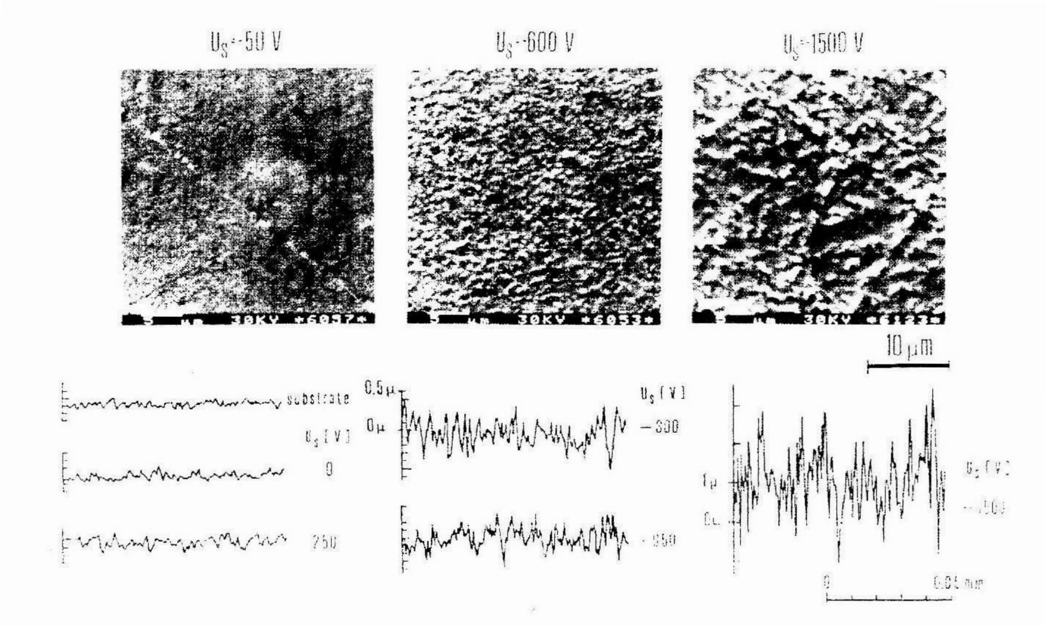

Fig. 2. Scanning electron micrographs showing the surface morphology of Ti *(99.5 %) films sputtered at three values of negative biases U_s. Surface roughness measurements corresponding to three roughness regions (from 0 to –250* V, *–300* V *to about –1000* V *and U_s higher than –1000* V*) are shown below.*

The sputter ion plating of films at high biases U_s of about -1500 V yielded films with a great (several μm) surface roughness. We believe that thin films with such a high surface roughness could be utilized in new applications where larger areas of the film surface are required. A high energy, ion flux bombardment could be also a new method of producing films with high surface porosity.

3.2. Nanocrystalline Ti-SS films

Nanocrystalline Ti based films can be produced by the ion bombardment of the growing film and/or by mixing of Ti and some immisible elements, for instance elements in stainless steel (SS) elements [6]. For the production of nanocrystalline Ti-SS films with a low content (about 10 % or lower) of SS elements in Ti films, the combined action of the ion bombardment and mixing of Ti and SS is necessary. Therefore, such films, sputtered at zero bias, exhibit well developed reflection lines and are polycrystalline. On the contrary, in the production of Ti-SS films with a high content (greater than 10 %) of the SS elements, the mixing Ti and SS elements is the dominant process which alone produces the nanostructured alloys films. Such Ti-SS films were used in this study.

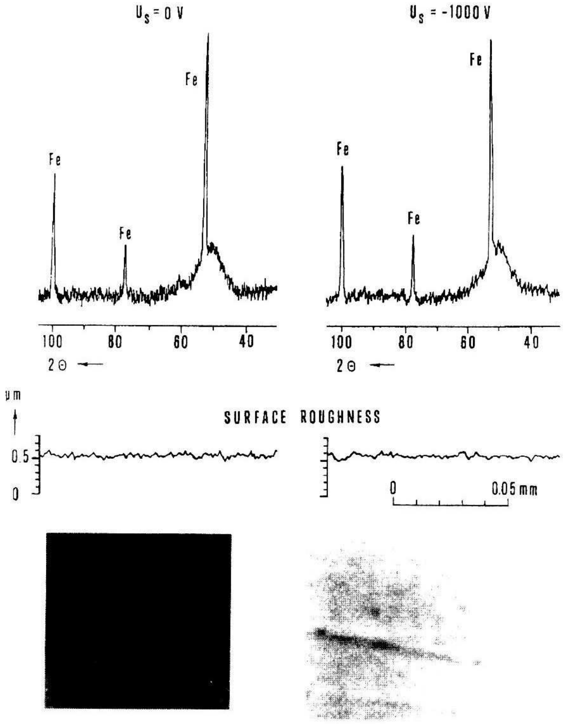

Fig. 3. The XRD patterns, surface roughness patterns and scanning electron micrographs of Ti-SS *films (46.2* Fe, *34.1* Ti, *11.8* Cr, *5.7* Ni, *O.2* Si, *1.0* Mn*) sputtered at two values of the negative substrate bias* U_s.

Ti-SS films are characterized by one very broad (FWHM=8.6°) low intensity reflection at $2\theta = 50°$. These films exhibit almost no change with bias U_s in the range from 0 to –1000 V (see Fig. 3). The size of the grains in the films, calculated from the Scherrer formula, is very small (about 1 nm). The formation of such grains in films, produced at high biases U_s of about –1000 V, shows that the mixing process of Ti and SS elements, resulting in a creation of nanostructured alloys, dominates over the crystal growth that was expected due to the heat delivered to the growing film by bombarding ions. For this reason the surface roughness of Ti-SS films does not change with increasing negative bias U_s and follows the roughness of the substrate. The nanocrystalline Ti-SS films have a very smooth surface. The surface morphology does not depend on the ion bombardment of the growing film and is the same for all films produced at arbitrary bias U_s used in our experiments (see Fig. 3).

4. *Conclusions*

The development of structure and surface morphology of polycrystalline Ti and nanocrystalline Ti-SS films, sputtered using the MSIP process on a substrate temperature of 200 °C and relatively low substrate ion current density, strongly differs. The results can be summarized as follows:

1. Pure Ti films deposited by the MSIP process on a steel substrate under an arbitrary negative bias U_s ranging from 0 to –1500 V are polycrystalline. The preferred orientation of crystallites varies with increasing negative bias voltage U_s from the (002) orientation at low biases (below 200 V) to the (110) orientation at high biases of about 300 V and higher.

2. Ti-SS films with a high content of SS elements are nanocrystalline, even if sputtered at zero bias. In this case, the mixing of Ti and SS elemets is the dominant process in the formation of a nanostructured films and no ion bombardment is necessary to destroy the growing grains. These films are characterized by a very broad (FWHM of about 10°) low intensity reflection for U_s ranging from 0 to –1000 V.

3. The surface roughness of polycrystalline Ti films depends on the ion bombardment of the growing film and increases with increasing negative bias U_s. At high U_s of about –1000 V and higher, the surface roughness reaches up to several micrometers. On the contrary, the surface roughness of nanocrystalline Ti-SS films does not depend on ion bombardment of the growing film and the film surface is always smooth even when Ti-SS films are sputtered at very high U_s of about –1000 V.

4. Nanostructured Ti-SS films have a smoother surface than the polycrystalline Ti films sputtered at zero bias. This is due to the large difference in the grain size of nanocrystalline and polycrystalline films, about 1nm and hundreds of nm, respectively

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STRUKTURA I POVRŠINA RASPRAŠENIH POLIKRISTALNIH I NANOKRISTALNIH Ti SLOJEVA

Proučavan je učinak ionskog bombardiranja na strukturu i oblik površine polikristalnog Ti (čistoće 99.5%) i nanokristalnog Ti-SS (nehrđajući čelik) tankih slojeva nanešenih procesom DC magnetronskog rasprašivanja. Istražen je učinak negativnog napona podloge na strukturu i oblik površine polikristalnih Ti i nanokristalnih Ti-SS slojeva. Nanokristalni Ti-SS slojevi imaju glatkiju površinu nego polikristalni Ti slojevi dobiveni rasprašivanjem na naponu nula volta. Razlog je velika razlika u veličini zrna nanokristalnih (oko 1 nm) i polikristalnih (stotine nm) tankih slojeva.