Research of niobium thin films with a predetermined thickness produced by RF magnetron sputtering

D A Polonyankin[1], A I Blesman[2], D V Postnikov[1], A I Logacheva[3], I A Logachev[4], A A Teplouhov[1] and A A Fedorov[5]
[1]Associate Professor, *Physics Department*,
Omsk State Technical University (OmSTU), 11 Mira Avenue, Omsk, 644050, Russia
[2]Head of the Physics Department, *Physics Department*,
Omsk State Technical University (OmSTU), 11 Mira Avenue, Omsk, 644050, Russia
[3]Deputy Director, *New Metallurgical technologies institute,*
Joint Stock Company Kompozit, 4 Pionerskaya St., Korolev, 141070, Russia
[4]Head of sector, *New Metallurgical technologies institute,*
Joint Stock Company Kompozit, 4 Pionerskaya St., Korolev, 141070, Russia
[5]Associate Professor, *Department of Mechanical engineering technology*,
Omsk State Technical University (OmSTU), 11 Mira Avenue, Omsk, 644050, Russia

E–mail: nano@omgtu.ru

Abstract. Niobium and niobium thin films are widely used in various fields of modern science and technology: in the electronics industry, in a nuclear medical imaging technique, in the information technology, in superconducting cavities technology etc. The grain size of thin niobium films depends on its thickness and the film's stoichiometry can be varied as a function of thickness. Thus the problem of thickness control has a great practical importance in all fields of niobium films application. The focus of this study was to perform an experimental calibration of STC–2000A deposition controller for niobium target on ADVAVAC VSM–200 setup and to conduct a grain size, roughness and stoichiometry research by scanning electron microscopy, X–ray diffraction and laser interference microscopy of niobium films produced by RF magnetron sputtering with the thickness range from 200 nm to 400 nm and 50 nm step.

1. Introduction

Niobium thin films produced by RF magnetron sputtering are widely used in various fields of modern science and technology. Niobium films have important potential applications in the electronics industry due to the threshold current reduction in resistive switching devices (memristor) which are promising emerging nanodevices for future information technology [1]. Nb films and Nb–Nb_2O_5 multilayer coatings may be used in a nuclear medical imaging technique (positron emission tomography, PET) because of the following features: it may prevent the aqueous corrosion of the chamber's wall as diffusion barrier at routine production of [^{18}F] (radiotracer in scanning technique), and it can withstand overpressure with value of 4 MPa and resist the proton beam energy absorption in the entrance foils [2]. Nb also is very available material for a bipolar plate in proton exchange membrane fuel cell (PEMFC) because it has the best corrosion resistance among transition metals in sulfuric solution [3].

Nb is a highly interesting transition metal that exhibits superconducting properties [4] and as a refractory superconductor is still considered as a perspective material for fabrication of robust and

reliable Josephson devices for a number of key applications, such as high–speed superconducting digital circuits (RSFQ), mm–wave receivers and sub–millimeter wave mixers, and as voltage standard [5]. Niobium films are also applied for superconducting radiofrequency (SCRF) devices, such as superconducting cavity–stabilized oscillators (SCSO) [6], superconducting resonant cavities for particle accelerators [7], step–edge superconducting junctions with films thickness ranges from 25 to 200 nm [8] and superconducting quantum interference devices (SQUIDs) with nanobridge junctions [9]. Superconducting properties of niobium are also used in processors of quantum computers (superconducting qubit) [10]. Photocathodes with high quantum efficiency and electrical compatibility with superconducting cavities technology usually made of niobium [11].

Critical temperature of superconducting Nb transition is usually lower for thin films than in bulk sample and depends on various parameters such as thickness [12], stoichiometry [1] and grain size [13]. At the same time the grain size of thin niobium films depends on its thickness [2] and film's stoichiometry can be varied as a function of thickness [1].

Thus the problem of thickness control has a great practical importance in all fields of niobium films application. Magnetron sputtering seems to be the most suitable method to produce Nb films (i.e. a nanostructured material with optimal physical properties) [14] that cause the RF magnetron sputtering selection as an experimental method for thin niobium films deposition.

The focus of this study was to perform an experimental calibration of STC–2000A deposition controller for niobium target on ADVAVAC VSM–200 setup and to conduct a research of thin niobium films by scanning electron microscopy, X–ray diffraction and laser interference microscopy.

2. Calibration of STC–2000A deposition controller for niobium target

There are two experimental methods of thickness measurement: the first is online thickness control during the sputtering process by using deposition controller of magnetron system, and the second is the film's thickness evaluation by approved methods such as ellipsometry, profilometry, X–ray diffraction analysis etc. If necessary, these methods can be combined for data verification.

Calibration of STC–2000A deposition controller for niobium target was performed using three different material parameters: density, Z–Factor and tooling. Density and Z–Factor are material factors of niobium that take the values of 8.576 gm/cc and 0.493 correspondently. Tooling is a deposition system geometry correction (location of sensor to substrates, see figure 1). Initially the tooling was set to 100%.

Niobium deposition was done in RF mode at source power of 100 W under $3.4 \cdot 10^{-3}$ mbar argon pressure and pre ionic chamber cleaning for 10 minutes. Then tolling (T) was calculated by expression (1) after niobium film sputtering with displayed thickness of 5000 angstroms for relative position of sensor and soda–lime silicate glass ($Na_2O \cdot CaO \cdot 6SiO_2$) substrate which was sonicated in isopropyl alcohol prior to film deposition.

$$T = \frac{Measured\ thickness}{Displayed\ thikness} \times 100\ \% \qquad (1)$$

Niobium thin film thickness of 216 nm was obtained on the base of "film–substrate" line scanning profile by energy dispersive spectroscopy (EDS) performed with JED–2300 analyzer of JEOL JCM–5700 (see figure 2), thus calculated tooling had a value of 0.43%. As can be seen from figure 2, the substrate includes oxygen (shown in red), sodium (light green), silicon (turquoise) and calcium (black) that corresponds to composition of the soda–lime silicate glass. Niobium, which is film's element, is marked in dark blue.

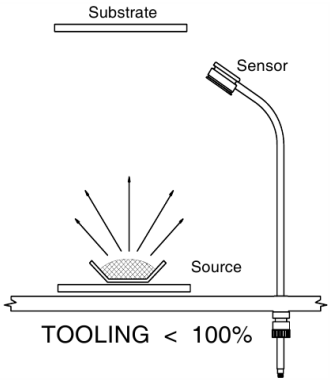

Figure 1. The relative position of the sensor and the substrate.

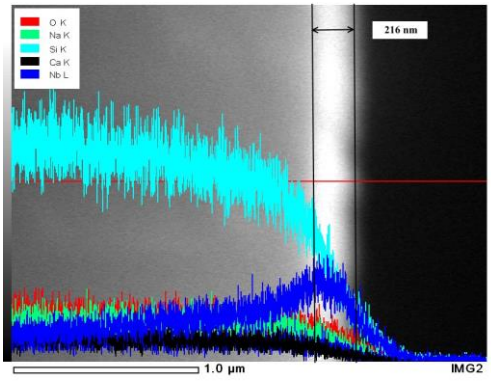

Figure 2. Line scanning profile of niobium thin film on soda–lime silicate glass substrate.

3. Experimental details

Nb films with the thickness range from 200 nm to 400 nm and 50 nm step were prepared by using the ADVAVAC VSM–200 setup at high vacuum RF mode with source power of 100 W under $3.4 \cdot 10^{-3}$ mbar argon pressure and pre ionic chamber cleaning for 10 minutes. Thickness range selection is caused by the reason that such thicknesses are far from quantum size effect region [15] and bulk properties are expected. Thin films were sputtered with pure argon (Ar) gas by using Nb target (99.95% in purity, 50 mm in diameter).

The XRD investigation of niobium films with thickness in the range from 200 nm to 400 nm and 50 nm step was carried out by a sliding X–ray beam method (fixed tube angle θ=5°) in Bragg–Brentano configuration with 2θ from 30° to 80° and Cu Kα radiation (λ=1.54 Å) using XRD–7000 diffractometer (Shimadzu). Shimadzu software allowed obtaining the peaks position and the according full–width at half–maximum (FWHM) for all 2θ angles.

Qualitative and quantitative analysis of thin niobium films was performed by scanning electron microscopy (SEM) with energy dispersive spectroscopy (EDS) and allowed to determine the composition of the surface layers.

The analysis of surface roughness has been provided by laser interference microscopy (LIM) performing root mean square (RMS) values in dependence of the film's thickness. Laser interference microscope MIM–340 (Shvabe) is designed for non–contact non–destructive investigations of submicron structures with nanometer level resolution in the field of materials science.

4. Results and discussion

The results of XRD and SEM with EDS investigations of thin niobium films are shown on the figures 3 and 4. The surface composition of 200 nm thickness niobium film is represented in table 1. LIM phase image of 200 nm niobium film's surface with roughness line profile is provided on the figure 5.

As seen from mixed XRD pattern it contains the peaks reflected by niobium crystal lattice and only lines related to planes with [(110), (211)] Miller indices was detected. The intensity corresponding to the grains oriented in the (110) plane surpasses in times similar intensity rates for the (211) plane. Moreover, as the film growth, signals reflected by the (211) plane does not practically change; while the intensity appropriate to the (110) orientation substantially increases. The shape of all peaks indicates a high crystallinity degree of sputtered films. Thus we can conclude that niobium films with the thickness range from 200 nm to 400 nm are polycrystalline with body–centred cubic structure of bulk Nb and crystallographic plane orientation of their grains preferential have (110) texture.

Applying a sliding X–ray beam allowed us to register scattering radiation only from niobium crystallite grains, because due to this method film's thickness is greater than the x–ray penetration depth so that a signal coming from the substrate is not observed.

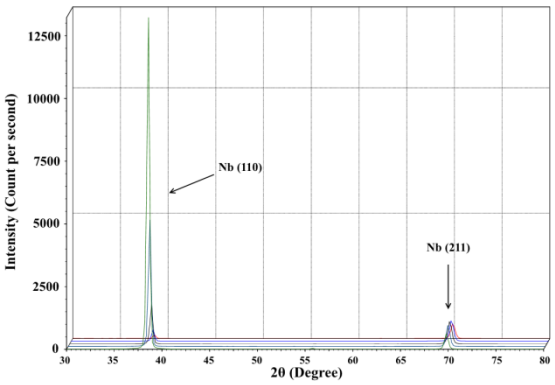

Figure 3. Mixed XRD pattern for niobium films with thickness in the range from 200 nm to 400 nm and 50 nm step.

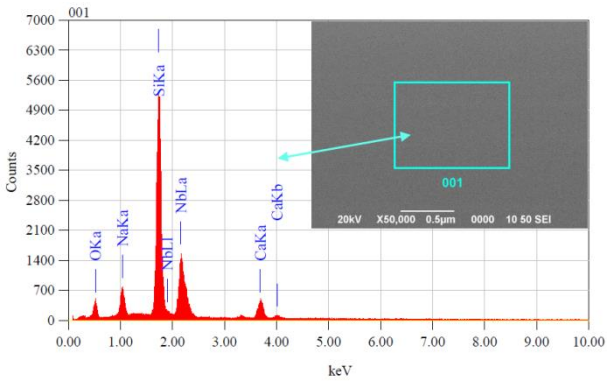

Figure 4. SEM image and EDS results of niobium film with 200 nm thickness on soda–lime silicate glass substrate.

In proof of niobium film's content but not completely coinciding to XRD method SEM results besides information of niobium concentration (19.03 % of atoms) include data about substrate's composition (proportions of Na, Ca, Si and O are presented in table 1). It can be explained by the penetration depth of the electron beam which exceeds even the highest thickness of film (400 nm).

Table 1. The surface layer composition of 200 nm niobium film.

Element	Energy (keV)	Mass %	Error %	Atom %
O	0.525	3.55	0.04	8.76
Na	1.041	4.78	0.04	8.22
Si	1.739	41.14	0.01	58.27
Ca	3.690	5.80	0.07	5.72
Nb	2.166	44.73	0.02	19.03

The roughness RMS values were calculated by using MIM–Visualizer program on the base length of 80 nm after bonding together four surface profiles by MIM–Series software for each film thickness.

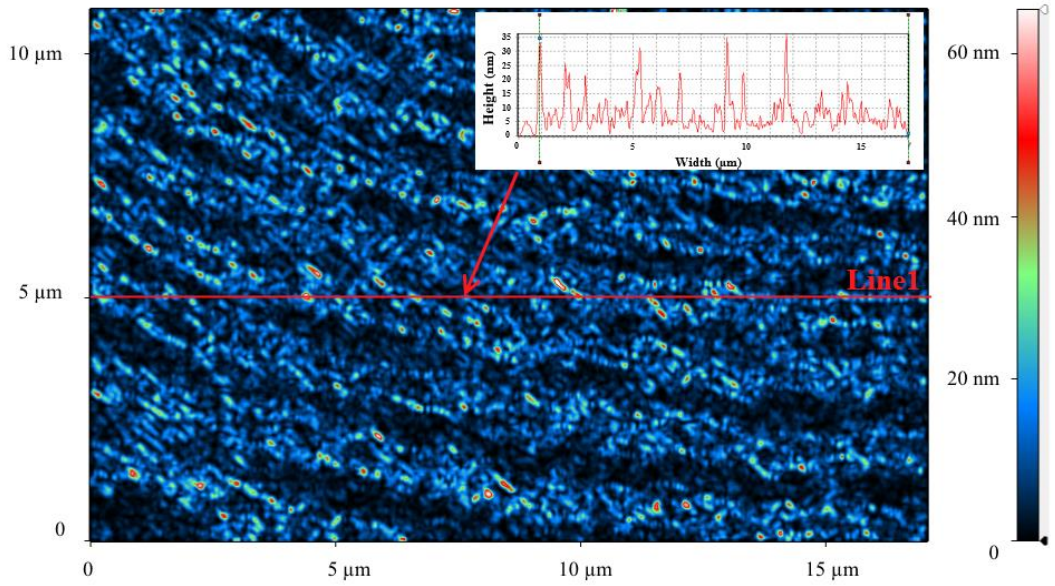

Figure 5. LIM phase image of 200 nm niobium film's surface with roughness profile along Line 1.

The average roughness of niobium films ranges from 4.49 nm (for 200 nm film thickness) to 6.86 nm (for 350 nm film thickness) in dependence of the film's thickness (see figure 6). All films produced by magnetron sputtering refer to highest (14) roughness level.

On the basis of XRD data according to the Debye–Scherrer formula the average grain size was calculated in dependence of the film's thickness (see figure 6). The average grain size increases with the growth of film thickness and ranges from the minimum 16 nm size (for 200 nm film thickness) to maximum value of 20 nm (for 350 nm film thickness). The roughness and the average grain size dependences from the film thickness can be explained by island mechanism [16] when adhesion force and energy to the substrate less than the mutual binding energy of the deposited atoms [17] and films properties with the thickness growth become close to the bulk material.

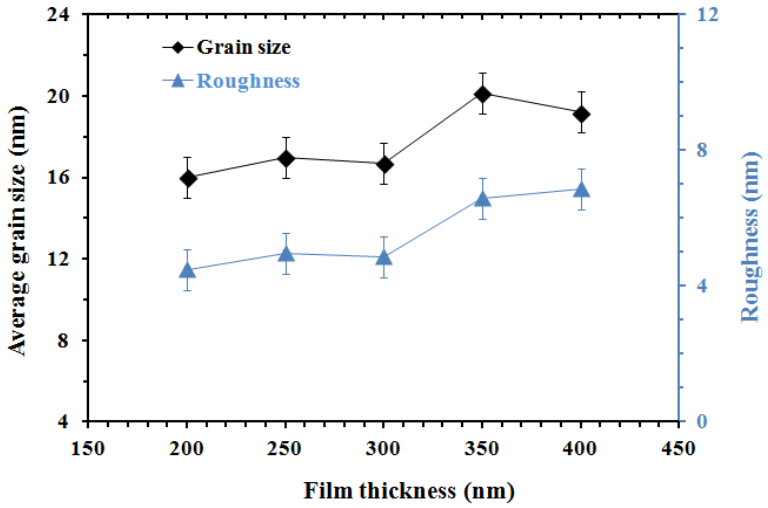

Figure 6. The average grain size and the roughness dependences from the thickness of niobium films.

5. Conclusion

Calibration of STC–2000A deposition controller for niobium target on ADVAVAC VSM–200 setup was performed using density (8.576 gm/cc), Z–Factor (0.493) and experimentally calculated tooling value (0.43%). Niobium films produced by RF magnetron sputtering with the thickness range from 200 nm to 400 nm are polycrystalline with body–centred cubic structure of bulk Nb and crystallographic plane orientation of their grains preferential have (110) texture. The average roughness of niobium films ranges from 4.49 nm (for 200 nm film thickness) to 6.86 nm (for 350 nm film thickness). The average grain size increases with the growth of film thickness and ranges from the minimum 16 nm size (for 200 nm film thickness) to maximum value of 20 nm (for 350 nm film thickness) which corresponds to the island mechanism of the films growth.

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