Study of the mechanical characteristics of the Al$_2$O$_3$ film on the LiNbO$_3$ substrate

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Abstract. The microhardness of a thin aluminum oxide film with a thickness of 80 to 240 nm on the surface of a lithium niobate single crystal was determined by the method of nanoindentation. The hardness and Young's modulus for a film deposited by dc-magnetron sputtering of an aluminum target in a gas (Ar+O$_2$) medium were more than 30 GPa and 300 GPa, respectively. The dependences of the film thickness on the mechanical properties of a layered system, in particular hardness and elastic modulus, were established.

1. Introduction

Aluminum oxide (Al$_2$O$_3$) is a widespread and inexpensive material that is used in various branches of science and technology. Aluminum oxide is applied in the form of a powder of various sizes as an abrasive material and reinforcing filler in metallurgy of alloys and the manufacture of ceramic products [1]. Synthetic and natural single crystals of aluminum oxide with different phase composition and impurities are used in the jewelry industry and high-energy optics (laser optics) [2].

Thin films of this material obtained by various methods have nanoscale elements of the structure and surface morphology. For example, aluminum oxide layers produced by the sol-gel method usually have a porous surface and a thickness of the order of hundreds of nanometers, which is necessary in the manufacture of filters and catalysts [3]. Modern technologies for the deposition of thin films allow the production of continuous, non-porous layers of dielectric materials with a thickness from a few nanometers to several micrometers. These methods include vacuum condensation techniques of the deposition of thin films, in particular PVD and CVD. One of the varieties of the PVD process is magnetron sputtering, which can be performed at direct (DC) or alternating (AC) current [4]. Modern PVD processes are characterized by reproducibility of parameters, high adhesion of deposited materials to the substrate, wide possibilities for the creation of composite materials, the possibility of alloying and relative simplicity of the process [5].

With all the variety of methods of production and applications of thin films of aluminum oxide, the question of studying their mechanical characteristics, in particular the hardness and modulus of elasticity of the "film-base" layered structure, remains relevant. According to a brief review, the microhardness of a LiNbO$_3$ single crystal reaches 5–10 GPa [6,7]. The value of microhardness of aluminum oxide depends on the method of its production and is in the range from 3 to 30 GPa [8,9].

The characteristics of the aluminum oxide film are studied in detail, which is required when creating high-quality dielectrics, protective and anti-reflective layers in solar cell elements [10]. However, the mechanical characteristics of the resulting layers are given insufficient attention. For example, in the layered system under consideration, hardness, elasticity, and wear resistance are important characteristics that determine their possible use in aggressive operating conditions, in particular in the open air or aerospace, as coatings for solar panels.
2. Methodology

The aluminum oxide film was deposited on the polished surface of lithium niobate single crystals (LiNbO₃), the shape of which was represented by parallelepipeds sized 10×11×6 mm. The normal to the deposition plane of the film was an angle of 36° with the positive direction of the crystallographic axis Y.

For PVD, an aluminum target (99.99 %) was used, which was installed in a DC magnetron. In this case, the gas mixture (Ar+O₂) with the 1:1 ratio of the partial pressure of gases was applied at a pressure inside the vacuum chamber \( P = 1.47 \text{ Pa} \). The deposition was conducted at a discharge current \( I = 50 \text{ mA} \), the discharge current density \( j \approx 50 \text{ A/m}^2 \), and the substrate heating temperature \( t = 80 ^\circ \text{C} \). The maximum value of magnetic induction on the target surface was about 25 mT. The films were deposited at a duration \( t \) of 24, 48, and 72 min, which corresponded to a film thickness of 80, 160, and 240 nm.

Samples of the series "1", "2" and "3" prepared for the study of hardness and elasticity modulus were fixed on a rectangular profile using thermoplastic glue (Figure 1).

![Figure 1. Samples of the layered "LiNbO₃ – Al₂O₃" system with different film thicknesses of 80 nm (1), 160 nm (2) and 240 nm (3) placed on an aluminum substrate to test the mechanical properties.](image)

The hardness \( H \) and elasticity modulus \( E \) were determined according to the Oliver-Farr method (ISO 14577-1: 2015) [11,12]. The data of mechanical properties were presented in diagrams of applied force \( P \) and penetration depth of the indenter \( h \), which were obtained on the mechanical tester "NANOUEA Ergonomic Workstation" with loads \( P \) on the Berkovich indenter of 5, 20 and 200 mN.

3. Results

Analysis of the loading diagrams showed that the hardness data referred to the "LiNbO₃ – Al₂O₃" layered system. The maximum penetration depth of the indenter exceeded the oxide film thickness in samples "1", "2" and was comparable to the film thickness for sample "3" (Figure 2).

With a minimum load \( P = 5 \text{ mN} \) and penetration of the indenter into the layered structure to a depth of not more than \( h = 100 \text{ nm} \), the hardness \( H \) reached maximum values above 32 GPa for all three samples (Figure 3). A maximum hardness of about 104±15 GPa was achieved for the film with a thickness of 240 nm. Thus, the resulting layered structure was characterized by the presence of a thin and hard oxide film on a softer substrate.
Figure 2. A graph showing dependencies of hardness of sample surfaces with different thicknesses of the oxide film at various loads.

The increase in hardness for sample "3" was associated with a lower influence of the substrate, since the depth of penetration of the indenter in this case was about 1/3 of the thickness of the oxide film. This was indirectly confirmed by the growth of the elasticity modulus \( E \) at a constant indentation depth for all samples. With a load on the indenter of 20 mN, the Young's modulus gradually fell with the growing film thickness and took values from 342±17 to 306±21 GPa. At a load of 5 mN, a smooth growth of the Young's modulus values with increasing film thickness was observed and for samples "1", "2" and "3" it was 141, 220 and 301 GPa, respectively.

Figure 3. A graph showing the maximum depth of penetration of the indenter into the samples depending on the oxide film thickness under various loads.
Of practical interest was the structure of sample "2" with an oxide layer thickness of 160 nm. Such film is used as a quarter-wave anti-reflection single-layer coating on a LiNbO₃ crystal for a wavelength of light $\lambda = 632.8$ nm. The hardness of the resulting coating at a level of 30 GPa allows the prediction of high wear resistance and the strength of the coating to external mechanical effects and indicates the predominant content of aluminum oxide in the form of a stable solid $\alpha$-phase without additional stabilization by impurities [13].

4. Conclusion

Thus, thin Al₂O₃ films were deposited on LiNbO₃ crystals by magnetron sputtering at a direct current. The hardness and elasticity modulus of the "LiNbO₃ – Al₂O₃" layered system were determined at different loads and film thicknesses. The results led to the conclusion about the possibility of their use in the form of thin anti-reflection Al₂O₃ films and protective layers for softer crystalline materials, which are the basis of modern solar energy devices.

Acknowledgments

The authors express their gratitude to Dr. Zakharevich A.M. and Dr. Skaptsov A.A. for the conducted nanoindentation tests and discussion of the results. The oxide films were deposited using the equipment of NPF "Piezon" of Yuri Gagarin State Technical University of Saratov, Russia.

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