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Features of influence of grain orientation and size on mechanical properties of Al/Mo thin film membranes

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For the first time, idea of the influence of grain orientation and size on mechanical properties of thin-film membranes had been presented.Testing was carried out using the example of thin-film Al/Mo materials on a Si wafer using blowing method. It had been experimentally confirmed that the horizontal size of Mo grains varies in the range from 27 to 67 nm, the size of Al grains varies from 166 to 502 nm, and the grains are oriented in a mutually perpendicular direction. The vertical grain size coincides with the film thickness for Mo at thicknesses up to 1130 nm and for Al at thicknesses up to 1000 nm. By varying the deposition sequence of Al and Mo layers, an increase in the critical burst pressure and biaxial elastic modulus by more than 50/

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One of the most important tasks in the field of mechanics and structural design is the determination of mechanical stresses in structural elements and mechanisms [1]. For example, a thin-film membrane made using silicon MEMS micro- and nanotechnology plays a key role in the development of physical quantity transducers (sensors). Thus, improving the mechanical properties of the membrane material makes it possible to improve the output characteristics of the sensors.

The effect of reduction of the thickness of a single layer (while maintaining the total thickness) can be used as technological method for increasing mechanical strength [2,3]. A single layer means one layer from a set of layers of the same thickness that make up the total thickness of a thin film (membrane). The effect is achieved by reducing the number of defects in the volume of films. The thickness of a single layer of material correlates with the grain size of the material. Increasing the number of boundaries (the number of constituent layers) between materials while maintaining the overall thickness allows reducing the size of defects owing to the formation of sliding barriers. Other factors of increasing mechanical strength include a decrease of grain size because of a change of the atomic ratio between the elements in the alloy [4], a decrease of the surface defects [5], transition from rectangular to polygonal (round) membrane shape, doping of the film material with copper atoms [6], silicon [7]. Thus, the developer can choose a balance between the time spent on the process and the required amount of mechanical strength of the material.

It is known that the reduction of the magnitude of internal mechanical stresses can improve the characteristics of the device [8] (in our case, increase the mechanical strength). The use of a high-strength membrane allows expanding the upper measurement range of membrane-type devices, such as pressure sensors, gas or air flow sensors. Thin films were formed by magnetron method on a Si substrate at a temperature of 180°C in this study, the vacuum level is 0.2mbar in the working chamber (MAGNA unit). The parameters of the film formation process are similar for molybdenum and aluminum, the deposition rate for aluminum is 50 nm/min, for molybdenum 62 nm/min.

A set of thicknesses of aluminum and molybdenum was analyzed using the Stony method [9] (Table. 1) and two samples of thin films on a Si substrate with mechanical stresses close to zero were selected: 71 MPa for an Al film with a thickness of 316 nm, 14 MPa for a Mo film with a thickness of 495 nm.

Fig. 1, *a* schematically shows the direction of external impact (gauge pressure) relative to the orientation of the grains of the membrane material, schematically shows the distribution of mechanical stresses across the membrane, indicates a local area with mechanical stresses close to the maximum value. Thus, for the molybdenum material, the external action is co-directed with the orientation of the grains. The external impact is perpendicular to the grain orientation for the aluminum material.

The value of the mechanical strength (critical rupture pressure) of the material will increase under this type of external impact with an increase of the contact area of the grain and the substrate. The maximum mechanical stresses are known to occur at the membrane/substrate interface and the probability of material destruction along the intergranular boundaries is high. The maximum value of mechanical strength will be achieved when the middle of the grain is located at the membrane/substrate boundary. Thus, the development of technological operations that

Table 1. Dependence of mechanical stresses on film thickness

Al thin film		Mo thin film	
Thickness, nm	Mechanical stresses, MPa	Thickness, nm	Mechanical stresses, MPa
100	−538	330	−994
200	−339	430	−105
316	71	495	14
600	89	830	118
1000	2	1130	80

allow controlling the orientation of grains during the film formation has a promising potential. It is desirable to use a set of alternating layers with horizontal and vertical grain orientation in the case of active operation of the device under conditions of severe vertical and horizontal loads.

A qualitative comparative assessment of the probability of deformation can be performed by controlling the grain size and the area of the membrane, on which the values of mechanical stresses are close to the maximum, i.e. exceed the value in the central region of the membrane. The grain size of molybdenum varies in the range from 27 to 67 nm, and the grain size of aluminum varies from 166 to 502 nm according to experimental data (Fig. 1, *b* and *c*). A microimage of the molybdenum film was acquired using Quanta FEI 3D scanning-electron microscope (SEM) with an accelerating voltage of 10 kV, a focal distance of 7.2 mm, and an angle of inclination of the sample -2°. The transmission electron microscopy (TEM) study was performed using Titan Themis 200 electron microscope with an accelerating voltage of 200 kV. According to other researchers, the size (width) of aluminum grains varies from 500 to 1500 nm [10], the average size (width) of molybdenum grains ranges from 11.7 to 48.5 nm [11]. Thus, the aluminum grains are wider than molybdenum grains. This means that the Al film (compared to the Mo film) has fewer boundaries on the same membrane area, and therefore the probability of critical deformation is lower.

552 nm of aluminum was deposited on a Mo film and 350 nm of molybdenum was deposited on an Al film for minimizing internal stresses in the film, ensuring maximum safety margin and resistance to various types of loads to form a structure with a set of alternating layers having vertical (molybdenum) and horizontal (aluminum) grain orientation. Then, a membrane was formed from Al−Mo by photolithography and plasma chemical etching of the Si substrate.

The roughness was measured using a Wyko NT9300 optical profilometer. The area of 0.84×3 mm was scanned for all types of samples (pure silicon wafer with crystallographic orientation (100) (Si), thin aluminum film on silicon wafer (Si−Al), thin molybdenum film on silicon wafer (Si−Mo), thin films of molybdenum and aluminum on silicon wafer (Si−Mo−Al and Si−Al−Mo). Fig. 2 shows the dependence of the roughness (R_a) on the type of sample. Therefore, the initial plate has the lowest roughness of 6.0 nm, the roughness for a sample with one type of film ($R_a = 6.2$ nm for molybdenum and $R_a = 13.7$ nm for aluminum) will be directly proportional to the horizontal grain size, the film roughness for a sample with two types depends on the type of material of the upper measured layer ($R_a = 11.3$ nm for the upper layer of molybdenum and $R_a = 19.5$ nm for the upper layer of aluminum).

The blowing method for measuring the mechanical properties of thin films is used on a test bench, which includes a line with the possibility of supplying air (vacuum, argon or other gas) at a pressure of up to 7 atm, filter regulator MC104-D00, proportional pressure regulator ER104-5PAP, cylinder-receiver with analog pressure gauge, Veeco Wyko NT9300 optical profilometer, samples for the study, control board. Figure 3 shows a test bench for measuring the mechanical properties of thin films. The cylinder receiver acts as a surge tank to smooth the gas pressure drops, as well as for ensuring the overall stability of the system. The supplied air enters the filter regulator MC104-D00 with a filter element made of HDPE (high density polyethylene) membrane type with a filtration degree of 25 μm according to ISO 8573-1:2010. The pressure regulator is responsible for maintaining a constant and controlled level of overpressure. Several pressure gauges are located at various points of the test bench for ensuring real-time monitoring of pressure levels. There are also built-in safety valves for automatic relief of overpressure in case of rupture of the membrane. The software for the Arduino Mega 2560 board allows controlling the pressure supply and relief. The accuracy of the overpressure supply is 0.01 atm.

According to the blowing technique, the calculation of the biaxial modulus of elasticity $E/(1-\mu)$ is performed from the dependence of the deflection of the membrane w on the overpressure P according to the formula [3]:

$$\frac{E}{1-\mu} = \frac{Pa^4}{C_2 h w^3}, \tag{1}$$

where P — overpressure, h — membrane thickness, w — membrane deflection, a — membrane radius, E — Young's modulus, μ — Poisson's ratio, C_2 — coefficient.

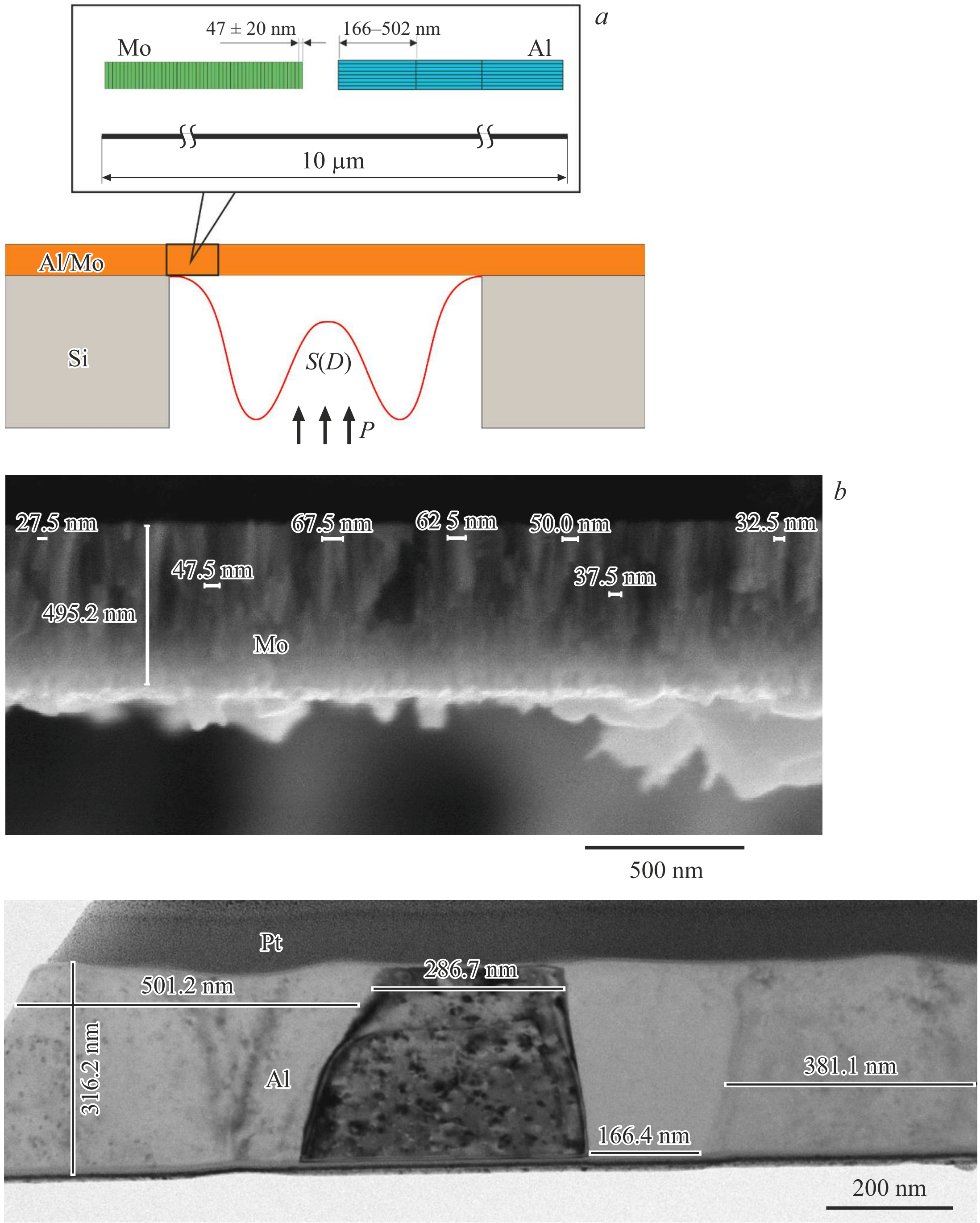

Figure 1. *a* — mutual orientation of the grains of the material and external impacts; *b* —SEM-image of the molybdenum film; *c* —TEM-image of the aluminum film.

Table 2. Study of mechanical properties

Diameter, mm	Critical pressure, atm		Biaxial modulus of elasticity, GPa	
	Si–Al–Mo	Si–Mo–Al	Si–Al–Mo	Si–Mo–Al
1.00	2.66	1.72	603	291
0.75	3.64	2.24	615	490
0.50	5.77	3.32	691	400

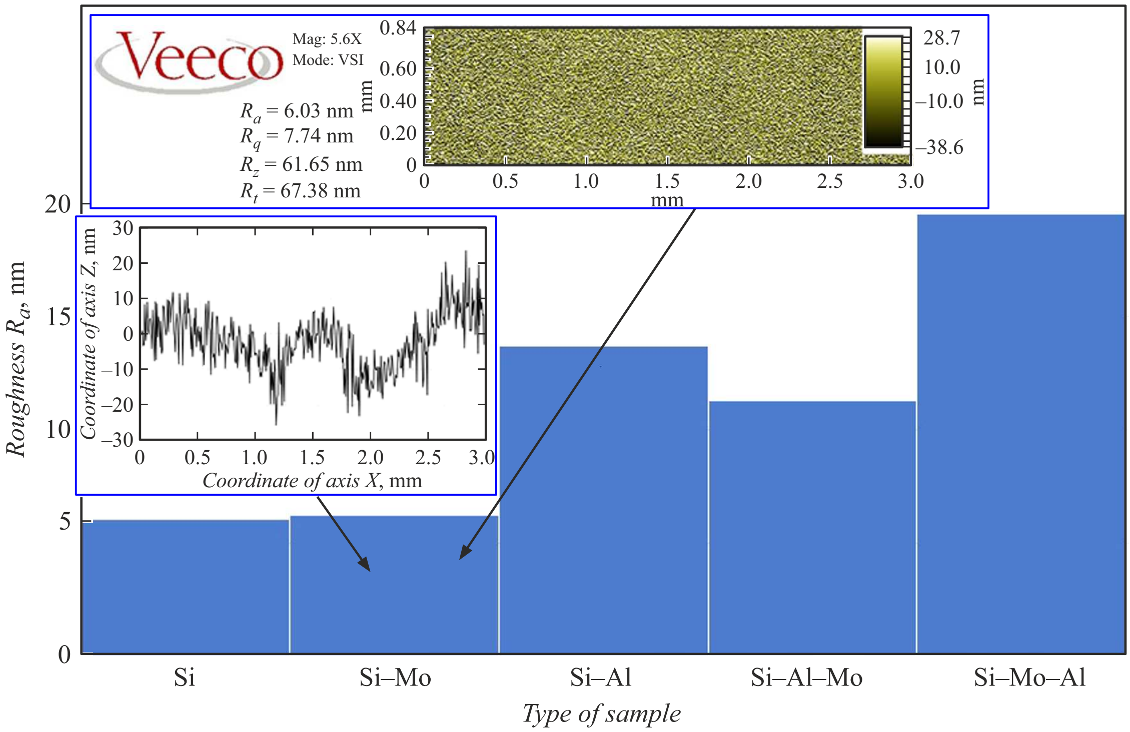

Figure 2. The dependence of roughness on the sample type. The relief distribution and automated calculation of the roughness value (R_a) using the software of the Veeco Wyko NT9300 optical profilometer are demonstrated on the inserts using the Si–Mo sample as an example.

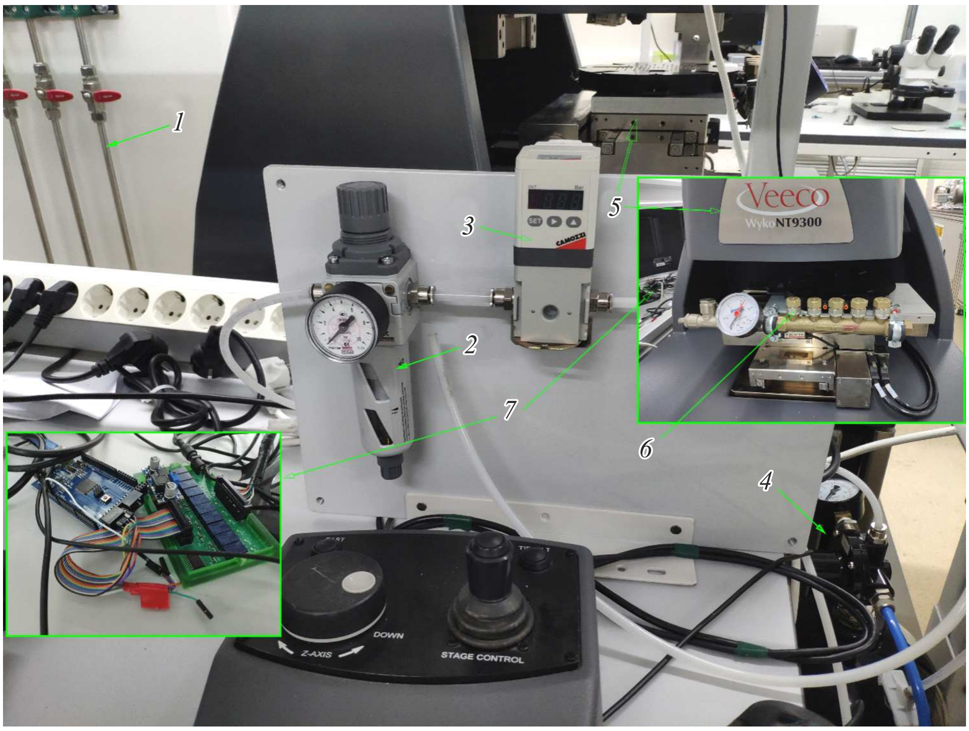

Figure 3. A test bench for measuring the mechanical properties of thin films. *1* — main line with the possibility of supplying air (vacuum, argon or other gas) under pressure up to 7 atm, *2* — filter–regulator MC104-D00, *3* — proportional pressure regulator ER104-5PAP, *4* — cylinder receiver with analog pressure gauge, *5* — optical profilometer Veeco Wyko NT9300, *6* — samples for the study, *7* — control board.

$C_2 = 8/3$ is usually used when working with round membranes [12]. The results of the study of mechanical properties are listed in Table 2.

The biaxial modulus of elasticity $E/(1-\mu)$ is 636 ± 54 GPa for the structure of Si–Al–Mo (the Al film is located on the Si substrate, the Mo film is located on the Al film), it is equal to 394 ± 102 GPa for the structure of Si–Mo–Al.

Thus, the paper presents the idea of the impact of grain orientation and size on the mechanical properties of thin-film membranes. It is necessary to form grains (grain boundary) perpendicular to the direction of external action, it is necessary to increase the contact area of the grain of the material and the substrate, strive to position (form) the middle of the grain in the area of maximum mechanical stresses for increasing the mechanical strength (critical rupture pressure) of the material. It is necessary to apply layers with alternating grain orientation to increase the margin of mechanical strength and expand the list of bearable loads. The mechanical properties vary depending on the sequence of formation of layers on the substrate. The membrane rupture critical value of the structure can be increased by more than 50%: from 1.72 to 3.32 atm for the structure of Si–Mo–Al and from 2.66 to 5.77 atm for the structure of Si–Al–Mo depending on the diameter, i.e., with the correct sequence of formation of layers, the membrane consisting of a layer of Al (316 nm) and a layer of Mo (350 nm), withstands greater critical pressure than a membrane consisting of a Mo (495nm) layer and an Al (552 nm) layer. Similarly, it is possible to increase the biaxial modulus of elasticity by more than 50%: 394 GPa for the structure of Si–Mo–Al and 636 GPa for the structure of Si–Al–Mo.

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Conflict of interest

The authors declare that they have no conflict of interest.

References

[1] Yu.P. Borodin, V.V. Kesaev, A.N. Lobanov, S.A. Ambrozevich, Tech. Phys. Lett., **49** (10), 52 (2023).

[2] N.A. Dyuzhev, E.E. Gusev, T.A. Gryazneva, A.A. Dedkova, D.A. Dronova, V.Yu. Kireev, E.P. Kirilenko, D.M. Migunov, D.V. Novikov, N.N. Patyukov, A.A. Presnukhina, A.D. Bakun, D.S. Ermakov, Nanotechnologies in Russia, **12** (7-8), 426 (2017). DOI: 10.1134/S1995078017040073.

[3] N.A. Dyuzhev, E.E. Gusev, M.A. Makhiboroda, Mech. Solids, **57** (5), 1044 (2022). DOI: 10.3103/S002565442205017X.

[4] J.H. Liu, J.X. Yan, Z.L. Pei, J. Gong, C. Sun, Surf. Coat. Technol., **404**, 126476 (2020). DOI: 10.1016/j.surfcoat.2020.126476

[5] M.G. Mueller, M. Fornabaio, G.Žagar, A. Mortensen, Acta Mater., **105**, 165 (2016). DOI: 10.1016/j.actamat.2015.12.006

[6] M.P. Kalashnikov, V.P. Sergeev, V. Neyfeld, AIP Conf. Proc., **1623** (1), 221 (2014). DOI: 10.1063/1.4901483

[7] K. Nakamura, H. Ohashi, Y. Enta, Y. Kobayashi, Y. Suzuki, M. Suemitsu, H. Nakazawa, Thin Solid Films, **736**, 138923 (2021). DOI: 10.1016/j.tsf.2021.138923

[8] G.A. Shneerson, V.V. Titkov, K.V. Voloshin, Tech. Phys. Lett., **49** (5), 11 (2023). DOI: 10.21883/TPL.2023.05.56017.19510.

[9] A.V. Novak, V.R. Novak, A.A. Dedkova, E.E. Gusev, Semiconductors, **52** (15), 1953 (2018). DOI: 10.1134/S1063782618150095

[10] S.S. Manohin, V.I. Betekhtin, A.G. Kadomtsev, M.V. Narykova, O.V. Amosova, Yu.R. Kolobov, D.V. Lazarev, Phys. Solid State, **65** (1), 126 (2023). DOI: 10.21883/PSS.2023.01.54986.492.

[11] H. Zhao, J. Xie, A. Mao, A. Wang, Y. Chen, T. Liang, D. Ma, Materials, **11** (9), 1634 (2018). DOI: 10.3390/ma11091634

[12] K.S. Chen, K.-S. Ou, in *Handbook of silicon based MEMS materials and technologies* (Elsevier, 2015), p. 398. DOI: 10.1016/b978-0-323-29965-7.00017-8

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