Peculiarities of the Influence of Nanostructuring of $[SiO_2/Si_3N_4]_n$ Multilayer Membranes on Its Thermal and Mechanical Properties

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***Abstract*— The effect of nanostructuring on the mechanical and thermal properties of $[SiO_2/Si_3N_4]_n$ multilayer membranes, where n is the number of repetitions, was studied. It also was theoretically found that, when the membrane thickness is decreased, the mechanical deformation of the layers at a given pressure must take into account the transition from linear to nonlinear stress mechanism. This corresponds to the experimental results of testing the strength of round SiO_2 (600 nm)/Si_3N_4 (130 nm)/SiO_2 (400 nm)/Si_3N_4 (130 nm) multilayer membranes of different diameters of 1 mm, 1.4 mm, 1.8 mm respectively, within the measurement error. The good agreement between the numerical and analytical calculation of the equivalent heat capacity, thermal conductivity, and density of $[SiO_2/Si_3N_4]_n$ multilayer membrane of different thickness and composition (for n = 8, 16, 28) is demonstrated. A decrease in the equivalent thermal conductivity of $[SiO_2/Si_3N_4]_n$ membrane with increasing n is shown, which corresponds to the predominant heat propagation along the boundary of the layers. The results obtained can be used in the development of nanostructured $[SiO_2/Si_3N_4]_n$ multilayer membranes for various applications, such as Micro-Electro-Mechanical Systems (MEMS), bio-, gas-, pressure-sensors, X-ray sources and ultra-strong thin coatings.**

Keywords—Multilayer membranes; nanostructuring; silicon oxide;silicon nitride; mechanical stress; geometric nonlinearity; maximum deformation; heat capacity; thermal conductivity

I. Introduction

Thermal and mechanical parameters of thin films used in nanoelectronics, sensors, and MEMS/NEMS (Micro-/Nano-Electro-Mechanical-Systems) play an important role for its correct work. Variation of these parameters will greatly affect the stability of the devices, depending on their purpose, which is critical and must be taken into account in the development of their design. It is known that nanostructuring of thin films, one of the variants of which is the formation of multilayer composite nanostructures with alternation of nanoscale layers, can lead to a noticeable improvement in their separate properties, which differ from the properties of bulk materials [1]. At present, thin dielectric multilayer composite membranes find wide application in the field of micro and nanoelectronics, in particular, in the development of piezoelectric nanowarchers, deformable mirrors for adaptive optics, microfocus X-ray tubes, thermal MEMS transducers of physical quantities such as gas flow sensors, accelerometers on thermal effects, pressure sensors, etc. [2,3]. Particularly attractive from this point of view are composite dielectric membranes of the type $[SiO_2/Si_3N_4]_n$, where n is the number of repetitions of the two-layer film $[SiO_2/Si_3N_4]$, as they provide good mechanical strength, have good adhesion and are compatible with silicon CMOS technology [4]. However for definition of mechanical strength of such "sandwiches" made from $[SiO_2/Si_3N_4]_n$, a full-fledged physical model is needed that allows one to determine the nature of the change in the mechanical properties of a multilayer structure upon transition to nanometer dimensions, and also predict membrane deformation at operating pressures on its surface, which should correspond to the obtained experimental data. In addition to the above, for these membranes the experimental measurement of the total thermal properties (thermal conductivity, heat capacity) is a rather complex task, and direct numerical modeling is required to understand the effect of nanostructuring on the heat distribution in such membranes, depending on the composition and number of layers. The second section of the article deals with the features of the change in the mechanical properties of a circular membrane $[SiO_2/Si_3N_4]_2$ with a decrease in its transverse dimensions, where the appearance of nonlinear deformation effects should be taken into account. Section III compares the experimental and theoretical results of studies of deformation of membranes of various diameters in the range of operating pressures not exceeding its mechanical strength. In the IV section, the analytical and numerical model of heat distribution in a multilayer composite membrane $[SiO_2/Si_3N_4]_n$ is considered for the values n = 18, 16, 28, on the basis of which

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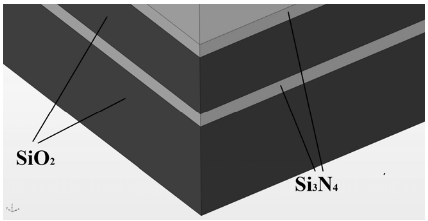
conclusions are drawn about the effect of nanostructuring on its thermal properties. Chapter V summarizes the conclusions of the article.

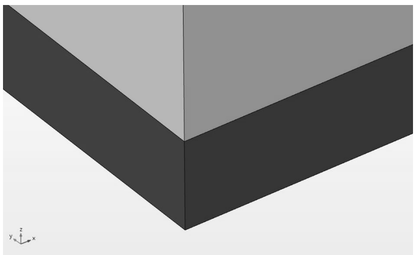
II. The Simulation of Mechanical Properties of Thin Multilayer Membranes: the Effects of Geometric Nonlinearity

To examine the effect of nanostructuring on the mechanical properties of structural elements of transducers, we will investigate the influence of the thickness of a single-layer silicon (Si) membrane and the multilayered membrane of the SiO_2-Si_3N_4-SiO_2-Si_3N_4 type used in thermal converters on the maximum deformation and mechanical stress arising during the application of constant pressure on the bottom wall of the membrane, as shown in Fig. 1.

Using the COMSOL Multiphysics software package, parametric modeling of thin-layer membranes of various compositions was made: 1) a single-layer silicon membrane and 2) a four-layer circular SiO_2-Si_3N_4-SiO_2-Si_3N_4 membrane. The purpose of the simulation was to reveal the dependence of the mechanical properties of the membrane (maximum displacement, stresses in the structure, and stresses of rupture) on the pressure on the membrane, taking into account the change in membrane thickness h and variation of parameters such as Young's modulus and Poisson's ratio.



a)



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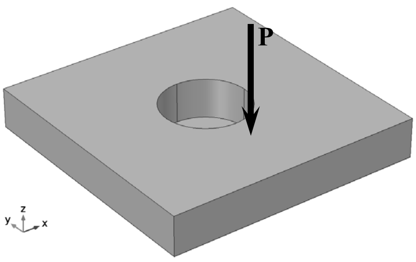


c)

Fig. 1 - a) Structure of a circular four-layer membrane; b) a single-layer silicon membrane; c) general view of the membrane on a silicon substrate

With small deflections of the membrane ($w_{\max} < h$) the displacement of the membrane can be described by expression [2]:

$$w(r) = \frac{Pa^4}{64D}\left[1 - \left(\frac{a}{r}\right)^2\right]^2 \quad (1)$$

where r – is the radius of the circular membrane, a – is the distance from the center of the membrane, and the maximum displacement in the center of the membrane $w_{\max}$ can be calculated by the formula:

$$w_{\max} = \frac{Pa^4}{64D} \quad (2)$$

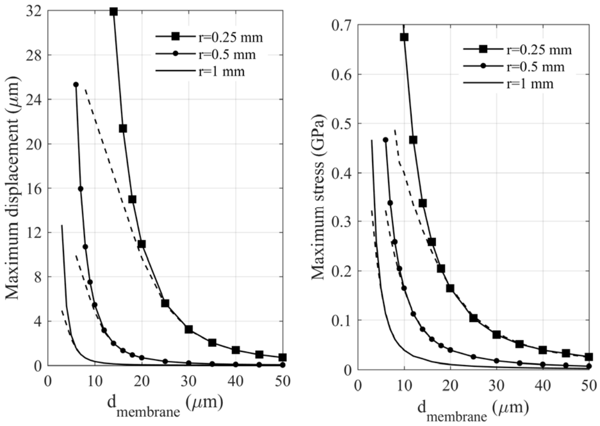
where $D = \frac{Eh^3}{12\left(1-\nu^2\right)}$ as a result we have:

$$w_{\max} = \frac{3Pa^4\left(1-\nu^2\right)}{16Eh^3} \quad (3)$$

where P – pressure on the membrane, E – Young's modulus, ν - Poisson's ratio, h – membrane thickness.

The single-layer silicon membrane has the following geometric parameters: radius - 0.25, 0.5 and 1 mm, thickness varied from 3 to 50 µm. The following values of the membrane material parameters were used in the calculations: Young's modulus $E = 200\,GPa$ and Poisson's ratio $\nu = 0.28$. The dependencies of the maximum displacement and the maximum stress of the silicon membrane from the membrane thickness, calculated using both the linear and nonlinear solver, are shown in Fig. 2.

Turning to Fig. 2, we can say that there is a certain maximum displacement of the membrane, to which the dependence curves obtained with the help of a linear and nonlinear solver coincide. In this case, formula (3) is valid. At the same time, as the thickness of the membrane decreases, when the deviation is of the same order of magnitude as the thickness, deviations from the linear law of deformation of the membrane arise, which is easily seen from the graphs in Fig. 2. This necessitates the introduction of a nonlinear term in the membrane deformation equation, which allows one to adequately describe the deformation and the mechanical stresses that appear in the membrane in the process of applying external pressure, which is close to reality.



(a) (b)

Fig. 2 - Comparison of the mechanical properties of a silicon membrane calculated using a linear (solid line) and nonlinear (dashed line) solver a) the dependence of the maximum displacement of the Si membrane on the thickness of the membrane; b) the dependence of the maximum stress of the Si membrane on the membrane thickness

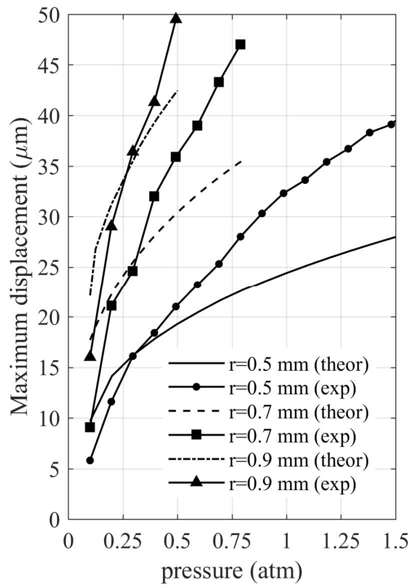


Fig. 3 - Experimental and theoretical dependence of the maximum displacement of a circular four-layer membrane on the applied pressure

III. The Deformation of [SiO2/Si3N4/SiO2/Si3N4] Multilayer Membrane Under Constant Pressure: Comparison of Experimental and Theoretical Study

Next, we present the results of modeling the four-layer circular membrane with SiO_2-Si_3N_4-SiO_2-Si_3N_4 structure for its mechanical properties. Membrane has the following geometric parameters: radius - 0.5, 0.7 and 0.9 mm; the thickness of the lower SiO_2 layer is 0.6 μm; the thickness of both Si_3N_4 layers is 0.13 μm; the thickness of the top layer of Si3N4 is 0.4 μm. In the calculations, the following values of the membrane material parameters were used: for Si3N4 Young's modulus $E = 250\,GPa$ and Poisson's ratio $\nu = 0.23$, for SiO_2 Young's modulus $E = 70\,GPa$ and Poisson's ratio $\nu = 0.17$.

In Fig. 3 shows the theoretical and experimental dependences of the maximum deflection of a circular four-layer membrane of radius 0.5, 0.7 and 0.9 mm from the applied pressure on the membrane. The theoretical curve was obtained using the COMSOL Multiphysics software package.

The theoretical data obtained were tested by experiment. In the course of the experiment, a single-crystal silicon wafer with a diameter of 150 mm with a thickness of 460 μm was studied with a set of four alternating dielectric layers of silicon oxide and nitride. The membrane is a circle with a diameter of 1 mm, 1.4 mm and 1.8 mm. The silicon base of the chip is a square with a side of 6 mm. Further, increasing the pressure on the compressor, the dependence of the deflection of the membrane on the excess pressure was obtained. Under the deflection of the membrane, the authors consider the distance from the silicon surface to the top of the membrane [3].

According to experimental data, the critical pressure, that is, the pressure at which the membrane ruptures, is: 0.5 atm for a membrane with a radius of 0.9 mm; 0.8 atm for a membrane with a radius of 0.7 mm; 1.6 atm for a membrane with a radius of 0.5 mm. Thus, we can say that there is a certain regularity of the critical pressure $P_{\max}$ from the radius of the membrane r, namely $P_{\max} \sim \frac{1}{r^2}$

IV. The Influence of Nanostructuring of $[SiO2/Si3N4]_N$ Multilayer Membrane on Its Thermal Properties: Analytical and Numerical Model

To simplify the calculations, in the simulation of multilayer membranes of the type $[SiO_2/Si_3N_4]_n$, where n is the number of repetitions of a two-layer film [SiO2 / Si3N4] with layer thickness of about 25 nm, a study was carried out to replace the multilayer structure by a single-layer structure with specified parameters. In this model, the following parameters were calculated for the equivalent material: equivalent thermal conductivity, equivalent heat capacity and equivalent density.

Two fundamentally different cases of heat transfer across the membrane were considered, namely the movement of heat along the membrane layers and the transfer of heat is normal to the plane of the membrane.

For the case of heat transfer along the plane of the membrane, the authors [5] obtained a formula for calculating the equivalent longitudinal thermal conductivity:

$$K_{long} = \frac{k_1 \cdot h_1 + k_2 \cdot h_2 + \cdots + k_n \cdot h_n}{h_{total}} \quad (4)$$

where $k_1, k_2, ..., k_n$ is the thermal conductivity of the n-th layer, $h_1, h_2, ..., h_n$ is the thickness of the n-th layer, h_{total} is the thickness of the membrane.

Based on this formula, the equivalent longitudinal thermal conductivity of composite dielectric membranes of the type [SiO_2 / Si_3N_4]$_n$ was calculated, where n is the number of repetitions of a two-layer film, in this case n = (8,16,28). The results are shown in Table I.

TABLE I. DEPENDENCE OF EQUIVALENT LONGITUDINAL HEAT CONDUCTIVITY FROM THE NUMBER OF LAYERS IN THE MEMBRANE TYPE [SIO2/SI3N4]$_N$

Number of Layers (pieces)	Equivalent Thermal Conductivity (J/m·K)
8	7.3
16	7.02
28	6.71

In the case of heat transfer normally to the membrane surface, the equivalent perpendicular thermal conductivity was calculated by the formula derived by the authors:

$$K_{perp} = \frac{h_{total}}{\frac{h_1}{k_1} + \frac{h_2}{k_2} + \cdots + \frac{h_n}{k_n}} \quad (5)$$

According to this formula, the equivalent perpendicular thermal conductivity of membranes with a different number of layers was calculated. The results are shown in Table II.

TABLE II. DEPENDENCE OF EQUIVALENT PERPENDICULAR HEAT CONDUCTIVITY FROM THE NUMBER OF LAYERS IN THE MEMBRANE TYPE [SIO2/SI3N4]$_N$

Number of Layers (pieces)	Equivalent Thermal Conductivity (J/m·K)
8	2.35
16	2.29
28	2.96

In the COMSOL Multipysics modeling environment, a simulation of the case was carried out, where the heat transfer simultaneously occurs in the two directions simultaneously. Equivalent thermal conductivity and density were calculated as the average value over the entire volume of the membrane. The results of the simulation are given in Table 3.

TABLE III DEPENDENCE OF EQUIVALENT THERMAL CONDUCTIVITY, EQUIVALENT HEAT CAPACITY AND EQUIVALENT DENSITY FROM THE NUMBER OF LAYERS IN THE MEMBRANE TYPE [SIO2 / SI3N4] N IN THE CASE OF THE HEAT DISTRIBUTION BY TWO DIRECTIONS

Number of Layers (pieces)	Equivalent Heat Capacity (J/kg·K)	Equivalent Thermal Conductivity (J/m·K)	Equivalent Density (kg/m^3)
8	722.9	8,84	2412
16	723.3	8,48	2400
28	719.2	8,07	2523

Theoretical calculations showed good convergence of numerical and analytical values of the equivalent heat capacity, thermal conductivity of a multilayer membrane. A decrease in the equivalent thermal conductivity of the membrane is shown with an increase in the number of layers, which corresponds to the prevailing heat spread along the boundary of the layers.

V. RESULT AND DISCUSSION

As a result of this work, the mechanical and thermal properties of multilayer membranes of the type [SiO_2/Si_3N_4]$_n$ were investigated. Theoretical dependences of the maximum displacement of a multilayer membrane of the SiO_2-Si_3N_4-SiO_2-Si_3N_4 type on the applied pressure are obtained, which have good convergence with the experimental data. Also, the dependence of the critical pressure on the membrane on the radius of the membrane are identified: $P_{max} \sim \frac{1}{r^2}$.

Using the example of a single-layer silicon membrane, it was found that the theoretical calculation of the mechanical deformation of layers must take into account the transition from a linear law to a nonlinear law in the case when the deflection of the membrane is greater than its thickness.

The influence of the number of layers of a multilayer membrane of the type [SiO_2/Si_3N_4]$_n$ on the analytical values of the equivalent heat capacity, thermal conductivity and membrane density is considered. Theoretical calculations showed good convergence of numerical and analytical values of the equivalent heat capacity, thermal conductivity of a multilayer membrane. It was found that an increase in the number of layers leads to a decrease in the thermal conductivity of the membrane, which corresponds to the prevailing heat spread along the boundary of the layers.

The results obtained in this work will be used in further studies in the development of nanostructured multilayer membranes, the scope of which includes X-ray sources, biosensors, thin coatings and MEMS devices.

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