Development and investigation of micro- and nanostructures of metamaterials to form the necessary characteristics and coefficients of piezoelectric elements

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Abstract. The development and research of micro and nanostructures for the manufacture of ultrasonic piezoelectric elements has been carried out. The structures obtained in this work have practical applications for the manufacture of piezoelectric and piezoelectric elements, in particular, for using in liquid flowmeters as receivers and emitters of an ultrasonic signal. A structure of nanocells was obtained that was different from standard piezoelectric elements (disk, cylinder), but with the same coefficients, characteristics, and radiation pattern.

1. Introduction
The piezomaterials are of great interest because of their unique properties since the discovery of the direct and inverse piezoelectric effect of single crystals by the Curie brothers in 1880 and 1881. And the discovery of the possibility of polarizing ceramic material with an electric field in 1946 led to the widespread distribution of piezoelectric and piezoelectric elements. The ability of piezoelectric materials to convert mechanical energy into electrical energy and vice versa allows them to be used in ultrasonic measurements, in pressure measurements, in medicine, in flow detectors, seismic sensors and energy collection systems. In addition, applying an AC voltage to the material causes it to vibrate and thus creates mechanical waves with the same frequency as the electric voltage, which can be used in micro-positioning devices, such as attenuators, scanning tunnelling microscopes, etc. Similarly, if mechanical vibration is used, a charge of a proportional size and the same frequency will be generated [1]. In some problems, piezoelectric materials are indispensable, since without this physical process it will be impossible to make studies or measurements. Copper and aluminum are used as a metal layer in the study.

But all the characteristics, coefficients, and piezoelectric constants of existing standard piezoelectric elements, such as a disk, cylinder, etc., are dictated by their structure and chemical composition. This leads to the fact that the properties of the piezoelectric elements, such as the radiation pattern or conversion coefficient, remain constant for the whole wide range of tasks. It is necessary to find such technical solutions in which the maximum response of the piezoelectric element is achieved, or to seek a compromise between the efficiency of the piezoelectric element and the complexity of the entire system design. In non-trivial problems, the necessary radiation pattern, which will correspond to the best signal emission result, cannot be obtained with standard piezoelectric
elements. Even the addition of impurities to the crystallographic structures of piezoelectric materials cannot lead to the possibility of dynamically adjusting the piezoelectric constants in certain directions, since the set of alloying components is limited, and any changes to the geometry of standard piezoelectric elements, in an attempt to obtain the desired characteristics, will lead to a change in the electrical and acoustic properties. One way to avoid losing the necessary parameters is to use metamaterials with a specific geometry to create piezoacoustic and piezoelectric elements. In this case, it will be possible to select material for a given geometry, and not vice versa [1].

With a certain structure of the connection of the nanocells (3 - 200 nm) of the piezoelectric materials, electromechanical bonds are formed, in which the piezoelectric properties are improved by changing parameters such as the electromechanical coupling coefficient, conversion coefficient, quality factor, piezoelectric module, etc. Due to these improvements, the efficiency of piezoelectric elements is significantly increased. These nanocells are three-dimensional (3D) structural nodes, as shown in Figure 1 [1].

![Figure 1. Three-dimensional structural units [1].](image)

Nanocomposite piezoelectric metamaterials from these structures achieve a high conversion coefficient and a piezoelectric voltage coefficient, and also have a high flexibility of characteristics, which is not achievable using standard piezoelectric and piezoelectric elements [3].

When using these structures, the field of application of piezoelectric elements significantly expands and their efficiency increases several times. There is the possibility of manufacturing piezoelectric elements with the necessary geometry for the user based on the task.

The purpose of this work is to obtain new structures to form the necessary characteristics.

2. Piezoelectric elements
One of the standard versions of piezo-acoustic elements are discs of various diameters (figure 2).

![Figure 2. Standard piezoacoustic element with a diameter of 4 mm for a frequency of 1 MHz.](image)

This element has certain characteristics and coefficients, which are key parameters. For this type of piezoelectric elements, there can be the following modes, presented in table 1[12].

The main coefficients and characteristics for piezoelectric elements, which determine their properties and are determined by the structure, geometry and impurities [2]:
1. Dielectric constant
2. Conversion coefficient
3. Dielectric loss factor
4. Quality factor of the elastic system
5. Frequency constant
6. Electromechanical coupling coefficient
7. Coefficient of the piezoelectric charge
8. Piezoelectric voltage factor
9. Coefficient of elastic compliance
10. Rate of aging
11. Curie point

| Table 1. Piezoelectric Element Mods [12] |
|----------------------------------------|
| Vibration mode                        | Dimensions | Constants to be calculated |
|                                       | L - length | W - width | D - diameter | Piezoelectric | Mechanical |
| Radial mode                           |            |           | D > 10 Th    | $k_p, \varepsilon_{33}^S, \varepsilon_{33}^T$ | $\sigma^E, S_{12}, Q_p$ |
| Thickness extension mode              |            |           | D > 10 Th    | $k_T, \varepsilon_{33}^S$ | $C_{33}^E, C_{33}^E, S_{13}, Q_T$ |

To describe these constants, it is necessary to consider elements as 3D objects with three possible directions of action of forces and three rotation axes, as shown in Figure 6. Also in this figure, directions (1 - 6) are numbered [3].

All characteristics and constants are indicated in accordance with the position of the electrodes on the cell, the direction of the applied voltage or load, the direction of shear, etc. Examples of these designations are presented in table 2 [12].

Dielectric constant (K) - the relative dielectric constant is defined as the ratio of the dielectric constant of the material to the dielectric constant of free space. Usually this is measured well below mechanical resonance. The dielectric constant is obtained from measurements of static capacitance at a
frequency of 1 kHz using a standard impedance bridge. This constant is calculated by the following formula [2, 12]:

$$K_T = \frac{\varepsilon_T}{\varepsilon_0} = \frac{C_T}{\varepsilon_0 A}$$  \hspace{1cm} (1)

where $\varepsilon_T$ is the constant voltage permeability ($F/m$), $\varepsilon_0$ is the free space permittivity ($8.854 \times 10^{-12} F/m$), $A$ is the surface area ($m^2$), $T$ is the distance between the electrodes (m), $C$ - capacity (f).

Table 2. Designations [12]

| Relative dielectric constant $\varepsilon^s_3/\varepsilon_0$ and $\varepsilon^T_3/\varepsilon_0$ | $K^S$ | All strains in the material are constant or mechanical deformation is blocked in any direction.
| $K^T$ | Electrodes are perpendicular to 3 axis.
| $K_1$ | Electrodes are perpendicular to 1 axis.

Electromechanical coupling factor

| $d_{15}$ | Stress or strain is equal in all directions perpendicular to 3 axis.
| $d_{16}$ | Electrodes are perpendicular to 1 axis.

Piezoelectric charge coefficient

| $d_{33}$ | Applied stress, or piezoelectrically induced strain is in 3 direction.
| $d_{36}$ | Electrodes are perpendicular to 3 axis.

Piezoelectric voltage coefficient

| $g_{31}$ | Applied stress, or the piezoelectrically induced strain is in 1 direction.
| $g_{36}$ | Electrodes are perpendicular to 3 axis.

Elastic compliance

| $S_{33}$ | Compliance is measured with closed circuit.
| $S_{56}$ | Stress or strain is shear around 3 direction.
| $S_{11}$ | Strain or stress is in 3 direction.
| $S_{13}$ | Stress or strain is in 1 direction.

Dielectric loss coefficient - dielectric loss coefficient is defined as the loss tangent (tan d). The loss factor is the ratio of conductivity to the perception of a parallel equivalent circuit of a ceramic element. The loss factor can be measured directly using an impedance bridge.

The quality factor of an elastic system (Q) is the ratio of reactance to resistance in a series equivalent circuit representing a piezoelectric resonator. The coefficient $Q_m$ is also related to the sharpness of the resonant frequency. This value is calculated using the following formula [2, 12]:

$$Q_m = \frac{f_r^2}{2\pi f_r Z_m C^T (f_a^2 - f_r^2)}$$  \hspace{1cm} (2)

where $f_a^2$ is the anti-resonance frequency (Hz), $f_r^2$ is the resonance frequency (Hz), $Z_m$ is the minimum impedance at $f_r$ (Ohm), $C$ is the capacitance (F).

In another way, the coefficient $Q_m$ can also be determined using the equation [3]:

$$Q_m = \frac{f_r}{f_1 - f_2}$$  \hspace{1cm} (3)

Frequency constants (N) - (frequency constant N) is the product of the resonant frequency and the linear measurement that determines the resonance. N is also equal to half the speed of sound in the same direction. The resonance modes for a standard disc are shown schematically in table 1.

Frequency constants for standard elements are presented in table 3 [12].

Electromechanical coupling coefficient (k) - the coefficients describe the ability of a piezoelectric element to convert energy from electrical to mechanical and vice versa. The square of the electromechanical coupling coefficient is defined as the ratio of the accumulated converted energy of one type (mechanical or electrical) to the input energy of the second type (electrical or mechanical).
The index shows the relative directions of electrical and mechanical quantities and the type of vibrations [4].

**Table 3. Frequency constants [12]**

| Mode type, element type  | Coefficient calculation                        |
|--------------------------|------------------------------------------------|
| Transverse mode, thin bar| \( N_{31} = f_r \cdot L \)                       |
| Radial mode, disc        | \( N_p = f_r \cdot D \)                           |
| Thickness mode, disc     | \( N_T = f_r \cdot T \)                           |
| Length mode, cylinder    | \( N_{33} = f_r \cdot L \)                       |
| Shear mode, plate        | \( N_{15} = f_r \cdot T \)                       |

where L is the length (m), D is the diameter (m), T is the thickness (m).

Electromechanical coupling coefficient can be calculated for various modes [2, 12]:

\[
k_{31} = \sqrt{\frac{\pi}{f_a}} \cdot \frac{1}{2f_r \pi f_a - \tan\left(\frac{\pi f_a}{2f_r}\right)}
\]

(4)

\[
k_p \approx \sqrt{\frac{2.51 f_a - f_r}{f_a} - \left(\frac{f_a - f_r}{f_a}\right)^2}
\]

(5)

\[
k_t = \sqrt{\frac{\pi f_r \cot\left(\frac{\pi f_r}{2f_a}\right)}{2f_a}}
\]

(6)

\( k_{33} \) and \( k_{15} \) can be calculated similarly to \( k_t \), using the corresponding resonant frequencies.

The piezoelectric charge coefficient (\( d \)) is the ratio of the electric charge generated per unit area to the applied force (C/N or m/V).

The constants \( d \) are calculated by the equations [2, 12]:

\[
d = k \sqrt{\varepsilon T \varepsilon E} (\text{C/N})
\]

(7)

\[
d_{31} = k_{31} \sqrt{\varepsilon_{33} T \varepsilon_{11} E}
\]

(8)

\[
d_{33} = k_{33} \sqrt{\varepsilon_{33} T \varepsilon_{33} E}
\]

(9)

\[
d_{15} = k_{15} \sqrt{\varepsilon_{11} T \varepsilon_{55} E}
\]

(10)

Piezoelectric stress coefficient (\( g \)) is the ratio of the generated electric field to the applied mechanical stress (V m/N).

The constants \( g \) are calculated by the equations [2, 12]:

\[
g = \frac{d}{\varepsilon T} (\text{V m/N})
\]

(11)

\[
g_{31} = \frac{d_{31}}{\varepsilon_{33} T}
\]

(12)

\[
g_{33} = \frac{d_{33}}{\varepsilon_{33} T}
\]

(13)

\[
g_{15} = \frac{d_{15}}{\varepsilon_{11} T}
\]

(14)

Elastic compliance coefficient (\( S \)) - Young's modulus describes the mechanical properties of stiffness and is expressed as the ratio of stress to strain. In a piezoelectric material, mechanical stress causes an electrical response that counteracts the resulting deformation. The value of Young's modulus
depends on the direction of stress and strain and electrical conditions [5]. The inverse value of Young's modulus $Y$ is the elastic compliance $s$, which can be calculated as follows (table 1) [2, 12]:

$$s = \frac{1}{Y} = \frac{\text{strain}}{\text{stress}} = \frac{1}{\rho v^2} \left( \frac{m^2}{N} \right)$$  \hspace{1cm} (15)

$$s_{33}^D = \frac{1}{4\rho f_a^2 L^2}$$ \hspace{1cm} (16)

$$s_{33}^E = \frac{1}{s_{33}^D}$$ \hspace{1cm} (17)

$$s_{11}^E = \frac{1}{4\rho f_a^2 L^2}$$ \hspace{1cm} (18)

$$s_{11}^D = s_{11}^E (1 - k_{33}^2)$$ \hspace{1cm} (19)

$$S_{55}^D = \frac{1}{4\rho f_a^2 T h^2}$$ \hspace{1cm} (20)

$$S_{55}^E = \frac{s_{55}^E}{1 - k_{15}^2}$$ \hspace{1cm} (21)

Aging rate ($Z$) - an indicator of the change in certain parameters of the material over time.

$$Z = \frac{1}{(\log t_2 - \log t_1)} \left( \frac{P_2 - P_1}{P_1} \right)$$ \hspace{1cm} (22)

where $t_2, t_1$ is the number of days after polarization, $P_2, P_1$ are the measured corresponding values of interest [3].

3. Modeling

Figure 4 shows a model of a standard piezoelectric element, which is used as a receiver and transmitter of an ultrasonic signal.

![Figure 4. Piezoelectric element.](image)

Figure 5. Microcell.
Figure 5 shows a micro-cell, which is used as a structural unit for constructing a new piezoelectric element (this cell was used in [1], a microscope image of the structure from these cells is shown in Figure 6 [1]).

The piezoelectric element shown in Figure 7 was constructed from these cells with the same geometric parameters as the standard one. In this sample, the upper and lower platforms of the model were chosen as electrodes for positive and negative potentials.

The standard model (Figure 4) and designed (Figure 7) were modeled under conditions of creating a potential difference between their surfaces. Figure 8 shows the deformation diagrams of both elements when applying a voltage of 5V.
4. Conclusion
A theoretical study of the constants and coefficients for piezoelectric elements was carried out, as well as the influence of various physical properties on the characteristics and response of piezoelectric and piezoelectric materials. It was found that the dielectric constant, conversion coefficient, dielectric loss coefficient, Q factor of the elastic system, frequency constant, electromechanical coupling coefficient, piezoelectric charge coefficient, piezoelectric stress coefficient, elastic compliance coefficient, aging rate and Curie point for the piezoelectric element depend on its structure and composition.

The processes of the piezoelectric effect for a standard piezoelectric element with a diameter of 4 mm and a thickness of 2 mm were simulated, as well as a piezoelectric element constructed from microcells with preserved geometric parameters. A unit cell option was proposed for piezoelectric material structures from which a non-standard element was assembled.

This element allows you to achieve different characteristics and efficiency compared to standard piezoelectric elements, while maintaining the same overall dimensions. The use of various configurations of microcells to create structures of piezoelectric materials allows you to change the properties, parameters and characteristics of piezoelectric elements made from these structures.

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