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Elastic properties of some clay minerals

A new method for the quantitative estimation of the elastic properties of clay minerals based on the principles of crystal-chemical systematization of clay minerals elastic properties was developed. An accurate mathematical method has been devised to approximate elastic properties of a random symmetry medium to those of a similar transversely isotropic elastic medium. A first successful attempt was made at a quantitative assessment of the elastic constants of all clay mineral constituents in transversely isotropic approximation, which fully agrees with experimental data on these minerals. Extensive research has been carried out into the anisotropy parameters of clay mineral elastic waves and their correlation with the structural features of minerals. This method can be used for approximating the elastic constants of other rock-forming minerals.

Key words: clay mineral, elastic constant, transversely isotropic approximation, stereo-projection, elastic anisotropy, phase velocity.

Własności sprężyste niektórych minerałów ilastych

Opracowano nową metodę oceny ilościowej sprężystych własności minerałów ilastych w oparciu o zasady krystaliczno-chemicznej systematyzacji tych własności. Nowa, dokładna matematyczna metoda ma na celu przybliżenie własności sprężystych ośrodka o losowej symetrii do tych o podobnym sprężystym ośrodku poprzecznie izotropowym. Pierwszej udanej próby dokonano przy ilościowej ocenie stałych sprężystych wszystkich komponentów minerałów ilastych w poprzecznie izotropowym przybliżeniu, co jest w pełni zgodne z danymi eksperymentalnymi dotyczącymi tych minerałów. Przeprowadzono wszechstronne badanie parametrów anizotropowych fal sprężystych minerałów ilastych i ich korelacji ze strukturalnymi cechami minerałów. Metoda ta może zostać wykorzystana do przybliżenia stałych sprężystych innych minerałów w formacjach skalnych.

Słowa kluczowe: minerał ilasty, stała sprężysta, przybliżenie poprzecznie izotropowe, stereo-projekcja, sprężysta anizotropia, prędkość fazowa.

Introduction

Research into anisotropy of elastic properties and seismic waves of clay minerals is of great interest in terms of oil and gas deposits exploration and applying seismic methods and techniques in the assessment of abnormally high pressures at great depths, as well as geophysical monitoring of such hazards as landslides, mud flows, soil subsidence, etc.

The work [9] has convincingly proved that the entire natural history of oil is associated with clay rocks. Being a component of clay rocks, organic matter turns into petroleum hydrocarbons due to the catalytic effect of rock-forming clay minerals. Argillaceous rocks located in the path of

hydrocarbons migration, contribute to deposits forming and prevent them from destruction.

Hydrocarbon deposits in clay collectors have been discovered in the menilite complex of the Carpathian Mountains (Ukraine) and the Bazhenkivska complex of West Siberia. These findings generate great scientific interest and stimulate further research into similar deposits as likely reserves of oil and gas [9].

It is predominantly the crystal-chemical structure of clay minerals and their specific patterns of interaction with water that determine clay properties such as plasticity, swelling

caused by water saturation, and shrinking caused by drying out. This accounts for the abnormal solidity and deformation behavior of clay rocks – their dilution resulting from dynamic external action, sharp decrease in strength caused by humidification, bellying of clay rocks under unbalanced load on the base rock. The biggest disaster in Europe associated with the disruption of the Malpas dam (France) in 1959 was proved to have resulted from ignoring the anisotropy of elastic and mechanical properties of clays [23]. The elastic properties anisotropy was determined by a high level of structuring in clay particles and extensive macro- and micro fracturing of clay rocks on which the dam had been built. Following intensive humidification caused by rain, clay rocks lost their solidity and collapsed under the dam’s weight.

Theoretical and experimental research into clay mineral elastic properties suggests that conventional theoretical

approaches and techniques fail to meet modern standards. Despite extensive research in this sphere being undertaken in many countries around the world, there is very little accurate data on the elastic properties of rock-forming clay minerals.

In fact, today only two clay minerals – muscovite and kaolinite – have been analyzed thoroughly enough to derive experimental data on a complete elastic constant matrix in monoclinic and triclinic approximated symmetry [2, 3, 7, 13, 24].

A new method of quantitative estimation of the elastic properties of clay minerals based on the principles of crystal-chemical systematization of clay minerals elastic properties [11, 12, 15, 16–19] was developed. An accurate mathematical method has been devised to approximate elastic properties of a random symmetry medium to those of a similar transversely isotropic elastic medium [10].

Structural anisotropy of the elastic properties of clay minerals

The major mineral constituents of clays are foliated minerals of the kaolinite-serpentine group [19, 20], the most common of these being kaolinite, halloysite, minerals of serpentine-lizardite group, and chrysotile, a fibrous mineral. Fine-grained mica in clays (illite) is similar to muscovite and biotite in its structure and composition. Smectites (the most important of them is montmorillonite) are extremely fine-grained and prone to swelling. Vermiculites and chlorites also occur as particles whose size is typical to clay particles.

Experimental data on the anisotropy of clay mineral elastic properties are scarce and rather controversial. That may be put down to a lack of mono-crystals of sufficient size and perfect quality, as well as the complexity of experimental research, with foliated minerals developing a perfect cleavage along (001). Atomic acoustic spectroscopy (DFT method) is known to have been directly applied to study all the components of the elastic constants matrix for a triclinic kaolinite approximation [22] (C_{mn} in GPa):

178.0 ± 8.8	71.5 ± 7.1	2.0 ± 5.3	-0.4 ± 2.1	41.7 ± 1.4	-2.3 ± 2.7
	200.9 ± 12.8	-2.9 ± 5.7	-2.8 ± 2.7	19.8 ± 0.6	1.9 ± 1.5
		32.1 ± 2.0	-0.2 ± 1.4	1.7 ± 1.8	3.4 ± 2.2
			11.2 ± 5.6	-1.2 ± 1.2	12.9 ± 2.4
				22.2 ± 1.4	0.8 ± 2.4
					60.1 ± 3.2

The above data show that the accuracy of estimating the kaolinite C_{11} and C_{22} elastic constants is over 8%, and the inaccuracies of C_{13} and C_{23} elastic constants are twice their absolute values. Moreover, the value of C_{23} elastic constant has been proven to be abnormally lower than zero ($C_{23} = -2.9$ GPa).

The matrix of kaolinite elastic pliabilities is as follows (S_{mn}, TPa^{-1}):

10.8	-2.0	-1.9	3.0	18.9	1.0
	5.9	0.6	1.6	1.5	0.0
		31.9	-2.7	-5.6	-2.4
			121.1	12.9	26.0
				82.9	2.7
					22.4

The matrix components of tensor linear compression are derived from the formula:

$$\beta_{ji} = S_{ijkk}$$

and presented below in a laboratory system of coordinates (β_{ji}, TPa^{-1}):

6.9	-1.4	14.8
	4.5	1.9
		30.6

The eigenvalues and eigenvectors of kaolinite tensor linear compression are as follows (β_m, TPa^{-1}):

$$\beta_1 = -1.0 (0.837; 0.356; -0.413)$$

$$\beta_2 = 5.2 (0.335; 0.933; 0.125)$$

$$\beta_3 = 37.7 (-0.431; 0.033; -0.901)$$

Stereo-projection isolines of Young’s modulus equal values $E(\vec{l})^{-1} = S_{ijkl}l_i l_j l_k l_l$ (Fig. 1) show that the difference between its maximum (more than 181 GPa) and minimal (less than 29 GPa) values in the directions that do not coincide with crystallographic axes is over 152 GPa.

The abnormally low negative value of the kaolinite C_{23} elastic constant evidently requires more accurate defining. The negative value of C_{23} might be caused by the ambiguity of the choice of the triclinic symmetry minerals coordinate system [10].

More experimental data on the elastic properties of mica have been given in transversely isotropic approximation, mica being similar to illite in its composition and structure [2, 3, 7].

Extensive experimental research on mica showed that despite the big size of mono-crystals available, making samples from foliated silicate crystals is an extremely difficult task, because of their cleavage foliation. As a result, the elastic wave velocity measurements were carried out by mechanical compression of samples with special clamps [3]. Mica was assumed to have a pseudo-hexagonal symmetry when measured, although in fact its structure has a monoclinic symmetry.

There is weak anisotropy of the elastic properties to be found in (001) mica crystal plane shown by the interatomic bond distribution analysis in the mica package with an ideal structural model [14, 18]. A schematic foliation structure based on conventional hexagons was used. When directions of the weakened and the stronger bonds in tetrahedral and octahedral crystal layers fail to coincide, their disorientation can be stated to imply disorientation of the stronger interatomic bonds, which accounts for pseudo-hexagonal symmetrization and a weak elastic anisotropy in (001) direction.

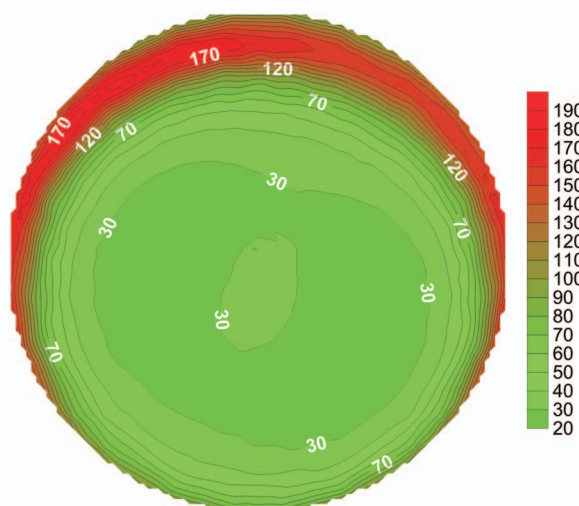


Fig. 1. Stereo-projection of Young's modulus isolines (E , GPa) of the triclinic kaolinite

The Mandelstam-Brillyuenivskiy scattering method [24] was used to measure the elastic constant matrix for a muscovite crystal with a monoclinic symmetry. Comparing our results with the previous research results [2, 3, 7] in transversely isotropic approximation, showed that C_{11} , C_{33} and C_{66} elastic

constants vary up to 5%. Besides, data on C_{12} in previous works were 15% underestimated, while for C_{13} and C_{44} the discrepancy turned out to be more than 30%.

Figure 2 represents a stereo-projection of the monoclinic muscovite Young's modulus isolines. As can be seen from the data, its symmetry is very close to the transversely isotropic symmetry. Young's modulus value actually only depends on the θ angle.

To estimate the accuracy of the transversely isotropic approximation of the monoclinic crystal elastic constant matrix the transversely isotropic anisotropy coefficient [2, 10] was used, which characterizes the deviation of the muscovite elastic anisotropy from the transversely isotropic medium most similar to it. Analytically, the elastic constants of a transversely isotropic medium can be derived from the following condition [10, 21]:

$$\left[(\Lambda_{il} - \Lambda_{il}^{(t)})^2 \right]_c = \min \quad (1)$$

where $\Lambda_{il} = C_{ijkl} n_j n_k / \rho$, $\Lambda_{il}^{(t)} = C_{ijkl}^{(t)} n_j n_k / \rho$; $C_{ijkl}^{(t)}$ – the elastic constants of an unknown transversely isotropic medium, which is similar to the elastic medium of the muscovite monoclinic crystal with elastic properties c_{ijkl} and density ρ ; C – tensor trace.

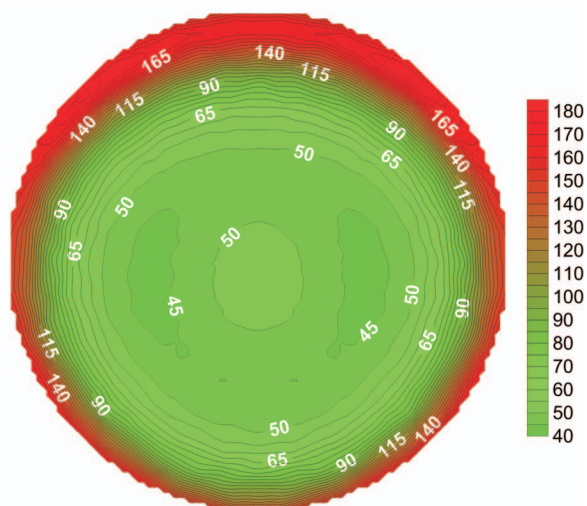


Fig. 2. Stereo-projection of Young's modulus isolines (E , GPa) of the monoclinic muscovite

To solve this equation the covariant form of Kristoffel's tensor was used when choosing the coordinate system [10]:

$$\Lambda_{il}^{(t)} = a_0 \delta_{il} + a_1 n_i n_l + a_2 e_i e_l + a_3 c_i c_l \quad (2)$$

where $c_i = [\vec{e}\vec{n}]_i$; n_i – components of the wave normal vector; e_i – components of the vector, which determines the direction of the main symmetry axis of the transversely isotropic medium.

The solution of equation (1) yields the unknown factors:

$$a_0 = \frac{(1+n_3^2)[\bar{e}\bar{c}]\Lambda[\bar{e}\bar{c}] - \bar{n}\Lambda\bar{n} + n_3^2\bar{e}\Lambda\bar{e}}{2n_3^2} \quad (3)$$

$$a_1 = \frac{\bar{n}\Lambda\bar{n} - n_3^2\bar{e}\Lambda\bar{e} - [\bar{n}\bar{e}]^2[\bar{e}\bar{c}]\Lambda[\bar{e}\bar{c}]}{2n_3^2[\bar{n}\bar{e}]^2} \quad (4)$$

$$a_2 = \frac{(1-2n_3^2)\bar{n}\Lambda\bar{n} + n_3^2\bar{e}\Lambda\bar{e} - [\bar{n}\bar{e}]^2[\bar{e}\bar{c}]\Lambda[\bar{e}\bar{c}]}{2n_3^2[\bar{n}\bar{e}]^2} \quad (5)$$

$$a_3 = \frac{3n_3^2\bar{c}\Lambda\bar{c} - n_3^2\Lambda c + \bar{n}\Lambda\bar{n} - [\bar{e}\bar{c}]\Lambda[\bar{e}\bar{c}]}{2n_3^2} \quad (6)$$

In order to compare the elastic properties of the monoclinic and transversely isotropic symmetry of the muscovite, the value of the differential coefficient of the relative mean- isotropic elastic anisotropy squared transversely (A_t) was used. This coefficient can be derived from the following formula [10, 21]:

$$A_t = \left[\frac{(\Lambda^2)_c - (\Lambda^{(t)})_c}{(\Lambda^2)_c} \right]^{1/2} \cdot 100\% \quad (7)$$

where $(\Lambda^2)_c = \Lambda_{11}^2 + \Lambda_{22}^2 + \Lambda_{33}^2 + 2(\Lambda_{12}^2 + \Lambda_{13}^2 + \Lambda_{23}^2)$;
 $(\Lambda^{(t)})_c = \Lambda_{11}^{(t)2} + \Lambda_{22}^{(t)2} + \Lambda_{33}^{(t)2} + 2(\Lambda_{12}^{(t)2} + \Lambda_{13}^{(t)2} + \Lambda_{23}^{(t)2})$.

To determine the coefficient of the integrated relative mean- isotropic elastic anisotropy squared transversely ($A_i^{(t)}$), averaging has to be made for all the likely directions of the wave normal in Formula (7) (let us denote averaging as the angular brackets $\langle \rangle$):

$$A_i^{(t)} = \left[\frac{\langle \Lambda^2 \rangle_c - \langle \Lambda^{(t)2} \rangle_c}{\langle \Lambda^2 \rangle_c} \right]^{1/2} \cdot 100\% \quad (8)$$

Elastic constants of transversely isotropic medium, which are the most proximate to the elastic properties of the monoclinic muscovite crystal, are derived from the following correlations [10]:

$$\begin{aligned} C_{11}^{(t)} &= g_1 + g_5 \\ C_{33}^{(t)} &= g_1 + 2g_2 + g_3 + g_4 \\ C_{44} &= C_5 + g_1 + g_2 \\ C_{66} &= g_1 + g_5 \\ C_{13} &= g_3 + g_1 + g_2 \end{aligned} \quad (9)$$

where: $g_1 = \frac{1}{112}(213d_1 - 90d_2 - 28d_3 + 77d_4 - 84d_5)$

$g_2 = \frac{1}{112}(-225d_1 + 114d_2 + 28d_3 - 105d_4 + 84d_5)$

$$g_3 = \frac{1}{4}(-5d_1 + 2d_2 + d_3 - 2d_4 + 2d_5)$$

$$g_4 = \frac{1}{16}(55d_1 - 30d_2 - 8d_3 + 35d_4 - 20d_5)$$

$$g_5 = \frac{1}{8}(-15d_1 + 6d_2 + 2d_3 - 5d_4 + 8d_5)$$

$$d_1 = \frac{1}{3}\langle \Lambda \rangle_c$$

$$d_2 = \frac{15}{2}\langle n_3^2 \Lambda \rangle_c + \langle \bar{e}\Lambda\bar{e} \rangle$$

$$d_3 = 15\langle \bar{n}\Lambda\bar{n} \rangle$$

$$d_4 = 15\langle n_3^2\bar{e}\Lambda\bar{e} \rangle$$

$$d_5 = \frac{15}{2}\langle [\bar{n}\bar{e}]^2\bar{c}\Lambda\bar{c} \rangle$$

It has to be noted that in reference [10] one formula is inaccurate. The formula should be written as follows:

$$\langle [\bar{n}\bar{e}]^2\bar{c}\Lambda\bar{c} \rangle = \frac{2}{15}(5g_1 + g_2 + 4g_5)$$

In a similar way, the integral (A_i) and differential (A_d) coefficients of elastic anisotropy can be calculated, which indicate how the specified anisotropic medium with an arbitrary symmetry differs, in an average or certain direction, from the most similar to it isotropic medium. To do this the isotropic medium tensor should be entered into the formula (1):

$$A_i = \left[\frac{\langle \Lambda^2 \rangle_c - \langle \Lambda_m^2 \rangle_c}{\langle \Lambda^2 \rangle_c} \right]^{1/2} \cdot 100\%$$

$$\Lambda_m = a + b\bar{n} \cdot \bar{n} \quad (10)$$

where: $a = \frac{1}{2}(\langle \Lambda \rangle_c - \langle \bar{n}\Lambda\bar{n} \rangle)$; $b = \frac{1}{2}(3\langle \bar{n}\Lambda\bar{n} \rangle - \langle \Lambda \rangle_c)$.

Finally, the following correlation for the integral coefficient of elastic anisotropy was obtained:

$$A_i = \left[\frac{\langle \Lambda^2 \rangle_c - \langle \Lambda_m^2 \rangle_c}{\langle \Lambda^2 \rangle_c} \right]^{1/2} \cdot 100\%$$

Table 1 represents the results of numerical calculations of the muscovite and kaolinite elastic constants in a transversely isotropic approximation.

Numerical calculations of the integral coefficient of transversely isotropic elastic anisotropy showed that its value for a muscovite crystal with a monoclinic symmetry is only 2.4%. This suggests that, in general, transversely isotropic approximation of the elastic properties matrix

of muscovite with a monoclinic symmetry is close to the experimental results. Anisotropy of muscovite in both cases is essential and exceeds 40%, but there is only a minor difference between the integral coefficients of the elastic anisotropy of monoclinic and transversely isotropic ap-

proximation (Table 1). The elastic anisotropy of kaolinite is 55.8% and the difference in anisotropy coefficients between triclinic and isotropic-lateral approximation is much larger, which can be explained by the presence of negative values of C_{23} .

Table 1. Elastic constants (C_{mn} , GPa) and pliabilities (S_{mn} , TPa⁻¹) and integral coefficients of the muscovite and kaolinite elastic anisotropy, according to experimental studies and their transversely isotropic approximation

Type of mineral	Muscovite	Muscovite	Muscovite	Kaolinite	Muscovite (mechanical layered model)	
Elastic constants (in the numerator) and elastic pliabilities (in the denominator)	$\frac{C_{11}}{S_{11}}$	$\frac{181.0}{6.9}$	$\frac{185.0}{5.98}$	$\frac{178.0}{6.04}$	$\frac{191.6}{6.17}$	$\frac{179.2}{6.3}$
	$\frac{C_{22}}{S_{22}}$	$\frac{178.4}{6.6}$	$\frac{185.0}{5.98}$	$\frac{178.0}{6.04}$	$\frac{192.6}{6.17}$	$\frac{179.2}{6.3}$
	$\frac{C_{33}}{S_{33}}$	$\frac{58.6}{18.7}$	$\frac{58.6}{8.0}$	$\frac{54.9}{18.9}$	$\frac{32.1}{31.2}$	$\frac{58.5}{18.3}$
	$\frac{C_{44}}{S_{44}}$	$\frac{16.5}{62.0}$	$\frac{18.0}{55.6}$	$\frac{12.2}{81.9}$	$\frac{16.7}{59.9}$	$\frac{6.4}{156.2}$
	$\frac{C_{55}}{S_{55}}$	$\frac{19.5}{58.8}$	$\frac{18.0}{5.6}$	$\frac{12.2}{81.9}$	$\frac{16.7}{59.9}$	$\frac{6.4}{156.2}$
	$\frac{C_{66}}{S_{66}}$	$\frac{72.0}{14.2}$	$\frac{66.7}{14.0}$	$\frac{67.2}{14.8}$	$\frac{58.2}{17.2}$	$\frac{63.5}{15.7}$
	$\frac{C_{12}}{S_{12}}$	$\frac{48.8}{-2.0}$	$\frac{51.7}{-1.5}$	$\frac{42.4}{-1.34}$	$\frac{75.2}{-2.4}$	$\frac{52.2}{-1.6}$
	$\frac{C_{13}}{S_{13}}$	$\frac{25.6}{-2.4}$	$\frac{19.3}{-1.47}$	$\frac{14.5}{-1.24}$	$\frac{-3.3}{0.4}$	$\frac{21.9}{-1.7}$
	$\frac{C_{23}}{S_{23}}$	$\frac{21.2}{6.6}$	$\frac{19.3}{-1.47}$	$\frac{14.5}{-1.24}$	$\frac{-3.3}{0.4}$	$\frac{21.9}{-1.7}$
	$\frac{C_{15}}{S_{15}}$	$\frac{14.2}{6.3}$	-	-	-	-
	$\frac{C_{25}}{S_{25}}$	$\frac{11}{5.1}$	-	-	-	-
	$\frac{C_{35}}{S_{35}}$	$\frac{1}{-1.9}$	-	-	-	-
	$\frac{C_{46}}{S_{46}}$	$\frac{-3.2}{4.5}$	-	-	-	-
	A [%]	41.9	41.8	45.3	$\frac{51.4}{55.8}$	46.1
$A_j^{(j)}$ [%]	2.4	0	0	25.3	0	
Note	Experimental data [24]	Transversely isotropic approximation	Experimental data [2, 3]	Transversely isotropic approximation	Transversely isotropic approximation	

Note: There is an integral value of the elastic anisotropy coefficient of kaolinite with a triclinic symmetry in the denominator.

Quantitative estimation of clay minerals elastic constants

When developing the method of quantitative estimation of clay minerals elastic constants, first of all, the crystal-chemical characteristics of their structure [2, 7, 18, 19, 23] should be taken into account. A clay mineral foliated structure is based on aluminum-silica-oxygen matrixes with a hexagonal pattern

of circles (Fig. 3) referred to as mica matrixes. Therefore, such minerals as kaolinite, montmorillonite and chlorites are slightly different in the structure of layers (but not the matrixes) and in interlayer space filling.

Two types of structural elements (Fig. 4) can be distinguished

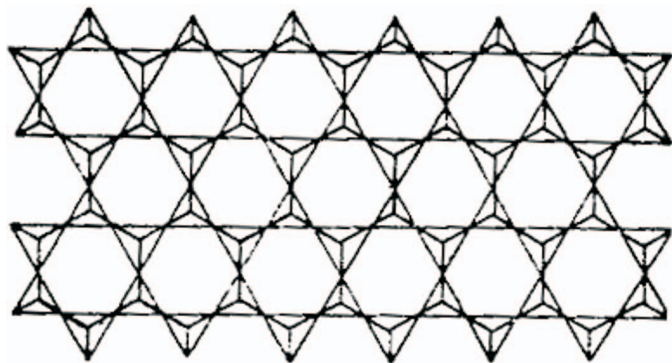


Fig. 3. Tetrahedral mica matrix in the clay mineral foliated structure

in clay mineral crystal structure [19]. The first element is a silica-oxygen tetrahedron (one silica atom and four equidistant atoms of oxygen). Single tetrahedrons are connected with each other to form a continuous two-dimensional tetrahedral matrix. The second structural element is an octahedron that consists of six oxygen atoms or hydroxyl groups. Atoms of

aluminum, magnesium or iron are in the center of the octahedron. When combining, octahedrons make up an octahedral matrix, where the adjacent octahedrons have a common edge, their oxygen atoms (hydroxyl groups) forming two parallel planes.

The structural layer of clay minerals may consist of two (one tetrahedral and one octahedral), three (two tetrahedral and one octahedral) or four matrixes (two tetrahedral and two octahedral). Therefore, ratios 1:1, 2:1 and 2:1:1 for the structural layers of clay minerals were accepted as showing correlations between the number of octahedral and tetrahedral matrixes in one layer (Fig. 5).

Foliated structure of clay minerals reflects the two-dimensional distribution of the strongest bonds in the structure [19]. There are weaker molecular or ionic links between layers. As links in layer planes are much stronger, their deformations are smaller. Elastic constants anisotropy tends to show the following correlations between elastic constants: $C_{11} \approx C_{22} > C_{33}$ [18].

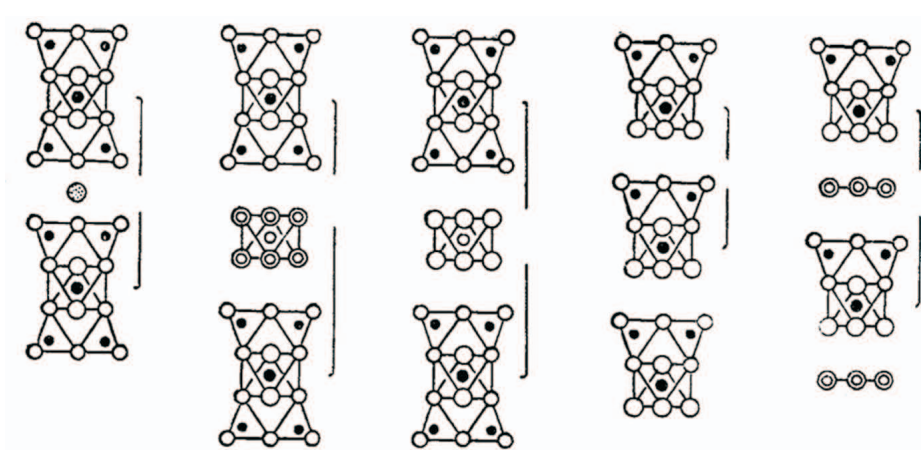


Fig. 4. Schematic image of the clay mineral structure
a – mica, b – montmorillonite, c – chlorite, d – kaolinite, e – halloysite

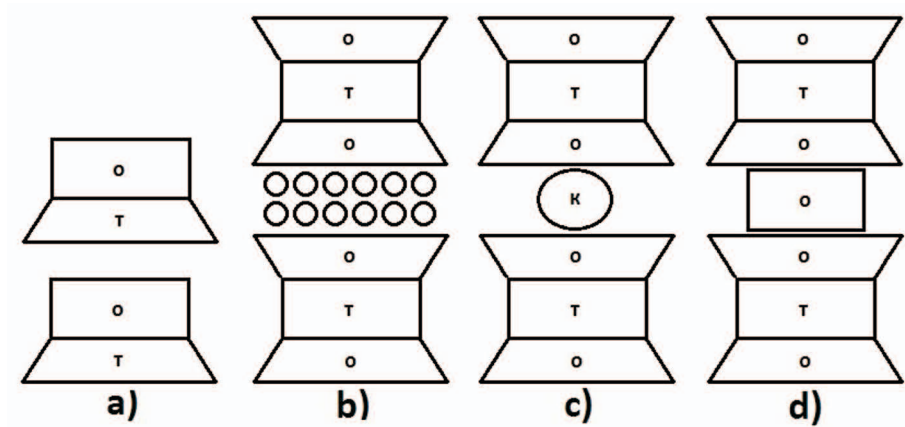


Fig. 5. Diagrammatic representation of the clay mineral structural types

a – structural type 1:1 (kaolinite); b – structural type 2:1 (montmorillonite); c – micaceous structural type (illite); d – structural type 2:1:1 (chlorite); O – octahedral matrix; T – tetrahedral matrix; small circles – water molecules in the interlayer space of minerals structure; oval circles – cation-compensators in the interlayer space of a mineral structure

Foliated structure of clay minerals is intensified by a perfect cleavage lengthwise (100). The cleavage was considered as a mechanical defect which has a significant influence on the clay minerals elastic constants anisotropy. A macroscopic mechanical model of clay minerals foliated structure was suggested as an aggregate of disoriented plates with "hard" contact weakened by the cleavage lengthwise (100). The mechanical defect caused by the cleavage was simulated as an almost zero concentration and fissure opening.

The effective elastic constants of such a foliated model can be calculated by the method developed in our paper [21]:

$$C_{ijkl} = \langle C_{ijkl} \rangle - \langle C_{ijm3} T_{m3n3}^{-1} C_{n3kl} \rangle + \langle C_{ijm3} T_{m3n3}^{-1} \langle E_{n3m3}^{-1} \rangle^{-1} \langle T_{m3p3}^{-1} C_{p3kl} \rangle \rangle \quad (11)$$

where: T_{m3n3}^{-1} – matrix inverted from the following matrix

$$T_{m3n3} = \begin{bmatrix} C_{1313}^{(q)} & C_{1323}^{(q)} & C_{1333}^{(q)} \\ C_{2313}^{(q)} & C_{2323}^{(q)} & C_{2333}^{(q)} \\ C_{3313}^{(q)} & C_{3323}^{(q)} & C_{3333}^{(q)} \end{bmatrix} \quad (12)$$

made up of the components of the elastic constants tensor $C_{ijkl}^{(q)}$ for q layer

$\langle E_{n3m3} \rangle^{-1}$ – matrix inverted from the following matrix

$$\langle E_{n3m3} \rangle = \begin{bmatrix} \langle C_{1313} \rangle & \langle C_{1323} \rangle & \langle C_{1333} \rangle \\ \langle C_{2313} \rangle & \langle C_{2323} \rangle & \langle C_{2333} \rangle \\ \langle C_{3313} \rangle & \langle C_{3323} \rangle & \langle C_{3333} \rangle \end{bmatrix} \quad (13)$$

made up of statistically averaged components of elastic constant layers matrix; $\langle \rangle$ – angular brackets stand for statistical averaging [20].

Numerical calculations of the effective elastic constants were made for the mechanical foliated model of muscovite structure that consists of 21 alternating layers with a monoclinic elastic symmetry and planar fissures. The elastic constants of muscovite in monoclinic approximation were borrowed from paper [24] (Table 1). The concentration and the elastic constants of the planar fissure were considered as infinitesimal when modeling the effective elastic constants. The plates of muscovite are disoriented at the angle of 30 degrees in our model.

The following effective elastic constants were devised from calculations (C_{mn}^* , GPa):

177.0	51.1	23.7	-0.1	-0.2	1.1
	177.2	23.2	0.3	0.1	-1.1
		58.5	0.0	0.0	0.0
			6.4	0.0	0.2
					65.6

The following correlations between the elastic constants in the model proved to be correct: $C_{11}^* \approx C_{22}^* > C_{33}^*$; $C_{44} = C_{55} < C_{66}$; $C_{13} \approx C_{23} < C_{12}$; $C_{12} > 0$, $C_{13} > 0$ and $C_{23} > 0$.

An integral coefficient value of elastic anisotropy is 46% (Table 2), so this value is typical of foliated minerals (mica).

Thus, the structure, plate disorientation and oriented mechanical defects caused by cleavage – all of these cause a nearly planar transversely isotropic symmetry of the elastic constants of minerals with foliated structure.

Crystal-chemical systematization of the minerals elastic constants is based on the isostructural principle [2, 6, 7, 8, 16, 17]. A review of elastic constants systematics [6, 11, 12, 14–19, 28] revealed a direct correlation between the elastic constants and interatomic bond parameters in isostructural minerals. The elastic constants anisotropy depends on the orientation of major structural elements of the mineral and the strength of bonds between them. The methods of quantitative estimation of clay minerals elastic constants must be based on the correlation between the elastic constants and the values of interatomic bonds in respective directions of the mineral structure.

Berch F. [8, 12] pioneered developing the system of correlation between the elastic waves velocity in minerals and rocks and their density. Berch made an empirical generalization based on the dependence of longitudinal and transverse wave velocities on density under hydrostatic compression. Today this generalization is known as Berch's formula [8, 28]: if the value of an average atomic mass \bar{M} is constant, the longitudinal wave velocity V_p is a linear function of density.

$$V_p = a(\bar{M}) + b(\rho) \quad (14)$$

where: $a(\bar{M})$, b – empirical coefficients, ρ – density. F. Berch [8] also pointed out that for isostructural minerals data on velocities with different \bar{M} are located on straight lines, intersecting the directions of \bar{M} constants on the density-velocity diagram.

Andersen & Nafe [5, 6] established a logarithmic correlation between elastic modules and average atomic volume and discovered corresponding regressive correlations for oxides and silicates. K. Katahara [13] used the correlation between $V_{ij} = \sqrt{C_{ij}/\rho}$ and density to predict the elastic constants of clay minerals such as kaolinite, illite and chlorite.

A multiple regression correlation was found between the complex parameter $\sqrt{\frac{C_{mn}}{\rho}}$ and density ρ and average atomic mass \bar{M} to predict clay minerals elastic constants:

$$\lg \left(\sqrt{\frac{C_{mn}}{\rho}} \right) = A \ln \rho + B \ln \bar{M} + C \quad (15)$$

Table 2. Estimation of elastic constants (C_{mn}) and elastic pliabilities (S_{mn}) of foliated minerals in transversely isotropic approximation, according to the multiple regression formula of correlation between the complex parameter $\sqrt{C_{mn}/\rho}$, density and the average atomic mass of minerals

The name of the mineral and its crystal-chemical formula	Density of the mineral 10^3 [kg/m ³]	Average atomic mass	The elastic constants C_{mn} , GPa (in the numerator) and elastic pliabilities S_{mn} , TPa ⁻¹ (in the denominator)						The integral coefficient of elastic anisotropy, A_i [%]	Remark
			$\frac{C_{11}}{S_{11}}$	$\frac{C_{33}}{S_{33}}$	$\frac{C_{44}}{S_{44}}$	$\frac{C_{66}}{S_{66}}$	$\frac{C_{12}}{S_{12}}$	$\frac{C_{13}}{S_{13}}$		
The structural type 1:1										
Kaolinite $Al_4[Si_4O_{10}](OH)_8$	2.52	15.19	$\frac{175.5}{6.6}$	$\frac{49.4}{21.6}$	$\frac{12.1}{82.6}$	$\frac{58.4}{17.1}$	$\frac{58.7}{-2.0}$	$\frac{18.9}{-1.7}$	45.3	1
			$\frac{191.6}{6.2}$	$\frac{32.1}{31.2}$	$\frac{16.7}{59.9}$	$\frac{58.2}{17.2}$	$\frac{75.2}{-2.4}$	$\frac{-3.3}{0.4}$	51.4	2 from [21]
			$\frac{171.5}{6.5}$	$\frac{52.6}{21.9}$	$\frac{14.8}{67.6}$	66.3	$\frac{38.9}{-1.0}$	$\frac{27.1}{-2.8}$	42.0	2 from [21]
Halloysite $Al_4(H_2O)_4[Si_4O_{10}](OH)_8$	2.10	12.49	$\frac{149.5}{11.8}$	$\frac{23.0}{114.0}$	$\frac{10.9}{91.7}$	$\frac{45.8}{21.8}$	$\frac{57.8}{0.9}$	$\frac{38.4}{-21}$	46.2	1
Antigorite $Mg_6[Si_4O_{10}](OH)_8$	2.6	15.40	$\frac{182.7}{6.2}$	$\frac{63.5}{16.2}$	$\frac{13.3}{75.2}$	$\frac{60.4}{16.6}$	$\frac{61.9}{-2.0}$	$\frac{14.7}{-1.0}$	43.4	1
Lizardite $Mg_6[Si_4O_{10}](OH)_8$	2.55	15.40	$\frac{177.2}{6.5}$	$\frac{51.7}{20.5}$	$\frac{12.2}{82.0}$	$\frac{59.3}{16.9}$	$\frac{58.6}{-2.0}$	$\frac{18.2}{-1.6}$	45.1	1
Dykyte $Al_4[Si_4O_{10}](OH)_8$	2.62	15.19	$\frac{186.6}{6.1}$	$\frac{74.9}{13.6}$	$\frac{14.5}{70.0}$	$\frac{60.6}{16.5}$	$\frac{65.5}{-2.1}$	$\frac{12.4}{-0.7}$	41.6	1
Chrysotile $Mg_6[Si_4O_{10}](OH)_8$	2.55	15.40	$\frac{177.2}{6.5}$	$\frac{51.7}{20.5}$	$\frac{12.2}{82.0}$	$\frac{59.3}{16.9}$	$\frac{58.6}{-2.0}$	$\frac{18.2}{-1.6}$	45.1	1
The structural type 2:1										
Montmorillonite $Na_x(H_2O)_4\{Al_2[Al_xSi_{4-x}O_{10}](OH)_2\}$	2.55	13.27	$\frac{195.3}{6.2}$	$\frac{126.7}{7.9}$	$\frac{21.3}{46.9}$	$\frac{56.3}{17.8}$	$\frac{82.6}{-2.6}$	$\frac{7.0}{-0.2}$	27.2	1
Illite $K_x(H_2O)_x\{Al_2[Al_xSi_3O_{10}](OH)_{2-x}(H_2O)_3\}$	2.79	16.55	$\frac{194.9}{5.8}$	$\frac{87.4}{11.6}$	$\frac{14.1}{70.9}$	$\frac{66.3}{15.1}$	$\frac{62.4}{-1.8}$	$\frac{10.9}{-0.5}$	41.0	1
			$\frac{179.9}{5.9}$	$\frac{55.0}{18.8}$	$\frac{11.7}{85.5}$	$\frac{70.0}{14.3}$	$\frac{39.9}{-1.2}$	14.5	45.8	2 from [21]
The structural type 2:1:1										
Chlorite $Mg_4(MgAl)_2[(SiAl)_2Si_2O_{10}](OH)_8$	2.69	15.44	$\frac{192.5}{5.9}$	$\frac{90.0}{11.2}$	$\frac{15.4}{64.9}$	$\frac{62.5}{16.0}$	$\frac{67.5}{-2.1}$	$\frac{10.4}{-0.4}$	39.8	1
			$\frac{181.8}{6.2}$	$\frac{106.8}{9.7}$	$\frac{11.4}{87.7}$	$\frac{62.5}{16.0}$	$\frac{56.8}{-1.8}$	$\frac{20.3}{-0.8}$	36.3	2 from [21]
Klinchlore	2.711	15.44	177.4	93.7	11.2	55.4	66.2*	–	–	Incomplete data of [3]
Leikhtenbergite	2.65	15.44	184.3	119.67	11.8	69.2	45.9*	–	–	Incomplete data of [3]
The structural type: micas										
Muscovite $K\{Al_2[AlSi_3O_{10}](OH)_2\}$	2.893	18.97	$\frac{188.8}{5.7}$	$\frac{56.6}{18.6}$	$\frac{9.9}{101.0}$	$\frac{71.9}{13.9}$	$\frac{44.9}{-1.2}$	$\frac{17.9}{-1.4}$	46.7	1
			$\frac{185.3}{5.9}$	$\frac{56.8}{18.3}$	$\frac{12.7}{78.7}$	$\frac{68.7}{14.6}$	$\frac{47.9}{-1.4}$	$\frac{15.3}{-1.2}$	45.3	2
Biotite $K\{(Mg_1Fe)_3[AlSi_3O_{10}](OH)_2\}$	3.097	21.11	$\frac{196.1}{5.4}$	$\frac{61.5}{16.9}$	$\frac{9.1}{109.9}$	$\frac{79.7}{12.5}$	$\frac{36.8}{-0.9}$	$\frac{17.0}{-1.2}$	47.1	1
			$\frac{189.4}{5.6}$	$\frac{54.8}{19.1}$	$\frac{6.0}{166.7}$	$\frac{77.7}{12.9}$	33.9	$\frac{16.9}{-1.4}$	49.3	2

ect. Table 2

The name of the mineral and its crystal-chemical formula	Density of the mineral 10^3 [kg/m ³]	Average atomic mass	The elastic constants C_{mn} , GPa (in the numerator) and elastic pliabilities S_{mn} , TPa ⁻¹ (in the denominator)						The integral coefficient of elastic anisotropy, A_i [%]	Remark
			$\frac{C_{11}}{S_{11}}$	$\frac{C_{33}}{S_{33}}$	$\frac{C_{44}}{S_{44}}$	$\frac{C_{66}}{S_{66}}$	$\frac{C_{12}}{S_{12}}$	$\frac{C_{13}}{S_{13}}$		
Phlogopite K{Mg ₃ [AlSi ₃ O ₁₀](OH) ₂ }	2.868	18.97	$\frac{186.3}{5.8}$	$\frac{51.6}{20.7}$	$\frac{9.5}{105.3}$	$\frac{71.4}{14.0}$	$\frac{43.6}{-1.2}$	$\frac{19.6}{-1.8}$	47.4	1
			$\frac{183.8}{6.0}$	$\frac{53.0}{21.5}$	$\frac{5.9}{169.5}$	$\frac{74.6}{13.4}$	$\frac{34.6}{-0.7}$	$\frac{26.5}{-2.6}$	47.8	2
Xanthophyllite Ca{Mg ₂ Al[Al ₃ SiO ₁₀](OH) ₂ }	3.100	19.03	$\frac{210.2}{5.1}$	$\frac{116.1}{8.6}$	$\frac{13.6}{73.5}$	$\frac{76.9}{13.5}$	$\frac{56.3}{-1.4}$	$\frac{8.6}{-0.3}$	39.4	1
			231.4	99.7	23.1	80.6	–	–	–	Incomplete data [3]

Note: * C_{12} – elastic constants calculated in transversely isotropic approximation; 1 – multiple regression data; 2 – transversely isotropic approximation from inversion data.

The value of multiple regression coefficients for different elastic constants C_{mn} and their trusted limits are presented in Table 3.

A physical principle underlies the value of the complex parameter $\sqrt{C_{mn}/\rho}$ for C_{11} , C_{33} , C_{44} , C_{66} diagonal components of the elastic constants matrix in transversely isotropic approximation: these are the values of phase velocities of the waves with longitudinal or transverse polarization along respective crystallographic directions of clay minerals, correlating with strength of bonds between atoms in these directions [2, 3, 7, 16, 17]. Quantitative estimation (Formula 15) of the elastic constants of rock-forming clay minerals are shown in Table 2. The obtained results agree with experimental data represented in Table 3.

The following correlations between the elastic constants of all clay minerals are true: $C_{11} = C_{22} > C_{33}$, $C_{66} > C_{44} = C_{55}$, $C_{12} > C_{13} = C_{23}$. For the first time the constants were devised in transversely isotropic approximation for such important minerals as halloysite, dykrite, antigorite, lizardite, chrysotile, montmorillonite, illite, chlorite and xanthophyllite. Reliable data have been obtained on transversely isotropic approximation of the kaolinite elastic constants. These data were found to agree with estimation data suggested by other researchers [11, 26].

The elastic pliabilities S_{mn} , GPa⁻¹ have been calculated for all rock-forming minerals in transversely isotropic approximation, which allows to estimate linear and volumetric compressibility of minerals (Table 2). The value of integral coefficient of clay minerals elastic anisotropy ranges from 27.2% to 50.4%. Montmorillonite has the lowest value of anisotropy coefficient (27.2%) resulting from water in the interlayer space weakening molecular bonds. With water saturation increasing, the elastic constants tend to increase and the value of anisotropy coefficient tends to decrease [2].

Table 3. Coefficients of linear regression correlation between the complex parameter $\ln(\sqrt{C_{mn}/\rho})$ logarithm and logarithms of density ($\ln \rho$) and average atomic mass \bar{M} of foliated minerals

Parameter	Regression equation coefficients			The trusted limits of predicted parameter estimation
	$\lg(\sqrt{C_{mn}/\rho}) = A \ln \rho + B \ln \bar{M} + C$			
	<i>A</i>	<i>B</i>	<i>C</i>	
$\ln(\sqrt{C_{11}/\rho})$	0.391	-0.326	2.741	±0.0129
$\ln(\sqrt{C_{33}/\rho})$	4.838	-3.013	5.215	±0.0340
$\ln(\sqrt{C_{44}/\rho})$	1,829	-1.892	4.241	0.0610
$\ln(\sqrt{C_{66}/\rho})$	1.120	-0.021	0.173	±0.0020
$\ln(\sqrt{C_{13}/\rho})$	-5.902	3.223	-2.305	±0.0180

Anisotropy parameters of rock-forming clay minerals elastic waves

Elastic mineral symmetry determines spatial changes of all the anisotropy parameters of elastic waves [2]: indicatrices of isoline stereo-projections of quasi-longitudinal, “quick”

and “slow” quasi-transverse waves, and elastic anisotropy differential coefficient.

The stereo-projection of the triclinic kaolinite quasi-

longitudinal wave isolines (E , GPa) (Fig. 6) shows a local minimum along the crystallographic axis (001). The stereo-projection and X_1X_3 coordinate plane are almost symmetrical. The maximal values of the quasi-longitudinal wave phase are in X_1X_2 plane.

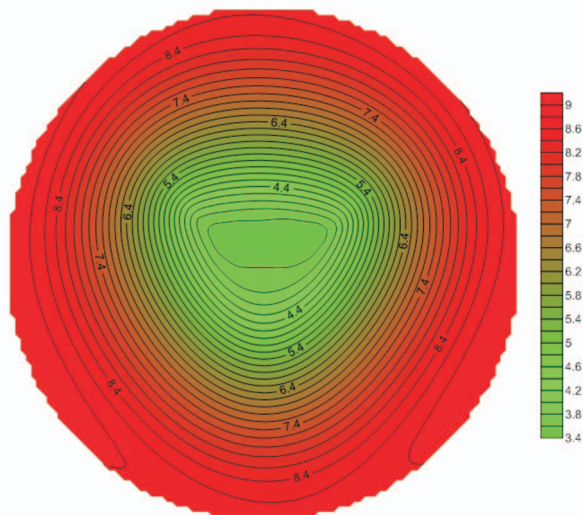


Fig. 6. Stereo-projection of the triclinic kaolinite quasi-longitudinal wave isolines (in km/s)

The isoline stereo-projection of difference between “quick” and “slow” phase velocities of quasi-longitudinal waves (in km/s) of the triclinic kaolinite (Fig. 7) shows about seven local extremums around (001) direction, there being about 7 acoustic axes.

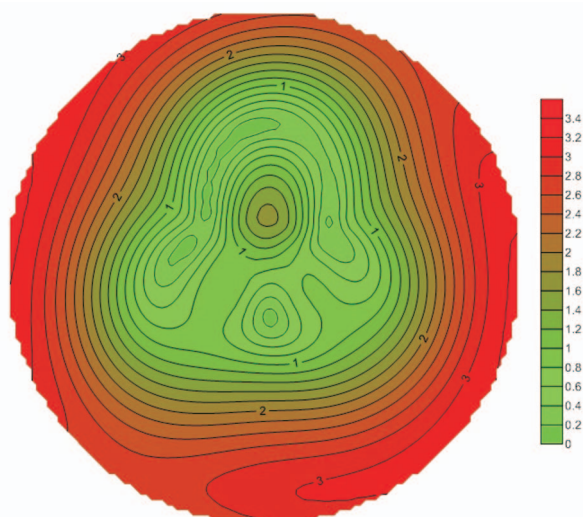


Fig. 7. Stereo-projection of difference between the triclinic kaolinite “quick” and “slow” quasi-transverse wave isolines (in km/s)

The acoustic axis is neither parallel to C axis of kaolinite. The greatest difference between “quick” and “slow” quasi-transverse waves is to be found in X_1X_2 plane (over 2.8 km/s),

which results from the intensive splitting of transverse waves in the kaolinite crystal.

There are three directions with maximal values (over 55% (Fig. 8)) of elastic anisotropy differential coefficients. Yet, around (001) direction, there are two directions with minimal values (less than 20%) of the elastic anisotropy coefficient.

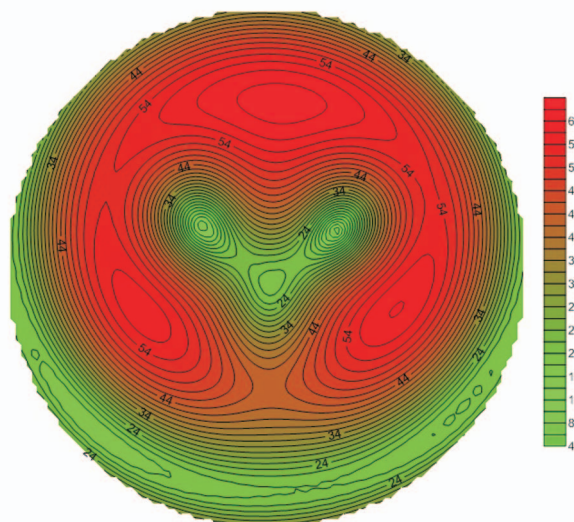


Fig. 8. Isoline stereo-projection of the triclinic kaolinite elastic anisotropy differential coefficient (A_a , %)

The transversely isotropic approximation of kaolinite elastic symmetry accounts for all the parameters of anisotropy showing cylindrical symmetry (Fig. 9). Extremal directions of anisotropy parameters, which are typical of triclinic approximation, get leveled. Instead of three extremal directions of elastic anisotropy differential coefficients, there appears a whole zone of its extremal values (over 55%).

A schistic structural motive also appears on the stereo-projections of anisotropy parameters of the monoclinic muscovite (Fig. 10). Around muscovite crystallographic C axis there are about six acoustic axes, with the value of difference between “quick” and “slow” quasi-transverse phase waves exceeding 2.4 km/s in X_1X_2 plane. However, this value is lower than that of the kaolinite (Fig. 6 and 10).

Transversely isotropic approximation of clay minerals elastic constants requires certain conditions for there being special directions deviating from the symmetry elements. These directions involve a number of correlations between elastic constants [2, 10]. For the longitudinal normal (the direction of a transverse wave propagation, which is non-parallel to the elements of the mineral symmetry) – the following condition is necessary and sufficient [10]:

$$-1 < \frac{g_2}{g_1} < 0 \tag{16}$$

So apart from (001) direction and X_1X_2 plane, longitudinal

waves proper can propagate in directions with θ_p angle for the clay crystals with transversely isotropic approximation. θ_p angle is formed by the longitudinal normal and the main axis of transversely isotropic symmetry {axis ∞ parallel to (001)}. The cosine of θ_p is derived from the formula:

$$\cos \theta_p = \sqrt{-\frac{g_2}{g_1}}$$

For acoustic axes along which the velocities of quasi-transverse waves are identical, the following condition is necessary:

$$0 < \frac{(g_5 - g_2)(g_5 - g_3)}{g_5^2 + g_4(g_5 - g_2)} < 1 \quad (17)$$

Table 4 shows values of acoustic axis angle θ_a . This angle is formed by the acoustic axis and the main axis of the mineral symmetry [10]:

$$\cos \theta_a = \sqrt{\frac{(g_5 - g_2)(g_5 - g_3)}{g_4(g_5 - g_3) + g_5^2}} \quad (18)$$

As is evident from the data in Table 4, the values of longitudinal normal angles vary from 6.3° to 36°. Values of angles forming acoustic axes are much higher: θ_a varies from 30.2° to 59.3°.

Moreover, montmorillonite has the highest values of angles θ_p and θ_a .

A comparison of the predicted and experimental data on phase velocities indicatrices (Fig. 11, 12) in the transversely isotropic approximation shows that, according to experimental data, negative values C_{13} and C_{23} account for abnormally extreme values on the indicatrices. Predicted estimations give an opportunity to correct the inaccurate estimates of C_{13} and C_{23} given in reference [22].

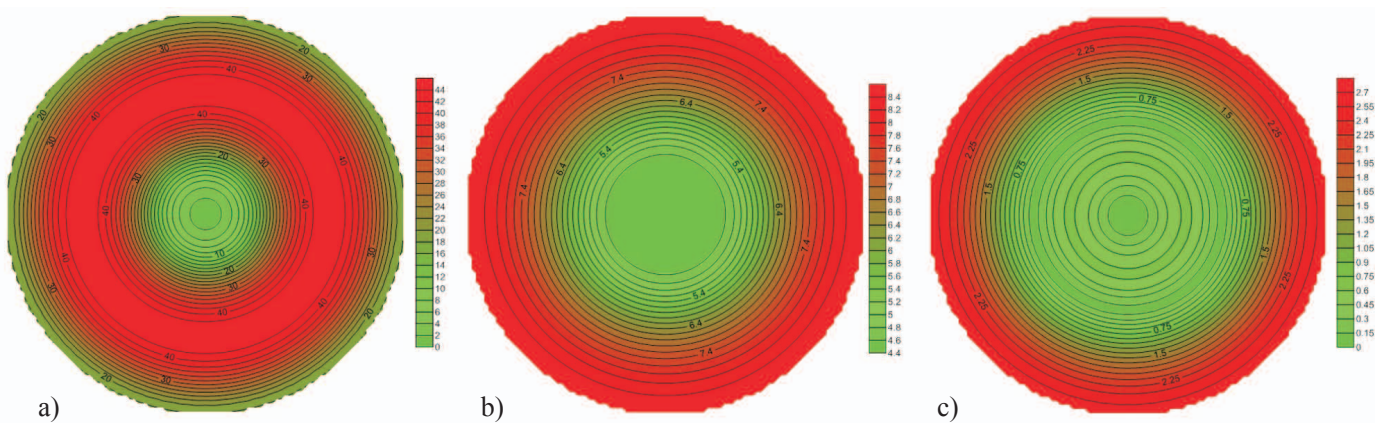


Fig. 9. Isoline stereo-projection of the triclinic kaolinite anisotropy parameters in transversely isotropic approximation
a – elastic anisotropy coefficient (A_w , %); b – quasi-longitudinal wave (in km/s); c – difference between “quick” and “slow” quasi-transverse waves (in km/s)

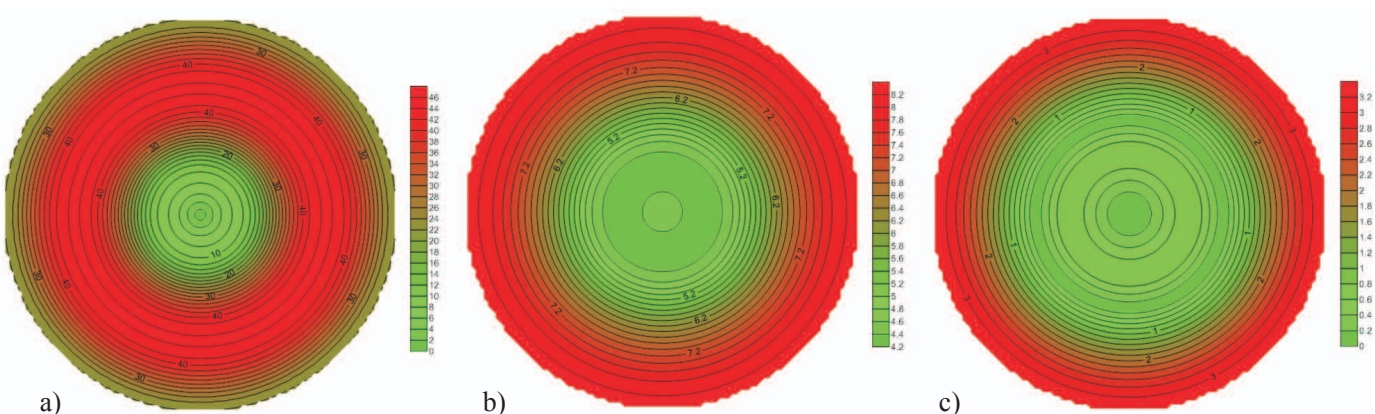


Fig. 10. Stereo-projection of the monoclinic muscovite anisotropy parameters isolines
a – elastic anisotropy coefficient (A_w , %); b – quasi-longitudinal wave (in km/s); c – difference between “quick” and “slow” quasi-transverse waves (in km/s)

Table 4. Parameters of acoustic anisotropy of clay mineral elastic constants (transversely-isotropic approximation) – the angles of longitudinal normal (θ_p) and acoustic axes (θ_a) deviating from the directions of mineral symmetry elements

Type of mineral	Angle θ_p , degrees	Angle θ_a , degrees	Note
Structural Type 1:1			
Kaolinite	12.3	38.4	multiple regression data
	6.3	30.2	transversely isotropic approximation [11, 28]
	6.3	32.6	transversely isotropic approximation [2]
Halloysite	6.3	32.6	multiple regression data
Antigorite	21.6	43.9	multiple regression data
Lizardite	14.6	39.3	multiple regression data
Dykite	25.6	47.4	multiple regression data
Chrysotile	14.6	39.3	multiple regression data
Structural Type 2:1			
Montmorillonite	36.0	59.3	multiple regression data
Illite	29.1	48.9	multiple regression data
	19.1	38.6	transversely isotropic approximation [11, 26]
Structural Type 2:1:1			
Chlorite	29.6	50.5	multiple regression data
	34.1	52.5	transversely isotropic approximation [11, 26]
Structural Type: Micas			
Muscovite	19.5	38.8	multiple regression data
	18.4	39.2	transversely isotropic approximation from inversion data
Biotite	22.0	39.1	multiple regression data
	21.9	38.1	transversely isotropic approximation from inversion data
Phlogopite	16.5	36.8	multiple regression data
	17.6	36.4	transversely isotropic approximation from inversion data
Xanthophyllite	34.2	51.3	multiple regression data

Note: θ_p – angle between the main symmetry axis and longitudinal normal deviating from symmetry elements of the mineral structure.
 θ_a – angle between the main symmetry axis and acoustic axis deviating from the symmetry elements of the mineral structure.

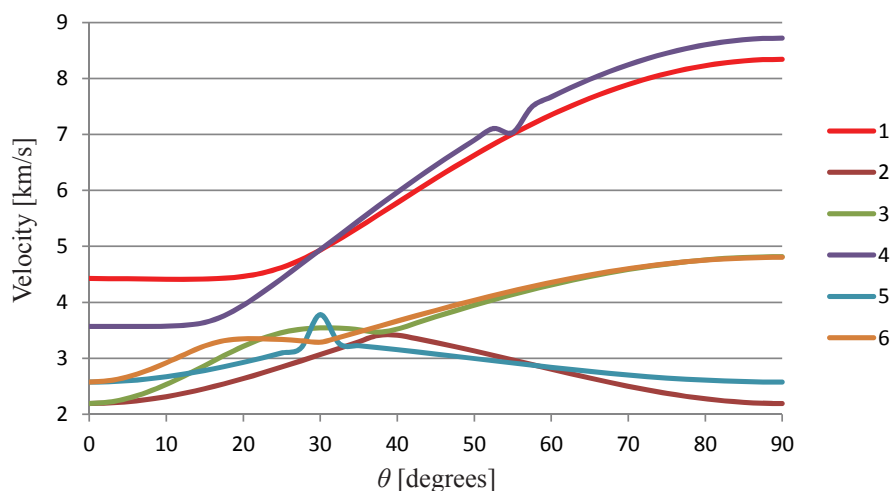


Fig. 11. Kaolinite indicatrices of phase velocities of quasi-longitudinal and quasi-transverse waves in transversely isotropic approximation, according to experimental data and a predicted estimation of regression correlation

1 – quasi-longitudinal wave according to the predicted data; 2 – “slow” quasi-transverse wave according to the predicted data; 3 – “quick” quasi-transverse wave according to the predicted data; 4 – quasi-longitudinal wave according to the experimental data; 5 – “slow” quasi-transverse wave according to the experimental data; 6 – “quick” quasi-transverse wave according to the experimental data.

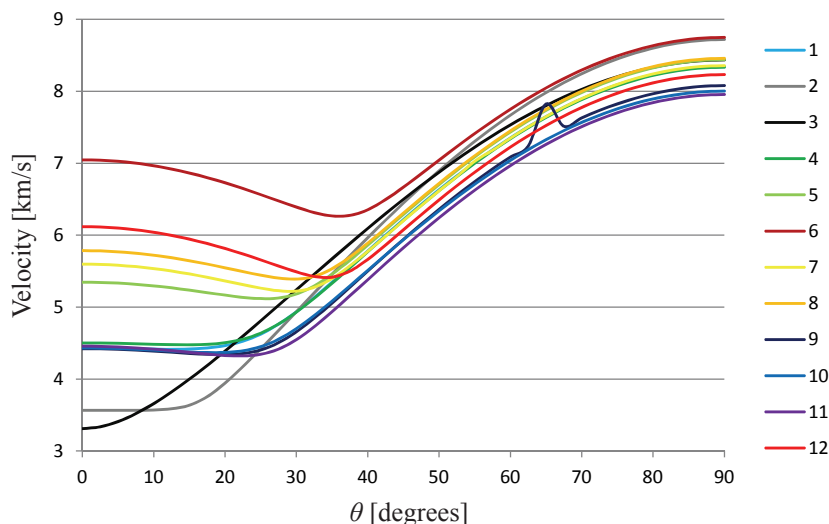


Fig. 12. Indicatrices of phase velocities of the clay minerals quasi-longitudinal waves (transversely isotropic approximation)

1 – kaolinite according to multiple regression data; 2 – kaolinite according to experimental data; 3 – halloysite according to multiple regression data; 4 – lizardite according to multiple regression data; 5 – dykrite according to multiple regression data; 6 – montmorillonite according to multiple regression data; 7 – illite according to multiple regression data; 8 – chlorite according to multiple regression data; 9 – muscovite according to multiple regression data; 10 – muscovite according to experimental inversion data; 11 – biotite according to multiple regression data; 12 – xanthophyllite according to multiple regression data.

Conclusion

A new method based on the principles of crystal-chemical classification of the elastic properties of isostructural minerals [16, 17, 25, 26, 27] proves to be efficient in the quantitative estimation of the clay minerals elastic constants.

A first successful attempt was made at a quantitative assessment of the elastic constants of all clay mineral constituents in transversely isotropic approximation, which fully agrees with experimental data on these minerals.

An analytical method of approximating the elastic constant matrix of arbitrary symmetry minerals was devised via a transversely anisotropic elastic medium. The approximation approach provides for a quantitative assessment of errors.

Extensive research has been carried out into the anisotropy parameters of clay mineral elastic waves and their correlation with the structural features of minerals. According to experimental data on kaolinite [17], the negative component of the elastic constant can be assumed to account for the extremums on the indicatrices of quasi-longitudinal and “slow” quasi-transverse waves. However, these extremums cannot be determined by the crystal-chemical structure of the mineral. To clarify the elastic constants of kaolinite, further experimental research has to be undertaken.

This method can be used for approximating the elastic constants of other rock-forming minerals.

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