Submitted: July 15, 2023 Revised: November 28, 2023 Accepted: May 23, 2024

Influence of adhesive interaction on the sound speed in layered composites

R.-R.V. Razakova ^{1⊠} D. R.A. Turusov ^{1,2} **D**

¹ Moscow State University of Civil Engineering, Moscow, Russia

² N.N. Semenov Federal Research Center for Chemical Physics, Russian Academy of Sciences, Moscow, Russia

 \boxtimes chernova riorita@mail.ru

ABSTRACT

There are the adhesive interaction parameters influence between soft polymer and steel layers to the velocity of the longitudinal sound wave in this paper. Numerical and experimental tests of the sound speed in two composite and one polymer samples are considered. The mixture formula was used to determine the elastic parameters in numerical calculations with contact layer. The model, which included the contact layer, allows calculating the layered composites elastic modulus with certain geometric and mechanical parameters of adhesive interaction. Authors have compared the results of numerical calculations and physical experiment. It was found that the adhesive interaction parameters in the calculation of layered structures is necessary to consider because the adhesive connecting polymer layers with the steel layers have experience comprehensive stretching which affects to the elastic and acoustic parameters of the samples.

KEYWORDS

layered materials • contact layer • Young's modulus • Poisson ratio • adhesion contact • tunable materials material properties • polymer

Citation: Razakova RRV, Turusov RA. Influence of adhesive interaction on the sound speed in layered composites. *Materials Physics and Mechanics*. 2024;52(3): 13–21. http://dx.doi.org/10.18149/MPM.5232024_2

Introduction

One of the most important tasks of the national economy branches is improving structures quality and reliability. This task is especially important in the military, construction, energy and other industries, where structural destruction can lead to human casualties, as well as global financial losses. Therefore, it is necessary to develop methods for analyzing building materials. In aviation and other leading industries in the 1950s, designers began to use multilayer structures, which are rigidly interconnected metal and polymer layers. Since then, the development of materials science has progressed greatly and now scientists and engineers focused their efforts on creating tunable materials that can be layered, cellular ant etc. Most scientific researches in composite materials field are focused on possible technical applications, but at the same time, there are a shortage of scientific researches dedicated to the analysis of the material parameters influence, topologies, deformations and stiffness effects on the elastic wave's propagation in various artificial composite materials. A large number of numerical and physical experiments which significance to science were carried out by Mary C. Boyce. Some of them were presented in $[1,2]$. It was paid attention to the development of tunable materials, analyze the influence of geometry, pattern and topology of structures in

heterogeneous materials on the physical and mechanical characteristics in these scientific publications. Engineering applications require high sound-insulating properties, safe, lightweight, as well as strong and durable from layered materials. The layered composites interlayer strength was studied in $[3,4]$. However, the authors do not describe the influence of the adhesive interaction parameters, which is one of the fundamental values for ensuring the required strength. In $[5]$, the authors presented studies for optimizing laminated materials with a viscoelastic damping layer by the finite element method. It was done by changing the position of the damping layer. The authors did not indicate how adhesion modeling was performed in the ABAQUS system, although this is very important for optimizing the strength of such structural materials, as evidenced by the works of $[6-8]$. In these articles, the authors used the finite element method to model the layered structure. This method has been worked out with a fairly high accuracy, but solution of the layered structure stress-strain state problem leads to singular stresses at the corner points of the gluing surface $[9]$. There are some ways to bypass singularities [\[10,11\],](#page-7-5) but these methods are often not verified by physical experiments. The contact layer method theory avoids such problems as infinite shear stresses that occur at the boundaries between layers and at the corner points of gluing. It has been verified by numerous experiments, which are presented in [\[12\].](#page-8-0)

The contact layer method has been tested in numerical experiments, which aimed at determining the sound speed in layered material in the present investigation. The influence analysis on the sound speed of some physico-mechanical and geometric characteristics of the composite materials components was done with the framework of the theory method possibilities. This work was supposed to compare the results of a physical experiment with the numerical experiments data.

Materials and Methods

In $[13]$, it was proposed a mathematical model (2) for calculating the effective Young's modulus of the layered sample, which include the contact layer and its parameters. We proposed the method for calculating the sound wave speed in a layered composite in this paper, which included the main mechanical parameters and sample cross-sectional dimensions. It is important to note that in the contact layer method, the interaction between the layers of the adhesive and the substrate was carried out with the contact layer (Fig. [1\)](#page-2-1), which is a system of oriented normally to the contact surface short thin elastic rods-bonds. In this layer, there is not any direct contact of the rods with each other and, therefore, there are not any normal stresses σ_x and σ_z . Short rods perceive shear stresses σ_{yx} , σ_{zy} , σ_{xz} and normal stress σ_{y} . The most important physical feature in the layered structure model (steel + polymer) is that more than 95 % of the polymer adhesive layer is fully stretched (or compressed). Therefore, it follows that instead of the usual polymer elastic modulus, there is the modulus of all-round volumetric tension works. The proportion of fully stretched adhesive depends on the concentration zone with of the edge effect: if it is the smaller, the proportion of fully stretched adhesive is greater. The bulk modulus *K* is related to Young's modulus *E* and Poisson's ratio *μ* by the relation: $K=\frac{E}{2G}$. (1)

$$
K = \frac{1}{3(1-2\mu)}
$$

This specialty included in Eq. [\(2\),](#page-2-0) which makes mathematical model more accurate than the mixture formula use. This rule is especially critical for layered structures where the polymer is relatively soft. This theory has been successfully tested in research for calculating the adhesive interaction of materials by Vladimir. I. Andreev, Robert A. Turusov and other [\[14,15\].](#page-8-2)

Fig. 1. Schematic diagram which is showing the model of the contact layer

The equation for calculating the effective Young's modulus $E_{eff.l.b.}$ according to contact layer theory can be represented in the following form [\(2\):](#page-2-0)

$$
E_{eff.l.b.} = \left[\left(\frac{V_0}{E_0} + \frac{V_1}{E_1} \right) - \frac{2 \cdot \left(\frac{\mu_0}{E_0} - \frac{\mu_1}{E_1} \right)^2}{\frac{(1 - \mu_1)}{E_1 \cdot V_1} + \frac{(1 - \mu_0)}{E_0 \cdot V_0}} \cdot \left(1 - \frac{\tanh(\nu)}{\nu} \right) \right]^{-1}, \tag{2}
$$

where E_0 and E_1 are Young's modules of the substrate and adhesive, respectively; μ_0 , μ_1 are Poisson's ratios of the substrate and adhesive, respectively; V_0 , V_1 are the relative volume fractions content of the substrate and adhesive, respectively.

Fig. 2. The investigated composite specimens: 1- epoxy, 2- three-section, 3- layered

The parameter *ν* in Eq[. \(2\)](#page-2-0) characterizes the contact layer and determined as follows: $\nu = \frac{\omega \cdot L}{2}$ $\frac{D \cdot L}{2}$; ω = $\sqrt{G_h \left[\frac{1-\mu_0}{E_0 h_0} \right]}$ $\frac{1-\mu_0}{E_0h_0} + \frac{2(1-\mu_1)}{E_1h_1}$ $\left[\frac{1-\mu_1}{E_1h_1}\right]$; L is the length of the section side; G_h is a stiffness of the contact layer; E_0 , E_1 are Young's modules of the substrate and adhesive; μ_0 , μ_1 are Poisson's ratios of the substrate and adhesive; h_0 , h_1 are thickness of the substrate and adhesive; The experimental study has been done on three rods, which are presented in Fig. [2.](#page-3-0) Initial data in the experimental study: $E_0 = 210,000$ MPa; $\mu_0 = 0.3$; $L = 10$ mm; μ_1 = 0.5; G_h , E_1 are changes during polymer hardening. The geometric dimensions of the rods are shown in the Fig. [2.](#page-3-0) In composite rods, the relative proportions of polymer and steel were 15 and 85 %, respectively.

Layered rod elastic modulus calculating according to Eq. [\(2\)](#page-2-0) and a composite rod according to Reuss model which based on rule of mixtures. This rule has been presented in Eq. [\(3\):](#page-3-0)

$$
E_{mix} = \frac{E_0 \cdot E_1}{V_0 \cdot E_1 + V_1 \cdot E_0}.\tag{3}
$$

The data from theoretical calculations obtained using Eqs. (2) and (3) were compared with the data from physical experiments. The experimental data which was used in this work has been published by the Semenov Institute of Chemical Physics Russian Academy of Sciences employees in [\[16\].](#page-8-3) The experiment included the acoustic resonance method, which is quite famous method to the non-destructive testing $[17-20]$. It is important to be noted that in acoustic methods it has been used elastic waves (longitudinal, shear, surface, normal, bending) of a wide frequency ultrasonic range, which have emitted in a continuous or pulsed mode. Elastic waves propagate different speeds in the sample material. Elastic waves are attenuated according to the parameters of the material. When a sound wave propagates in a transversally isotropic material at the boundaries of interfacial surfaces (in the area of adhesive interaction between layers), processes such as refraction, reflection, and scattering can occur. It affects the amplitude, phase, and other parameters.

There is a lot of scientific evidence that the section geometric dimensions, which are much less than rod length, are very important for ultrasonic control of the rod parameters. However, all this evidence is for rods with a homogeneous structure. Equation [\(2\)](#page-2-0) could be used for makes it possible to include the length of the rod section *L*, the contact layer and the Poisson's ratio. Therefore, it becomes possible to see the effect of the section size on the speed of transmitted sound and check the effect of the elastic modulus and Poisson's ratio of a viscoelastic polymer (epoxy). The relationship between Young's modulus and the speed of sound is expressed by the following Eq. [\(4\):](#page-3-1)

$$
c = \sqrt{\frac{E}{\rho}},\tag{4}
$$

where *E* is Young's modulus; *ρ* is a density (it is constant in this work).

Results

It has been shown the sound speed in composite samples, which were shown in Fig. [2,](#page-3-0) versus the sound speed in the polymer in Fig. [3.](#page-5-0) Also, it can be found that with an increase of Young's modulus of polymers, the layered and three-section rods samples sound speed increases too. As a result of the experiment, a significant discrepancy between Young's modulus of the layered and three-section (composite) rod was obtained. It has been shown in Fig. [3.](#page-5-0) The sound speed in a sample with thin polymer layers begins to increase sharply, exceeding the speed in a three-section rod many times over. As can be found

from the graph and numerical information (Table [1\)](#page-5-1), the difference between the characteristics of the layered and three-section sample is significant. The result indicates that the interlayer effect is greater in a layered rod than in a three-section. It is worth recalling that in two composites, rods have the same volume fractions of polymer and steel (15 % polymer, 85 % steel). The obtained result could be explained by a physical and mechanical feature behavior of thin polymer layers, which enclosed between steel layers. Based on the relation for the bulk modulus (1) , which is part of Eq. (2) , it follows that the bulk modulus *K* can be much larger than Young's modulus. Also, it could be found that the closer Poisson's ratio is to the limit value of 0.5, the greater *K* and more the resistance of the layered structures stretching or compressing. The equation for determining the effective Young's modulus of a layered rod [\(2\)](#page-2-0) includes the indicated physical dependence. The considered layered rod was created because a layer of polymer is glued to a solid steel layer. The transverse dimensions of the layers are the same and shown in Fig. [2.](#page-3-0) As a result, a tensile force can be applied to the rod across the layers along the rod, i.e. across the layers. In this case, the tensile stresses in all layers are the same. Since the tensile stresses are the same, the material with the lower young's modulus (i.e. polymer adhesive) is deformed first. When any rod is stretched, its transverse dimensions are reduced. These phenomena are reflected by Poisson's ratio. For isotropic material, its value is usually greater than zero, but less than 0.5. If it is equal to 0.5, then it is a comprehensively in extensible or comprehensively incompressible material. Also, it means that its volume does not change during deformation. But with other Poisson's ratios, the material changes its volume when there is a deformation. The interconnection between volumetric relative strain Δ*V*/*V* and confining pressure (tension) reflects the volumetric tension (compression) modulus *K* = Δ*P*/(Δ*V*/*V*). It is related to Young's modulus by Poisson's ratio in elasticity [\(1\)](#page-1-0). There is the following statement: when Poisson's ratio *μ* is close to 0.5, the value of bulk modulus tends to infinity. Obviously, as a result of applying a tensile force to the rod, the softer material responds to tension. In that situation, it is a polymer adhesive, but when there is a stretched process, it must reduce its transverse dimensions. However, because of the adhesion process to a rigid and practically non-deformable substrate (i.e. steel), the polymer cannot reduce its transverse size. As the result, the polymer adhesive became fully stretched. It happens during compression, too. Those, instead of Young's modulus, the modulus of all-round tension (compression) *K* operates, which can significantly exceed Young's modulus. Especially with Poisson's ratio close to 0.5. Solving the problem of the stress-strain state of a thin layer of adhesive using the contact layer method $[12,13]$ $[12,13]$ makes it possible to determine the size and magnitude of stress concentration near the edge. The rest of the inside part of the adhesive is completely stretched. The result of these phenomena presented in Fig. [3](#page-5-0) for a sound speed.

Equation [\(2\)](#page-2-0) reflects not only volume fractions effect, Young's modulus, and Poisson's ratios of the polymer and steel, but also the stiffness of the contact layer *G*h. The parameter *G*^h is the shear modulus ratio of the contact layer to its thickness. The data presented in Table [1](#page-5-1) shows that if contact layer stiffness increase, the effective Young's modulus and the sound speed increases too. The contact layer stiffness changes its parameters at the beginning of gluing quite rapidly, but over time this process slows down. The stiffness of the contact layer *G*^h was calculated by using the function which was obtained from the approximation of experimental data: $G_h = -145.6 \cdot {E_1}^2 + 6438 \cdot E_1 - 33250.$

		The speed of sound, m/s			
Experimental data				Theoretical methods data	
E_1 , MPa	Contact layer stiffness G_h , MPa/mm	Layered composite	Three-segment composite	Model taking into account the contact layer	Reuss model
6	136.4	129.962	87.655	98.518	76.396
7	4681.6	223.996	92.591	235.773	82.516
8	8935.6	278.537	101.082	286.401	88.212
9	12898.4	325.586	108.914	323.182	93.562
10	16570.0	368.029	112.625	353.127	98.621
11	19950.4	385.623	119.704	378.701	103.434
12	23039.6	401.020	123.090	401.091	108.031
13	25837.6	409.839	126.386	420.974	112.441
14	27968.4	418.472	129.598	438.773	116.684

Table 1. Experimental and theoretical data of testing the sound in rods speed

Fig. 3. Experimental and analytical study comparison of the three-section and layered rods sound speed and epoxy adhesive Young's modulus

There are the graphs and initial data for each case of the investigated composite rods by acoustic resonance method in this part of article. The processing of the measurement results and the necessary numerical experiments were performed by using a computer program which was written in the Matlab R2020b.

In the Fig. [4](#page-6-0) it has been shown that the sound speed increased significantly when the size of the section was changed. This process occurs in connection with an increase of the composite Young's modulus. It was calculated by Eq. (2) . The length is also taken into account the coefficient *ν* in Eq. [\(2\).](#page-2-0) It characterizes the contact layer of the considered object.

Fig. 4. Dependence of the sound speed and the size of the square section *L*. Initial data of the calculation case: *L* varies from 10⁻⁹m to 0.01 m; E_0 = 210,000 MPa; E_1 = 500 MPa; μ_0 = 0.3; μ_1 = 0.44; ρ_0 =7800 kg/m3; G_h = 25000 MPa/m; $ρ_1$ =1200 kg/m³

The sound speed upraises with an increase of Young's modulus of the polymer, since the interatomic interaction becomes much stronger. Soft viscoelastic polymer layers of epoxy resin have relatively high damping properties, and in the process of hardening one can observe their decrease. It has accompanied by an increase in Young's modulus and the speed of sound. The speed of sound in crystalline solids has characterized by anisotropy, i.e. the dependence on the propagation direction. In anisotropic bodies, there are different distances between atoms in different directions, different values of the interaction force, and, consequently, different properties. All these characteristics affect the speed of propagating sound. Polymeric materials have a viscoelastic nature, which affects too.

Fig. 5. Dependence of the sound speed passing through the composite and Poisson's ratio of polymer. lnitial data: L = 0.01 m; E₀ = 210,000 MPa; E₁ = 500 MPa; μ₀ = 0.3; μ₁ varies from 0 to 0.5; ρ₀ =7800 kg/m³; G_h = 25000 MPa/m; ρ_1 = 1200 kg/m³

It has been shown in Fig. [5](#page-6-1) that with an increase in Poisson's ratio of the polymer to the limiting value of 0.5, the sound speed increases since the layered structure, acquires a greater ability to resist compression. Poisson's ratio which included in Eq. [\(2\)](#page-2-0) must be used for accuracy of the calculations. The size of the square section *L* and Poisson's ratio do not affect the value of Young's modulus, which was calculated by Reuss model, and therefore the speed of sound remains unchanged. It was shown in Figs. [3](#page-5-0) and [4.](#page-6-0)

Conclusion

We can conclude that the sound speed in a rod with a layered structure depends on many parameters, which must be included. It is important to note that the analytical formula for determining the effective Young's modulus with the contact layer model has a fairly good agreement with the results of a physical experiment and can be considered accurate. The formula of the effective Young's modulus, which included the contact layer for determining the sound speed, gives possible to carry out the numerical experiments by changing the size of the rectangular section and the mechanical properties values of the two materials constituent layered composite. It has been believed that as smaller the rectangular section size of the rod in relation to the length as the results of nondestructive testing are better, but it was found from the presented results, this rule does not match in the case of a layered composite rod. With a significant change in the modulus of elasticity depending on the section size, a significant synergistic effect can be found but with small cross-sectional dimensions, it can be lost.

References

1. Galich PI, Fang NX, Boyce MC, Rudykh S. Elastic wave propagation in finitely deformed layered materials. *[Journal of the Mechanics and Physics of Solids](https://www.doi.org/10.1016/j.jmps.2016.10.002)*. 2017;98: 390-410.

2. Li Y, Kaynia N, Rudykh S, Boyce M. Wrinkling of Interfacial Layers in Stratified Composites. *[Advanced](https://www.doi.org/10.1002/adem.201200387) [Engineering Materials](https://www.doi.org/10.1002/adem.201200387)*. 2013;15(10): 921-926.

3. Coogan TJ, Kazmer DO, Prediction of interlayer strength in material extrusion additive manufacturing. *[Additive Manufacturing](https://www.doi.org/10.1016/j.addma.2020.101368)*. 2020;35: 101368.

4. Nsengiyumva W, Zhong S, Lin J, Zhang Q, Zhong J, Huang Y. Advances, limitations and prospects of nondestructive testing and evaluation of thick composites and sandwich structures: A state-of-the-art review. *[Composite Structures.](https://www.doi.org/10.1016/j.compstruct.2020.112951)* 2021;256: 112951.

5. Zhang G, Zheng H, Zhu X. Optimization of composite plates with viscoelastic damping layer for high sound transmission loss under stiffness and strength constraints. *[Composite Structures](https://doi.org/10.1016/j.compstruct.2022.116563)*. 2023;306: 116563. 6. Gong M, Zhang H, Wu J. CZM analysis and evaluation of influencing factors on interlayer adhesion of

asphalt mixture with double-layer continuous pave. *[Construction and Building Materials](https://www.doi.org/10.1016/j.conbuildmat.2021.124211)*. 2021;302: 124211. 7. Mukherjee B, Batra RC, Dillard DA. Effect of confinement and interfacial adhesion on peeling of a flexible plate from an elastomeric layer. *[International Journal of Solids and Structures](https://www.doi.org/10.1016/j.ijsolstr.2016.09.004)*. 2017;110-111: 385-403.

8. Mohanty S, Das RR. Laminated FRP composite patch design for arresting joint fractures (Adhesion/cohesion) in resin-infilled centrally inclined V-notched Al-6061-T6 plate. *[Theoretical and Applied](https://www.doi.org/10.1016/j.tafmec.2022.103491) [Fracture Mechanics.](https://www.doi.org/10.1016/j.tafmec.2022.103491)* 2022;121: 103491.

9. Dionísio JMM, Ramalho LDC, Sánchez-Arce IJ, Campilho RDSG, Belinha J. Fracture mechanics approach to stress singularities in composite adhesive joints. *[Composite Structures](https://www.doi.org/10.1016/j.compstruct.2021.114507)*. 2021;276: 114507.

10. Fu Z, Xi Q, Gu Y, Li J, Qu W, Sun L, Wei X, Wang F, Lin J, Weiwei Li, Xu W, Zhang C. Singular boundary method: A review and computer implementation aspects. *[Engineering Analysis with Boundary Elements.](https://www.doi.org/10.1016/j.enganabound.2022.12.004)* 2023;147: 231-266.

11. Bhowmick S, Liu GR. On singular ES-FEM for fracture analysis of solids with singular stress fields of arbitrary order. *[Engineering Analysis with Boundary Elements](https://doi.org/10.1016/j.enganabound.2017.10.013)*. 2018;86: 64-81.

12. Turusov RA. *Adhesive Mechanics.* Moscow: Moscow State University of Civil Engineering Publication; 2016. (In Russian)

13. Andreev V, Tsybin N, Turusov R. Layered composite and contact layer. Effective modulus of elasticity. *[E3S Web of Conferences.](https://www.doi.org/10.1051/e3sconf/20199704071)* 2019;97: 04071.

14. Turusov RA, Kuperman AM. Mechanical behavior of an adhesive in the state of thin interlayers between rigid substrates. *[Doklady Physical Chemistry.](https://www.doi.org/10.1134/S0012501611030043)* 2011;437: 38-40.

15. Tsybin N. Merged contact layer in adhesion mechanics. *[E3S Web of Conferences](https://www.doi.org/10.1051/e3sconf/202128101048)*. 2021; 281.

16. Turusov RA, Berlin AA. Adhesion and Adhesive Mechanics. *News of Materials Science. Science and Technology*. 2016;2: 17-23. (In Russian)

17. Ivanova Y, Partalin T, Georgiev I. Comparison of NDT techniques for elastic modulus determination of laminated composites. *[Studies in Computational Intelligence](https://www.doi.org/10.1007/978-3-319-65530-7_8)*. 2018;728: 79-89.

18. Ivanova Y, Partalin T, Mihovski M. Experimental study of the ultrasonic wave propagation in materials with mechanical stresses. *Journal of Theoretical and Applied Mechanics*. 2010;40(2): 71-76.

19. Zhang Z, Guo S, Li Q, Cui F, Malcolm AA, Zhongqing Su, Liu M. Ultrasonic detection and characterization of delamination and rich resin in thick composites with waviness. *[Composites Science and Technology.](https://www.doi.org/10.1016/j.compscitech.2020.108016)* 2020;189: 108016.

20. Chaki S, Krawczak P. Non-Destructive Health Monitoring of Structural Polymer Composites: Trends and Perspectives in the Digital Era. *[Materials.](https://www.doi.org/10.3390/ma15217838)* 2022;15: 7838.

About Authors

Rio-Rita V. Razakova

PhD Student (Moscow State University of Civil Engineering, Moscow, Russia)

Robert A. Turusov

Doctor of Physical and Mathematical Sciences Chief Researcher (N.N. Semenov Federal Research Center for Chemical Physics, Russian Academy of Sciences, Moscow, Russia)