

MODULATION OF VISCOELASTICITY AS A RESULT OF INFORMATION READING BY ULTRASONIC FIELD OF POLYVINYL CHLORIDE–METAL-NANODISPERSED FILLER SYSTEM

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The results of studying the viscoelastic properties and mechanical relaxation processes of systems based on polyvinyl chloride (PVC) containing 0–3.00 vol. % of nanodispersed copper powder obtained by the physicochemical method (ph/ch) and the method of electric conductor explosion (ECE) were presented. The viscoelasticity modulation was carried out at a frequency of 0.4 MHz in the temperature range $298\text{ K} \leq T \leq T_g$. Based on the phenomenological Maxwell–Frenkel approach, the processes of compression–tension, shear, and bulk deformation of the composite were investigated. It has been shown that the viscoelastic properties of the system are due to the structural changes, energy and entropic influence of the boundary layer (BL). The maximum changes in the value of the viscoelastic characteristics of the PVC system occur in the range of changes in the Cu content of 0.05–1.50 vol. %, which expands the scope of the material in thermal and dynamic (mechanical) fields.

Keywords: viscoelasticity, structural element, information, relaxation, modulation

1. Introduction

In polymer physics, two types of interaction between a system and the external environment are traditionally studied: the exchange of energy and matter. In the second half of the 20th century, the third type of interaction was discovered – the exchange of information [1]. It was concluded that it was advisable to further study, using cybernetic methods, the relationship between dynamics, kinetics and information in polymeric systems [2]. It has been shown that macromolecules are both carriers and consumers of information, and this allows them to be interpreted as cybernetic converters of information into energy. Consideration of issues related to the dissipation of macro-

molecular energy led to the creation of molecular cybernetics [1], which was used to establish fundamentally new capabilities of polymers. This is due to the fact that macromolecules have an ‘eternal’ text written on them: polymer molecules can enter into chemical reactions with environmental molecules without losing their integrity (the number of links in the main chain remains constant), but the number of ways of dynamic self-organization increases. Thus, macromolecules that already possess a certain amount of information can interact with the environment, increasing the previous amount of information. This makes it possible to show [3] that the technology of forming, modifying (filling, plasticizing, etc.) and processing polymers, obtaining composites with

an adjustable set of properties on their basis, also refers to the use, change and processing of information. For a polymeric system, any change in structure leads not only to a new mutual arrangement and the method of packing macromolecules, but also causes a change in their conformations. Therefore, the act of polymerization is a more significant event than a phase transition, since from the moment when a macromolecule appears, the system has the ability to self-assemble and self-improve. We will analyze, along with the structure, the configuration information, which is discrete, taking into account changes in the structure formation under the influence of external factors (nanodispersed filler, temperature (T), content (φ) and type of ingredients, force field). It should be noted that the process of associative reading of information has been mastered only at the level of factual knowledge (certain quantitative and qualitative characteristics of systems), which is not enough to predict, obtain and exploit newly created materials. The amount of constructive knowledge is usually not formalized and does not have a complete logical form. The information obtained needs to be specified in each cluster of constructive knowledge for the purpose of its practical application.

Accordingly, the aim of this work is to investigate the molecular kinetic nature of PVC-based composites in the temperature range covering the glassy and transitional (highly elastic) states, as well as modulation processes associated with changes in viscoelastic characteristics (dynamic moduli, energy dissipation, viscosity) depending on the content of nanodispersed Cu of a different physicochemical nature of its production and the presence of a boundary layer. At the same time, a heterogeneous polymer system (HPS) is subjected to longitudinal, volumetric and shear deformation at a frequency of 0.4 MHz.

2. Experimental part

PVC of the C-6359-M brand, with a molecular weight of $62.5 \cdot 10^3 \text{ kg} \cdot \text{mol}^{-1}$, was chosen as the object of study, since in the $[-\text{CH}_2\text{CHCl}-]_n$ link, more than half of the mass is accounted for by the chlorine atom, which makes it possible to trace the interaction of the macromolecule with the active centres of the surface of the metal nanodispersed

filler. In addition, the production of PVC-based polymeric materials is growing steadily, and their properties are, in most cases, well understood, which makes it easier to interpret the mechanisms of relaxation processes in composites. At the same time, intensive searches are underway for active modifiers of the material's set of properties, which are widely used in various fields of science and technology.

Nanodispersed Cu powders were chosen as a PVC filler because there are active centres on their surface that are capable of donor–acceptor interaction between metal atoms and carbon [1]. It is characteristic that there is also a known law of change in the thickness of the oxide film, which does not lead to the destruction of the bonds of PVC structures with the active centre of the Cu surface (ions) [3]. This can reduce internal stresses and change the free volume of the material, which will expand the range of use of the composite in scientific and industrial areas of electronic engineering [4].

Two types of PVC systems were studied, in particular: the first one contained nanodispersed Cu powder obtained by the physical and chemical method (pc/ph) [5] using an ultrasonic field that ensured the presence of particles with a size of $(35 \pm 1) \text{ nm}$, which, according to Joint Committee on Powder Diffractions Standards, Powder Diffraction File (JCPDS, PDF-2), corresponds to copper crystallites. The second is the electric explosion of the conductor (ECE) [5], during which it was in extreme non-equilibrium conditions associated with a rapid heating of the metal and a subsequent cooling of dispersion products in the presence of electromagnetic fields, ionization and diffusion. The average size of Cu particles determined by X-ray diffraction analysis (XRD) and Debye–Scherrer–Selyakov [6] was $(45 \pm 1) \text{ nm}$. The volume content of the filler varied in a range of $0 \leq \varphi \leq 3.00 \text{ vol. \%}$, and the T - p mode was carried out at $T = 403 \text{ K}$, $p = 10.0 \text{ MPa}$.

The study of the propagation velocities of longitudinal (v_l) and transverse (v_t) ultrasonic waves (US), as well as their corresponding attenuation coefficients (α_l , α_t), was carried out at a frequency of 0.4 MHz using an experimental US setup [7]. The measurement error of the velocities was 0.5–1.05, and the absorption coefficients were 8.0%. The density of the samples ρ was determined by hydrostatic weighing [7] with an accuracy of 0.2%.

3. Model and asymptotics of its description

To study PVC systems, we used the interval of stress p and strain ε changes at which the linear viscoelasticity of high molecular weight systems (HMS) is observed [8]. One way to describe the viscoelastic properties of polymers is to consider the viscosity coefficients R_i and shear modulus G_i in the form of active resistance and capacitance $C_k = G_k^{-1}$ in the substitution schemes. In accordance with the way they are connected, there are heterogeneous models (e.g. Maxwell, Kelvin, Frenkel, etc.) that can be used to determine the relaxation time τ_i , the dissipation of the energy of body structures [9] in the form of internal friction $\text{tg}\delta$. In PVC, only fluctuating structural elements with a finite lifetime τ_i can exist – microblocks (or supernets), which exhibit various forms of mobility in a wide temperature range [10]. As a model, the microblocks are represented by point masses $M_i(\bar{i}; \bar{n})$ in the form of atomic groups $[\text{CH}_2]_n$ and $[\text{CHCl}]_n$, which are united by inter- and intramolecular bonds through which the relaxation properties of the system encoded in the conformation of the macromolecule are transmitted to the macrolevel. At the same time, it is possible to record both the elastic and viscous response of the composite to the action of ultrasonic vibrations [4]. To describe the behaviour of GPCs in such a dynamic mechanical field, we assume that the total deformation of the body consists of its elastic, highly elastic, and viscous components as a combination of elastic (Hookean) and viscous (Newtonian) elements. For the analytical description of the body deformation process, we use an electrical circuit, where the total change in the pressure Δp and the volume strain ε of the system is analogous to the relationship between the voltage U and the amount of electricity q in a circuit [8]. We assume that the change in the values of Δp and ε of the composite under the action of an ultrasonic field occurs in the $\gamma(t) = \gamma_0 \exp(i\omega t)$ mode, and we identify the Maxwell's relaxation time τ_i with the settled life of the structural element. Taking into account that the temperature changes in the viscoelastic properties of the composite are studied in the glassy state and the transition region, we consider the Maxwell medium in the form of the corresponding compounds $R_i C_k$, which correspond to polyvinyl chloride and metal nanodispersed filler system (PVC + MS) [10].

According to the Maxwell–Frenkel scheme, this is equivalent to the moduli of comprehensive compression K_1, K_2 , shear C_1, C_2 , viscosity η_1, η_2 ; $C_1 = K_1^{-1}$; $C_2 = K_2^{-1}$; $R_1 = \eta_1$; $R_2 = \eta_2$. Consider the corresponding electrical circuit (Fig. 1), at the input of which there is a source of ultrasonic vibrations $u_1(t)$, which, according to Maxwell, is equivalent to the pressure of the wave $P = P_0 \exp i\omega t$. Then,

$$Z_2 = \frac{R_2}{1 + i\omega\tau_2} = \frac{R_2 - iR_2\omega\tau_2 C_2}{1 + \omega^2\tau_2^2}, \quad (1)$$

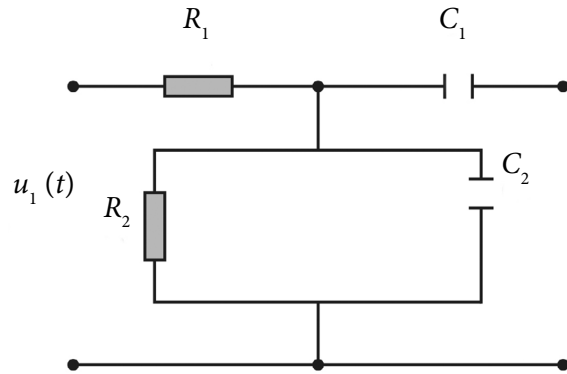


Fig. 1. Equivalent circuit diagram of the Maxwell–Frenkel model.

where $\tau_2 = R_2 C_2$ is the relaxation time of the MS structure elements, and Z_2 is the total complex resistance of MS ($R_2; C_2$).

Define the total resistance of the circuit $C_1 Z_2$ as

$$\begin{aligned} Z_3 &= \frac{1}{i\omega C_1} + \frac{R_2 - i\omega R_2^2 C_2}{1 + \omega^2\tau_2^2} = \\ &= \frac{\omega C_1 R_2 - i(R_2\omega^2\tau_2 C_1 + 1 + \omega^2 C_2^2)}{\omega C_1 (1 + \omega^2\tau_2^2)}, \end{aligned} \quad (2)$$

and the total resistance of the circuit $R_1 Z_3$ in the form Z_4 ,

$$\begin{aligned} Z_4 &= R_1 + Z_3 = \\ &= R_1 + \frac{1}{i\omega C_1} + \frac{R_2 - i\omega R_2^2}{1 + \omega^2\tau_2^2} = \frac{\omega^3 C_1^2 [(R_1 + R_2) + \omega^2 R_1 \tau_2^2]}{\omega^2 C_1^2 (1 + \omega^2\tau_2^2)} - \\ &- i \frac{\omega C_1 [1 + \omega^2\tau_2^2 (C_1 + C_2)] \omega C_1}{\omega^2 C_1^2 [(R_1 + R_2) + \omega^2 \tau_2^2 R_1]} = \eta'_1 - \eta''_2 \end{aligned} \quad (3)$$

where

$$\begin{aligned} \eta'_1 &= \text{Re}(\eta) = \text{Re}(Z_4) = \frac{R_1 + R_2 + R_1 \omega^2 \tau_2^2}{1 + \omega^2 \tau_2^2} = \\ &= \frac{R_2 + R_1 (1 + \omega^2 \tau_2^2)}{1 + \omega^2 \tau_2^2}, \end{aligned} \quad (4)$$

$$\begin{aligned}\eta'_2 &= \text{Im}(\eta) = \text{Im}(Z_4) = \\ &= -\frac{1 + \omega^2 \tau_2 (C_1 + C_2) R_2}{\omega C_1 (1 + \omega^2 \tau_2^2)}.\end{aligned}\quad (5)$$

Accordingly, the modulus of complex viscosity $|\eta|$ of the system PVC + nanodispersed metal is

$$|\eta| = \sqrt{\eta_1'^2 + \eta_2''^2}, \quad (6)$$

at the phase angle ('loss angle' [11]) δ between the force (shear stress) and the strain, which is defined by Eq. (7):

$$\text{tg} \delta = \frac{G''}{G'} = \frac{K''}{K'} = \frac{\eta_1'}{\eta_2''}. \quad (7)$$

The calculations show that the amount of energy dissipation during the deformation of the composite by an ultrasonic wave is equal to

$$\frac{1}{\text{tg} \delta} = \frac{1 + \omega^2 \tau_2 R_2 (C_2 + C_1)}{\omega C_1 [(R_1 + R_2) + \omega^2 \tau_2^2 R_1]}. \quad (8)$$

Let us study Eq. (8) in the case when

1) $C_1 \ll C_2$ and $R_1 \ll R_2$, then we have that

$$\text{tg} \delta^{-1} = \frac{1}{\omega \tau_1}, \quad (9)$$

where $\tau_1 = R_1 C_1$ is the relaxation time of PVC as a polymer matrix of the composite.

That is, in this case, the viscoelastic properties of a PVC composite can be described using the Focht model [8].

2) $C_1 \gg C_2$ and $R_2 \gg R_1$, then, respectively,

$$\frac{1}{\text{tg} \delta} = \omega \tau_{12}, \quad (10)$$

where $\tau_{12} = C_1 R_2$ is the relaxation time of the PVC-MS structures.

Thus, having the results of experimental studies of the dependences K_i ; $\eta_i = f(T)|_{\varphi_i}$ or K_i ; $\eta_i = f(\varphi)|_T$, it is possible to determine the relaxation characteristics of a heterogeneous system. Thus, based on Eq. (7), it follows that the complex viscosity of PVC + nanodispersed metal systems, the modifying effect of which is manifested in the form of MS, is equal to η = shear stress (K)/shear strain rate, i.

$$\left(\frac{d\varepsilon}{dt} \right) = \frac{K}{i\omega} = \eta_1' - i\eta_2'', \quad (11)$$

where K is the complex shear modulus, which is the ratio of stress to strain. At the same time, $K = K' + iK''$, where K' is the elastic shear modulus, which determines the amount of energy accumulation of the ultrasonic field in the composite, and K'' is the shear modulus characterizing the energy dissipation ($K'' = \omega \eta_1$). According to Eq. (7), we have that

$$\text{tg} \delta = \frac{K''}{K'} = \frac{\eta_1'}{\eta_2''}. \quad (12)$$

According to Eq. (9), it follows that due to entropic interactions and donor–acceptor bonds that occur between the active centres of nanodispersed copper as a PVC filler, the deformation of the PVC+Cu system can be described by the Focht model [8]. This indicates the validity of Hooke's and Newton's laws, respectively, in the temperature range $T < T_c$ and $T_{\text{dissip.}} \geq T \geq T_c$.

We will use this approach to predict the behaviour of the PVC + MS in the ultrasonic and temperature fields. Accordingly, we will determine the value of the dynamic modulus of elasticity ($K_2 = C_2^{-1}$) and viscosity ($R_2 = \eta_2$) of the MS of PVC + 0.50 vol. % Cu (ECE).

Provided that

$$\begin{aligned}\eta &= \frac{(R_1 + R_2) + \omega^2 R_1 \tau_2^2}{1 + \omega^2 \tau_2^2} - \\ &- i \frac{1 + \omega^2 \tau_2 (C_1 + C_2) R_2}{\omega C_1 (1 + \omega^2 \tau_2^2)},\end{aligned}\quad (13)$$

and

$$\text{tg} \delta = \frac{[(R_1 + R_2) + \omega^2 R_1 \tau_2^2] \omega C_1}{1 + \omega^2 \tau_2 (C_1 + C_2) R_2}, \quad (14)$$

$$\eta^2 = \eta'^2 + \eta''^2, \quad (15)$$

it follows that

$$\begin{aligned}R_2^2 \{ [\omega^2 C_2 (C_1 + C_2) \text{tg} \delta] - \omega^3 R_1 C_1 C_2^2 \} - \\ R_1 \omega C_1 + (\text{tg} \delta - R_1 \omega C_1) = 0.\end{aligned}\quad (16)$$

This makes it possible to determine the value of R_2 by solving Eq. (16). After appropriate calculations and mathematical transformations, we have the following:

$$R_2 = \frac{C_1}{2\omega C_2^2 \operatorname{tg} \delta}. \quad (17)$$

From Eq. (16), we obtain

$$\eta + \omega^2 R_2^2 C_2^2 \eta = R_1 + R_2 + \omega^2 R_1 R_2^2 C_2^2. \quad (18)$$

Solving Eq. (18) with respect to C_2 , we have

$$C_2 = \left[\frac{R_1 + R_2 - \eta}{\omega^2 R_2^2 (\eta - R_1)} \right]^{1/2}. \quad (19)$$

The knowledge of the viscosity value (R_2) and the value of C_2 inverse to the shear modulus (K_2^{-1}) makes it possible to determine the relaxation time τ_2 of the structure of the PVC boundary layer formed by nanodispersed Cu powder obtained by the ECE method. Using Eqs. (18) and (19), we have

$$\tau_2 = \frac{C_1}{2R_2 \omega^2 C_2^2 \operatorname{tg} \delta} \sqrt{\frac{R_1 + R_2 - \eta}{\eta - R_1}}. \quad (20)$$

Based on the results of experimental studies of the PVC + 0.50 vol. % Cu system, knowing the values of ω , C_1 , R_1 and η , calculations showed that ($R_2 = 5.0 \cdot 10^3$ Pa·s, $C_2 = 1.0 \cdot 10^{-9}$ Pa⁻¹) the structure of the MS is loosened compared to the original PVC.

4. Results and discussion

The results of the study showed that the values of the moduli K_i , G_k , as well as the corresponding viscosity coefficients $R_1 R_2$ (η_1 , η_2) and dissipative losses $\operatorname{tg} \delta$ depend on the type and content of nanodispersed copper in the composite (Fig. 2) and its temperature (Fig. 3).

It is characteristic that for all PVC systems in a range of 298–353 K, a nonlinear change in the viscoelastic properties of the material is observed, indicating a different mechanism of the deformation process under tension–compression, shear and volume change. For example, for the original PVC, in a temperature range of 308–328 K, there is β -relaxation, which does not appear under shear deformation. This indicates that PVC microblocks with point masses M_1 ($23.3 \cdot 10^{-27}$ kg) and M_2 ($80.5 \cdot 10^{-27}$ kg) form a system of parallel planes that alternate in metabolic processes (diffusion, viscosity, heat transfer [11]). Significant differences in the energy values of the inter- and intramolecular interaction of structural elements and the existence of MS, at $\varphi > 0.10$ vol. % Cu, characterizes the presence of β transition at different types of deformation. The results obtained are in a satisfactory agreement with the calculated viscosity values (Eq. (3)), which indicates the realization of conformations that correspond to more elongated PVC structures, and the rigid and dense networks of PVC interactions prevent the tight packing of polymer chains. This is also confirmed by the change in

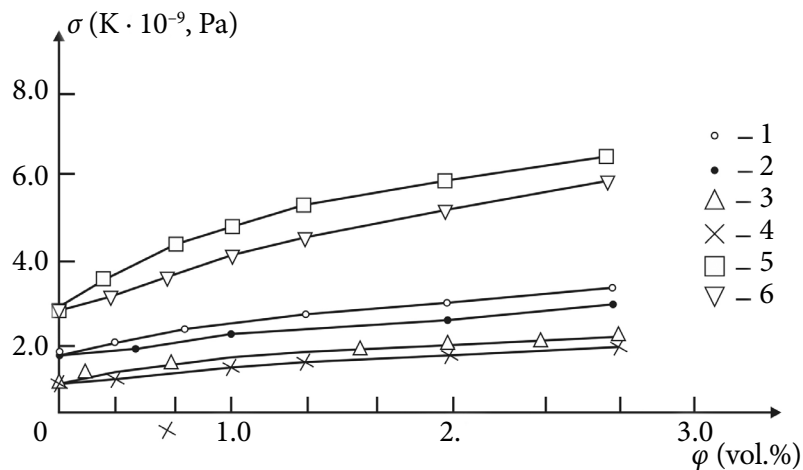


Fig. 2. Concentration dependences of the shear moduli (1, 4) and elasticity (5, 6) of PVC systems at $T = 303$ K (1) ph/ch; 303 K (2) ECE; 338 K (3) ph/ch; 338 K (4) ECE; 308 K (5) ph/ch; 308 K (6) ECE.

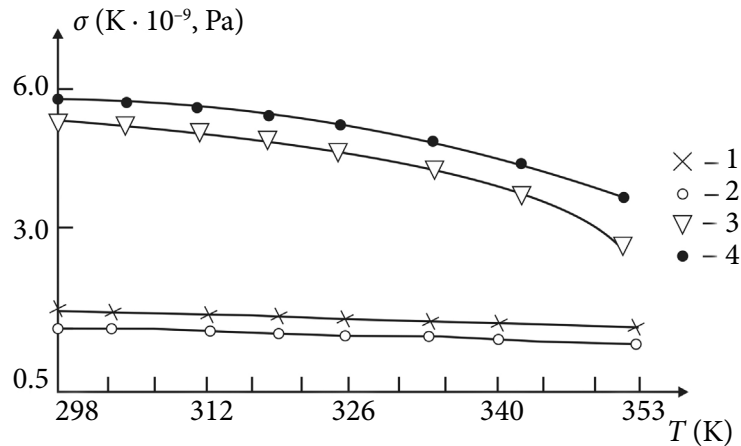


Fig. 3. Temperature dependence of the shear moduli (1, 2) and elasticity (3, 4) of PVC systems at (1) PVC + 0.05 vol. % Cu ph/ch; (2) PVC + 0.05 vol. % Cu ECE; (3) PVC + 0.05 vol. % Cu ECE; (4) PVC + 0.05 vol. % Cu ph/ch.

the value of energy dissipation during the deformation of the composite by an ultrasonic wave (Fig. 4) and the nature of the dependence of the elastic modulus ($K_2 = C_2^{-1}$) and viscosity ($R_2 = \eta_2$) of MS.

At the same time, nanodispersed copper obtained by the ph/ch method, in comparison with the ECE method, more actively affects the magnitude of changes in the viscoelastic characteristics of the material (Figs. 2–4), shifting the relaxation spectrum on the scale $\ln \tau - T$ to the region of the highly elastic state [9]. The introduction of nanodispersed copper into PVC increases the settled life time (τ_1)

of the structural element, especially intensively in a range of $0.20 \leq \varphi \leq 0.45$ vol. % Cu, causing characteristic changes in the value of the elastic modulus, viscosity, and energy dissipation (Figs. 2–4). Thus, at $T \geq 343$ K and $\varphi \geq 0.10$ vol. % Cu (ph/ch) in the composite, there is an increase in the value of energy dissipation (Fig. 4) of the ultrasonic field, indicating the dominant participation of the filler and the MS in dissipating the energy of mechanical vibrations. At the same time, the uniform excitation of a plane wave by the spherical surface of the PVC-MS system is disturbed [10].

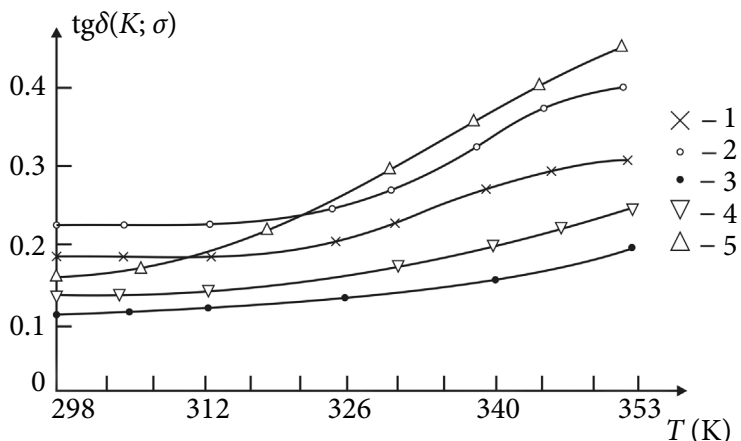


Fig. 4. Temperature dependence of $\text{tg} \delta$ of PVC systems at (1) $K \parallel$ PVC + 0.10 vol. % Cu ph/ch; (2) $K \parallel$ PVC + 0.10 vol. % Cu ECE; (3) $|\sigma|$ PVC + 2.0 vol. % Cu ph/ch; (4) $|\sigma|$ PVC + 2.0 vol. % Cu ECE; (5) the result of calculation according to Eq. (7) for the PVC + 0.05 vol. % Cu ECE system.

It has also been established that at $T_0/4 \leq t \leq T_0/2$ (where $\omega = 2\pi/T_0$) as a result of elastic (ε_0) and viscoelastic (ε_2) deformation of the composite by an ultrasonic wave, the structural elements of the PVC system undergo a modulating effect accompanied by energy dissipation, the resulting value of which is proportional to the area $\Delta S = f(T)_\varphi$ [1]:

$$\Delta S = A \left(\frac{1}{G} + \frac{2}{\pi K} \right) p_0. \quad (21)$$

To clarify other reasons for the nonlinear change in the value of viscoelastic moduli (Figs. 2–4) in the temperature range $298 \text{ K} \leq T \leq 353 \text{ K}$, we use Eq. (21) as a characterization of the relationship between the elastic and viscoelastic components of the deformation of dynamic body structures. Calculations were performed according to Eq. (22):

$$N = \frac{\Delta S_{T_3} - \Delta S_{T_4}}{\Delta S_{T_4}}, \quad (22)$$

where $\Delta S_{T_3(T_4)}$ is the area characterizing the energy loss of the ultrasonic field at $T_3 = 353 \text{ K}$ and $T_4 = 298 \text{ K}$, showed that for the original PVC $N = 30\%$, and for the PVC + 0.10 vol. % Cu (ECE) $N = 36\%$ and 32% for Cu (ph/ch). It is characteristic that for all PVC systems, the value of N increases nonlinearly with the increase in the filler content, reaching higher values for the system containing nanodispersed Cu powder obtained as a result of ECE as a filler. The most intense increase in the value of N occurred in a range of 0.10 (ECE); 0.20 (ph/ch) vol. % Cu $\leq \varphi \leq 0.50$ vol. % Cu with the subsequent entry of the value N into the region of linear change in the dependence $N = f(\varphi)_T$. Relative to the initial PVC in the studied region of changes φ , T the viscoelastic component of the system at $0.50 \leq \varphi \leq 2.00$ vol. % Cu (ECE) increased by 12% and Cu (ph/ch) by 8%.

5. Conclusions

Using the Maxwell–Frenkel model, the modulation of the viscoelasticity of a PVC composite containing nanodispersed copper powder as a filler obtained by electric explosion of a conductor and by a physicochemical method was found. It has been shown that its activity in the temperature range $298 \leq T \leq T_c$ depends on the content

and method of obtaining the nanodispersed filler. The maximum effect of nanodispersed copper on the viscoelastic properties of PVC is observed in a range of $0.05 \leq \varphi \leq 1.50$ vol. % Cu (ph/ch) and $0.15 \leq \varphi \leq 2.30$ vol. % Cu (ECE). In the case of compression–tension, shear, and bulk deformation, the relaxation moduli, energy dissipation, and viscosity coefficient are satisfactorily described by the theory, which allows us to calculate the Maxwellian relaxation time of the system's structures. The viscoelastic characteristics of the boundary layer are determined and its role in changing the relaxation time of the structural elements in a range of $2.30 \cdot 10^{-6} - 1.00 \cdot 10^{-5}$ is clarified, the values of viscoelastic moduli $1.60 \cdot 10^9 - 6.00 \cdot 10^9 \text{ N} \cdot \text{m}^{-2}$, $0 \leq \text{tg} \delta \leq 0.4$, and other viscoelastic characteristics of the composite at $298 \text{ K} \leq T \leq 353 \text{ K}$ and $0 \leq \varphi \leq 3.00$ vol. % Cu. The associative reading of the information of the initial ingredients (PVC, Cu, T - p) using a model approach to describe the PVC composite as a parametrically controlled object allows us to develop elements of the material creation algorithm and the conditions of its operation as acoustic delay lines, dampers, and matching devices.

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POLIVINILCHLORIDO IR JO NANODISPERSGUOTO METALINIO UŽPILDO SISTEMOS KLAMPIOJO TAMPRUMO MODULIAVIMAS KAIP INFORMACIJOS NUSKAITYMO ULTRAGARSINIŲ LAUKŲ REZULTATAS

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