

**INFLUENCE OF NANODISPERSE METAL FILLERS  
ON THE VISCOELASTIC PROPERTIES AND PROCESSES  
OF MECHANICAL RELAXATION OF POLYMER SYSTEMS**

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**Keywords:** *structural element, nanocomposites, glass-transition temperature, relaxation, reversible deformation*

*The results of research into the viscoelastic properties and processes of mechanical relaxation of polyvinylchloride (PVC) containing Cu nanoparticles obtained by means of electroerosion crushing and electrohydraulic destruction of agglomerates of disperse Cu in the presence of an ultrasonic field are presented. It is shown that, in the case of longitudinal shear deformation at a frequency of  $0.4 \times 10^6 \text{ s}^{-1}$  over a wide range of temperatures and content of ingredients, viscoelastic phenomena depending on structural changes in the PVC system occur. An analysis of quantitative results of the elastic and viscoelastic deformation of a body is carried out taking into account the energy and entropy components of interaction of the polymer and filler at their interface.*

**Introduction**

The results obtained for the deformation thermodynamics of glassy polymers [1] showed that the classical model of forced high-elasticity cannot describe many experimental results [2]. It became clear the model have to be abandoned, because the law of distribution of the excess energy accumulated between the elastic and inelastic deformation of a body had not been established. Therefore, the question on the accumulation and dissipation of energy and on the work of deformation of glassy polymers in a dynamic mechanical field has become the key one. It is revealed that the growth of internal energy of polymer during its loading is associated with origination of local mechanically excited states (restructuring) accompanied by inelastic

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deformations. But the role of a nanodisperse metal filler, its content, and temperature in the process of structural excitation of composite remains unclarified. Also, the deformation processes of the materials have to be investigated both in the regions of glass-transition temperature  $T_g$  and high-temperature ones (above the glass-transition temperature). Moreover, the interrelation between the strain state and the structure, relaxation properties, and flexibility of molecular chains has not been established. This especially concerns the analysis of the structure of filled polymers. Currently, investigations into the structural formations of layers bordering on the metal filler in composites with a flexible-chain polymeric matrix [3], which will allow one to calculate the dynamic moduli as functions of external power, energy, and entropy actions, are only at the beginning. Such systems can widely be employed in technics, medicine, and food-processing industry.

The purpose of this work is to investigate the viscoelastic properties of PVC as functions of the content of a nanodisperse Cu filler (i.e., PVC systems) over a wide temperature range, to evaluate the role of structural compressibility of a composite in the relaxation state, and to establish a relationship between the mechanical and structural relaxation in the ultrasonic region of deformation of the composite.

## Experiment

**Objects of research.** The basis for the production of composites in the temperature–pressure ( $T$ – $p$ ) mode was a suspension-polymerized PVC of trademark S-6359-M (GOST 14332–76) (Closed Joint Stock Company “Caustic”, Bashkortostan) with a molecular weight of  $1.4 \cdot 10^5$ . The nanodisperse Cu particles were prepared in two stages. Initially, the metal was reduction from a salt by the chemical method in an ultrasonic field, followed by washing the colloidal system. This allowed us to obtain a metallic deposit with particles sizes of about 100 nm. After primary cleaning, the suspension was placed in a reactor, where the nanodisperse metal was obtained in the liquid phase passing an alternating electric current. The voltage selected ensured the origination of electric discharges between electrodes, which caused the electroerosion grinding and electrohydraulic destruction of agglomerates of the disperse metal. The subsequent mixing of the suspension was carried out by an ultrasonic field, which ensured uniformity of Cu particles. An X-ray phase analysis was used to determine the average size  $d$  of the nanodisperse Cu particles. X-ray diffraction patterns of PVC–nanodisperse metal composite specimens of thickness  $7.0 \cdot 10^{-4}$  m and an  $\alpha = \text{Al}_2\text{O}_3$  reference specimen with particle sizes of 10–40  $\mu\text{m}$  were obtained under the same conditions on a DRON-3 diffractometer in the step scan mode of a photon counter by using a nickel  $\beta$ -filter of  $\text{CuK}_\alpha$  radiation with wavelength  $l = (1.542 \pm 0.002) \cdot 10^{-10}$  m at an operating voltage of 24 kV and strength of anode current of 20 mA in the range of angles  $34 \leq 2\theta \leq 78^\circ$ . Diffraction peaks corresponding, according to the JCPDS PDF2 standards, to Cu crystallites and indicate the absence of CuO and  $\text{Cu}_2\text{O}$  crystallites, were observed on the X-ray diffraction patterns of all specimens.

The size  $d$  was determined as

$$d = \frac{1.33\lambda}{\beta_d \cos \theta},$$

where  $\lambda$  is wavelength,  $\beta_d$  is expansion of the diffraction peak, and  $\theta$  is the Bragg angle. The processing of experimental data was carried out by using an X-Ray Scanner, X-Ray Graphic 1.28, and Origin 6.0 programs. Calculations showed that  $d$  did not depend on the volume content of filler and equaled to  $35 \pm 2$  nm.

The volume content of PVC varied in the range  $0 < \varphi \leq 5.0\%$ . Specimens for the investigation were prepared at  $T = 403$  K and  $p = 10.0$  MPa.

**Research methods.** Propagation speeds of the longitudinal,  $v_l$ , and shear,  $v_t$ , ultrasonic waves and the corresponding absorption coefficients  $\alpha_l$  and  $\alpha_t$  were evaluated at a frequency of 0.4 MHz using an experimental setup [4]. By means of a measuring differential cuvette, pulse method, and rotating plate, in one step, we determined the values of  $v_l$ ,  $v_t$ ,  $\alpha_l$ , and  $\alpha_t$  of PVC-based systems in the interval of temperatures  $298 \text{ K} \leq T \leq 353 \text{ K}$ . In this method, the longitudinal and shear waves, propagating with speeds  $v_l$  and  $v_t$  at absorption coefficients  $\alpha_l$  and  $\alpha_t$  were excited in a specimen on the same equipment by turning the specimen through corresponding angles around the axis perpendicular to the wave propagation direction. The transmitting medium was a silicon oil of trademark PFMS-4.

The values of  $v_l$ ,  $v_t$ ,  $\alpha_l$ , and  $\alpha_t$  in the heterogeneous polymer system (HPS) were calculated by the relations

$$v_l = \frac{v_p d}{d \pm \Delta\tau_l v_p}, \quad (1)$$

$$v_t = \frac{v_p d}{\sqrt{d^2 + (\Delta\tau_t v_p)^2 - 2d\Delta\tau_t v_p \cos \alpha}}, \quad (2)$$

$$\alpha_{l,t} = \frac{1}{d} \ln \frac{A_0}{A_{l,t}}, \quad (3)$$

where  $d$  is specimen thickness,  $\Delta\tau_i$  ( $i = l, t$ ) is the transmission time of ultrasonic signal, and  $v_p$  is the speed of ultrasonic wave in liquid at the research temperature.

With account of the known values of  $v_l$ ,  $v_t$ ,  $\alpha_l$ , and  $\alpha_t$ , the real and imaginary parts of the elastic moduli of HPS were determined on the basis of Stokes equation [5], which describes the process of wave motion for vibrations of small amplitudes in an isotropic viscoelastic medium:

$$\mu' = \frac{\rho v_t^2 \left(1 - \frac{\alpha_t^2 v_t^2}{\omega^2}\right)}{\left(1 + \frac{\alpha_t^2 v_t^2}{\omega^2}\right)^2}, \quad (4)$$

$$\mu'' = \frac{2 \frac{\alpha_t v_t}{\omega} \rho v_t^2}{\left(1 + \frac{\alpha_t^2 v_t^2}{\omega^2}\right)^2}, \quad (5)$$

$$E' = k' + \frac{4}{3} \mu' = \rho v_l^2 \frac{1 - \left(\frac{\alpha_l v_l}{\omega}\right)^2}{\left[1 + \left(\frac{\alpha_l v_l}{\omega}\right)^2\right]^2}, \quad (6)$$

$$E'' = k'' + \frac{4}{3} \mu'' = \rho v_l^2 \frac{2 \frac{\alpha_l v_l}{\omega}}{\left[1 + \left(\frac{\alpha_l v_l}{\omega}\right)^2\right]^2}, \quad (7)$$

$$\nu = \frac{\left(2 - \frac{v_l}{v_t}\right)^2}{2 \left[1 - \left(\frac{v_l}{v_t}\right)^2\right]}, \quad (8)$$

where  $E'$ ,  $\mu'$ , and  $k'$  are the real parts of moduli of the longitudinal and shear waves and of the bulk modulus, respectively;  $E''$ ,  $\mu''$ , and  $k''$  are the corresponding imaginary parts;  $\nu$  is the Poisson ratio;  $\omega$  is the frequency of ultrasonic vibrations. Measurement errors of the quantities  $v_l$  and  $v_t$  were 0.5 and 1.0% and of their absorption coefficients — 8.0 and 10.0%, respectively. The density  $\rho$  of specimens and temperature coefficient of volumetric expansion  $b$  were determined by the method of hydrostatic weighing with an accuracy of 0.2% at a heating rate of 3 K/min [4].

**Structural compressibility of PVC systems.** As a typical representative of amorphous polymers, PVC has microblocks in the form of fluctuating structural elements with finite lifetimes  $\tau_i$ . The microblocks can exist for a rather long time, and the macromolecules themselves exhibit various forms of mobility over wide temperature ranges [6]. Using the statistical approach [7], PVC can be endowed with a relaxation spectrum [6], i.e., with a distribution of relaxator function in  $\tau_i$ , which is interpreted as the possibility of the system to give an elastic or inelastic response to an external action (ultrasonic vibrations in the case considered). In this case, according to the principle of time–temperature equivalence, the lifetime of individual structural elements, such as relaxators, is described by the Boltzmann–Arrhenius formula

$$\tau_i = \tau_0 \exp\left(-\frac{\delta S}{k}\right) \exp\left(\frac{U}{kT}\right) \quad (9)$$

at  $0 < T < T_g$  and the Williams–Landel–Ferry equation

$$\tau_i = \tau_0 \exp\left[\frac{AB}{[1-(T_g - B)/T]T}\right] \quad (10)$$

at  $T \geq T_g$  [8], where  $\tau_0$ ,  $A$ , and  $B$  are constants,  $U$  is the activation energy,  $S$  is entropy, and  $k$  is the Boltzmann constant.

From relations (9) and (10), it follows that, with change in temperature and at  $\omega = 4 \cdot 10^5 \text{ s}^{-1}$ , the relaxation spectrum of the PVC system is shifted and deformed allowing one to separate out the temperature ranges of relaxation ( $\dots \gamma, \beta, \alpha$ ) transitions. In this case, the region of relaxation transitions of the system can be changed not only by the parameters  $T$  and  $\omega$ , but also by ingredients, affecting the magnitudes of  $\tau_i$  [9]. However, despite the progress achieved, today, there are no a sufficient number studies allowing one to assess the entire diversity of contributions of external factors to the mechanism of interaction of a polymer with a nanodisperse metal filler and its role in the formation of viscoelastic properties of composites [10]. In the case of PVC systems, if they are regarded as a set of structural subsystems, beginning with the side groups of repeating units and ending with microblocks (or supermeshes [6]), in which they connect the composite as a whole, the presence of various types of interaction with the active centers of filler surface, at  $T < T_g$ , causes immobility of mesh nodes. In this case, the internal friction, caused by the vibrations of structural elements, can be of two types [10] and is accompanied by changes in the viscoelastic properties of the material.

In the case of distribution of ultrasonic waves in the PVC systems considered, the strains and stresses as functions of time are determined by using the multiplier  $\exp(j\omega t)$ , and the dynamic viscosity is [8]

$$\eta_* = \frac{G_*}{j\omega}, \quad (11)$$

where  $j = \sqrt{-1}$  and  $G_* = G' + jG''$ . From here, it follows that  $\eta_* = \frac{G''}{\omega} + \frac{jG'}{\omega}$ . The quantity  $\eta = \frac{G''}{\omega}$ , as the dynamic viscosity, characterizes the mechanical losses (internal friction) [6].

Two kinds of deformations in the stressed state of the system take place: volumetric and shear ones [11]. Therefore, we will characterize the polymer by two types of relaxation processes [8]. In general, the volumetric relaxation, associated with the compressibility and volume changes of polymer flows, proceeds in other conditions than shear deformation, where the volume does not change. However, the relaxation processes under these types of deformation in nanodisperse metal systems, have not been studied yet. This especially concerns the volumetric viscoelasticity of composites, because the complicated influence of structural relaxation process have an affect on it. The only method allowing one to simultaneously determine the viscoelastic (highly elastic) material properties under the both types of deformations is the acoustic method proposed, where the absorption coefficient  $\alpha_i (i = l, t)$  is related to the shear  $\eta_t$  and volumetric  $\eta_V$  viscosities by the relations [5]

$$\alpha_l = \frac{\omega^2}{2\rho V_l^3} \left[ \left( \frac{4}{3} \eta_t + \eta_V \right) + \lambda \frac{C_p - C_V}{C_p C_V} \right], \quad (12)$$

$$\alpha_t = \frac{\omega^2}{2\rho V_t^3} \eta_t. \quad (13)$$

It was found that, for polymer composites, the energy dissipation due to thermal conductivity  $\lambda$  can be neglected at  $\omega \cong 10^6 \text{ s}^{-1}$  [12], which simplifies the calculation of  $\eta_V$ . An analysis shows that the Maxwell model [8] allows one to use the concept of relaxation time  $\tau = \eta/E$  as a material constant at  $T = \text{const}$  to characterize the viscoelastic properties of the materials. It should be noted that, in the classical elasticity theory of amorphous bodies, the influence of the parameters  $\eta_l$  and  $\eta_V$  is neglected. However, any system relaxes according to its own laws, regardless of its excitation mode [13]. In this case, under the action of ultrasonic waves, the heterogeneous system is disturbed from the quasi-equilibrium state by mechanical forces. In structural relaxation, no external forces operate on the system, and it goes into a new nonequilibrium state by changing the temperature  $T$ . In the studies carried out, the matrix polymer (PVC) was subjected to simultaneous actions of a nanodisperse metal filler, temperature, and ultrasonic vibration with a frequency  $\omega$ . However, the relation between the relaxation time  $\tau$  of individual simple relaxation processes and the mobility of structural elements of the PVC systems, determined by the ultrasonic method, was not fully investigated. For this purpose, using the concept of volumetric viscoelasticity [11], we present the total shear strain  $S$  induced by the action of an ultrasonic field of stress  $P$  as the sum of pure elastic  $\varepsilon_1$  and viscoelastic  $\varepsilon_2$  strains, which vary according to the laws [14]

$$\varepsilon_1 = \frac{1}{G_1} P, \quad \varepsilon_2 = \frac{1}{G_2} P \left(1 - e^{-t/\tau_2}\right), \quad (14)$$

where  $t$  is the time equal to the period of action of the ultrasonic field;  $\tau_2 = \eta_V \left(\frac{1}{K} + \frac{1}{G}\right)$  is the lifetime of a structural element in the supramolecular structural formation [6].

The total time of the structural formation of PVC-based systems is

$$\tau = \tau_1 + \tau_2. \quad (15)$$

Relations (14) and (15) allow us to determine the ratio of the pure elastic strain of the system to the viscoelastic strain in the form

$$\frac{\varepsilon_1}{\varepsilon_2} = \frac{G_2}{G_1} \left(1 - e^{-t/\tau_2}\right)^{-1}, \quad (16)$$

where  $G_1$  and  $G_2$  are the shear moduli at  $T < T_g$  and  $T > T_g$ , respectively.

## Results and Their Discussion

In Figs. 1 and 2, the temperature and concentration (at  $T = \text{const}$ ) vs. time  $\tau = G''/(\omega E)$  are presented. It is typical that an insignificant nonmonotonic increase in  $\tau$  is seen for all composites in the range  $298 \text{ K} \leq T \leq 353 \text{ K}$ . At the same time, in the concentration relation  $\tau = f(\varphi)_T$ , a characteristic reduction in  $\tau$  is evident, which is especially significant at  $0 < \varphi \leq 0.5\%$ .

In Figs. 3 and 4, the volumetric  $h_V$  and shear  $h_I$  viscosities vs. temperature  $T$  and filler content  $\varphi$  of PVC-based systems according to relations (12) and (13) are shown. It turned out that  $h_V$  and  $h_I$  nonmonotonically decreased with rise in temperature both for the initial PVC and the heterogeneous systems on its basis. In this case, the values of  $h_V$  exceeded the values  $h_I$  1.6-2.0 times over the entire temperature range (298-353) K considered. This fact indicates that a greater number of structural elements of the polymer participated in the volumetric strain of the PVC system in the dissipative process of pulse propagation than in shear. The concentration dependence  $\eta_{V,I} = f(\varphi)|_T$  shows a nonlinear increase in the quantities, whose values are reduced in an isothermal cut of characteristic, while retaining a practically parallel translation of the curves. Knowing the values of  $h_V$  and  $h_I$ , the relaxation times  $\tau_V$  and  $\tau_I$  for the volumetric and shear strains of PVC systems can be calculated. It turned out that their values were close to each other and equal to  $(2.24-2.76) \cdot 10^{-6} \text{ s}$  at  $T = 298 \text{ K}$ , while remaining higher than those corresponding to the Maxwell relaxation, which lie in the range  $(1.3-0.9) \cdot 10^{-7} \text{ s}$  for similar systems and temperatures. The character of changes in these values indicates that the composites possess elasticity properties, which is characteristic of viscoelastic bodies [6].

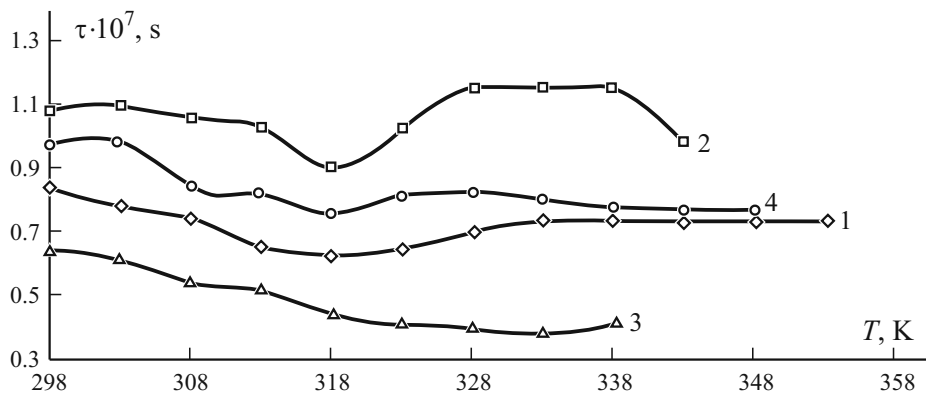


Fig. 1. Maxwell relaxation time  $\tau$  vs. temperature  $T$  for the initial PVC (1) and PVC systems with  $\phi = 0.05$  (2), 1.0 (3), and 5.0 vol.% (4).

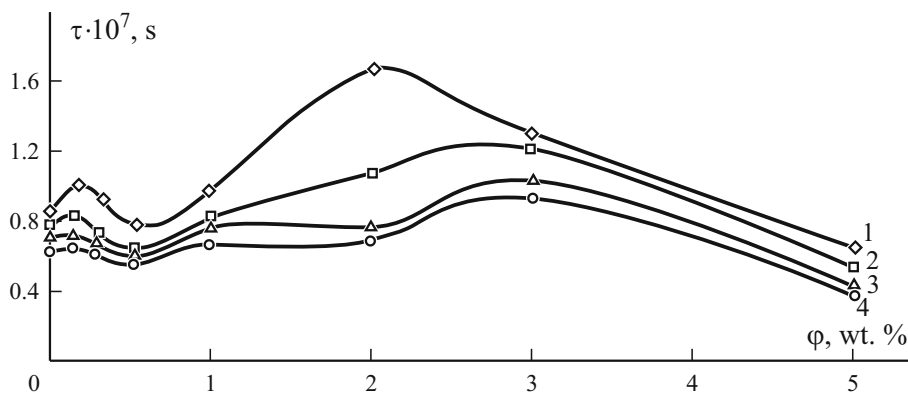


Fig. 2. Maxwell relaxation time  $\tau$  vs. volume content of nanodisperse filler  $\phi$  at  $T = 298$  (1), 308 (2), 318 (3), and 328 K (4).

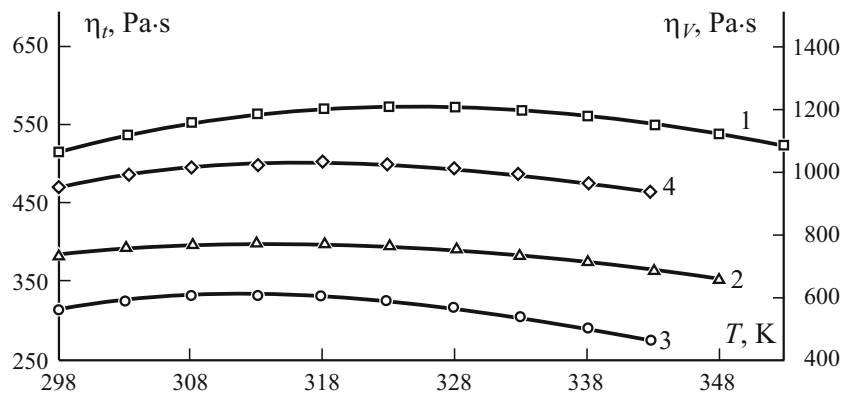


Fig. 3. Viscosities  $\eta_t$  (1, 2) and  $\eta_V$  (3, 4) vs. temperature  $T$  for the initial PVC (1) and PVC systems with  $\phi = 0.1$  (2), 0.3 (3) and 5.0 vol.% (4).

Further, let us investigate the viscoelastic properties of the material and determine, based on relation (16), the part of the pure elastic strain  $\varepsilon_1$  of the body relative to the viscoelastic strain  $\varepsilon_2$ . Previously, the effective relaxation time  $\tau_2$  (14) as the lifetime of a structural element in the supramolecular formation was calculated. It was equal to  $4.1 \cdot 10^{-6} \leq \tau_2 \leq 3.01 \cdot 10^{-5} s$  for the PVC systems at  $T = 298 K$  and  $0 \leq \phi \leq 5.0$  vol.%. According to relation (16) at  $T = 298 K$ ,  $\varepsilon_1 / \varepsilon_2 = 0.61$  for the initial

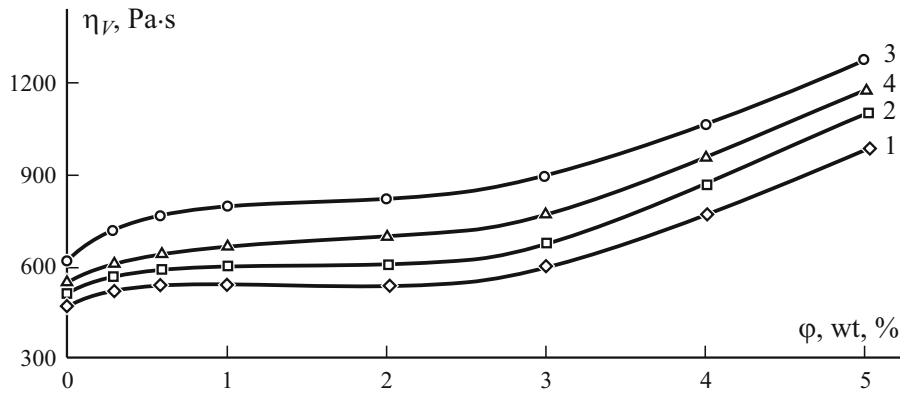


Fig. 4. Viscosity  $\eta_V$  vs. volume content of nanodisperse filler  $\varphi$  at  $T=298$  (1),  $308$  (2),  $318$  (3), and  $328$  K (4).

PVC, and 0.58 and 0.20 for the PVC systems with  $\varphi=0.05$  and  $0.05$  vol.%, respectively, i.e., the ratio  $\varepsilon_1/\varepsilon_2$  varied according to the same law as the structural part of Maxwell relaxation  $\tau$  considered. This means that, with increasing content of the nanodisperse metal filler, loosening of the structural formations of the composite took place, which manifested as an increase in the viscoelastic part of strain. The nonlinear relation  $\varepsilon_1/\varepsilon_2 = f(\varphi)$  is connected with the state of the interphase layer, in which the packing of macromolecules is changed [15], as well as with the distribution of the character of nanodisperse metal [13].

Let us analyze the mobility of structural elements of PVC at  $T < T_g$  by using relation (9) and considering that  $\delta S/k \cong 0$  [6]. Knowing the Maxwell relaxation time  $\tau$  and that  $\tau_0 = 3 \cdot 10^{-12}$  s [4], the activation energy  $U$  of Cl atom of PVC, which limits the mobility of the repeating link of the matrix, can be determined. It turned out that, at  $T=298$  K and  $0 \leq \varphi \leq 5.0$  vol.%, the value of  $U$  was in the range  $(4.6-7.0) \cdot 10^{-20}$  J, corresponding to the energy of intermolecular interaction of PVC [16].

Results of an analysis of relation (16) showed that the part of the viscoelastic strain of the composite could be regulated with the help of the nanodisperse filler and temperature. According to the data given in [10], a viscoelastic deformation occurs in an energetically nonequilibrium structure [11]. Knowing the values of  $T_g$ ,  $E$ ,  $G$ ,  $K$ , and Poisson ratio  $\gamma$ , and the fraction of fluctuating volume  $f$  ( $f=2.5 \cdot 10^{-2}$  for PVC), the volume of atomic excitation of the material [17] is calculated:

$$V_l = \frac{3(1-2\gamma)T_g}{fE} = (39-43) \cdot 10^{-30} \text{ m}^3.$$

In this case, the energy  $W = RT_g \ln\left(\frac{1}{f}\right) = 1.8 \cdot 10^{-20}$  J coincides with the enthalpy of atomic excitation [17]

$H = ST_g = W/T_g = 5.9 \cdot 10^{-23}$  J. Knowing the volume of atomic excitation  $V_l$  and the fraction of the fluctuation volume  $f$ , the number  $n$  of excited atoms in the unit of volume whose displacement relative to the quasi-equilibrium state ensures the viscoelastic deformation of composite can be calculated:

$$n = \frac{f}{V_l} = 6 \cdot 10^{26} \text{ m}^{-3}.$$

Such a volume of atomic excitation  $V_l$  and the number of excited atoms  $n$  enables one to realize the mobility of PVC elements (first of all, H and Cl atoms, whose amounts are  $3.6$  and  $1.2 \cdot 10^{27}$  g/mol, respectively) not only in the region of pure elastic, but also viscoelastic deformation.



## Conclusions

Thus, using the acoustic spectroscopy and principle of time–temperature equivalence, it was established that two kinds of deformation (volumetric and shear) proceeded in the stressed state of PVC systems. Consideration of the dynamic viscosity (both shear and volumetric) showed that the structural compressibility of composites is a property which is closely connected with their volumetric viscosity. The deformation of PVC-based systems was considered as consisting of a pure elastic component and a viscoelastic one increasing according to the relaxation law, i.e., the same law as the structural part of volumetric relaxation. The introduction of 0-5.0 vol.% nanodisperse copper into PVC changed the viscoelastic properties of composites nonlinearly in the temperature range  $298\text{ K} < T \leq 353\text{ K}$ . Both the elastic and viscoelastic parts of deformation of the system occur due to the loosening action of the nanodisperse filler in the form of a boundary layer and the volume of atomic excitation by means of local fluctuation of enthalpy. This finally leads to an intense increase in energy dissipation, which allows one to use PVC systems containing 0.05-0.30 vol.% nanodisperse metal filler as damper devices able to operate effectively in the temperature range  $298\text{ K} \leq T \leq 353\text{ K}$ . At  $0.3 < \varphi \leq 5.0\text{ vol.}\%$ , the activity of the filler in formation of the viscoelastic properties of PVC systems decreases.

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