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Coordination Bonds as Information Systems of Polyvinyl Chloride–Nanodispersed Copper in an Ultrasonic Field

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Using computational methods, the influence of the donor–acceptor interplay between the structural elements of polyvinyl chloride (PVC) and nanodispersed Cu-filler (at $298\ \mathrm{K} \leq T \leq T_g + 10\ \mathrm{K}$, $0 \leq \varphi \leq 0.30$ vol.% Cu, $\omega = 0.4$ MHz) on the change in the values of dynamic moduli (E, K, G), viscosity (μ), internal friction (tgδ), and relaxation times (τ_i) of the system. Based on the Maxwell–Frenkel model, the ways to regulate the behaviour of PVC composite in the ultrasonic and thermal fields are indicated.

З використанням обчислювальних методів з’ясовано вплив донорно-акцепторного взаємочину між елементами структури полівінілхлориду (ПВХ) і нанодисперсного Cu-наповнювача (за умов $298\ \mathrm{К} \leq T \leq T_g + 10\ \mathrm{К}$, $0 \leq \varphi \leq 0{,}30$ об.% Cu, $\omega = 0{,}4$ МГц) на зміни величин динамічних модулів (E, K, G), в’язкости (μ), внутрішнього тертя (tgδ), часів релаксації (τ_i) системи. На основі моделю Максвелла–Френкеля зазначено шляхи реґулювання поведінки ПВХ-композита в ультразвуковому та тепловому полях.

Key words: nanosystems, coordination bonds, structural element, dissipation.

Ключові слова: наносистеми, координаційні зв’язки, структурний елемент, дисипація.

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1. INTRODUCTION

It has been established that polyvinyl chloride (PVC) filled with nanodispersed metal can be used as a matrix for displays, acoustic delay lines, and structural elements [1]. However, there have been no systematic studies of the viscoelastic properties of PVC composites containing nanodispersed copper obtained by the method of rapid conductor electric explosion (RCE) at different types of deformation (ε_i ($i = 1, 2, 3$), content (φ) of ingredients, and a wide range of temperatures (T). At the same time, the most valuable and complete information on the nature of the effect of nanodispersed metal on PVC structure formation can be obtained on the basis of the results of ultrasound physics (molecular acoustics) in the mega- and kilohertz frequency range [2]. Today, it has become obvious that the law of distribution of the internal energy of the system between the elastic and viscous components of the composite deformation has not yet been established, the role of the surface and the interstructural interaction of amorphous polymer–metal on the formation of viscoelastic properties of the material has not been clarified, and there is no complete structural and statistical model that can explain the physics of the processes of momentum and energy transfer in the composite and indicate ways to control them.

Accordingly, the aim of the study is to investigate the viscoelastic properties of PVC–nanodispersed metal systems using phenomenological approaches and elements of the molecular kinetic theory of polymers as a condensed state of matter, taking into account the physical chemistry of the surface metal (Cu)–dielectric (PVC), and to determine the limits of practical use of the composite.

2. MODEL. THE EFFECT OF VISCOELASTICITY IN A COMPOSITE

The development of non-ionic technology [3] requires the study of the viscoelastic properties of flexible chain polymers filled with nanodispersed metal powders. At the same time, it is necessary to investigate the directed effect of the active centres of the filler surface on the distribution of the composite free energy between elastic and inelastic deformation of such heterogeneous polymer systems (HPS). The solution to this problem requires the use of the Boltzmann's superposition principle, which allows describing the behaviour of HPS by a system of differential equations and using elements of the dual quasi-linear body model to determine changes in the mobility of structures in force and energy fields. The model representations of the PVC system are based on the statement about the fluctuating structure of the existence of microblocks [4], the elements of which are characterised by a certain lifetime τ_{ik}. Characteristically, depending on T, macromolecules themselves can exhibit different forms of mobility. This makes it possible to represent the PVC macromolecule as a one-dimensional crystal-like 'pearl necklace' [5] with the lattice value of the wave number, $k_D = (6\pi^2 N)^{1/3}$ (N is the number of structural elements of a body with volume V). Calculations have shown that $k_D = 1.6 \cdot 10^{10}$ m^{-1} is commensurate with the size of the monomeric link $[-CH_2 \cdot CHCl-]_n$ PVC. Since the Debye frequency $\omega_D = v_t k_D$ is equal to $2.4 \cdot 10^{13}$ s^{-1}, consequently, the condition $\omega_D >> \omega$ is fulfilled that allows, according to the Williams–Landel–Ferry principle, to study the relaxation spectrum of PVC–Cu at $\omega = 0.4$ MHz [2].

It has been established [6] that, in order to regulate the viscoelastic properties of a composite in a targeted manner, it is necessary to create an appropriate covalent network in it, in which there are areas for dissipating the thermal energy of dynamic structural elements, whose movement is realised due to the thermal energy of the structons.

The active centres of the surface of nanodispersed copper powder were chosen as a model for this coordination bond network, which contribute to changes in the mobility of the structures in accordance with different inter- and intramolecular forces [7]. This suggests that the material possesses the property of bulk viscoelasticity under the action of an ultrasonic field of low intensity, the deformations are small ($\varepsilon = \Delta V/V \ll 1$) and are described by the Frankel–Maxwell model [8]. The result of inter- and intramolecular interactions between structural elements is manifested in the form of viscosity (damper (R_i)), and springs characterise Hookean deformation (ε). The analytical description of the model (as a kind of connection of dampers and springs) was carried out using the method of electrical analogy [8], which allows the relaxation time of PVC structures exposed to the action of active Cu surface centres to be identified with the waiting time τ_i of the transition across the potential barrier [9] and to determine the viscoelastic properties of the transition layer (TL) of the PVC–Cu system. First of all, let us take into account the fact that coordination bonds of the donor–acceptor-type interaction occur between them, which is due to the acceptor properties of PVC macromolecules containing polar chlorine (Cl) groups [5]; Cu ions can accept unshared elemental pairs of Cl atoms and thus form a stable energy bond [10].

Let us assume that the surface density of donors and acceptors per 1 m^2 is N_D and N_A, respectively. The occurrence of adsorption bonds of the PVC macromolecule with the Cu surface, when forming a polymer composite in the T–p mode, contributes to additional structuring of the system, limiting the mobility of polymer links near the surface of the nanodispersed filler. As a result, this causes a change in the conditions of the relaxation processes and delays the establishment of a quasi-equilibrium state of PVC structure formation near the Cu surface, increasing the free volume of the composite [11].

Thus, the PVC macromolecules are only in contact with a part (S_1) of the total surface (S) of the filler:

$$S_1 = \frac{n_1}{n_1 + n_2} S, \qquad (1)$$

where n_1, n_2 are the number of atoms of the polymer matrix and filler per unit volume, respectively. Considering that the interaction between the components, depending on the distance (d_i) between the structural elements, is determined, as a first approximation, by the Boltzmann's function [8], the number of PVC-atoms' 'bounds' to the surface of the filler is

$$N_{sl} = S_1 N_1 \exp\left(\frac{-U(x)}{kT}\right), \qquad (2)$$

where N_1 is the number of PVC kinetic elements per surface unit, $U(x)$ is the energy of interaction between component atoms at the PVC–Cu interface. The values of S_1, N_1 were calculated based on the change in the value of the heat capacity for the unfilled (ΔC_a) and filled (ΔC) composite [11]; $U(x)$ was calculated by the Morse's method [8]. This difference between the potential energies of the electron in the donor and acceptor centres is defined as

$$W(n) = W_0\left(d_1, d_2, \varepsilon_1, \varepsilon_2\right) - e\Delta V, \qquad (3)$$

where ΔV is the potential difference between the centres of the electron pits in the donor and acceptor centres created by the field of all other unreacted pairs.

Since the density of acceptor centres located on the surface of nanodispersed copper is greater than that of donor centres [12], we assume that the electron of the donor centre is transferred to the nearest acceptor centre. In this case, the quasi-equilibrium state of the composite is determined from the condition of minimum free energy with respect to the number of reacted donor–acceptor pairs [6]. This allowed us to determine the limits of parameter changes: $W_0(d_1, d_2, n_1, n)_2 \approx (1.0–1.5)\cdot 10^{-19}$ J, $d_1 + d_2 \approx 10^{-10}–10^{-9}$ m, $n_1 \approx 10^{13}–10^{14}$, which provide a change in the shear modulus at the interface of the order of $(0.8–2.0)\cdot 10^9$ N·m^2 at $T_1 = 343$ K, $T_2 = 313$ K, respectively.

On the equivalent Frankel–Maxwell substitutional scheme [8], the relationship between the change in pressure (Δp) of the U/W field caused by the action of a low-intensity generator in the mode $p = p_0 \exp(j\omega t)$ and the volumetric strain was established by introducing the viscosity coefficients R_1, R_2 and viscoelastic moduli (and/or compression coefficients C_1, C_2) K_i, G_i ($i = 1, 2$) of PS and PVC, respectively.

To derive the rheological equation of state of the composite, the mechanical model of the Kneser's medium is represented as the Kelvin's electrical model [2] for the PS structures (R_1, C_1) and Maxwell's one [8] for the original PVC (R_2, C_2).

Let us describe the proposed model using the method of symbolic representation through the potential difference U_i ($i=1, \ldots, n$) and the system charge q. After the appropriate mathematical transformations, we obtain the following relations:

$$\mathrm{Re}\left|\dot{Z}\right| = \mathrm{Re}\left|\dot{\eta}\right| = \frac{R_1 + R_2 + R_2\omega^2\tau_1^2}{1 + \omega^2\tau_1^2}, \tag{4}$$

$$\mathrm{Im}\left|\dot{Z}\right| = \mathrm{Im}\left|\dot{\eta}\right| = -j\frac{1 + \omega^2\tau_1^2 + \omega^2\tau_1 R_1 C_2}{\omega C_2\left(1 + \omega^2\tau_1^2\right)}, \tag{5}$$

where $\tau_1 = R_1C_1$ is the relaxation time, C_1, C_2 are the compressibility coefficients ($C_1 = \chi_1^{-1}$, $C_2 = \chi_2^{-1}$).

Given that the sample has the shape of a flat cylinder of infinite dimensions ($l >> \lambda$) and is deformed by an ultrasonic wave (λ) propagating in a medium, whose dimensions are significant compared to the wavelength, we have:

$$\dot{\eta} = \dot{Z} = \mathrm{Re}\left|\dot{\eta}\right| + \mathrm{Im}\left[\dot{\eta}\right] = \eta' - j\eta'', \tag{6}$$

where $\dot{\eta}$ is the dynamic viscosity of the composite equivalent to the total resistance of the system $\dot{Z}$.

At the same time, the composite undergoes a transverse deformation with a frequency ω, which causes a phase shift δ between stress and strain equal to

$$\mathrm{tg}\delta = \frac{G''}{G'} = \frac{\eta'}{\eta''} = \frac{\left(R_1 + R_2 + R_2\omega^2\tau_1^2\right)\omega C_2}{1 + \omega^2\tau_1^2 + \omega^2\tau_1 R_1 C_2}. \tag{7}$$

Let us analyse the limiting cases of changes in the value of tgδ, if: 1) $R_1 >> R_2$, then, $\mathrm{tg}\delta = (\omega\tau_1)^{-1}$, *i.e.*, the behaviour of the PS is described by the Maxwell's model [8]; 2) $R_2 >> R_1$, then, dynamic change $\mathrm{tg}\delta = \omega\tau_2$ characterises the energy exchange of PVC in the volume and is described by the Feucht's model [2]. In the studied region of ingredient content, the viscoelastic properties of PVC composites obey the relationship (7), which indicates the relationship between elastic (C_1, C_2) and viscous (R_1, R_2) deformation.

3. OBJECTS OF STUDY AND METHODOLOGY OF THE EXPERIMENT

We studied redeposited PVC of the KSR-676 brand with MM $1.4 \cdot 10^5$ in the $[-CH_2 \cdot CHCl-]_n$ linkage, where more than half of the mass belongs to the chlorine atom. This allows us to trace its interaction with the active surface centres of nanodispersed copper (Cu) as a polymer filler. In addition, the results of the study of the PVC–Cu composite can serve as the basis for model representations of flexible-chain and heterogeneous systems (HS) obtained on their basis. PVC, as a multitonnage polymer, requires new active modifiers, the search for which is ongoing [5].

The nanodispersed copper powders were obtained by the method of electric explosion of the conductor (EEC). In this case, a copper conductor was placed in the mass of the initial PVC, the rapid electric explosion of which made it possible to obtain a homogeneous metal–polymer mixture (PVC–Cu), from which a composite was formed in the T–p mode ($T = 410$ K, $p = 10.0$ MPa) [3].

The linear dimensions (∅) of the filler, determined by the method of Debye–Scherrer x-ray diffraction analysis [13], were $(45 \pm 2) \cdot 10^{-9}$ m.

The volume content of nanodispersed copper powder, due to the lack of segregation, the formation of a phase topology, and the high activity of low filler contents [5], varied in the range of $0 < \varphi \leq 0.30$ vol.% Cu with a step of 0.05 by volume.

To conduct ultrasonic measurements of the viscoelastic properties of the composite, samples of the following dimensions were used: diameter of $3 \cdot 10^{-2}$ m, height of $(8–10) \cdot 10^{-3}$ m. The propagation velocity of the longitudinal v_l and transverse v_t U/W waves, as well as their corresponding absorption coefficients α_l, α_t, were determined at a frequency of 0.4 MHz using an experimental setup [11]. Using a measurable differential cuvette, the values of v_l, v_t α_l, α_t for PVC systems in the range $298\,\text{K} \leq T \leq 358\,\text{K}$ were determined on one sample using the pulse method in conjunction with the rotating plate method. Based on the Stokes equation [2], knowing the propagation velocity and energy absorption coefficients of the U/Z field, the real and imaginary parts of the elastic moduli and energy dissipation of the system were calculated. The measurement error of the v_l and v_t values was of 0.5–1.0%, and the absorption was of 8–10%, respectively.

The density of the samples (ρ) was determined by hydrostatic weighing with an accuracy of 0.2% at a heating rate of 3 K/min.

4. RESULTS AND DISCUSSION

Figure 1 shows the results of the concentration dependence of tgδ, longitudinal (E) and transverse (μ) U/W waves in PVC systems. It is characteristic that a nonlinear dependence of the energy dissipation value is observed in the entire region of nanodispersed copper content ($0 < \varphi \leq 0.30$ vol.%) and temperature range ($T = 303–333$ K). At the same time, the value of $tg\delta_t$ ($> tg\delta_l$), in both the concentration region and temperature one, changes with external factors. It is characteristic that the value of tgδ of PVC systems in the studied range of φ and T exceeds the corresponding characteristics of the original PVC. This indicates the presence of quantitative changes in the values of intra- and intermolecular interactions, which affects the mobility of the composite structure elements.

In the case of the original PVC, in the range of $308 \text{ K} \leq T \leq 328$ K, under longitudinal deformation, the β-relaxation region is manifested, which, at a content of $0.05 < \varphi \leq 0.30$ vol.% Cu, is characteristic of shear deformation. An intensive increase in the value of mechanical energy losses during deformation of the composite also occurs in the range of 333–358 K. The obtained results allow a differentiated (Δtgδ) approach to the analysis of the energy dissipation of the PVC system, which is due to the presence of nanodispersed copper as PVC filler. It has been found that the value of Δtgδ at different types of deformation of the composite by the U/W field increases in the entire range of changes in φ and T. Characteristically, when the temperature increases above 323 K (φ = const), the intensity of growth of $\Delta tg\delta = f(\varphi)_T$ decreases.

Taking into account the influence of low nanodispersed filler content ($\varphi \leq 0.30$ vol.% Cu) on the change in the structure of PVC sys-

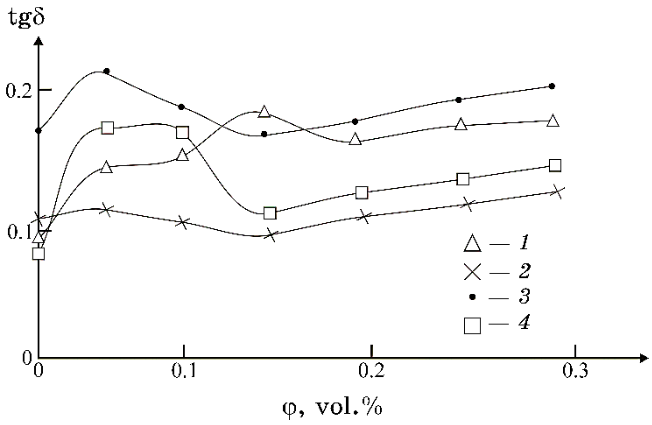

Fig. 1.

tems and the condition that $\varnothing << \lambda$ (U/Z), we calculate the effect of donor–acceptor interaction between the system structure units on the value of the viscoelastic characteristics of the material. Calculations performed according to relations (2), (3) showed that the value of the strength of a single donor–acceptor interaction in the PVC–Cu system is of $2.7\cdot10^{-11}$ N. This, according to the Kirkwood–Riseman model [13], corresponds to a shear modulus of $3.8\cdot10^{9}$ $N\cdot m^{-2}$ as a result of the relative percentage of chlorine atoms in the PVC macromolecule and the average value of the strength of the donor–acceptor interaction.

The knowledge of the value of the dissipation energy of U/W vibrations (Fig. 1) in the original PVC ($tg\delta_1$) and composite ($tg\delta_2$) makes it possible, according to the conservation law, to determine the mechanical energy losses caused by the presence of donor–acceptor interaction forces in the form of

$$\Delta tg\delta = tg\delta_2 - tg\delta_1, \qquad (8)$$

Considering the presence of donor–acceptor bonds in the PVC + Cu system as an electrical model of the Kelvin's environment [8] RC_2, we have that

$$\Delta tg\delta = \frac{\mathrm{Re}|\dot{\eta}_2|}{\mathrm{Im}|\dot{\eta}_2|} = \omega\tau_2 = \omega\eta_2 G_2^{-1}. \qquad (9)$$

This makes it possible, knowing the value of the shear modulus and the dissipation of the U/W field energy, according to Eq. (9), to determine the dynamic viscosity of the composite, taking into account the donor–acceptor interaction in the system:

$$\eta_2 = \frac{\Delta tg\delta}{\omega} G_2. \qquad (10)$$

Figure 2 shows the concentration dependence of the dynamic viscosity of the PVC + Cu system under compression–tensile deformation under isothermal conditions ($T_1 \neq T_2$). As the content of the nanodispersed filler increases, the value of $\eta_2 = f(\varphi)|_T$ changes nonlinearly with a tendency to increase and a plateau region at $\varphi > 0.05$ vol.% Cu. For the longitudinal and shear strains, the condition $\eta_l > \eta_t$ is observed to be fulfilled at $T = \text{const}$. This indicates that a change in the amount of movement of the composite structures between adjacent infinitesimal elements of the medium volume occurs, when their velocities are different under shear and/or tensile–compressive deformation. The internal friction forces in a PVC composite between structural units due to the presence of donor–acceptor bonds in the case of shear deformation are smaller (Fig. 2)

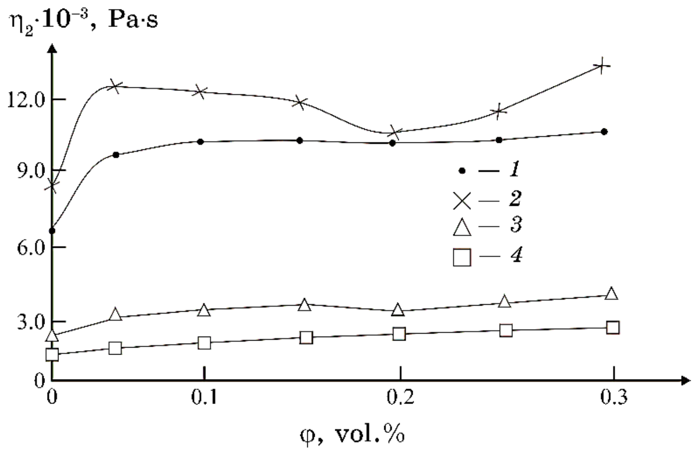

Fig. 2.

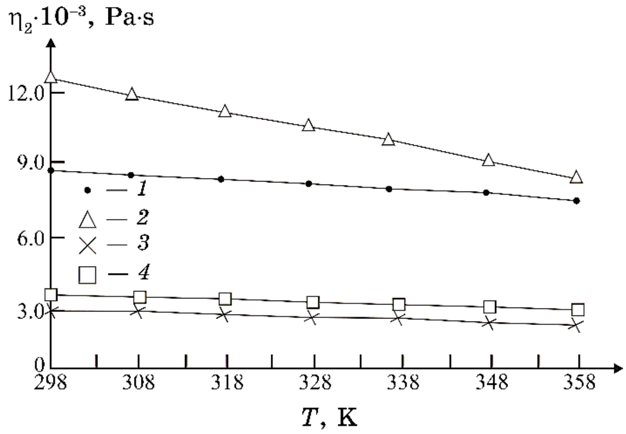

Fig. 3.

than in the tensile–compression process. With an increase in temperature (Fig. 3), the nature of the relationship between the values of $h_l > h_t$ is preserved, and the value itself decreases monotonically. This dependence is also typical for other Cu concentrations and temperature regions.

The results of the study of the temperature and concentration dependence of the dynamic moduli and viscosity per unit volume of the PVC system, which was directly affected by the donor–acceptor bond between the ingredients, allow, using the relations (5)–(7), to calculate the viscoelastic properties of the composite as a condensed body. This, in turn, makes it possible to trace the nature of the relaxation processes in the PVC system due to the formation of a donor–acceptor bond between nanodispersed copper powder and a chlorine atom of the polymer matrix. It should be noted that such studies in the megahertz range of the GCS field with simultaneous determination of the real and imaginary parts of Young's and shear moduli, volume strain, and viscosity allow us to determine not only the dynamic characteristics but also the range of composite performance in wide ranges of changes in T and φ. The nature of the deviation of the calculated values of the viscoelastic characteristics ($tg\delta_i$, η_i, E_i, G_i ($i = 1, 2$)) for PVC + Cu systems from their experimental data will allow us to assess the relaxation state of the material.

Figure 4 shows the temperature and concentration dependences of the value of the viscoelastic Young's modulus and volumetric compression of PVC systems. It is characteristic that, for all composites in the range of $298\ K \leq T \leq 358\ K$ and $0 < \varphi \leq 0.30$ vol.% Cu, a nonlinear change in values with a relaxation β-transition in the region of $308\ K \leq T \leq 328\ K$. At the same time, the most intense decrease in the value of $E = f(T)|_\varphi$ occurs with a minimum at $\varphi = 0.30$ vol.% Cu. Comparison of the experimental results of the dynamic viscosity with the calculated values according to Eqs. (4), (5) and (6), where $\eta = (\eta'^2 + \eta''^2)^{1/2}$, showed that, in this region of the volume content of the ingredient, they are in satisfactory agreement with each other. As the temperature increases, the action of the acceptor centres of the filler surface, as well as the increase in the mobility of Cl^- atoms of PVC, reduce the effectiveness of the donor–acceptor interaction on the relaxation properties of the composite. Due to thermal fluctuations, the time of detachment of the Cl^- atom from the ac-

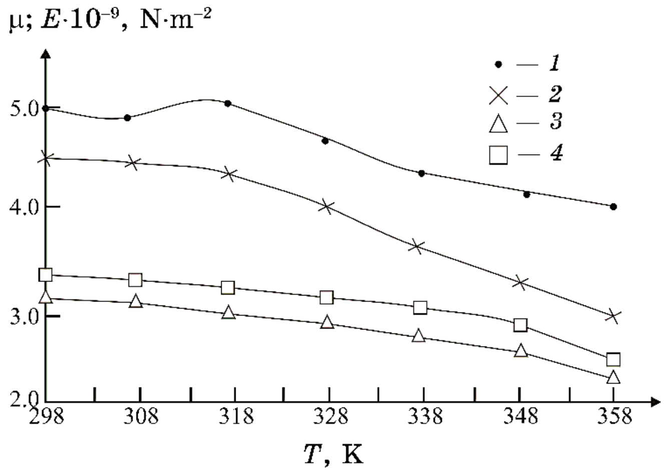

Fig. 4.

tive centre of the nanodispersed copper surface becomes shorter than the period of action of the external alternating voltage of the U/V source [14], *i.e.*,

$$\tau \leq \tau_0 \exp\left(-\frac{U}{kT}\right), \quad (11)$$

where $\tau_0^{-1} = 1.0 \cdot 10^{12}$ s^{-1} [8]. The equality sign in expression (11) corresponds to the temperature (T_0), starting from which there is an effective reduction in the influence of the filler on the value of the viscoelastic characteristics of the material. The calculations showed that the value of T_0 lies in the region of 400 K, indicating an increase in the thermal stability of the PVC system.

The analysis of the dependence $\tau = f(T)_\varphi$ (relation (11)) states that the Arrhenius-type equation is valid for the centre of the maximum β-transition at $T = T_m$ [8]:

$$\omega\tau_i = \omega\tau_0 \exp\left(\frac{U_m}{kT_m}\right) = 1; \quad (12)$$

moreover, at $\varphi = 0.30$ vol.% Cu, the relaxation time of the structures in the region $300\ \text{K} \leq T \leq 331\ \text{K}$ varies in the range of $1.8 \cdot 10^{-7}$–$1.2 \cdot 10^{-9}$ s. At $T = 313$ K, the value of U_m is of $5.3 \cdot 10^{-20}$ J, which corresponds to $\tau_i = 1.7 \cdot 10^{-7}$ s. For the original PVC, at the same temperature, the value of $\tau_i = 8.9 \cdot 10^{-9}$ s. Thus, the presence of the donor–acceptor interaction Cu^+–Cl^- at the PVC–Cu interface reduces the mobility of the structural elements of PVC systems, increasing their lifetime as an independent ‘morphosis’, *i.e.*, observed by the U/Z method at a frequency of 0.4 MHz.

If we consider that the value of the bulk viscosity and viscoelastic moduli due to donor–acceptor bonds is η_2, G_2, and the polymer matrix η_1, G_2, then, a change in the quasi-equilibrium conditions due to the energy of the U/W field will cause a non-equilibrium state with a stability criterion [6]:

$$K_c = \frac{\eta_2}{\eta_1 + \eta_2}, \quad (13)$$

The calculations of the value of K_c, using relations (4)–(6), showed that, in the regions $0 < \varphi \leq 0.30$ vol.% Cu, $298\ \text{K} \leq T \leq 358$ K, it varies in the range of 0.58, 0.62, and 0.64 for the Young’s modulus, shear, and volumetric compression, respectively.

5. CONCLUSIONS

The studies show that, due to coordination bonds of the donor–

acceptor-type interaction at the interface of nanodispersed metal-flexible polymer in the regions $0 < \varphi \leq 0.30$ vol.% Cu, $298 \text{ K} \leq T \leq 358$ K, a nonlinear change in the viscoelastic properties of the PVC–Cu system occurs, and the composite resistance to dynamic deformation in the range of $\varepsilon_{1\,(\text{tensile–compressive strain})} > \varepsilon_{2\,(\text{shear strain})} > \varepsilon_{3\,(\text{volume strain})}$ increases. This increases the temperature range of the PVC system. The proposed computational methods and a Maxwell's mechanical model can be used to predict the behaviour of other condensed media under the influence of dynamic and temperature fields.

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