 DOI: 10.1002/masy.200950412

Model Theory of Visco-Elastic Properties and Relaxation Spectra of Two Different Interpenetrating Polymer Networks

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Summary: Dynamic viscoelastic models of the system of two different interpenetrating polymer networks with different topology and type of interactions were used for calculating spectra of relaxation times of the system under consideration. It was shown, that two branches of the relaxation spectrum appear for two models of interpenetrating networks with different components. One of the branches is the branch of the collective motion of double network consisting of two initial interacting networks. Parameters of this branch of relaxation spectrum are defined by both own elastic constants of each of interacting networks and by effective quasi-elastic interactions between two networks. This branch is the low frequency one and is described by broad relaxation time spectrum. The second branch is the high frequency one and characterizes mutual local motions of two interacting networks. The relaxation spectrum of this branch is comparatively narrow and depends on the quasi-elastic constants and mutual friction which is defined by the entanglements of the networks and by its effective rigidity. The second branch does not contain extremely large relaxation times for infinitely large networks.

Keywords: interpenetrating polymer networks; relaxation spectra

Introduction

The polymer materials based on the interpenetrating polymer networks or including them are intensively investigated at the last years. There are many studies of the kinetic of the synthesis of the interpenetrating networks and mechanical, dielectric, optical, NMR properties of these systems.[1–8] The interpenetrating networks build from two similar components were also investigated.[9–11]

The local interpenetrating effects may take place also in usual network systems, prepared by normal vulcanization of a polymer melt or cross-linked by other methods. Such interpenetrating networks effects can be obtained when the cross-linking process in dense polymer systems goes so that the distances between neighbouring junctions in space (belonging to different cells of the non-regular networks) are smaller then the distance between the neighbouring junctions belonging to a given chain. In other words, the small parts of the ''primary'' networks can be treated as ''interpenetrating'' ones.

The relaxation spectra of the interpenetrating network (or ''interpenetrating'' parts of the network which is uniform in average) and the dynamic modulus and other relaxation properties of these systems may be different from those for uniform homogeneous network models. We shall remind that in polymer networks either in homogeneous or in interpenetrating ones

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many types of relaxation processes exist with different time or space scales. The α - and β - relaxation process are connected with the local motions which are smaller or comparable as the motions of Gaussian subchains (or corresponding kinetic segments).[12,13] The next characteristic scale of the local motions is the collective motions of Gaussian subchains in the chain between junctions of the network. This type of motion is described by the well-known ''visco-elastic'' Kargin-Slonimskii-Rouse model.[14–17] But in the case of polymer networks a new basic kinetic unit appears, i.e. the chain between neighbouring junctions. This element consists of many Gaussian subchains (or corresponding visco-elastic kinetic units consisting of springs and viscous beads).[14,15,18,19]

It was shown[18,19] that in good approximation one can divide the intra-chain motions (between junctions) with Gaussian subchains acting as kinetic units and large scale inter-chain specific network motions. For this type of motion, the kinetic unit is represented by the whole many segmental chains between neighbouring junctions. The effective kinetic units are characterized by average elastic constant of the chain between junction and effective friction which included the total friction of all elements of the chains between junctions and the effective friction of the junctions.

The specific features of the relaxation properties of the interpenetrating networks (as for simple homogenous networks) depend on the scale and local types of motion. The short-range motions in the main chain or in the side chain of the interpenetrating networks appearing in high-frequency dielectric relaxation, NMR, and mechanical relaxation (β and γ processes) are mostly controlled by local intrachain interaction. Probably the α-processes which are sensitive to the interchain molecular interaction (free volume effects) may be sensitive especially to the interaction between neighbouring chains in the systems of different interpenetrating networks. But such effects are not specific network effects and may be also in non-network mixture of two types of macromolecules.

It means that our model describes long-time and low-frequency behaviour of the interpenetrating network. The comparison of the increments of intrachain and specific network interchain motions for the frequency or time dependence of the dynamic modulus and dynamic viscosity was made previously especially in the papers of Gotlib, Gurtovenko, Toshchevikov, Blumen.[18,19–27]

This increment depends strongly on the number (n) of the Gaussian subchains between the neighbouring junctions, and on the ratio of the friction coefficient of the network junctions and the intrachain segments. The characteristic parameters are the relaxation time of the chain between junctions τ_{ch} (in our model it is $\tau_{ch} \sim \zeta_{ch} \langle l_{ch}^2 \rangle / kT$) and the characteristic time of the subchain τ_{seg}. It was shown for example for the network with $n = 10$ Gaussian subchains in the chain between junction that the increment of interchain relaxation properties (as in our treatment) in the $G(\omega)$ is prevailing the intrachain segmental motion if $\omega\tau_{ch}$ is smaller than 0.1- 0.2.[18,19]

The case when $\zeta_{ch} \sim 3n\zeta_{seg}$ and $K_{ch} \sim K_{seg}/n$ (were K_{seg} is the elastic constant of Gaussian subchain) has been considered.[18,19] For $\omega\tau_{ch} \sim 0.6$ the increments of the intra- and interchain motion can be equal. In the case of high frequencies (if $1/\tau_{ch} < \omega < 1/\tau_{seg}$ or $\omega \geq 0.1\tau_{seg}$) increment of the intrachain motion is prevailing. The corresponding results were obtained for dynamic viscosity. But in this case (for $n = 20$) the increments of intrachain and interchain motion in the $\eta(\omega)$ are not strongly different for $\omega\tau_{ch} \geq 0.5$ the increment of intrachain motion is greater.[18,19]

In ref. 27 the dynamic moduli G' and G'' were also considered for the network where friction of the junction is not equal to $3\zeta_{ch}$ and may be greater or smaller than the friction of the segment. In the previous paper the ratio $\zeta_{jun}/\zeta_{ch} = 3$ was used because in this case the equation of motion have the simplest form. In ref. 27 we have considered G' and G'' for case when the

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number of subchain $n = 30$, and the ratio $\sigma = \zeta_{jun}/\zeta_{ch}$ can have different value (from 0.001 to 10^4), that is for the junction with great or small viscosities (in our case $\sigma = 3$). The frequency dependences G' and G'' were calculated and it was shown that for $\sigma \leq 3$ the frequency dependence of $G'(\omega)$ is not very sensitive to the value of σ except of the case of very small ω. But if σ is greater than 30 the low-frequency part of G' will have an additional low-frequency plateau due to very ''viscous'' junctions. Also the frequency behaviour of G'' was considered at different σ. It was shown that for $\sigma = 3$ as in our previous paper the frequency dependence of G'' is mostly determined by high-frequency motions, and will have the maximum $G''(\omega)$ at $\omega \sim 1/\tau_{seg}$ and the maximum of the low-frequency part in the G'' due to interchain motion will be overlapped by the relatively intensive part of G'' due to high-frequency intrachain motion. Only at $\sigma > 30$, we can separate the maximum of G'' due to interchain motion at low-frequency from the part of the $G''(\omega)$ due to intrachain motion. But if the network is very dense, and if the number of segments between junctions is small as well as the friction of the junctions is high as compared with the friction of segments, the low-maximum G'' of the intrachain motions can be separated (at small ω). It means that the results of our theory at $\sigma < 30$ can be applied to network with relatively short chains between junction or for very low-frequency motion where the increment of low-frequency motion is greater, but in this case the value of G'' may be very small.

The form and curvature of the $G''(\omega)$ at low frequencies may be sensitive to the increment of the low-frequency motion. The asymmetry of the G'' at small frequency may exist due to increment of low-frequency interchain motion. The increment of the low-frequency motion may be greater if the value σ is large enough ($\sigma \geq 30$, i.e. very viscous junction), and n is small. In this case the interchain relaxation is presumable and G'' also at greater ω will be controlled by interchain relaxation.

The behaviour of the network at low frequencies ($\omega\tau_{seg} \leq 1$–10) is controlled by the motions on the distances the scale of which is near to the dimensions of the chains between junction. Therefore the behaviour of G' and G'' at low frequencies may be essential for the investigation of the interchain interaction in interpenetrating networks with similar distances between junction in both networks. The specific feature of the network behaviour will be essential for these distances. The interactions of smaller scale are controlled by differences between small units (Gaussian subchain) of the interpenetrating networks and must be considered separately. In the case of the network with relatively short chains between junctions the effective kinetic unit is near to the chain between junction and in this case the theory described not only long time, but also relatively the short range motion.

The specific feature of 3-D networks polymer systems, the frequency and time dependencies of the long-time and long-scale network motion are presented by the average relaxation times $\langle\tau\rangle$ and the average inverse relaxation times for relatively short motions $\tau_{invers} = \langle 1/\tau\rangle^{-1}$. But for 3D networks these values are not strongly different one from another. We shall note that for the 1-D system (i.e. for separate chain) $\langle\tau\rangle$ increases strongly with the length of the chain ($\tau_{max} \sim n^2$) $\langle\tau\rangle \sim n$ and $\langle 1/\tau\rangle^{-1}$ for long chains doesn't depend on n. For 2-D networks $\langle\tau\rangle$ also increases with the number of cells in the network (with the dimensions of the network.) But for the infinitely great 3-D network low-frequency part of the network relaxation spectrum $\langle\tau\rangle$ and $\langle 1/\tau\rangle^{-1}$ have the same order of magnitude.

The interactions between the networks due to entanglements in our approach are described by effective quasi-elastic potential and viscous interactions between elements of neighbouring entangled cells of the two networks. Therefore only interchain relaxation spectra are considered. The motions are considered, the scale of which are near or greater then the average distance between nearest junctions

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of the interacting interpenetrating networks. The intrachain motions should be considered separately by more detailed dynamic models. The models of such types were used in many papers.[15,19–28] In our paper, two cubic visco-elastic dynamic models of interpenetrating networks with the same density of junction, but with different properties of both components are considering. The spectra of the relaxation times and average relaxation time $\langle\tau\rangle$ and $\langle 1/\tau\rangle^{-1}$ will be considered. These results may be used in future for calculation of frequency and time dependencies dynamic modulus for specific networks motions.

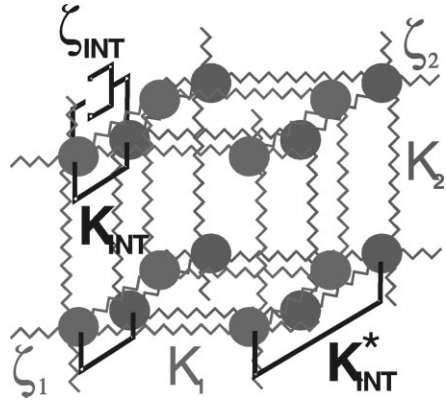

Figure 2.
Model II.

The Dynamic Models, Corresponding Relaxation Spectra and Average Relaxation Times

Two models of the interpenetrating networks with different types of interactions are presented in Figure 1 and 2. In the model I the junctions one of the networks are moving mostly inside the cells of the other network. The effective interacting force constant K_{int} between networks due to entanglements has the value between the elastic constants (K_1 and K_2) for both components. Also additionally friction interaction between networks may exist (ζ_{int}). The effective friction constants of two networks can be different ($\zeta_1 \neq \zeta_2$), also for networks with the same $K_1 = K_2$, i.e. with the same average length of the quasielastic segments between junctions.

In the model II the networks are strongly connected by the nearest positions of junctions, by specific entanglements between nearest neighbouring junctions. In this case the constants of the effective interaction between networks (K_{int} and ζ_{int}) describe mostly the interaction between regions of neighbouring junctions of the two networks.

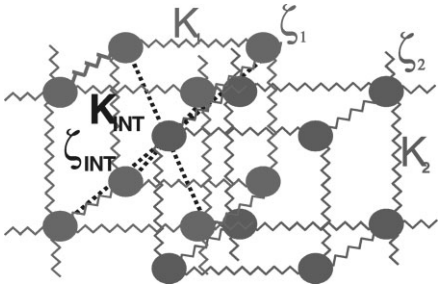

Figure 1.
Model I.

We note, that in theory we consider only the simplest case when the average distance between junctions of both interpenetrating networks is the same but the elasticity of both components is different ($K_1 \neq K_2$). But also in this case the properties of the whole complex system depend on many parameters: K_1/K_2, ζ_1/ζ_2, K_{int}/K_1, ζ_{int}/ζ_1, etc.

The equations of motion for both described models are considered. Equations of motion for model I of different interpenetrating networks are

$$\begin{aligned}
&\zeta_1 \frac{d}{dt}\left(x^{(1)}_{\alpha,\beta,\gamma}\right) + \zeta_{\text{int}} \frac{d}{dt}\Big(8x^{(1)}_{\alpha,\beta,\gamma} - x^{(2)}_{\alpha,\beta,\gamma} - x^{(2)}_{\alpha-1,\beta,\gamma} \\
&\quad - x^{(2)}_{\alpha,\beta,\gamma-1} - x^{(2)}_{\alpha-1,\beta,\gamma-1} - x^{(2)}_{\alpha,\beta-1,\gamma} \\
&\quad - x^{(2)}_{\alpha-1,\beta-1,\gamma} - x^{(2)}_{\alpha-1,\beta-1,\gamma-1} - x^{(2)}_{\alpha,\beta-1,\gamma-1}\Big) \\
&\quad + K_1\Big(6x^{(1)}_{\alpha,\beta,\gamma} - x^{(1)}_{\alpha-1,\beta,\gamma} - x^{(1)}_{\alpha+1,\beta,\gamma} - x^{(1)}_{\alpha,\beta-1,\gamma} \\
&\quad - x^{(1)}_{\alpha,\beta+1,\gamma} - x^{(1)}_{\alpha,\beta,\gamma-1} - x^{(1)}_{\alpha,\beta,\gamma+1}\Big) \\
&\quad + K^{*}_{\text{inter}}\Big(8x^{(1)}_{\alpha,\beta,\gamma} - x^{(2)}_{\alpha,\beta,\gamma} - x^{(2)}_{\alpha-1,\beta,\gamma} \\
&\quad - x^{(2)}_{\alpha,\beta,\gamma-1} - x^{(2)}_{\alpha-1,\beta,\gamma-1} - x^{(2)}_{\alpha,\beta-1,\gamma} - x^{(2)}_{\alpha-1,\beta-1,\gamma} \\
&\quad - x^{(2)}_{\alpha-1,\beta-1,\gamma-1} - x^{(2)}_{\alpha,\beta-1,\gamma-1}\Big) = 0
\end{aligned}$$

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$$\zeta_2 \frac{d}{dt}\left(x^{(2)}_{\alpha,\beta,\gamma}\right) + \zeta_{\text{inter}} \frac{d}{dt}\Big(8x^{(2)}_{\alpha,\beta,\gamma} - x^{(1)}_{\alpha,\beta,\gamma} - x^{(1)}_{\alpha-1,\beta,\gamma} - x^{(1)}_{\alpha,\beta,\gamma-1} - x^{(1)}_{\alpha-1,\beta,\gamma-1} - x^{(1)}_{\alpha,\beta-1,\gamma} - x^{(1)}_{\alpha-1,\beta-1,\gamma} - x^{(1)}_{\alpha-1,\beta-1,\gamma-1} - x^{(1)}_{\alpha,\beta-1,\gamma-1}\Big) + K_2\Big(6x^{(2)}_{\alpha,\beta,\gamma} - x^{(2)}_{\alpha-1,\beta,\gamma} - x^{(2)}_{\alpha+1,\beta,\gamma} - x^{(2)}_{\alpha,\beta-1,\gamma} - x^{(2)}_{\alpha,\beta+1,\gamma} - x^{(2)}_{\alpha,\beta,\gamma-1} - x^{(2)}_{\alpha,\beta,\gamma+1}\Big) + K^*_{\text{inter}}\Big(8x^{(2)}_{\alpha,\beta,\gamma} - x^{(1)}_{\alpha,\beta,\gamma} - x^{(1)}_{\alpha-1,\beta,\gamma} - x^{(1)}_{\alpha,\beta,\gamma-1} - x^{(1)}_{\alpha-1,\beta,\gamma-1} - x^{(1)}_{\alpha,\beta-1,\gamma} - x^{(1)}_{\alpha-1,\beta-1,\gamma} - x^{(1)}_{\alpha-1,\beta-1,\gamma-1} - x^{(1)}_{\alpha,\beta-1,\gamma-1}\Big) = 0$$

The equations of motion for model II with different interpenetrating networks have form

$$\zeta_1 \frac{d}{dt}\left(x^{(1)}_{\alpha,\beta,\gamma}\right) + K_1\Big(6x^{(1)}_{\alpha,\beta,\gamma} - x^{(1)}_{\alpha-1,\beta,\gamma} - x^{(1)}_{\alpha+1,\beta,\gamma} - x^{(1)}_{\alpha,\beta+1,\gamma} - x^{(1)}_{\alpha,\beta-1,\gamma} - x^{(1)}_{\alpha,\beta,\gamma+1} - x^{(1)}_{\alpha,\beta,\gamma-1}\Big) + \zeta_{\text{int}} \frac{d}{dt}\left(x^{(1)}_{\alpha,\beta,\gamma} - x^{(2)}_{\alpha,\beta,\gamma}\right) + K_{\text{int}}(x^{(1)}_{\alpha,\beta,\gamma} - x^{(2)}_{\alpha,\beta,\gamma}) + K^*_{\text{int}}\Big(6x^{(1)}_{\alpha,\beta,\gamma} - x^{(2)}_{\alpha-1,\beta,\gamma} - x^{(2)}_{\alpha+1,\beta,\gamma} - x^{(2)}_{\alpha,\beta+1,\gamma} - x^{(2)}_{\alpha,\beta-1,\gamma} - x^{(2)}_{\alpha,\beta,\gamma+1} - x^{(2)}_{\alpha,\beta,\gamma-1}\Big) = 0$$

$$\zeta_2 \frac{d}{dt}\left(x^{(2)}_{\alpha,\beta,\gamma}\right) + K_2\Big(6x^{(2)}_{\alpha,\beta,\gamma} - x^{(2)}_{\alpha-1,\beta,\gamma} - x^{(2)}_{\alpha+1,\beta,\gamma} - x^{(2)}_{\alpha,\beta+1,\gamma} - x^{(2)}_{\alpha,\beta-1,\gamma} - x^{(2)}_{\alpha,\beta,\gamma+1} - x^{(2)}_{\alpha,\beta,\gamma-1}\Big) + \zeta_{\text{int}} \frac{d}{dt}\Big(x^{(2)}_{\alpha,\beta,\gamma} - x^{(1)}_{\alpha,\beta,\gamma}\Big) + K_{\text{int}}\Big(x^{(2)}_{\alpha,\beta,\gamma} - x^{(1)}_{\alpha,\beta,\gamma}\Big) + K^*_{\text{int}}\Big(6x^{(2)}_{\alpha,\beta,\gamma} - x^{(1)}_{\alpha-1,\beta,\gamma} - x^{(1)}_{\alpha+1,\beta,\gamma} - x^{(1)}_{\alpha,\beta+1,\gamma} - x^{(1)}_{\alpha,\beta-1,\gamma} - x^{(1)}_{\alpha,\beta,\gamma+1} - x^{(1)}_{\alpha,\beta,\gamma-1}\Big) = 0$$

In these equations indexes (1) end (2) relate to the first and the second interpenetrating networks correspondingly. The indexes of α,β,γ are the numbers of network junctions in the cubic cell in three directions. The solution of these equations can be found in general form: $x^{(1,2)} \sim \exp[i(\alpha\theta_1 + \beta\theta_2 + \gamma\theta_3) - \lambda t]$ and leads to the quadratic equation for $\lambda(\theta_1,\theta_2,\theta_3)$, where $\theta_1,\theta_2,\theta_3$ are the phase shifts between displacements of the junctions of neigbouring cells. We obtain the equation for λ in the form:

$$\lambda^2 - b\lambda + c = 0$$

The parameters b and c are the functions of the elastic and viscous constants of the models. The roots of the quadratic equation determine the set of inverse relaxation times $\lambda_1 = 1/\tau_1(\theta_1,\theta_2,\theta_3)$, and $\lambda_2 = 1/\tau_2(\theta_1,\theta_2,\theta_3)$ for the normal modes with phase shifts $(\theta_1,\theta_2,\theta_3)$:

$$\lambda_{1,2} = b/2 \pm ((b/2)^2 - c)^{1/2}$$

The theory predicts the splitting of the two different relaxation spectra for two different (but ''independent'' networks) or two similar but ''independent'' relaxation spectra for similar networks on two new types of relaxation spectra. One of them is cooperative long time relaxation spectrum ($\tau_{\max} \to \infty$ for infinitely great network). The second finite relaxation spectrum describes the local relative motions of the interacting neighbouring junctions of both networks. The first type of collective relaxation describes the motion of ''new combined, united'' network consisted of two components, but the second branch of relaxation describes the relative mutual motion both components.

For the model I the values of b and c have the form:

$$b = \frac{(\zeta_2 + 8\zeta_{\text{int}})Z_1 + (\zeta_1 + 8\zeta_{\text{int}})Z_2 - 2\zeta_{\text{int}} + K^*_{\text{int}}G^2}{(\zeta_1 + 8\zeta_{\text{int}})(\zeta_2 + 8\zeta_{\text{int}})},$$

$$c = \frac{Z_1 Z_2 - (K^*_{\text{int}})^2 G^2}{(\zeta_1 + 8\zeta_{\text{int}})(\zeta_2 + 8\zeta_{\text{int}})}$$

where

$$Z_1 = 8K^*_{\text{int}} + 2K_1(3 - 3X(\theta);$$
$$Z_2 = 8K^*_{\text{int}} + 2K_2(3 - 3X(\theta)$$
$$G = 8\cos\left(\frac{\theta_1}{2}\right)\cos\left(\frac{\theta_2}{2}\right)\cos\left(\frac{\theta_3}{2}\right)$$

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In the case of model II the values of b and c have the form:

$$b = \frac{(\zeta_2 + \zeta_{\text{int}})(A_1 + B)}{\zeta_1\zeta_2 + \zeta_{\text{int}}(\zeta_1 + \zeta_2)} + \frac{(\zeta_1 + \zeta_{\text{int}})(A_2 + B)}{\zeta_1\zeta_2 + \zeta_{\text{int}}(\zeta_1 + \zeta_2)} + \frac{\zeta_{\text{int}} + B(\theta)}{\zeta_1\zeta_2 + \zeta_{\text{int}}(\zeta_1 + \zeta_2)}$$

$$c = \frac{(A_1 + B)(A_2 + B) - B(\theta)^2}{\zeta_1\zeta_2 + \zeta_{\text{int}}(\zeta_1 + \zeta_2)}$$

where

$$A_1 = 2K_1(3 - 3X(\theta)), A_2 = 2K_2(3 - 3X(\theta)),$$
$$B = K_{\text{int}} + 6K^*_{\text{int}}, B(\theta) = K_{\text{int}} + 6K^*_{\text{int}}X(\theta),$$
$$X(\theta) = \frac{1}{3}(\cos(\theta_1) + \cos(\theta_2) + \cos(\theta_3))$$

In the model I the relaxation time is a complicated function of $\cos\theta_1$, $\cos\theta_2$, $\cos\theta_3$ (see Figure 3 and 4) therefore we present (for visual) this dependences, λ ($\cos\theta_1$, $\cos\theta_2$, $\cos\theta_3$), as a function of the definite ways in the multidimensional space (θ_1, θ_2, θ_3) where $\cos\theta_1 = \cos\theta_2 = \cos\theta_3$ (see Figure 3 and 4). In the model II spectra the inverse times $\lambda = 1/\tau$ is a simple function of parameter $X(\theta) = (1/3)(\cos\theta_1 + \cos\theta_2 + \cos\theta_3)$ (see Figure 5 and 6).

The relaxation spectra of the polymer networks can be characterized by the

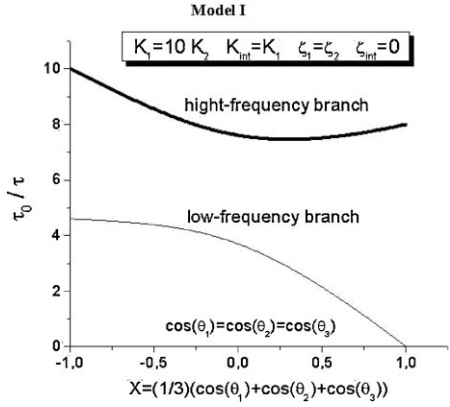

Figure 3.
The relaxation time spectra for model I at different parameters pointed in the Figure. Here $\tau_0 = \zeta_1/2K_1$, where ζ_1 and $2K_1$ are the parameters of the first network.

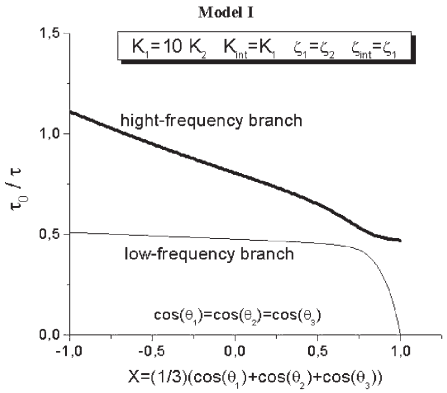

Figure 4.
The relaxation time spectra for model I at different parameters pointed in the Figure.

average values of the inverse relaxation times $\langle 1/\tau \rangle^{-1}$

$$\langle 1/\tau^- \rangle = (1/\pi)^3 \int_0^\pi \int_0^\pi \int_0^\pi \lambda(\theta_1, \theta_2, \theta_3) d\theta_1 d\theta_2 d\theta_3$$

and average times

$$\langle \tau^- \rangle = (1/\pi)^3 \int_0^\pi \int_0^\pi \int_0^\pi (1/\lambda(\theta_1, \theta_2, \theta_3)) d\theta_1 d\theta_2 d\theta_3$$

The inverse average time characterizes high-frequency dynamic behaviour of the network. The average time characterized low-frequency dynamic behaviour of the network. In contrast to one-dimensional

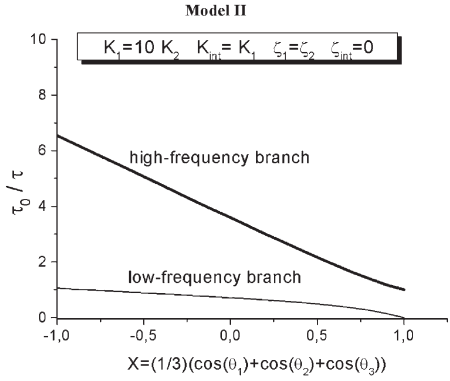

Figure 5.
The relaxation time spectrum of the model II at different parameters pointed in the Figure. Here $\tau_0 = \zeta_1/2K_1$.

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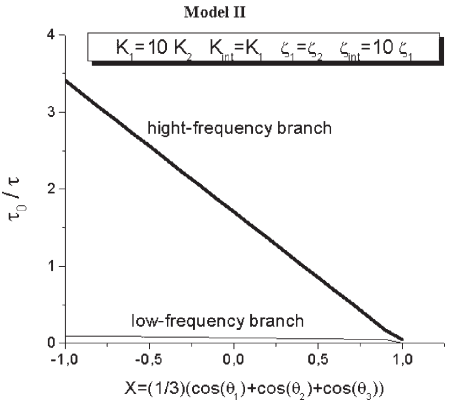

Figure 6.
The relaxation time spectrum of the model II at different parameters pointed in the Figure.

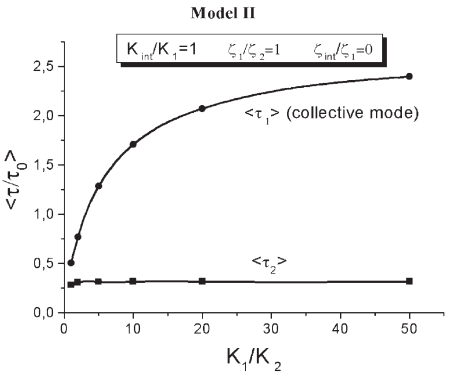

Figure 8.
Dependence of the average time of both branches of the relaxation spectrum on the parameter K_1/K_2 for the model II.

polymer systems (i.e. polymer chains) and two-dimensional polymer network systems the average time for tree-dimensional network is finite. It is due to a fast decrease of the density of the relaxation spectrum $L(\tau^-)$ in log-scale for high relaxation time $L(\tau^-) \sim \tau^{-3/2}$.[15] The analysis of the average $\langle\tau\rangle$ and inverse relaxation times $\langle 1/\tau\rangle^{-1}$ is restricted here by the investigation of these characteristics as the functions of the parameter only for the dynamic model II. The second model is more sensible to the interactions between networks. As it was shown in the previous paper of the authors devoted to the dynamics two similar interpenetrating networks the average and average inverse relaxation times of the branch describing collective motion were independent on the parameters of the interaction between networks.

The dependence of the average relaxation times $\langle\tau\rangle$ on the interaction parameter K_{int}/K_1 for the model II is presented in Figure 7. The increase of the interaction leads to the decrease of the relaxation times for both branches of the relaxation spectra. The average relaxation times of the branch of local (mutual) motions, τ_2, tend to 0 at $K_{int} \to \infty$

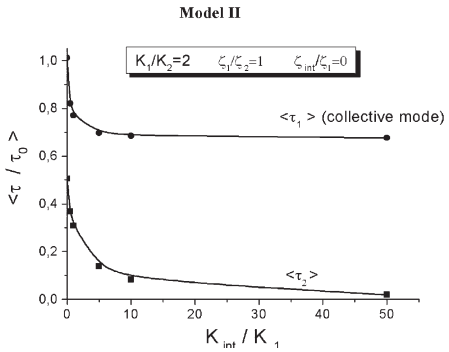

Figure 7.
Dependence of the average time of both branches of the relaxation spectrum on the parameter K_{int}/K_1 for the model II.

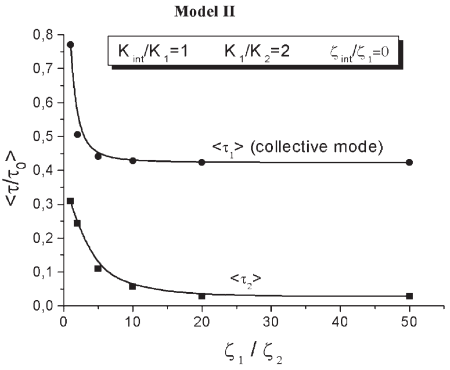

Figure 9.
Dependence of the average time of both branches of the relaxation spectrum on the parameter ζ_1/ζ_2 for the model II.

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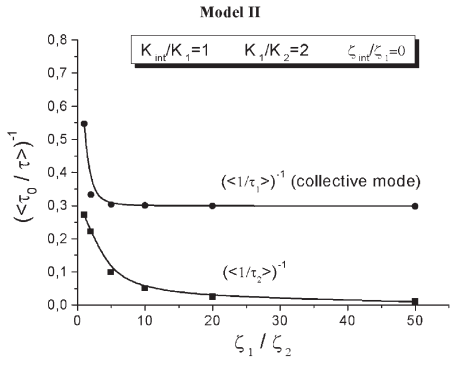

Figure 10.
Dependence of the average inverse time of both branches of the relaxation spectrum on the parameter ζ_1/ζ_2 for the model II.

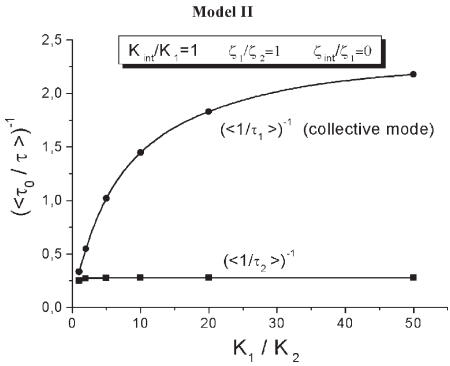

Figure 12.
Dependence of the average inverse time of the both branches of the relaxation spectrum for the model II on the parameter K_1/K_2.

($\langle\tau_2\rangle \sim 1/K_{int}$). The average time of specific collective network motion is characterized by finite asymptotic value at $K_{int} \to \infty$ (see Figure 7).

The dependence of $\langle\tau\rangle$ for both branches on the ratio K_1/K_2 at fixed K_{int} and ζ_1/ζ_2 is presented in Figure 8. The dependencies of the $\langle\tau\rangle$ on K_1/K_2 for both modes are different. The $\langle\tau\rangle$ for modes of local motion are practically constant. The $\langle\tau\rangle$ for collective mode is raising with raise K_1/K_2 and reaches the asymptotic value depending on parameter K_{int}/K_1. The theory also predicts that increase of the ratio ζ_1/ζ_2 leads to the decrease of the average relaxation times $\langle\tau\rangle$ (at fixed K_1/K_2, ζ_1, K_{int}) see Figure 9.

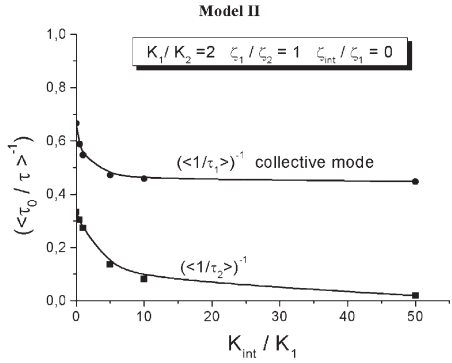

Figure 11.
Dependence of the average inverse time of the both branches of the relaxation spectrum for the model II on the parameter K_{int}/K_1.

The dependences of the average inverse relaxation time $\langle 1/\tau\rangle^{-1}$ are similar to these dependences for $\langle\tau\rangle$, see Figure 10–12 for both modes, because the relaxation spectrum of 3-D network is sufficiently narrow and the values $\langle 1/\tau\rangle^{-1}$ are close to $\langle\tau\rangle$.

Conclusion

1. The theory of the relaxation properties of similar and different interpenetrating polymer networks has been proposed considering the "long-scale" collective network relaxation processes. Interaction between networks is described (i) by the effective quasi-elastic potential depending on the entanglement between networks and (ii) by the interchain friction between networks.
2. Two types of the topology, i.e. of mutual disposition of the neighbouring junctions in the cubic model of every network, were considered. In model 1, a junction of a given network is presumably in the centre of the cell of other network. In model 2 the neighbouring junctions of different networks interact stronger than the next junctions.
3. Two branches of collective long-range motion exist. One branch is determined by correlated long-wave collective motion in both networks. The other branch is connected with relative displacements of

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neighbouring junctions of the two networks.

4. The behaviour of the average relaxation times $\langle\tau\rangle$ and average inverse relaxation times $\langle 1/\tau\rangle^{-1}$ as functions of the structures of the interpenetrating networks and of the parameters of the interactions between them was investigated. The relaxation spectra of the interpenetrating network will be useful for interpreting the frequency dependence of the dynamic modulus and other relaxation properties of these systems.

Acknowledgements: This work was carried out with the financial support of the Russian Foundation for Basic Research (project 08-03-00150-a).

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