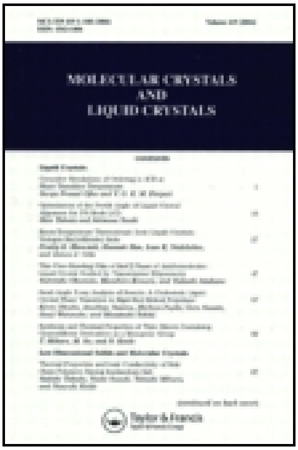

Molecular Crystals and Liquid Crystals

Hybrid Materials Based on Sequential Semi-Interpenetrating Polymer Networks

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Hybrid Materials Based on Sequential Semi-Interpenetrating Polymer Networks

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Peculiarities of formation kinetics of sequential semi-interpenetrating polymer networks based on crosslinked polyurethane with different cross-linking density and linear polystyrene, polybutylmethacrylate have been studied. Viscoelastic and thermophysical properties of sequential semi-IPNs were investigated by the method of dynamic mechanical analysis and differential scanning calorimetry. The experimental data show the dependence of the kinetic parameters of polymerization and relaxation properties on Mc.

Keywords: confinement; heat capacity jump; reaction kinetics; relaxation transitions; semi-interpenetrating polymer networks

INTRODUCTION

During last years a great interest is declared to the behavior of polymeric molecules in confined state [1,2]. Some situations are considered: properties of thin films at the interface with solid, behavior of solutions and polymers in thin pores. In all these cases the polymeric molecules are situated in a very restricted space whose dimensions are comparable to the size of macromolecules [3,4]. Restrictions imposed by the small volumes on the behavior of polymeric molecules are very important. The static and dynamic properties of confined fluids are of interest in many fields because of their importance in a variety of technological processes, including catalysis, chromatography etc. A considerable progress in understanding the dynamic and thermodynamic properties of molecular liquids in restricted geometry has been made in recent years [1–6]. In spite of

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this, up to now there were no investigations of the confinement effect on the reactions of macromolecules formation. The systems where such an effect may be expected are sequential semi-interpenetrating polymer networks (semi-IPN).

In such systems one of the components is synthesized in the pores of another network prepared preliminary. Such the systems were widely investigated [7–9]. However the most of the works was dedicated to the study of the mechanical, viscoelastic and other physical properties of the material. This method was used for increasing of the components compatibility of IPNs [7,10]. The processes of the phase separation proceeding (if any) in sequential IPNs were also not investigated in spite of the great deal of works dedicated to such a processes in simultaneous IPNs [11]. At the same time, the kinetics of the reaction of the formation of the second linear or cross-linked polymers in first networks was almost not studied; there are no data about the effect of the confined space on the reaction kinetics of the second polymer formed. The only exception are two works [12,13], where authors studied the kinetics of the polymerization of styrene and methyl methacrylate in beforehand prepared networks based on the methyl methacrylate copolymer with dimethacrylate tri-decaethyleneglycol and styrene with dimethacrylate ethylene glycol. It was found that a growth rate constant by polymerization of styrene and methyl methacrylate in the network coincides with the corresponding constants by polymerization of the same monomers in bulk. The termination constant is lower as compared with bulk polymerization, which is the reason of the fast appearance of the gel effect. In the same works the peculiarities of the molecular mass distribution of linear polymers formed in networks were analyzed [12,13].

Dynamic mechanical analysis (DMA) and differential scanning calorimetry (DSC) are the effective methods used for finding correlations between the structure and properties of polymer systems. From DMA dates the degree of segregation was calculated for estimation the level of microphase separation (or compatibility) in two-phase systems [14].

The aim of the present work was to establish the confinement effect on the reaction kinetics of linear polymers formed in the matrix of polyurethane networks and viscoelastic and thermophysical properties of sequential semi-IPNs.

EXPERIMENTAL

Semi-IPNs were produced by sequential method. 1st stage consisted of the synthesis of cross-linked polyurethane (PU) on the basis of poly(oxypropylene glycol) with molecular masses 500, 1000 and 2000 and triisocyanate adduct (2,4-,2,6 tolulene diisocyanate and trimethylol propane). Reaction was performed at 60°C up to full conversion of the functional groups (monitored by IR-spectroscopy). The catalyst concentration (dibutyltin laurate) was $1.4 \cdot 10^{-4}$ mol/l. Synthesized products were dried under vacuum up to constant weight. On the second staged the second component was introduced into the network by swelling it in monomer (butylmethacrylate (BMA) and styrene (St)). Initiator of radical polymerization of monomers (2,2′-azo-*bis*-isobutyronitrile) was dissolved in monomers before swelling. The amount of monomer introduced into the network by swelling was dependent on the cross-linking density; the identical conditions of reaction in each case were supported. Polymerization was performed at 60°C to full exhausting of double bonds. The initiator concentration was taken $2.5 \cdot 10^{-2}$, $5.0 \cdot 10^{-2}$ mol/l correspondingly for BMA, St. The amount of the second component in the network depends on the network density. In such a way semi-IPNs obtained using networks with different cross-linking density have various ratios of components. The amount of a monomer in the network was determined by the gravimetric method using the equation [15]:

$$\text{Component, } \% = \left[\frac{(m_{IPN} - m_{PU})}{m_{IPN}}\right] \cdot 100,$$

where component (%) is the amount of the second component in semi-IPN, m_{IPN} and m_{PU} are the masses of the specimens of semi-IPN and PU network. For each system the ratio of components in semi-IPN was estimated as an average value from 5 experiments (the measurement error no more 10%).

Table 1 presents the data on the equilibrium swelling degrees of PU networks with different M_c in BMA (q_{BMA}) and styrene (q_{St}).

TABLE 1 The Degree (q) and the Time (τ) of Equilibrium Swelling of PU in BMA and Styrene Dependence on M_c of the Network

		Buthylmethacrylate		Styrene	
Sample	M_c	q, %	τ, min	q, %	τ, min
PU-500	220	15	150	32	120
PU-1000	1170	70	134	93	75
PU-2000	5730	186	110	257	65

TABLE 2 Kinetic Parameters of Initial PBMA, PS Formation and in Semi-IPNs

Sample	Ratio, %	τ_a, min	α_a	W_{max}, min^{-1}	τ_{max}, min
PBMA	–	100	0.33	0.026	175
PU-500/PBMA	87/13	220	0.02	0.055	340
PU-1000/PBMA	59/41	90	0.03	0.072	220
PU-2000/PBMA	35/65	25	0.05	0.090	140
PS	–	590	0.48	0.021	840
PU-500/PS	76/24	60	0.05	0.023	210
PU-1000/PS	52/48	230	0.24	0.036	470
PU-2000/PS	28/72	540	0.45	0.045	760

These were found from the equation [15]:

$$q,\ \% = \left[\frac{(m_{swollen} - m_0)}{m_0}\right] \cdot 100,$$

where $m_{swollen}$ and m_0 are the masses of the swollen specimen and of initial one.

To characterize PU networks the molecular mass M_c of the chains between two junction points was determined using Flory-Rehner method. Values of M_c are presented in Table 1.

The kinetics of polymerization of monomers was studied at 60°C using differential calorimeter DAC-1-1A using the method described in detail in [16]. From the kinetic data the following parameters were determined: conversion of monomer at the onset of autoacceleration α_a, time of the onset of autoacceleration τ_a, maximum value of reduced

TABLE 3 Parameters of Relaxation Transitions and Degrees of Segregation for Sequential Semi-IPNs PU/PBMA and PU/PS

Composition, mass %	T_{g1}	T_{g2}	Max lg E''_1, MPa	Max lg E''_2, MPa	α
PU-2000	−30		1.70		
PU-1000	10		1.60		
PU-500	50		1.45		
PBMA		55	–	1.65	
PU-2000/PBMA 35/65	−30	50	1.10	1.50	0.22
PU-1000/PBMA 59/41	14	35	1.20	1.30	0.04
PU-500/PBMA 87/13	55		1.30		0
PS		100		1.65	
PU-2000/PS 28/72	50	100	1.50	1.62	0.34
PU-1000/PS 52/48	45	100	1.30	1.36	0.20
PU-500/PS 76/24	70		1.25		0

TABLE 4 Thermophysical Parameters of Relaxation Transitions of Initial PU and Semi-IPNs

Sample	Ratio PU/PBMA (PS), mass %	T_g, °C		ΔC_p	
		PU	PBMA (PS)	PU	PBMA (PS)
PU-500	100/0	19	–	0.80	–
PU-1000	100/0	−33	–	0.53	–
PU-2000	100/0	−60	–	0.62	–
PBMA	0/100	–	43	–	0.28
PS	0/100	–	79	–	0.40
PU-500/PBMA	87/13	29		0.51	
PU-1000/PBMA	59/41	35		0.30	
PU-2000/PBMA	35/65	37		0.27	
PU-500/PS	76/24	36		0.38	
PU-1000/PS	52/48	−11	90	0.24	0.24
PU-2000/PS	28/72	17	96	0.14	0.22

reaction rate W_{max} of polymer formation and the time τ_{max} of reaching W_{max}. The reduced reaction rate of PBMA and PS formation calculated from $W_{red} = V/[M]$, where $V = dM/dt$, M is the amount of monomer unreacted and t, the reaction time. The kinetic parameters are given in Table 2.

The study of viscoelastic properties was performed by the method DMA using a relaxometer with forced sinusoidal oscillations at a frequency of 100 Hz, in the temperature range from −70°C to 200°C, and at heating rate of 2°C/min. The glass transition temperature was determined according to the position of the maximum of loss modulus E''. The results are given in Table 3.

The study of relaxation transitions in initial PU and semi-IPNs was performed by the method DSC using a DSC-D differential scanning calorimeter at temperatures from −100 to 250°C and at heating rate of 2°C/min. The parameters of relaxation transitions are given in Table 4.

RESULTS AND DISCUSSION

Kinetic Investigations

Table 1 shows the characteristics of PU networks, which served as a matrix for the formation of semi-IPN. The increasing density of cross-links for PU based on POPG 500 (PU-500) and 1000 (PU-1000) may be explained by the formation of a network of strong physical (hydrogen) bonds, which contributes to the total network density. A more defective network is formed for PU based on POPG 2000

(PU-2000). It is connected with low reactivity of end OH-bond of POPG 2000. The equilibrium swelling of PU networks in monomers depends on the M_c of matrix, which determines various amounts of monomers in matrices with different M_c, i.e., the component ratios in semi-IPNs (Table 2).

Figure 1 shows the kinetic curves of the formation of pure PBMA (curve 1) and PBMA in matrix with different M_c (curves 2–4). It is seen that conversion of BMA at the beginning of autoacceleration (α_a) is one order lower in comparison with polymerization of pure BMA, being insignificantly increased with increasing M_c (Fig. 1, Table 2). The maximum value of reduced rate W_{red} formation of PBMA growths with increasing M_c. Simultaneously, the time of attainment of maximum W_{red} diminishes from 340 min for PU-500 to 140 min for PU-2000 (Fig. 2, Table 2).

As is known [17] in viscous media the role of diffusion processes in polymerization is very important. Diffusion may affect not only the chain termination but initiation reaction as well. Increasing viscosity of the media diminishes the constant of decomposition of the initiator and efficiency of initiation. The important role plays so called "cage" effects preventing the diffusion separation of a radical couple. It is probable, that by polymerization of BMA in PU-2000 diffusion limits only reaction of chain termination. Really, maximum value of W_{red} for BMA polymerization increases three times as compared with

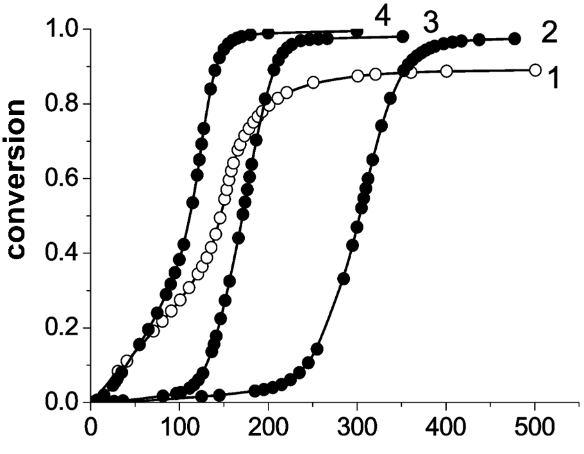

FIGURE 1 Kinetic curves of formation of initial PBMA (curve 1) and PBMA in PU matrix: PU-500 (curve 2), PU-1000 (curve 3), PU-2000 (curve 4).

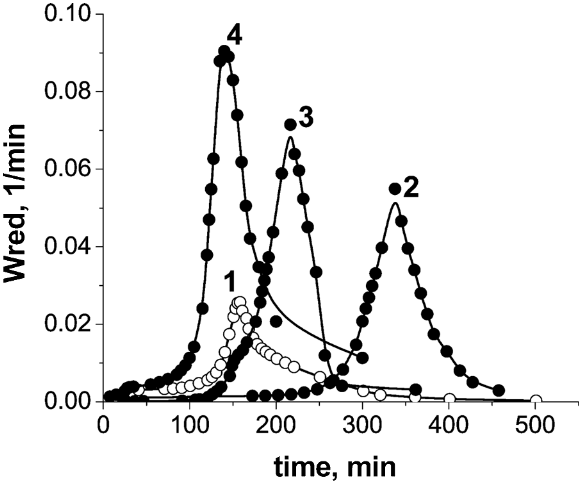

FIGURE 2 Dependences of reduced rate of polymerization W_{red} of initial PBMA (curve 1) and PBMA in PU matrix: PU-500 (curve 2), PU-1000 (curve 3), PU-2000 (curve 4).

polymerization of pure monomer. By polymerization of BMA in PU-500 the maximum of the time dependence of W_{red} broadens whereas the reaction rate diminishes. This effect may be related to essential decrease of the initiator decomposition rate and initiation efficiency. In this case the high density of the matrix network (4 times higher in comparison with theoretical one) impedes the outlet of radicals from the "cage" and diffusion may restrict not only the termination reaction but initiation. In [18] for polymerization of n-alkyl methacrylate it was shown that diminishing the termination constant is determined by lowering the free volume of the reaction media and by the appearance of topological entanglements leading to the formation of entanglement network. It was also established that by polymerization of styrene and MMA in beforehand prepared three-dimensional networks the termination constant is lower 3 orders as compared to bulk [12].

For styrene as the second component in semi-IPN the following regularities of the polymer formation in network cells were observed. From Figure 3 and Table 2 follows that styrene conversion at the point of autoacceleration and its time increase with increasing M_c. Values of α_a and τ_a are lower in comparison with polymerization of free monomer. The same effect was observed for BMA polymerization. The maximum value of W_{red} increases with increasing M_c as well like in BMA case but maximum is shifted to lower reaction times as compared with free styrene, as distinct from PBMA (Figs. 4 and 2).

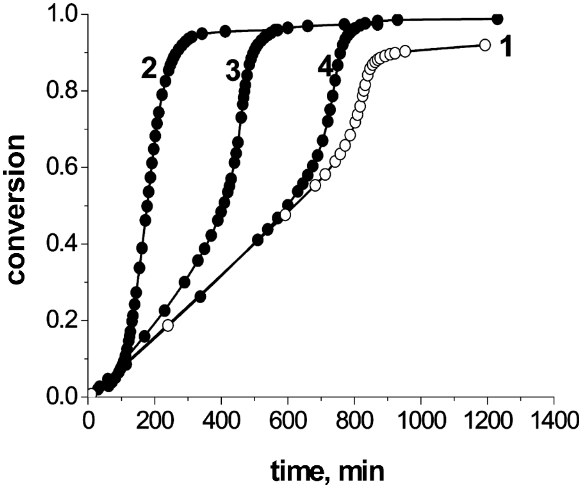

FIGURE 3 Kinetic curves of formation of initial PS (curve 1) and PS in PU matrix: PU-500 (curve 2), PU-1000 (curve 3), PU-2000 (curve 4).

With diminishing M_c there is observed the lowering of styrene amount in semi-IPNs and diminishing of maximum W_{red}. The time of the onset of autoacceleration and time of attainment of maximum

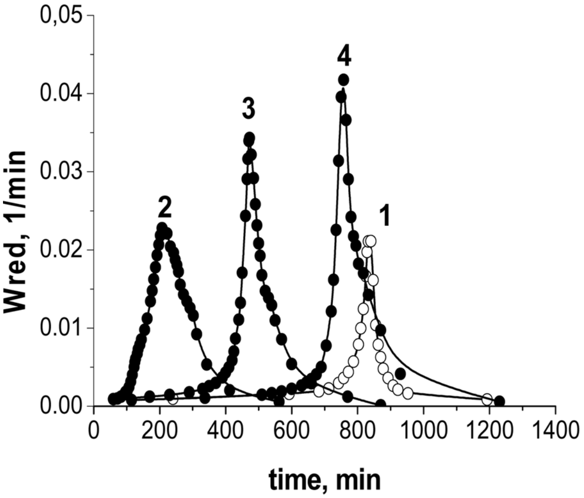

FIGURE 4 Dependences of reduced rate of polymerization W_{red} of initial PS (curve 1) and PS in PU matrix on time: PU-500 (curve 2), PU-1000 (curve 3), PU-2000 (curve 4).

W_{red} for PS in PU-500 is essentially lower as compared with pure styrene and styrene in PU-1000 and PU-2000. For this case it may be supposed that by polymerization of styrene in PU-500 the rate constant of diffusion controlled termination reaction sharply decreases, which leads to the prompt beginning of autoacceleration (60 min) as compared with pure styrene (590 min) and for PS formation in PU-1000 and PU-2000 (Table 2). In given case by PS formation, diffusion controls only termination reaction.

The experimental data confirm the expected dependence of the kinetics of polymerization of various monomers on the spacial restriction. We suppose that the differences in the reaction kinetics for monomers (BMA and styrene), which polymerization in various matrices are determined by the distinctions in the termination mechanism. Probably, the conditions of termination in confined space depend on the termination mechanism. The differences in reaction kinetics may be also connected to various dependence of the chain growth and termination of various monomers on the density of network, i.e., on the intranetwork space.

DMA-Measurements

Figure 5 demonstrates the temperature dependencies of the loss modulus E'' (a) and the elastic modulus E' (b) for networks PU-2000, PU-1000 and PU-500. The both values of the glass transition temperature and the elastic modulus in the rubber state (E_e) increase with decreasing MM of POPG of PU networks. According to the theory of rubber elasticity, the value of E_e is closely related to the average molecular mass between crosslinks (M_c), and as the E_e increases, the M_c decreases. These results have good agreement with calculated values M_c by Flory-Rehner method (Table 1). The both the increasing of T_g and the decreasing of M_c are evidence of the increasing of the crosslink density [19] of PU networks.

Figure 6 compares loss modulus vs. temperature plots of sequential semi-IPNs PU/PBMA with various compositions determed by PU crosslink densities (through changing of MM POPG). The results of their T_g are given in Table 3. The semi-IPNs PU-2000/PBMA 35/65 mass % and PU-1000/PBMA 59/41 mass % are two-phase polymer systems, as are indicated by the existence of two relaxation maxima (Figs. 6a,b). Information about phase miscibility was deduced from the number, location, and shape of the relaxation maximum. The inward shift of the maximum for PU-1000/PBMA 59/41 mass % (Fig. 6b) may be result of the improvement of compatibility of the semi-IPN components [20].

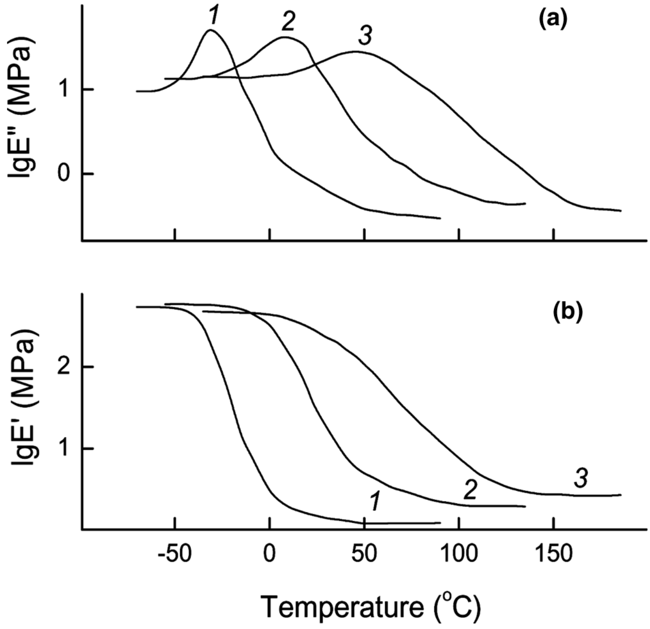

FIGURE 5 Temperature dependence of the loss modulus E″ **(a)** and elastic modulus E′ **(b)** of PU matrix: PU-2000 (curve 1), PU-1000 (curve 2) and PU-500 (curve 3).

For estimation the level heterogeneity in the studied systems were used the analytical expression for calculating the degree of segregation (α) in two-phase systems from the parameters of the relaxation maxima [14]. Parameter α is a relative value, which characterizes the level of microphase separation (or compatibility) of components in two-phase polymer system. For complete phase separation, the degree of segregation is $\alpha = 1$; for complete mixing, we have $\alpha = 0$. Values α for semi-IPNs are given in Table 3. Decreasing α for PU-1000/PBMA 59/41 mass % ($\alpha = 0.04$) compared with corresponding value for PU-2000/PBMA 35/65 mass % ($\alpha = 0.22$) points to significant enhancement the compatibility of components in sequential semi-IPNs PU/PBMA with increasing of the crosslink density of PU network. Figure 6c shows the one relaxation transition for sequential semi-IPN PU-500/PBMA 87/13 mass %. The existens of a single T_g for IPNs indicates evident compatibilization in the system [21].

Similar results were obtained for sequential semi-IPNs based on PU/PS. Figure 7 shows E″ vs. temperature plots for semi-IPNs PU/PS

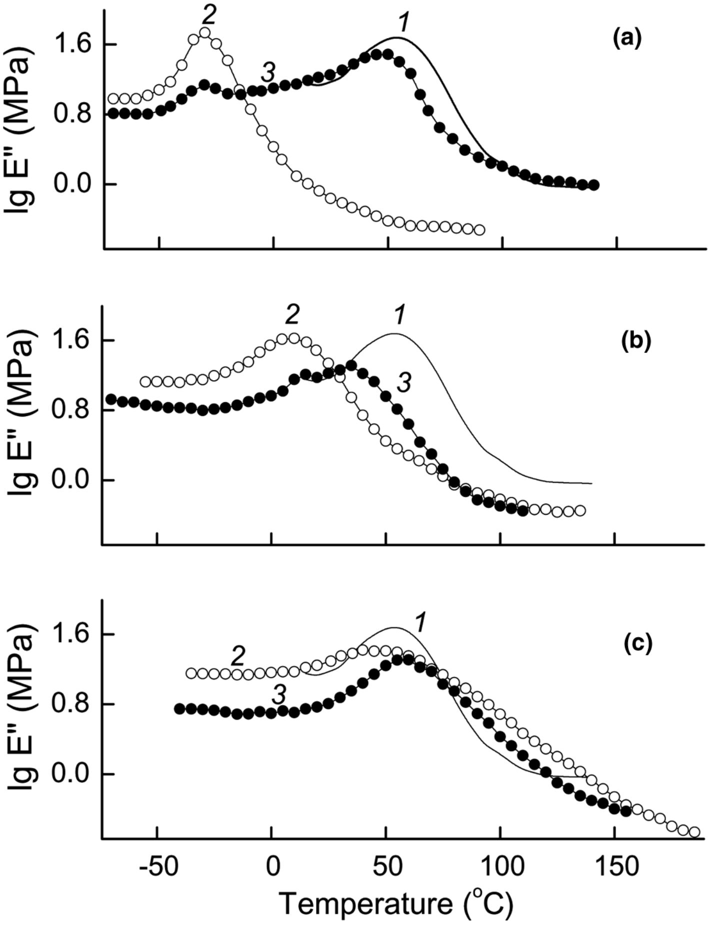

FIGURE 6 Temperature dependence of the loss modulus E″ of PBMA (curve 1), PU-2000 (curve 2**a**), PU-1000 (curve 2**b**), PU-500 (curve 2**c**) and semi-IPNs PU-2000/PBMA 35/65 mass % (curve 3**a**), PU-1000/PBMA 59/41 mass % (curve 3**b**) PU-500/PBMA 87/13 mass % (curve 3**c**).

PS (curve 3) with various compositions (a-c) in comparison with the curves of the individual PS (curve 1) and PU networks (curve 2). Obviously, the curves indicate phase-separated materials for both semi-IPNs PU-2000/PS 28/72 mass % (Fig. 7a) and PU-1000/PS 52/48 mass % (Fig. 7b). However, decreasing the value α for PU-1000/PS 52/48 mass % ($\alpha = 0.20$) compared with corresponding value for PU-2000/PS 28/72 mass % ($\alpha = 0.34$) were observed (Table 3).

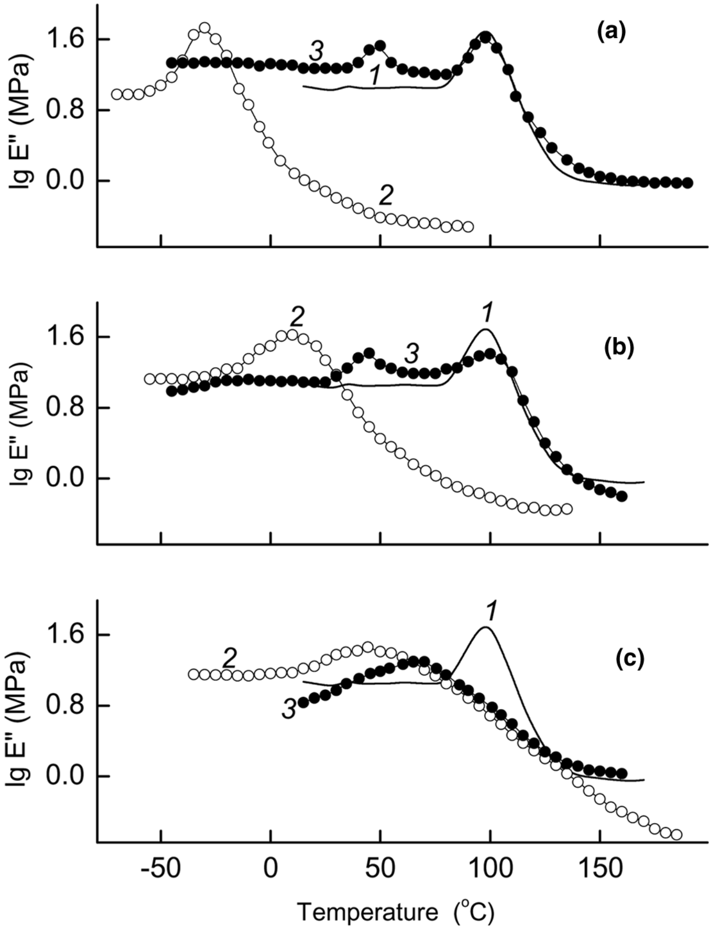

FIGURE 7 Temperature dependence of the loss modulus E″ of PS (curve 1), PU-2000 (curve 2**a**), PU-1000 (curve 2**b**), PU-500 (curve 2**c**) and semi-IPNs PU-2000/PS 28/72 mass % (curve 3**a**), PU-1000/PS 52/48 mass % (curve 3**b**), PU-500/PS 72/64 mass % (curve 3**c**).

It may be result of enhancement the compatibility of components in the system. With decreasing of value M_c of PU network (PU-500) only single broad relaxation maximum is present for semi-IPN PU-500/PS 76/24 mass %. (Fig. 7c).

For both semi-IPNs PU-500/PS and PU-500/PBMA the present of single relaxation maximum may be due to either to the formation of a thermodynamically compatible single-phase system or to a change in its morphology as a result of increased component compatibility and decreased dimensions of the resultant domains [20].

DSC – Measurements

The DSC data are given in Table 4 for the initial specimens of PU networks with different MM POPG are observed a change of heat capacity increments in the region glass temperature (from 0.53 to 0.80), which may be related to the oligoether component corresponding to PU network. The jump of heat capacity of PU networks decrease with increasing of MM POPG. It is related with growth of molecular mobility of segments of flexiblechain fragment. For PS the high-temperature jump of heat capacity at PS glass transition (79°C) and jump of heat capacity at PBMA glass transition (43°C) are observed.

The results of DSC measurements obtained for semi-IPNs composition PU/PBMA based on PU-500, PU-1000, PU-2000 show only one a jump of heat capacity in region glass temperature. The jump of heat capacity decrease with increasing of value M_c and respecting initial PU-component. With increasing the content of the rigid PBMA component in system value of ΔC_p decreases and value of T_g increases. The change of jump of heat capacity from 0,51 to 0,27 (Table 4) indicate on increasing the degree of system ordering at increasing of the intranetwork space (M_c PU networks).

The Experimental data show that one jump of heat capacity in region glass temperature for semi-IPNS based on PU-500/PS and two jumps of heat capacity for PU-1000/PS and PU-2000/PS are observed. With increasing of value M_c of PU network structure of sequential semi-IPNs changes from one-phase to two-phase polymeric systems (Table 4). Consequently, DSC studies show that polymerization of styrene in confined geometries lead to formation of semi-IPNs and these IPNs are two-phase polymeric systems with different levels of heterogeneity.

CONCLUSIONS

The investigation of the confinement effect on the reaction kinetics polymer formed in the matrix PU network and viscoelastic and thermophysical properties of sequential semi-IPNs allows to establish following regularities. It has been shown that with diminishing M_c of PU matrix there is observed the lowering of monomers (BMA and styrene) amount in semi-IPNs. It is established that reaction kinetics strongly depends on M_c of PU matrix. For styrene polymerization the main kinetic parameters ($\alpha_{a,}$ τ_a, W_{max} and τ_{max}) depend in symbate way on the

M_c, whereas for BMA polymerization there is direct correlation between α_a, W_{max} and M_c and reverse correlation between M_c and τ_a and τ_{max}. The differences in reaction kinetics may be also connected to various dependence of the chain growth and termination of various monomers on the density of network, i.e., on the intranetwork space.

The study of dynamic mechanical and thermophysical properties of sequential semi-IPNs show that with diminishing of M_c of PU matrix there is observed the enhancement components compatibility in sequential semi-IPNs as result of increasing of number of topological entanglements. By varying the crosslink density (M_c) in PU matrix, i.e. internetwork space in semi-IPN, it is possible to realize a purposeful regulation the structure of materials in wide range from two-phase system to one-phase polymer system practically.

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