Solid-State Relaxation Properties of Poly(α-methyl-α-n-propyl-β-propiolactone)/Poly(vinyl chloride) Miscible Blends

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ABSTRACT: Stress relaxation measurements were made at different temperatures on poly(α-methyl-α-n-propyl-β-propiolactone) (PMPPL), poly(vinyl chloride) (PVC), and PMPPL/PVC miscible blends in the linear viscoelastic region. Stress relaxation master curves were constructed for each blend at a reference temperature of $T_g - 25$. Time-temperature superposition was satisfactory, indicating that these mixtures are thermorheologically simple materials and suggesting their homogeneity. The stress relaxation master curves of the blends are intermediate between those of PMPPL and PVC. At short times, the influence of PMPPL is predominant. At longer times, the rapid decrease in modulus is similar to that seen in PVC, although this decrease is stopped by crystallinity in blends where PMPPL can crystallize. The same shift factor-temperature dependence was found for PMPPL, PVC, and their blends.

It is generally recognized that immiscible polymer/polymer blends form multiphase materials and exhibit poor mechanical properties as a result of poor adhesion between phases.[1,2] Mechanical properties of immiscible blends normally fall below those predicted by the rule of mixture[3]

$$M = V_1M_1 + V_2M_2 \qquad (1)$$

where V represents the volume fraction of polymer 1 or 2 and M the considered property, i.e., modulus, tensile strength, or toughness.

Miscible polymer/polymer blends form single-phase materials as a result of the presence of a specific interaction between two polymers.[4,5] It is expected that the mechanical properties of miscible blends will follow eq 1[3,6] or will be greater at particular blend compositions than the corresponding properties of either polymer in the unblended state. Indeed, such synergism in modulus and/or tensile strength has been observed in several instances.[7-16] Specific interactions between blend components cause volume contractions on mixing and a loss in the free volume of the blend.[9,14,15] This loss in free volume leads to higher modulus and strength values than predicted by eq 1.

Of course, semimiscible blends can also be found. They represent an intermediate situation in terms of phase dispersion between those of miscible and immiscible blends. Semimiscible blends also show separate phases. However, under proper phase dispersion and mixing conditions, semimiscible blends often exhibit mechanical properties that follow closely eq 1 or even show some synergism in modulus.[17-19]

The factors giving rise to synergism in stress-strain properties of miscible and semimiscible blends, as discussed above, should also influence all viscoelastic properties of these materials, although it has never been verified. In this paper, a study of the relaxation properties of miscible polymer/polymer blends is presented. For this study, we needed a mixture for which the miscibility was clearly established and a mixture for which mechanical properties could be measured at any composition (very fragile films cannot be easily studied by stress relaxation methods). In addition, we preferred amorphous blends, or at least blends where the degree of crystallinity was not too high in order to simplify the interpretation of the results, and blends having their glass transition temperatures, T_g, not too far above room temperature in order to simplify the measurements. We have then selected PMPPL/PVC blends, where PMPPL is poly(α-methyl-α-n-propyl-β-propiolactone) and PVC is poly(vinyl chloride). This mixture satisfies the above criteria.[20]

Table I
Glass Transition Temperature T_g, Melting Temperature T_m, Density, Degree of Crystallinity, and Enthalpy of Fusion of PVC, PMPPL, and Their Blends[a]

wt % PMPPL	T_g, K	T_m, K	density, g cm^{-3}	enthalpy of fusion, J g^{-1}	degree of crystallinity, %
0	337.5		1.39	0	0
25[b]	316.0		1.30	0	0
50	305.5	350	1.20	1	1
75[b]	291.0	360	1.16	12	13
100	275.0	370	1.07	27	25

[a] Reference 20. [b] Values interpolated from the data of ref 20.

It is the purpose of this paper to present stress relaxation measurements at different temperatures above and below T_g of PMPPL/PVC blends. Stress relaxation master curves will be constructed and the validity of the time-temperature superpositon principle will be verified.

Experimental Section

Details about the origin of the PMPPL and PVC samples used in this study can be found in ref 20. Let us just recall the structural formula of PMPPL, which is $-[CH_2C(CH_3)(C_3H_7)COO]-_n$.

Blends were prepared by casting films from tetrahydrofuran solutions. Solvent evaporation was conducted at room temperature. The resulting films were dried under vacuum until they reached constant weight.

PMPPL/PVC blends were previously analyzed by calorimetry[20] and a dynamic mechanical method.[21] Both measurements indicated miscibility, i.e., extensive mixing between PMPPL and PVC, since a single glass transition temperature was observed at any composition. The main characteristics of the blends and homopolymers are given in Table I. It can be seen that the 75/25 blend is semicrystalline. In other words, PMPPL crystallizes in this blend and the PMPPL crystals do not include PVC. The amorphous phase is made of PVC and of the remaining noncrystalline PMPPL.

All the test specimens were annealed at 320 K for 1 week before the mechanical measurements. An Instron tester, table model 1130, was used to obtain stress-strain curves and for relaxation measurements. Speed of testing was 0.05 cm/min. The temperature was regulated by an Instron environmental chamber and measured with a digital thermometer whose copper-constantan thermocouple was placed close to the sample. At higher temperatures, the temperature stability was of the order of 0.1 K.

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Table II
Tensile Properties of PVC, PMPPL, and Their Blends at 298 and T_g − 25 K[a]

	T = 298 K					T_0 = T_g − 25			
wt % PMPPL in blend	Young's modulus, MPa	yield stress, MPa	elongation at yield, %	ultimate stress, MPa	ultimate elongation, %	T_0, K	Young's modulus, MPa	ultimate stress, MPa	ultimate elongation, %
0	1800			33	1.7	312	1150	28	6.0
25	1195			24	4.3	293	1500	30	3.3
50	664	14.6	5.8	18	126	282	1600	22	6.7
75	204	7.7	16	18.2	587	266	1650	33	4.8
100	130	7.2	9.6	14.3	648	249	1850	38.	

[a] The parameters reported in this table are averages of four or five measurements. They do not correspond exactly to the curves shown in Figure 1, which are representative examples.

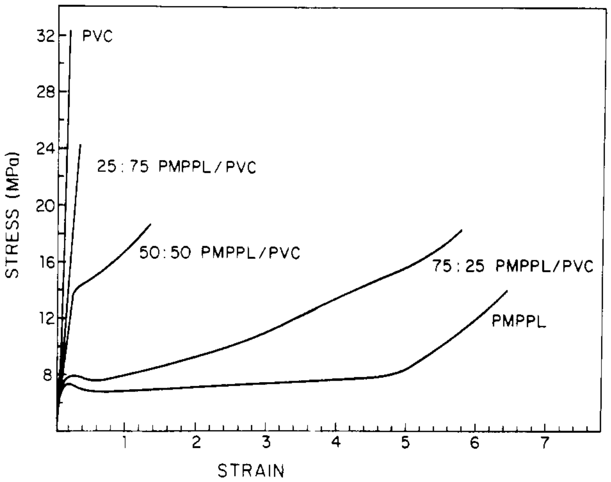

Figure 1. Representative stress–strain curves of PMPPL/PVC blends at 298 K.

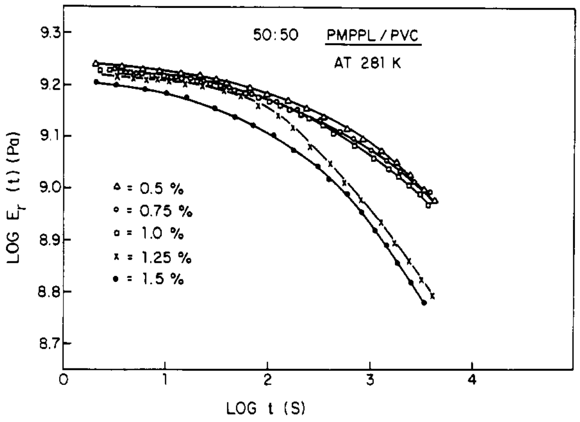

Figure 2. Stress relaxation curves at different strain levels for a 50/50 PMPPL/PVC blend at 281 K.

At lower temperatures, i.e., below 260 K, the temperature stability was of the order of 0.5 K.

Each test specimen was hung freely from the upper jaw of the Instron tester at the measuring temperature for 1 h before the relaxation measurements were made. The tensile strain was always kept within the limits of linear viscoelasticity that was determined experimentally. All series of measurements were made starting from room temperature to the highest measuring temperature and then from room temperature to the lowest measuring temperature. Calibration of the Instron tester was done before each series of measurements. This testing procedure yielded reproducible results.

Results and Discussion

1. Stress–Strain Behavior. The stress–strain curves of PMPPL, PVC, and PMPPL/PVC blends at 298 K are shown in Figure 1. As expected, PVC undergoes a brittle fracture at an elongation of 1.7%. In constrast, PMPPL shows a ductile behavior with a yield point and an ultimate elongation of 648%. Cornibert et al. reported similar results.[22]

PMPPL/PVC blends exhibit intermediate properties. The 75/25 PMPPL/PVC blend behaves similarly to PMPPL except that its stress softening region is reduced to elongations of less than 100% and that it is followed by a broad stress hardening region. In both cases, yielding and necking occur. The 25/75 blend behaves similarly to PVC with, however, lower modulus and tensile strength. The 50/50 blend initially behaves like PVC but it subsequently shows a slower increase of stress with strain which is similar to that observed for the 75/25 blend and PMPPL before break. This is a first indication of the dual behavior of the 50/50 blend at short and long times, which will be seen recurrently in this paper.

Table II gives a list of the stress–strain parameters calculated for PMPPL/PVC blends. If the experimental values of modulus are compared with those calculated from eq 1, it is found that the experimental values are much lower than those calculated except for the 25/75 blend, where agreement is found within experimental error. This surprising result, in view of those cited in ref 7–16, is due to the fact that the temperature of measurement of stress–strain properties was above T_g for PMPPL and the 75/25 blend and close to T_g for the 50/50 blend. Equation 1 is based on the assumption that the mixture is homogeneous and it does not take into account that some measurements were made below T_g whereas others were made above or close to T_g. In order to verify the validity of this explanation, we made stress–strain measurements at a temperature of T_g − 25 for all samples. The results are reported in Table II and they follow eq 1 within experimental error. No synergism could be observed.

Ultimate stresses of PVC, PMPPL, and their blends measured at 298 and T_g − 25 K are also shown in Table II. Ultimate stresses are much less sensitive to the temperature of measurement relative to T_g than the moduli; therefore, both series of measurements follow closely eq 1. However, no synergism was observed.

2. Linear Viscoelastic Region. Before stress relaxation measurements are made, the linear viscoelastic region of each specimen must be determined. In this study, two methods were used for that purpose: the hysteresis loop method[23] and the relaxation method.[24] In the former method, hysteresis loops are generated from very low strains to higher strains. The linear viscoelastic limit is found at the strain where permanent set appears. In the latter method, stress modulus, $E_r(t)$, is determined as a function of time t at a given temperature for different strain values. In the linear viscoelastic region, $E_r(t)$ is independent of strain. An example of the latter method applied to a 50/50 PMPPL/PVC blend is shown in Figure 2. Within experimental error, $E_r(t)$ is independent of strain for strain values equal to or lower than 1.0%. At

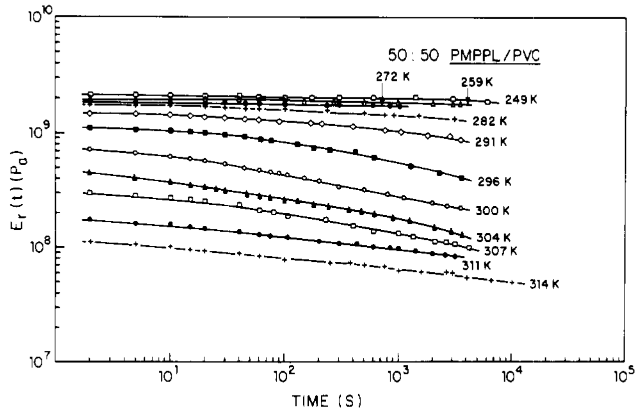

Figure 3. Stress relaxation curves of a 50/50 PMPPL/PVC blend.

1.5% elongation, the nonlinear viscoelastic region is reached at any time while at 1.25% elongation, the linear viscoelastic behavior is found at $t < 100$ s but the nonlinear viscoelastic behavior is observed at $t > 100$ s. This dependence of the linear viscoelastic limit upon time is generally found in high polymers.[25,26]

For PMPPL, previous measurements indicated a linear viscoelastic limit at 2.5% by the two methods cited above.[27] For PVC, measurements conducted at 305 and 283 K indicated a linear viscoelastic limit at 1.3% by the relaxation method at short times and the hysteresis loop method and at 1.0% by the relaxation method at long times.[28] These values agree with those reported by Jamshid.[29]

In view of the results cited above and since the linear viscoelastic limit is found at higher strain values at temperatures higher than T_g,[25,26] all stress relaxation data reported in this study obtained at a strain level of 0.6%, well within the linear viscoelastic limit.

3. Stress Relaxation Behavior. The stress relaxation behavior of PMPPL at temperatures between 222 and 309 K has been published in ref 27. It was shown that a stress relaxation master curve can be constructed for this polymer.

In this study, stress relaxation measurements of PVC and of PMPPL/PVC blends were made. A typical series of data is shown in Figure 3 for the 50/50 blend. At low temperatures, the relaxation modulus is of the order of $10^{9.1}$ Pa and relatively time insensitive, indicating a glassy polymer. At higher temperatures, the relaxation modulus changes rapidly with time from a value of about 10^9 Pa to one of about 10^8 Pa. This is the transition region. At high temperatures, the modulus changes less rapidly with time and has a value of about 10^8 Pa. This is the plateau region. Similar data for PVC and PMPPL/PVC blends of 25/75 and 75/25 compositions can be found in ref 28.

Using Ferry's reduction method,[30] we reduced all the results to an arbitrary temperature T_0, whose value was chosen as $T_0 = T_g - 25$. We neglected a correction for thermal expansion and for temperature variation of the elastic modulus due to rubber elasticity. We assumed $T_0\rho_0/T\rho$ to be unity, ρ_0 and ρ being the densities of the specimen at temperatures T_0 and T, respectively. For the 50/50 PMPPL/PVC blend, we composed the smooth stress relaxation master curve shown in Figure 4. All the relaxation curves satisfactorily superposed by sliding them along the time axis only, using an experimental shift factor a_T at each temperature T. Only a few points deviate from the master curve and these deviations are believed to be due to experimental error.

At short times ($\log t < 2$), the modulus values are typical of those usually found in the glassy region. At intermediate times ($2 < \log t < 12$), the decrease in modulus is due to the glass transition. At long times ($\log t > 12$), the decrease in modulus becomes less rapid. A constant value should eventually be reached, corresponding to the plateau region, although it is not observed in Figure 4 due to the limited time interval covered (measurements at higher temperatures could hardly be made).

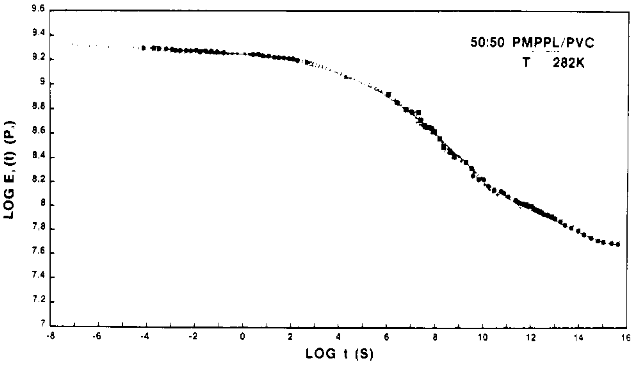

Figure 4. Stress relaxation master curve of a 50/50 PMPPL/PVC blend at a reference temperature of 282 K.

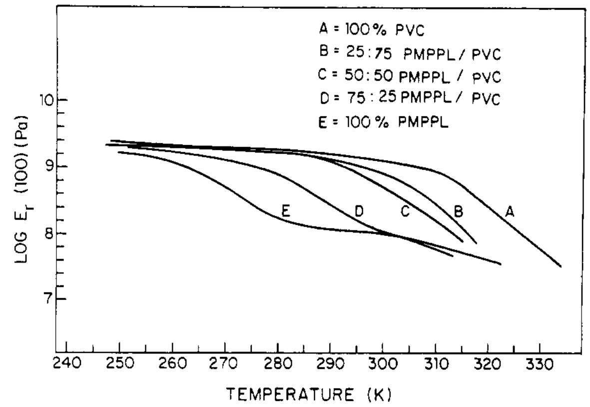

Figure 5. Stress relaxation modulus curves vs. temperature, measured at 100 s, of PMPPL/PVC blends and homopolymers.

Similar stress relaxation master curves, at $T_0 = T_g - 25$, have been obtained for PMPPL (using ref 27 original data), PVC, and 25/75 and 75/25 PMPPL/PVC composition data. The detailed master curves are given ref 28. In all cases, satisfactory time–temperature superposition has been obtained.

In the linear viscoelastic region, polymers that obey the time–temperature superposition principle are called thermorheologically simple materials whereas those that do not are called thermorheogically complex materials.[30,31] Normally, homogeneous materials are thermorheologically simple while heterogeneous materials, i.e., block copolymers, immiscible blends, and ionomers, are thermorheologically complex.[30-35] The satisfactory superposition of the stress relaxation curves of PMPPL/PVC blends at T_0 indicates that these blends are thermorheologically simple materials and suggests that they form homogeneous mixtures.

Discussion and Conclusion

Figure 5 shows the stress relaxation modulus, E_r, of PMPPL, PVC, and their blends, measured at 100 s, as a function of temperature. These curves were drawn from the data shown in Figure 3 and similar ones.[28] It is seen clearly that the transition region is shifted to higher temperatures with increasing PVC concentrations. These results confirm the calorimetric[20] and dynamic mechanical[21] measurements. They indicate the miscibility of the

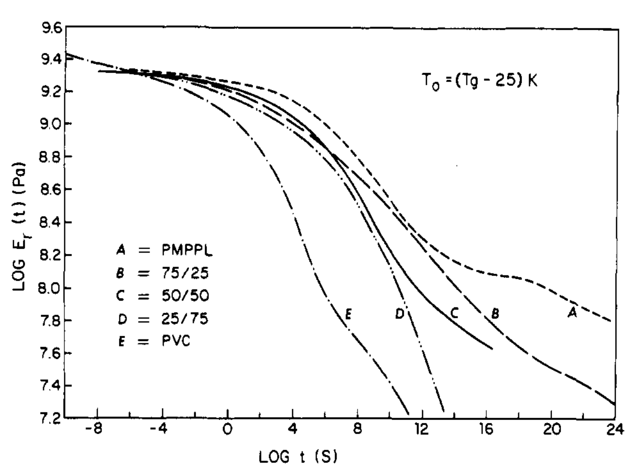

Figure 6. Comparison of the master curves of PMPPL/PVC blends and homopolymers at $T_0 = T_g - 25$.

PMPPL/PVC blends, i.e., extensive mixing of the two polymers.

Figure 6 shows the stress relaxation master curves of PMPPL, PVC, and PMPPL/PVC blends at a reference temperature $T_0 = T_g - 25$. A comparison of the different master curves becomes possible since the effect of T_g on the relaxation modulus is equal for all mixtures.

It is seen that the position and the shape of the master curves change systematically with blend composition. The observed behavior of PVC is that expected for a completely amorphous linear polymer where a rapid decrease of modulus from the glassy to the plateau region occurs. The behavior of the 25/75 PMPPL/PVC blend is different from that of PVC: the shape of the master curve remains similar but its position has been shifted to longer times despite the fact that both curves are plotted at T_g – 25. This result indicates that within the 10^0–10^8 s interval, the relaxation behavior of the 25/75 blend is influenced significantly by PMPPL. In this particular time interval, the relaxation modulus of the 25/75 blend is very close to that observed for PMPPL. It is only at times longer than 10^8 s that the rapid drop of modulus expected for an amorphous polymer occurs.

For PMPPL, the stress relaxation master curve at T_g – 25 resembles that of a semicrystalline polymer whose degree of crystallinity is low.[3,30] The glassy region where the modulus is almost constant is followed by a well-defined transition region, which is followed by a plateau region where the drop of modulus becomes less rapid. The plateau is due to the crystalline part of the polymer and it was suggested before that it corresponds to the thermal expansion of its crystalline portion followed by the loosening of the entanglements of its amorphous portion.[27] The behavior of the 75/25 PMPPL/PVC blend is very similar to that of PMPPL, except that the plateau is not as well defined due to its lower degree of crystallinity (Table I).

The behavior of the 50/50 PMPPL/PVC blend is intermediate between those of PMPPL and PVC. At short times ($t < 10^8$ s), it follows closely the PMPPL and the 75/25 PMPPL/PVC curves. At longer times, $E_r(t)$ drops rapidly and follows the 25/75 composition curve. And then, unexpectedly, at 10^{12} s, it forms the onset of a plateau, similarly to PMPPL and to the 75/25 PMPPL/PVC blend. This latter portion of the master curve is due to the small amount of crystallinity that remains in this blend.

In summary, all PMPPL/PVC stress relaxation master curves at T_g – 25 are deeply influenced by the presence of PMPPL. At short and intermediate times ($t < 10^{12}$ s), they follow closely the PMPPL curve and are shifted by roughly 4 decades of time as compared to the PVC curve. At long times, blends where PMPPL crystallized show the plateau seen for PMPPL even if only a small amount of crystallinity is present. Blends that are completely amorphous exhibit a rapid drop in modulus like PVC.

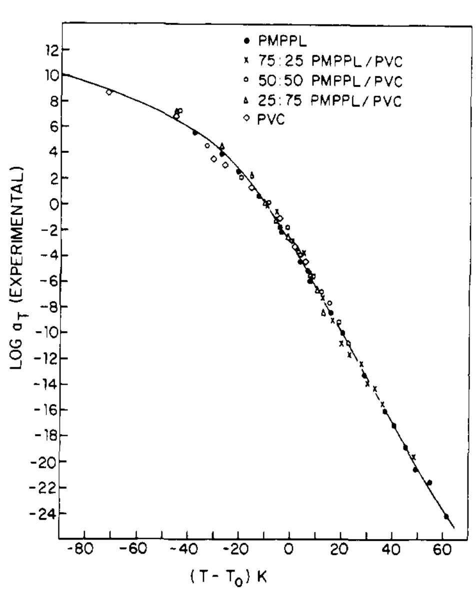

Figure 7. Shift factors vs. $T - T_0$ of PMPPL/PVC blends.

Hardly any data have appeared in the literature on the time–temperature superposition of miscible polymer/polymer blends. However, Cohen and Ramos examined the viscoelastic properties of polybutadiene/polyisoprene diblock copolymers that were homogeneous;[36] i.e., a single T_g was observed at each composition at a temperature intermediate between those found for the T_g's of the parent homopolymers. They observed linear changes in the storage and loss moduli in the rubbery region (the glassy and transition regions were not studied), which is in qualitative agreement with the data of Figure 6.

The experimental shift factors a_T for all specimens investigated were determined when constructing stress relaxation master curves.[28] They are reported in Figure 7 as a function of $T - T_0$. It is remarkable that a single curve is obtained. It indicates that despite different stress relaxation master curves, PMPPL, PVC, and PMPPL/PVC blends exhibit the same temperature dependence of their shift factors, independent of the amount of PMPPL of the blend. Figure 7 can then be considered as a shift factor master curve and the stress relaxation master curves of PMPPL/PVC blends of any composition can be constructed at T_0 using these experimental shift factors. In other words, assuming that this result is general for miscible polymer/polymer blends made of two amorphous polymers A and B or made of an amorphous polymer A and of a semicrystalline polymer B of low degree of crystallinity, the shift factors of A and B can be determined at T_0 and used to construct the master curves of any miscible A/B blends. A similar result was obtained for homogeneous block copolymers.[36] In the latter case, since the analysis was made in the rubbery and in the flow zones, the Williams–Landel–Ferry (WLF) equation (eq 2) satisfactorily expressed the results. In the present case, the analysis was done in the glassy, transition, and rubbery zones. The WLF equation does not apply below T_g and consequently it cannot be used to analyze our data.[27] An empirical function, shown in Figure 7, must be used instead. The occurrence of a shift factor master curve does not prove the homogeneity of PMPPL/PVC blends but

Table III
Constants C_1° and C_2° at a Reference Temperature T_0, the Fractional Free Volume f_0, and Its Thermal Expansion Coefficient α_f for Blends and Homopolymers

wt % PMPPL	C_1°	C_2°, K	f_0	α_f, K^{-1}	T_0, K
0	14.2	63	0.030	4.7×10^{-4}	312
25	15.5	65	0.028	4.3×10^{-4}	293
50	14.4	74	0.030	4.05×10^{-4}	282
75	13.3	67	0.032	4.7×10^{-4}	266
100	16.6	107	0.026	2.4×10^{-4}	249

it certainly leads to its suggestion. The WLF equation reads[30]

$$\log a_T = -\frac{C_1^\circ(T - T_0)}{C_2^\circ + (T - T_0)} \tag{2}$$

where C_1° and C_2° are two constants, which are now known not to be "universal", a_T is the shift factor at temperature T, and T_0 is the reference temperature. If eq 2 is applied to PMPPL/PVC data at temperatures above T_0 (eq 2 applies well above T_g and reasonably well between T_0 and T_g), then using Ferry's method,[30] one can calculate the constants C_1° and C_2°. They are reported in Table III along with the fractional free volume f_0 at T_0 and the thermal expansion coefficient αf since[30]

$$f_0 = 1/2.303C_1^\circ \tag{3}$$

and

$$\alpha f = f_0/C_2^\circ \tag{4}$$

All values of f_0 reported in Table III are close to 0.030, a value that is within the range 0.020–0.030 often seen.[30] All values of αf but one are close to a value of 4.8×10^{-4} K^{-1} proposed from the free volume theory.[30] The large C_2° and small αf values found for PMPPL are unexplained.

From the point of view of a scientist interested in the miscibility of polymer/polymer blends, the results presented in this study certainly indicate extensive mixing between the two polymers. Relaxation measurements indicate a single T_g at each composition without any indication of phase separation and without any significant broadening of the transition zone. Moreover, stress relaxation master curves can be constructed at any composition and the blends exhibit the same temperature dependence of their shift factors. PMPPL/PVC miscible blends are then proved to be thermorheologically simple materials and essentially homogeneous. It is, however, not known if this homogeneity extends to the molecular level.

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Registry No. PVC, 9002-86-2; PMPPL, 25188-77-6; PMPPL (repeating unit), 25265-42-3.

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