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to develop an extensive theory of the dynamics of polymer melts for the most usual chain lengths and concentration regimes.

Even if reptation does not occur in melts at all, nevertheless the reptation model would be important for many polymer problems, such as short chains in a melt of very long chains or the problem of a single mobile chain in a network. Therefore even the data of the moving chain in the frozen environment are carefully analyzed. It turns out that the available theories of $S_n(k,t)$ and $g(t)$[10,11,13] have a rather qualitative character. It is necessary to find a more quantitative theory, especially for the connections between the two different scaling formulations ($x \propto k^2t^{1/2}$ or $x \propto k^2t^{1/4}$) and for the full crossover from Rouse-like to reptative behavior.

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Polymer–Polymer Diffusion in Melts

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ABSTRACT: The mutual diffusion coefficient for a miscible binary polymer mixture is strongly dependent on the composition of the mixture. This leads to very unusual concentration profiles. For two pure A and B blocks in contact at time $t = 0$, we predict a concentration profile $\phi_t(x)$ linear in the spatial coordinate x. The region of mixing should not have a diffuse tail, but has a finite size $2(D_0t)^{1/2}$. These features are attenuated if we start not from pure A and B but from an A-rich sample against a B-rich sample.

I. Introduction

Compatible polymers are largely studied because of the mechanical properties offered by blends of two (A and B) polymers. It is difficult to find two polymers that are compatible on a microscopic scale. In the liquid state, the free energy F of the A–B mixture is well described by Flory–Huggins theory. Per site, F is given by

$$\frac{F}{kT} = \frac{\phi}{N_A} \ln \phi + \frac{1 - \phi}{N_B} \ln (1 - \phi) + \chi\phi(1 - \phi) \quad (1)$$

where ϕ is the volume fraction of A and N_A and N_B are the degree of polymerization of A and B. From (1), one can derive the critical value of χ for segregation of A and B. In the symmetrical case ($N_A = N_B$), $\chi_C = 2/N$. For most A–B polymer pairs, χ is positive and larger than χ_C, and segregation occurs. The few cases that mix[2] are (a) almost chemically identical species (e.g., deuterated–nondeuterated pairs) and (b) some A–B pairs with specific interactions giving $\chi < 0$ (corresponding to a negative enthalpy of mixing). There is a growing interest in the *dynamics* of fluctuations in blends.[1,3] Our purpose here is to study the mutual penetration of two miscible polymers A and B. At time $t = 0$, consider that two liquid but viscous blocks of pure A and pure B are put into contact and examine the concentration profile $\phi(x,t)$ at later times t, assuming a negative interaction parameter χ *independent of ϕ*. This type of mixing has been studied by spectroscopic techniques.[4] In the standard interpretation of the data, the mutual diffusion coefficient D is supposed

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to be independent of ϕ, and this leads to the classical diffuse form involving error functions. In the present article, we take into account the strong dependence of D upon ϕ. Effects of this type have been reviewed by Crank.[9] But our case is special: as we shall see, in compatible mixtures, the diffusion processes are driven mainly by the enthalpy of mixing; for the pure materials, these mixing effects drop out and $D(\phi)$ nearly vanishes for both $\phi = 0$ and $\phi = 1$. This leads to a very anomalous profile.

Initially, we recall the derivation of D for both long chains[1] (where the reptation process is crucial) and small nonentangled chains.[5] In the second part, we calculate the concentration profile for the most important, symmetrical case ($\phi(-\infty) = \phi_0$, $\phi(+\infty) = 1 - \phi_0$). We also calculate profiles for the nonsymmetrical case where one block of A is put into contact with an A–B mixture. Finally, we compare our results to experimental data. A limitation of our analysis is the assumption that *the monomer–monomer friction coefficients are independent of* ϕ.

II. Dynamics of Polymer Blends

The relation between the A and B fluxes ($\vec{J}_A$ and $\vec{J}_B$) and the forces acting on monomers A and B can be written[1] as

$$\vec{J}_A = -\Lambda_A \vec{\nabla}(\mu_A + U)$$
$$\vec{J}_B = -\Lambda_B \vec{\nabla}(\mu_B + U) \qquad (2)$$

where Λ_A and Λ_B are Onsager coefficients to be discussed below.

μ_A and μ_B are the chemical potentials of A and B derived from (1):

$$\mu_A = F + (1 - \phi)\frac{\partial F}{\partial \phi}$$
$$\mu_B = F - \phi\frac{\partial F}{\partial \phi} \qquad (3)$$

U is a potential ensuring the incompressibility condition $\phi_A + \phi_B = 1$. For Fourier components ($q \neq 0$), this imposes $J_A + J_B = 0$, leading to $U = -(\Lambda_A\mu_A + \Lambda_B\mu_B)/(\Lambda_A + \Lambda_B)$. Inserting this result in (2), we arrive at

$$\vec{J}_A = -\Lambda\vec{\nabla}\mu \qquad (J_B = -J_A) \qquad (4)$$

where $\Lambda = \Lambda_A\Lambda_B/(\Lambda_A + \Lambda_B)$ and $\mu = \mu_A - \mu_B$ is the exchange chemical potential

$$\mu = \frac{\partial F}{\partial \phi} = \left[\frac{1}{N_A}\log\phi - \frac{1}{N_B}\log(1 - \phi) + \chi(1 - 2b)\right]kT \qquad (5)$$

Derivation of Λ. (a) Small Chains. If $N < N_e \sim 300$, the threshold value of N to have entanglements,[7] the chains behave as Rouse chains; i.e., $\Lambda_A = \phi_A\Lambda_{0_A}$ and $\Lambda_B = \phi_B\Lambda_{0_B}$, where Λ_0 is a monomer mobility. For simplicity, we assume *single-chain dynamics identical for A and B*; $\Lambda_{0_A} = \Lambda_{0_B} = \Lambda_0$. Using eq 4, we are led to

$$\Lambda = \phi(1 - \phi)\Lambda_0 \qquad (6)$$

where Λ_0 is independent of ϕ. The flux of A calculated from eq 4 and 5 defines the mutual diffusion coefficient

$$D = \phi(1 - \phi)\Lambda_0\left[\frac{1}{N_A\phi} + \frac{1}{N_B(1 - \phi)} + 2|\chi|\right]kT$$

with the flux relation

$$\vec{J}_A = -D\vec{\nabla}\phi \qquad (7)$$

As soon as ϕ and $1 - \phi$ are larger than $\phi_s = 1/2|\chi|N$ ($\sim 10^{-2}$), we can take

$$D = \phi(1 - \phi)\Lambda_0(2|\chi|)kT = \phi(1 - \phi)(2|\chi|)D_1 \qquad (8)$$

where $D_1 = kT\Lambda_0$ is a monomer diffusion coefficient. It follows that the mutual diffusion of small compatible chains is (1) large, of the order of monomer diffusion coefficient, and (2) dependent upon ϕ as soon as ϕ is larger than ϕ_s; $D \sim \phi(1 - \phi)$.

(b) Long Chains. Long chains are entangled and the motion of one chain is restricted by the topological constraints due to other chains. This leads to a decrease of the monomer mobility. Consider the case of a pure melt. One can describe the motion of a chain as trapped in a tube.[1] The diameter of the tube is $N_e^{1/2}a$ and the length is $L = NN_e^{-1/2}a$. The chain moves along the tube with a mobility $\Lambda_t = \Lambda_0/N$. As the chain moves along the tube length L, the center of mass is displaced only on a distance $R_0 = N^{1/2}a$. The effective mobility is then reduced by the ratio $(R_0/L)^2$:

$$\Lambda = N\Lambda_t\frac{R_0^2}{L^2} = \Lambda_0\frac{N_e}{N} \qquad (9)$$

The transport coefficient for the A–B mixture can be calculated from eq 4, noting that

$$\Lambda_A = \phi\Lambda_0\frac{N_e}{N_A}$$
$$\Lambda_B = (1 - \phi)\Lambda_0\frac{N_e}{N_B} \qquad (10)$$

In what follows we consider only the symmetrical case $N_A = N_B = N$, $\Lambda_{0_A} = \Lambda_{0_B}$. This leads to

$$\Lambda = \phi(1 - \phi)\Lambda_0 N_e/N \qquad (11)$$

The mutual diffusion coefficient deduced from (5) and (11) is now given by

$$D = \phi(1 - \phi)\Lambda_0\frac{N_e}{N}\left[\frac{1}{N\phi} + \frac{1}{N(1 - \phi)} + 2|\chi|\right]kT$$

For long chains, one can approximate D by

$$D = \phi(1 - b)D_1\frac{N_e}{N}(2|\chi|) \qquad (12)$$

The N^{-1} dependence of D upon N is consistent with the findings of the only existing experimental study.[4] Measurements of D as a function of N should be constant for $N \leq N_e$ and decreasing as N^{-1} for $N > N_e$.

For both entangled and nonentangled regimes, we expect D to be proportional to $\phi(1 - \phi)$. In the following section, we set

$$D = D_0\phi(1 - \phi)$$

where

$$D_0 = D_1(2|\chi|) \qquad N < N_e$$
$$D_0 = D_1(2|\chi|)\frac{N_e}{N} \qquad N > N_e \qquad (13)$$

Remark: For the general case $\Lambda_{0_A} \neq \Lambda_{0_B}$, $N_A \neq N_B$, D_0 is slightly dependent upon ϕ. This case is discussed in detail in the Appendix.

III. Concentration Profiles

At $t = 0$ we place a specimen of concentration ϕ_1 in contact with a second specimen of concentration ϕ_2. To calculate the concentration profile $\phi(x,t)$ of the junction as a function of time, must add to eq 7 the conservation law

$$\frac{\partial\phi}{\partial t} + \frac{\partial}{\partial x}J_A = 0 \qquad (14)$$

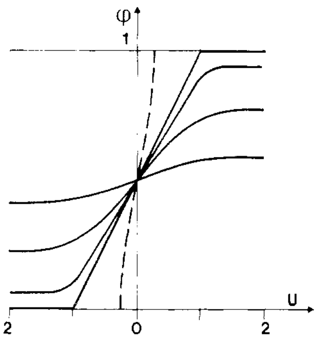

Figure 1. Concentration in nondimensional coordinates $u = x/(D_0t)^{1/2}$ for symmetrical boundary conditions. The curves are calculated numerically, starting from $\phi = {}^1/_2$ and varying the initial slope $\phi_0' = (\mathrm{d}\phi/\mathrm{d}u)_{u=0}$. A physical solution exists only for $\phi_0' < {}^1/_2$. The straight line ($\phi_0' = {}^1/_2$) is the profile for pure melts in contact.

Inserting eq 13, corresponding to the simple case $\Lambda_{0_A} = \Lambda_{0_B}$, $N_A = N_B$,

$$\frac{\partial\phi}{\partial t} = +\frac{\partial}{\partial x}D_0\phi(1-\phi)\frac{\partial\phi}{\partial x} \qquad (15)$$

The solution is defined by eq 15 and the boundary condition $\phi(-\infty) = \phi_1$, $\phi(+\infty) = \phi_2$. It is convenient, following Boltzmann, to set $u = x/(D_0t)^{1/2}$. Equation 15 becomes

$$-\frac{1}{2}u\frac{\mathrm{d}\phi}{\mathrm{d}u} = \frac{\mathrm{d}}{\mathrm{d}u}\left[\phi(1-\phi)\frac{\mathrm{d}\phi}{\mathrm{d}u}\right] \qquad (16)$$

Thus there exist solutions where $\phi(x,t)$ is a function only of *one* variable u.

(a) Pure A and B Blocks in Contact. Let us solve eq 16 with the boundary conditions $\phi(-\infty) = 0$ and $\phi(+\infty) = 1$. By symmetry, $\phi(u = 0) = {}^1/_2$. The solution of (16) is very surprising. It is just a straight line!

$$\phi = (u+1)/2 \qquad |u| < 1 \qquad (17)$$
$$\phi = 0 \qquad u < -1$$
$$\phi = 1 \qquad u > 1$$

This result can be checked directly on eq 16. Because $D(\phi)$ vanishes at both $\phi = 0$ and $\phi = 1$, we do not find the usual "tails" of diffusion problems. The mixed layer extends from $u = -1$ to $u = +1$ and has sharp edges. Beyond these points there is no mixing. The slope of the profile leads to a direct derivation of D_0.

(b) More General Symmetrical Case.

$$\phi(-\infty) = \phi_1$$
$$\phi(+\infty) = 1 - \phi_1$$

The solution of eq 16 satisfies $\phi(-u) = 1 - \phi(u)$. Then $\phi = {}^1/_2$ for $u = 0$. We have calculated numerically the profiles by starting from $\phi = {}^1/_2$, $u = 0$ and varying the slope $(\mathrm{d}\phi/\mathrm{d}u)_{u-0}$. In this way, we generate all the symmetrical solutions shown in Figure 1. When $(\mathrm{d}\phi/\mathrm{d}u)_{u=0}$ tends to ${}^1/_2$, we reach the limiting case (a). (There are no physical solutions for slopes $(\mathrm{d}\phi/\mathrm{d}u)_{u=0}$ larger than ${}^1/_2$.) Note that in the limit of very small differences $|\phi_1 - \phi_2| \ll 1$, we recover the classical solution of the linear diffusion equation.[8]

(c) Asymmetrical Boundary Conditions. We take now $\phi(-\infty) = \phi_1$ and $\phi(+\infty) = \phi_2$ with $\phi_2 \neq 1 - \phi_1$; the value $\phi(u = 0)$ is then different from ${}^1/_2$. To generate the asymmetrical solutions, we start from $\phi(u = 0) = \phi_0$, $(\mathrm{d}\phi/\mathrm{d}u)_{u=0} = \phi_0'$ for a given ϕ_0 and we generate many solutions by varying ϕ_0'. We show in Figure 2 the results obtained for $\phi_0 = 0.8$.

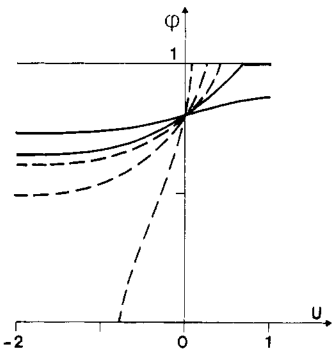

Figure 2. Concentration profile calculated for nonsymmetrical boundary conditions. We have arbitrarily chosen $\phi(u = 0) = 0.8$. The curve that corresponds to $\phi(+\infty) = 1$ reaches $\phi = 1$ with a finite slope ($\phi' = u/2$).

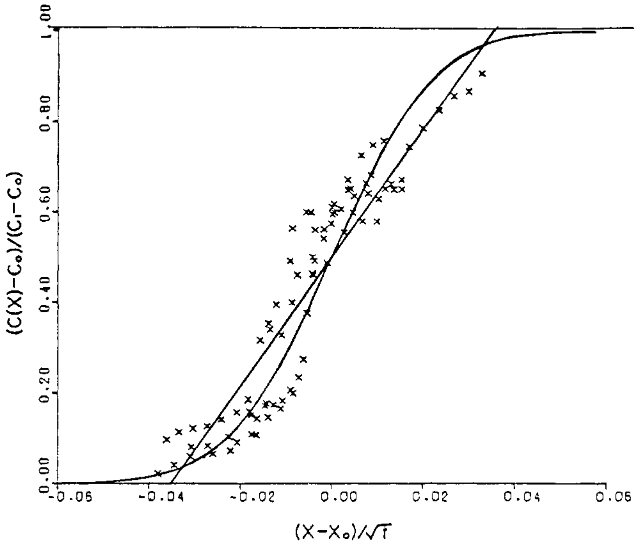

Figure 3. Master curve for the mutual diffusion in a blend of poly(vinyl chloride) (PVC) and poly(ϵ-caprolactone) (PCL) at 70 °C. The weight-average molecular weights are respectively 46 500 ($M_w/M_n = 2.5$)[10] and 37 200 ($M_w/M_n = 1.9$). x is measured in μm and time is in seconds. We have represented the straight line corresponding to $D = D_0\phi(1-\phi)$ and the classical curve[4,8] for $D = D_0$, independent of ϕ. The scattering of experimental data is due to the high polydispersity of the sample.

IV. Conclusion

To interpret the mixing process of two compatible polymers, one *must* take into account the concentration dependence of the diffusion coefficient.

(*a*) *For the simplest case considered* (χ and Λ_0 independent of ϕ, $N_A = N_B$), this leads to a very unusual diffusion profile, with a mixing region of finite site $2(D_0t)^{1/2}$. These unusual features are due to the fact that D tends to zero for $\phi = 0$ and $\phi = 1$. It has to be noticed that this remains true, even for χ and Λ_0 a function of ϕ. These features are attenuated if we start not from pure A and B but from two (different) mixtures.

(*b*) *The general case* ($\Lambda_{0_A} \neq \Lambda_{0_B}$, $N_A \neq N_B$) is analyzed in the Appendix. We again predict sharp edges for the diffuse region, with a slightly more complex profile in between the edges. But, from the slopes near the two edges, one can very simply extract the two parameters Λ_A and Λ_B.

For the compatible pair poly(vinyl chloride)/polycaprolactone the profile $\phi(x,t)$ has been obtained by Laurence et al.[4] using a refined probing technique. A typical profile is shown in Figure 3. It is indeed found that the profile is rather sharp. Of course, for practical samples, the sharp edges of the diffuse region are smeared

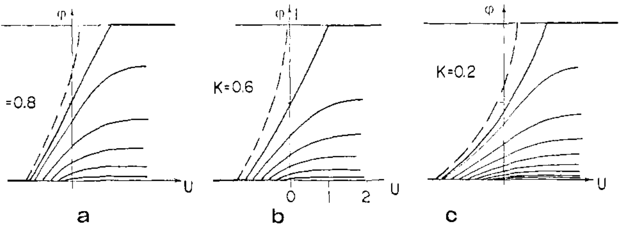

Figure 4. Concentration profiles for pure melts in contact for different values of k [$k = \Lambda_{0_B}/\Lambda_{0_A}$ ($N < N_e$) and $k = (\Lambda_{0_B}/\Lambda_{0_A})\cdot(N_A/N_B)$ ($N > N_e$)]. The curves in a–c are calculated by starting from $\phi = 0$ and varying $u(\phi = 0)$.

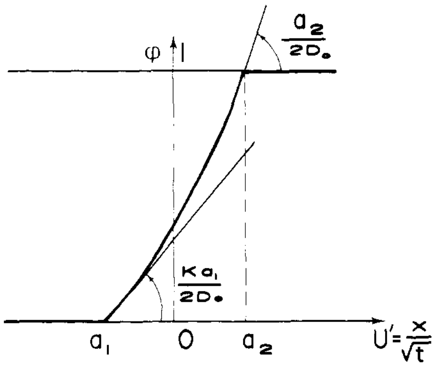

Figure 5. Experimentally one can determine ϕ as a function of $u' = x/t$. From the measure of the slopes $\mathrm{d}\phi/\mathrm{d}u'$ at $\phi = 0$ ($u' = a_1$) and at $\phi = 1$ ($u' = a_2$), one can derive completely the mutual diffusion coefficient $D(N_1,N_2,\Lambda_{0_A}/\Lambda_{0_B},\phi)$

out by polydispersity effects, since for long chains D_0 depends on N ($D_0 \sim N^{-1}$).

For the future, it will be interesting to work with *small chains* ($N < N_e$). In that case, we expect D_0 to be independent of N. This should lead to a linear concentration profile even for polydisperse samples.

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Appendix. Profile in the General Case ($\Lambda_{0_A} \neq \Lambda_{0_B}$, $N_A \neq N_B$)

The mutual diffusion coefficient of an A–B polymer mixture is given by eq 13 only in the symmetrical case ($\Lambda_{0A} = \Lambda_{0B}$, $N_A = N_B$). What happens when the mobility and the polymerization index of the two species are not equal? For $\Lambda_{0_A} \neq \Lambda_{0_B}$, $N_A \neq N_B$, the diffusion coefficient deduced from 4, 6, and 9 is given by

$$D = D_0\frac{\phi(1-\phi)}{\phi + k(1-\phi)} \tag{18}$$

where

$$D_0 = 2|\chi|kT\Lambda_{0_B}, \qquad k = \frac{\Lambda_{0_B}}{\Lambda_{0_A}} \qquad N < N_e$$

$$D_0 = 2|\chi|kT\Lambda_{0_B}\frac{N_e}{N_B}, \qquad k = \frac{\Lambda_{0_B}}{\Lambda_{0_A}}\frac{N_A}{N_B} \qquad N > N_e$$

Note that D is still independent of N_A and N_B for nonentangled chains and thus not sensitive to polydispersity effects. With the diffusion coefficient of eq 18, eq 16 is modified to

$$-\frac{1}{2}u\frac{\mathrm{d}\phi}{\mathrm{d}u} = \frac{\mathrm{d}}{\mathrm{d}u}\left[\frac{\phi(1-\phi)}{\phi + k(1-\phi)}\frac{\mathrm{d}}{\mathrm{d}u}\phi\right] \tag{19}$$

The case considered previously corresponds to $k = 1$. We have calculated the concentration profiles starting from $\phi = 0$ for three values of k (0.8, 0.6, and 0.2) The solutions are shown in Figure 4. The main conclusions of the simple case ($k = 1$) are not modified:

(1) For pure A and B blocks in contact, the size of the mixing zone is finite.

(2) The slopes $(\mathrm{d}b/\mathrm{d}u)_{\phi=0}$ and $(\mathrm{d}\phi/\mathrm{d}u)_{0=1}$ are finite.

(3) In the mixing region, the profile is not exactly a straight line, but there is a gradual change in slope between the limiting values at $\phi = 0$ and $\phi = 1$. From the experimental curve $\phi = f(x/t^{1/2})$, one can directly extract k and D_0 as shown in Figure 5. We set $u' = x/t^{1/2}$. We call a_1 the value of u' corresponding to $\phi = 0$ and a_2 the value of u' for $\phi = 1$. From eq 19, it is obvious that

$$\frac{\left(\dfrac{\mathrm{d}\phi}{\mathrm{d}u'}\right)_{u'=a_1}}{ka_1} = \frac{\left(\dfrac{\mathrm{d}\phi}{\mathrm{d}u'}\right)_{u'=a_2}}{a_2}$$

The value of D_0 is given by the slope at $\phi = 1$:

$$\frac{1}{u'}\left(\frac{\mathrm{d}\phi}{\mathrm{d}u'}\right)_{u'=a_2} = \frac{1}{2D_0}$$

To analyze an experimental curve, one should first extract k from the slopes $\mathrm{d}\phi/\mathrm{d}u'$ at $\phi = 0$ and $\phi = 1$. Inserting this value in eq 19, one can calculate numerically the full profile.

Registry No. Poly(vinyl chloride), 9002-86-2; poly(ϵ-caprolactone), 24980-41-4; poly[oxy(1-oxo-1,6-hexanediyl)], 25248-42-4.

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