

Viscometric Study on the Specific Interaction Between Proton-Donating Polymers and Proton-Accepting Polymers

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Received 8 March 2001; accepted 4 October 2001

ABSTRACT: The interpolymeric specific interaction between proton-donating polymers (PDPs) and proton-accepting polymers (PAPs) was studied by viscometry. The intrinsic viscosities and Schultz–Blaschke constants were measured for the component polymers as well as for the polymeric mixture systems. The estimation of the extent of interpolymeric interaction was done with a new criterion, β , based on the relationship between the degree of complexation (DA) and the theoretical specific viscosity of the polymeric mixture ($\eta_{sp,m,cal}$). This method shows the interpolymeric interaction predictions to be in agreement with those found by a viscosity-enhancement factor. According to the β criterion and viscosity enhancement factors, the octyl acrylate unit in the PDP/PAP mixture is found to make a positive contribution to the complexation between PDP and PAP attributed to its solvation and van der Waals force between side chains of the octyl acrylate unit on unlike macromolecular chains. © 2002 Wiley Periodicals, Inc. *J Appl Polym Sci* 85: 415–421, 2002

Key words: complexation; interpolymeric specific interaction; viscometry; proton donor; proton acceptor

INTRODUCTION

Because of the incorporation of functional groups in the polymeric backbone, interpolymeric specific interactions have long been known to result in unusual behavior and material properties that are dramatically different from those of the non-functional parent polymers.¹ These interactions include ion–ion coulombic interaction,² hydrogen bonding,^{3,4} and transition metal complexation.⁵

Specific interactions between polymers cause aggregation or complexation of the component polymer chains, resulting in solution viscosity variation. For studying interpolymeric interaction, viscometric techniques have been proven

and widely used as a relatively reliable and simple method compared to such demanding and time-consuming techniques as DSC, neutron scattering, light scattering,^{6,7} and nonradiative energy-transfer fluorospectroscopy.⁸

Intrinsic viscosity data for a ternary system composed of polymer–polymer–solvent have been employed to estimate the interaction between unlike polymers by some investigators.^{9,10} However, the effect of specific interaction on the intrinsic viscosity of mixtures is complicated as a result of such influencing factors as molecular weight, the structure and the shape of the mixtures, as well as the power of the solvent. Therefore, other theoretical considerations based on the Huggins equation, such as the Δb criterion^{9–13} and the Δk criterion,^{14–16} were developed to estimate the interaction of the two polymers. The effect of intrinsic viscosity, however, is not taken into account among these criteria, which are applicable to mis-

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Contract grant sponsor: National Natural Science Foundation of China; contract grant number: 59973016.

Journal of Applied Polymer Science, Vol. 85, 415–421 (2002)
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cible systems in which there is no strong interpolymeric specific interaction. For these reasons, a new criterion should be suggested to estimate the interpolymeric interaction level.

To date, there are few reports on interpolymeric specific interaction concerned with long-chain alkyl acrylate copolymers containing functional groups. On researching associating drag reducers, we found that the composition of macromolecular chains remarkably affected the interpolymeric interaction between proton-donating polymers (PDPs) and proton-accepting polymers (PAPs) in solutions. In our present study, poly(octyl acrylate-co-styrene-co-acrylic acid) and poly(octyl acrylate-co-styrene-co-4-vinylpyridine), with varying macromolecular chain compositions, were used as PDP and PAP, respectively. The effect of copolymer chain composition on interpolymeric interaction was studied by the viscometric technique.

THEORETICAL CONSIDERATIONS

Viscosity-Enhancement Factor

A viscosity enhancement factor, R , is defined as follows¹⁷:

$$R = (\eta_{sp,m,exp} - \eta_{sp,m,cal})/\eta_{sp,m,cal} \quad (1)$$

$$\eta_{sp,m,cal} = (\eta_{sp,1}C_1 + \eta_{sp,2}C_2)/C_0 \quad (2)$$

where $\eta_{sp,1}$ and $\eta_{sp,2}$ are the specific viscosities of polymer 1 and polymer 2 at concentration $C_0 = C_1 + C_2$, respectively; $\eta_{sp,m,cal}$ is the specific viscosity of a solution of two unlike polymers assumed without interpolymeric specific interactions; $\eta_{sp,m,exp}$ is the experimentally measured specific viscosity. When there are specific interactions in the mixture (and there should be little thermodynamic repulsion between the unlike polymers because of the similar backbone of both polymers), the complexed systems have specific viscosities that differ from those predicted by eq. (2). Therefore, R is the relative change of the viscosity ascribed to the specific interaction.

Interaction Parameter β of Polymeric Mixture Based on Schulz–Blaschke Equation

As is well known, except with very high molecular weight, data of viscosity measurements in the usual nonassociating dilute system where $[\eta]C$

$\ll 1$ are well described by Huggins equation, so that an easy linear extrapolation to zero concentration of the plot of η_{sp}/C versus C can be made. However, it often occurs that if η_{sp} is above approximately 0.7, the dependency of reduced viscosity on concentration is more or less curved. Model calculations based on the Baker equation¹⁸ indicate that, even if $\eta_{sp} < 0.7$, the concentration dependency of η_{sp}/C is curved, especially if the Huggins constant is above 0.5, so that it is difficult to extrapolate to zero concentration and therefore to accurately predict the intrinsic viscosity and Huggins constant. Consequently, the Huggins equation cannot be used as a basis for a general procedure for estimating $[\eta]$ and, particularly, the Huggins constant k' in the regime with comparatively wide concentration range.

A considerable number of equations have been proposed to describe the relationship between η_{sp}/C and C . Among them, the Schultz–Blaschke law, which is not bound to constant limits of a specific viscosity region and which also allows a wider range of concentrations than is possible with the method of Huggins, can be generally applied to the polymer solutions,¹⁹ expressed in the following Schultz–Blaschke equation:

$$\eta_{sp}/C = [\eta] + k_{SB}[\eta]\eta_{sp} = \frac{[\eta]}{1 - k_{SB}[\eta]C} \quad (3)$$

where k_{SB} is Schultz–Blaschke viscosity slope constant. For a solution of two unlike polymers (1 and 2) without interpolymeric specific interactions between them, the reduced viscosity can be calculated as a weight average of the reduced viscosities of the individual components, given by eq. (4),

$$\begin{aligned} \frac{\eta_{sp,m,cal}}{C_m} &= \frac{[\eta]_1x_1}{1 - k_1[\eta]_1C_m} + \frac{[\eta]_2x_2}{1 - k_2[\eta]_2C_m} \\ &\approx \frac{[\eta]_{m,cal}}{1 - k_{m,cal}[\eta]_{m,cal}C_m} \quad (4) \end{aligned}$$

where x_i and k_i are the weight fractions of the two polymers and the Schultz–Blaschke constants in the polymeric mixture, respectively; subscripts 1 and 2 correspond to polymer 1 and polymer 2; $[\eta]_{m,cal}$ is the weight average of the intrinsic viscosities of the two component polymers in solution; C_m is the sum of the concentrations of the two polymers; and $k_{m,cal}$ is the theoretical value of the Schultz–Blaschke constant of the polymeric

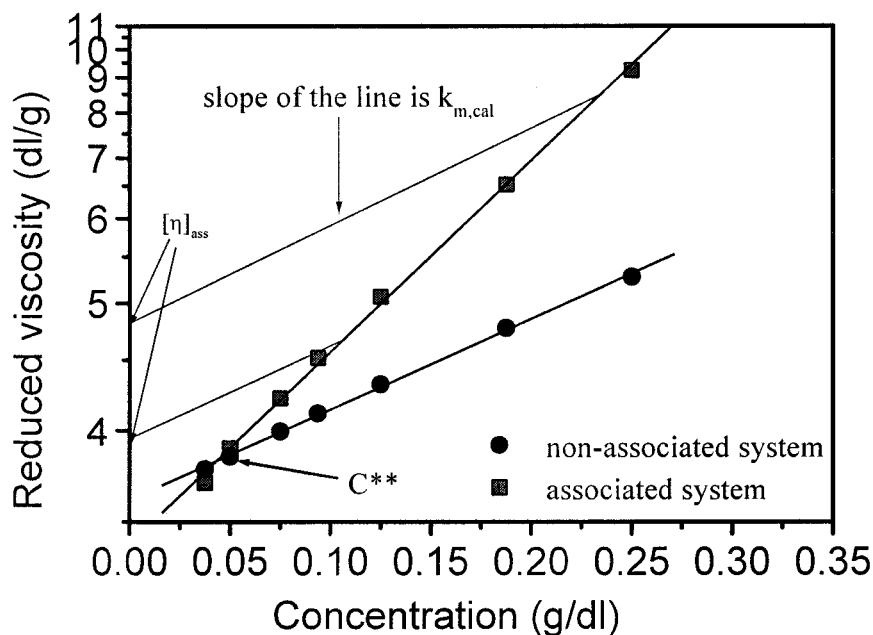


Figure 1 Schematic illustration of the method of obtaining the apparent intrinsic viscosity of associates, $[\eta]_{\text{ass}}$, by use of the Schultz-Blaschke equation.

mixture, which may approximate to an average calculated by eq. (5) within a tolerance of experimental error over the range of measured concentration,

$$k_{m,\text{cal}} = \frac{k_1[\eta]_1^2 x_1 + k_2[\eta]_2^2 x_2}{([\eta]_1 x_1 + [\eta]_2 x_2)^2} \quad (5)$$

It was previously reported that some values of R of the blend solutions are lower than zero in very dilute regions, whereas with the increase of the blend solution concentrations, R increases gradually and changes into a positive value.²⁰ Therefore, the R criterion acts as a measure of interpolymeric specific interaction, depending on the polymer level in solution. The extent to which the degree of complexation increases with polymer level in solution acts as a characterization of interpolymeric specific interaction in this study. It is assumed that the molecular complexes formed by the blended polymer molecules can be seen as a single macromolecule. As the blended polymer solution concentration increases, both the size of the complexes and the interactions between the molecular complexes increase. To depict the relationship of the degree of complexation DA with concentration, DA is defined as follows:

$$DA = \frac{[\eta]_{\text{ass}}^2}{[\eta]_{m,\text{exp}}^2} \quad (6)$$

where $[\eta]_{m,\text{exp}}$ represents the experimental intrinsic viscosity of the PDP/PAP complex, obtained by the linear extrapolation to zero concentration by means of the Schultz-Blaschke equation, and $[\eta]_{\text{ass}}$ is the apparent intrinsic viscosity of the complex at concentration C .

Similar to the empirical method suggested by Hara,²¹ the theoretical Schultz-Blaschke constant of the polymeric mixture $k_{m,\text{cal}}$ from eq. (5) was substituted into eq. (3) with the experimental data of reduced viscosities and concentrations to obtain $[\eta]_{\text{ass}}$, given as

$$\frac{\eta_{sp,m,\text{exp}}}{C} = \frac{[\eta]_{m,\text{exp}}}{1 - k_{m,\text{exp}}[\eta]_{m,\text{exp}}C} = \frac{[\eta]_{\text{ass}}}{1 - k_{m,\text{cal}}[\eta]_{\text{ass}}C} \quad (7)$$

Equation (7) is normalized by C and inverted, whereupon eq. (8) is obtained:

$$\frac{C}{\eta_{sp,m,\text{exp}}} = \frac{1}{[\eta]_{m,\text{exp}}} - k_{m,\text{exp}}C = \frac{1}{[\eta]_{\text{ass}}} - k_{m,\text{cal}}C \quad (8)$$

The procedure is schematically illustrated in Figure 1. (Note: the reduced viscosity is on the reciprocal scale, so the slope of the line is the Schultz-Blaschke constant.) Thus,

$$\frac{1}{[\eta]_{\text{ass}}} = \frac{1}{[\eta]_{m,\text{exp}}} - \Delta k C \quad (9)$$

where

$$\Delta k = k_{m,\text{exp}} - k_{m,\text{cal}} \quad (10)$$

A combination of eqs. (6) and (9) obtains:

$$DA = \left(\frac{1}{1 - \Delta k[\eta]_{m,\text{exp}}C} \right)^2 \quad (11)$$

By substituting eq. (4) into eq. (11), a new expression correlating DA with $\eta_{sp,m,\text{cal}}$ is deduced:

$$\begin{aligned} DA &= \frac{1}{(1 - \Delta k[\eta]_{m,\text{exp}}C)^2} \\ &= \frac{1}{\left[1 - \left(\frac{\Delta k[\eta]_{m,\text{exp}}}{[\eta]_{m,\text{cal}}} \right) \left(\frac{\eta_{sp,m,\text{cal}}}{1 + k_{m,\text{cal}}\eta_{sp,m,\text{cal}}} \right) \right]^2} \end{aligned} \quad (12)$$

where $\eta_{sp,m,\text{cal}}$ is a measure of the hydrodynamic volume fraction of polymer molecules without specific interaction in solution by the modified Einstein equation; the Guth–Simha–Gold equation,²² shown as eq. (13), can act as a scale of polymer level in solution:

$$\eta_{sp,m,\text{cal}} = 2.5\phi + 14.1\phi^2 \quad (13)$$

where ϕ is the hydrodynamic volume fraction of solute. It is obviously obtained that $DA = 1$ at $\eta_{sp,m,\text{cal}} = 0$ and DA increases with the increase in $\eta_{sp,m,\text{cal}}$ attributed to increasing ϕ . When $\eta_{sp,m,\text{cal}} = 1$,

$$DA = \frac{1}{\left[1 - \left(\frac{\Delta k[\eta]_{m,\text{exp}}}{[\eta]_{m,\text{cal}}} \right) \left(\frac{1}{1 + k_{m,\text{cal}}} \right) \right]^2} \quad (14)$$

At the same $\eta_{sp,m,\text{cal}}$ as 1, the larger the DA , the stronger the interpolymeric specific interaction. Therefore, a parameter β to estimate interpolymeric specific interaction may be obtained:

$$\beta = \left(\frac{\Delta k[\eta]_{m,\text{exp}}}{[\eta]_{m,\text{cal}}} \right) \left(\frac{1}{1 + k_{m,\text{cal}}} \right) \quad (15)$$

The β value can therefore be considered as a contribution expressing the ability to interpolymeric specific interaction in a given complexed system. In the complexed systems, the solution shows the significant nonlinear increase in reduced viscosity with the concentration. This is

caused by thermodynamic interaction (complexation), as a result of the incorporation of specific interaction, as well as inherent hydrodynamic interaction, and so on. β reflects the contribution made by interactions other than hydrodynamic interaction.

EXPERIMENTAL

Materials

Styrene (St), acrylic acid (AA), potassium persulfate, *n*-octanol, and sodium lauryl are chemical agents. 4-Vinylpyridine (VP) is a commercial product from Germany.

Styrene was washed with 10% sodium hydroxide to remove the inhibitor and then with distilled water. VP and AA were vacuum distilled before use. Octyl acrylate (OA) was prepared by direct esterification of acrylic acid with *n*-octanol.

Synthesis of Polymers

A series of PDPs and PAPs were synthesized by radical emulsion copolymerization. Potassium persulfate was used as the initiator and sodium lauryl as the emulsifier. The copolymers from the emulsion were isolated by precipitation in cold methanol and then dried under vacuum at 60°C. The apparent average molecular weights of copolymers were determined by static light scattering in toluene. The carboxyl content was determined by titration of PDP in toluene/methanol (4/1,v/v) solution to a phenolphthalein end point with methanolic sodium hydroxide. The VP content was determined by element analysis with an EA 1110 element analysis apparatus. The contents of St and OA in copolymers were calculated from JEOL FX90Q (JEOL, Peabody, MA) ¹H-NMR analysis in CDCl₃ at 30°C. The data of copolymer compositions and molecular weights for PDPs and PAPs are listed in Table I.

Viscosity Measurements

Blend solutions with various weight ratios of PDP to PAP were prepared by blending the two corresponding polymer solutions. Reduced viscosities of single polymer components and their polymer mixtures were measured at 30°C in toluene with the Ubbelohde viscometer. The kinetic energy correction was made with *n*-butanol and acetone.

Table I Chain Composition, Apparent Molecular Weight, Intrinsic Viscosities, and Schultz–Blaschke Constants of Copolymers

Copolymer ^a	Percentage by Weight				M_w^b	$[\eta]$ (dL/g)	k
	OA Unit	St Unit	AA Unit	4-VP Unit			
D2.1–50OA	50.0	47.9	2.1	—	78	2.25	0.286
D2.2–15OA	15.3	82.5	2.2	—	77	2.48	0.399
D3–St	—	97.0	3.0	—	50	2.07	0.444
A5.3–47OA	47.3	47.4	—	5.3	175	4.67	0.341
A5.8–16OA	16.2	78.0	—	5.8	172	4.91	0.353
A6.1–St	0	93.9	—	6.1	190	5.72	0.342

^a Dx–yOA denotes a PDP in which the AA unit content is $x\%$ and the OA unit content is $y\%$ by weight. Ax–yOA denotes a PAP in which the AA unit content is $x\%$ and the OA unit content is $y\%$ by weight. D(A)x–St denotes a binary copolymer composed of St and functional monomer in which $x\%$ is the content of the functional monomer unit.

^b The apparent M_w 's are determined on an 18-angular Dawn DSP laser photometer using polarized light of wavelength 633 nm from a He–Ne laser. The refractive index increments dn/dc were measured in toluene at 30°C with an Optilab DSP interferometric refractometer operating at 633 nm. The apparent M_w 's were obtained from the Zim plot.

RESULTS AND DISCUSSION

Before the discussion of data on polymeric mixtures, it is necessary to characterize the viscometric behavior of each polymer separately in toluene. Measurements of the reduced viscosities of the polymers were conducted and their respective plots were fitted through the Schultz–Blaschke equation. The intrinsic viscosities and Schultz–Blaschke constants for the different copolymers are listed in Table I.

The values of $[\eta]_{m,exp}$ of each polymeric mixture were determined by linear extrapolation to infinite dilution of the plots of C/η_{sp} versus C and the values of the Schultz–Blaschke constants $k_{m,exp}$ are the negative slopes of the plots. Table II lists the theoretical and experimental Schultz–Blaschke constants and intrinsic viscosities obtained by means of linear regression analysis as well as β values by eq. (15); the γ values are correlation factors in linear regression analysis. The correction factors are almost equal to unity,

Table II Theoretical and Experimental Schultz–Blaschke Constants, Intrinsic Viscosities, and β and R Values Obtained Accordingly

Polymer Mixture	F_{AA}^a	$k_{m,exp}$	$k_{m,cal}$	$[\eta]_{m,cal}$	$[\eta]_{m,exp}$	β	R	γ	C^* (g/dL)
D2.1–50OA/ A5.3–47OA	0.161	0.601	0.359	4.07	3.65	0.160	0.57	0.9975	0.11
	0.223	0.754	0.364	3.86	3.40	0.251	1.26	0.9991	0.09
	0.365	0.713	0.371	3.46	3.27	0.236	1.08	0.9996	0.05
	0.534	0.611	0.369	3.06	2.93	0.170	0.57	0.9992	0.06
	0.633	0.551	0.361	2.86	2.89	0.141	0.41	0.9992	—
D2.2–15OA/ A5.8–16OA	0.269	0.577	0.393	3.94	3.84	0.129	0.54	0.9932	0.06
	0.356	0.750	0.401	3.69	3.43	0.234	1.07	0.9994	0.04
	0.524	0.683	0.413	3.29	3.27	0.190	0.71	0.9968	0.01
	0.689	0.594	0.417	2.97	3.09	0.131	0.51	0.9988	—
D3–St/A6.1–St	0.378	0.486	0.411	4.05	3.68	0.048	0.08	0.9990	0.25
	0.485	0.615	0.433	3.65	3.16	0.110	0.19	0.9996	0.22
	0.546	0.776	0.444	3.43	2.95	0.197	0.38	0.9933	0.20
	0.614	0.689	0.455	3.21	2.82	0.141	0.26	0.9968	0.18
	0.735	0.628	0.470	2.82	2.59	0.099	0.12	0.9972	0.20

^a F_{AA} is the molar fraction of AA groups, referring to the ratio of moles of acrylic acid unit to moles of acrylic acid plus 4-vinylpyridine.

which shows the experimental values follow the typical linear relationship of the Schultz–Blaschke equation for the complexed systems.

It can be observed from the values of β in Table II that for each series of PDP/PAP mixtures with similar macromolecular chain composition, there exists a strong complexation among polymer molecules. These PDP/PAP mixtures with similar macromolecular chain composition have corresponding molar fractions of carboxyl (AA) groups of 0.22, 0.36, and 0.55 for D2.1–50OA/A5.3–47OA, D2.2–15OA/A5.8–16OA, and D3–St/A6.1–St, respectively. That is, as the OA unit content is decreased in PDP/PAP, the functional group stoichiometry (AA to VP ratio) at maximal β increases gradually up to 1, which is the ideal functional group stoichiometry for acid–base interaction.

According to the β values, the complexation between D2.1–50OA and A5.3–47OA is a little stronger than that between D2.2–15OA and A5.8–16OA but much stronger than that between D3–St and A6.1–St, although the contents of AA unit and 4-vinylpyridine unit in D3–St and A6.1–St are higher than that in the other two regimes, respectively. Because of similar macromolecular chain composition for each PDP/PAP, this may indicate that the octyl acrylate unit plays an important role in the interpolymeric specific interaction. In the opinion of Malik,²³ van der Waals forces between side chains of the long-chain alkyl acrylate unit on unlike macromolecular chains make a positive contribution to the complexation between PDP and PAP, which may establish an explanation for D2.1–50OA/A5.3–47OA having the strongest complexation among the blend systems in this study.

As is seen from Table II, values of $[\eta]_{m,\text{exp}}$ are decreased compared to those of $[\eta]_{m,\text{cal}}$ with the increase in interpolymeric specific interaction, which indicates R may be negative in a very dilute region. The PDP/PAP complex shows much stronger concentration dependency in toluene, whereas the reduced viscosity–concentration relationship for the polymer mixture without specific interaction is found to be almost linear. It is concluded that there must exist a crossover C^{**} when $[\eta]_{m,\text{exp}}$ is lower than $[\eta]_{m,\text{cal}}$, as shown in Figure 1. As mentioned above, if the intermolecular complex can be seen as a new macromolecule, the PDP/PAP solution behavior is similar to that of the sulfonated polystyrene ionomers, and the crossover C^{**} approximates the C^* region for the ionomer solution.²³ In a dilute region and

when there are strong specific interactions, the complexes are isolated from each other with the formation of a compact structure, which reduces the viscosity of the blend solution. When the blend solution concentration rises above C^{**} , the isolated complexes combine and lead to the formation of a gel-like intermolecular complex structure, which leads to the increase of the solution viscosity. However, for some systems such as D2.1–50OA/A5.3–47OA and D2.2–15OA/A5.8–16OA with $F_{\text{AA}} = 0.63$ and 0.69 , respectively, the crossovers do not exist, indicated by the case that $[\eta]_{m,\text{exp}}$ is higher than $[\eta]_{m,\text{cal}}$. We think this is because the interaction is diminished, thus leading to the formation of a comparatively loose associate. Consequently, we think that R may act as a measure of interpolymeric interaction among the blend systems only when the concentrations are higher than C^{**} , where molecular chains overlap.

The C^{**} values, where $R = 0$, are compiled in Table II. The values of R for PDP/PAP mixtures at 0.3 g/dL concentration, where the molecular chains should overlap sufficiently, were compiled in Table II. The variations of R exhibit a tendency similar to that of β , which shows the results of the β criterion are reasonable and valid for evaluating the extent of interpolymeric specific interaction.

In our experiment, when concentrations are below 0.2 g/dL, some values of R in the D3–St/A6.1–St system are negative. By comparison, the values of R in the case of D2.1–15OA/A5.8–16OA and D2.1–50OA/A5.3–47OA are generally positive at concentrations above 0.1 g/dL. The reason, we conclude, may be that solvation of side chains of the octyl acrylate unit stabilizes the associates formed by PDP and PAP. For interaction between D2.1–50OA and A5.3–47OA, associates may form a loose structure with many “free” side chains of octyl acrylate unit solvating the associates, whereas in the case of D3–St/A6.1–St, associates form a dense structure attributed to no solvation of side chains of the octyl acrylate unit, which results in a contraction or collapse of the component polymer chains and accordingly a decrease in the blended solution viscosity.

CONCLUSIONS

The association between PDP and PAP in toluene was studied by viscometry. An empirical relationship between degree of association (DA) and the-

oretical specific viscosity of the polymer mixture ($\eta_{sp,m,cal}$) was proposed, based on which a novel criterion used to characterize the ability to the interpolymeric specific interaction was deduced. In agreement with the viscosity-enhancement factor at a concentration at which molecular chains sufficiently overlap, the β criterion is valid for estimating interpolymeric specific interaction. It is observed that the octyl acrylate unit makes a beneficial contribution to complexation between PDP and PAP. This may be explained by the solvation of side chains of octyl acrylate unit on unlike macromolecular chains and the van der Waals forces between them.

The authors are indebted to the National Natural Science Foundation of China for financial support of this research (No. 59973016).

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