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# Scattering Properties and Phase Behavior of Mixtures of Cyclic and Linear Homopolymers and Copolymers

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ABSTRACT: The static scattering properties and phase behavior of mixtures of homopolymers and diblock copolymers are considered. The effects of architecture on these properties are investigated. In particular, the results obtained for mixtures with cyclic homopolymers and copolymers are compared with those obtained in the case of mixtures of linear chains. Depending on the concentration of copolymer, one may observe an interplay between macrophase and microphase separation transitions. This interplay is investigated for cyclic and linear chain systems. Mixtures made of cyclic copolymers show enhancement of compatibility towards phase separation. Phase diagrams are plotted to show the compatibilization. Effects of topological constraints on chain architecture and interactions are ignored for cyclic chain systems.

## Introduction

A few years ago, Hashimoto and co-workers[1,2] reported a study of the scattering properties and phase behavior of mixtures of homopolymers and diblock copolymers at several compositions. They observed that in the case of the pure diblock copolymer A−B, microphase separation takes place if the interaction parameter $\chi$ reaches a critical value $\chi_s$. This transition is followed by a microstructure emerging within the system which has a characteristic size $\lambda^* = 2\pi/q^*$. This size is the wavelength of the mode of maximum composition fluctuations, and $q^*$ is the wavevector at which the scattered intensity shows a peak in the homogeneous one-phase region. Adding parent homopolymers A and/or B, one observes a modification in the scattering behavior and melting of the microphase structure. This behavior is the result of a substantial increase in the wavelength $\lambda^*$ which eventually tends to infinity when $q^*$ tends to zero.

On the other hand, in the case of a blend of A and B homopolymers at the temperature of macrophase separation $T_s$, the blend phase separates via a spinodal decomposition mechanism. Adding a certain amount of copolymer induces enhancement of compatibility and homogenization of the mixture. This can be observed in scattering experiments through reduction of the forward scattered intensity as the concentration of copolymers is increased. Such a behavior was observed by others and is well documented in the literature.[3−5]

However, studies along these lines were usually made with linear polymers and relatively few investigations have been reported in the case of polymers with other architectures. In particular, little is known on the thermodynamic behavior and scattering properties of cyclic homopolymers and copolymers.

The purpose of the present paper is to report a theoretical study of such properties for mixtures of cyclic homopolymers and copolymers and compare the results with those obtained in the case of mixtures made of linear chains.

## Scattered Intensity and Spinodal Equation

The scattered intensity $I(q)$ for a system made of a blend A/B and a block copolymer A−B can be easily obtained within the framework of the random phase approximation (RPA).[3−6] If one considers that A and B monomers have an interaction parameter $\chi$ and a contrast factor $(a-b)^2$, one obtains

$$\frac{(a-b)^2}{I(q)} = \frac{S(q)}{W(q)} - 2\chi \tag{1}$$

where $q$ is the amplitude of the scattering wavevector which is related to the wavelength of incident radiation $\lambda$ and the scattering angle $\theta$ through $q = (4\pi/\lambda)\sin(\theta/2)$. The definitions of $S(q)$ and $W(q)$ depend upon the system under consideration. In view of the fact that the mixtures of interest in this work involve only two monomer species, it is convenient to think of a two by two matrix whose elements are the partial structure factors $S_{aa}(q)$, $S_{bb}(q)$, and $S_{ab}(q) = S_{ba}(q)$. Regardless of whether the chains are cyclic or linear, $S(q)$ represents the sum of the four partial structure factors:

$$S(q) = S_{aa}(q) + S_{bb}(q) + 2S_{ab}(q) \tag{2}$$

whereas $W(q)$ represents the determinant:

$$W(q) = S_{aa}(q)S_{bb}(q) - S_{ab}^{\;2}(q) \tag{3}$$

These quantities will be defined more precisely shortly after in this paper. Prior to detailed discussions of the predictions of eq 1, it is worthwhile to note that this is a well-known result which was derived for the first time by Leibler[3] using the random phase approximation (RPA) in the case of a diblock copolymer A−B. Later, it was generalized to copolymers in solution,[6] mixtures of homopolymers and copolymers,[1,2,5,7] rigid copolymers,[8] etc. Within the RPA, one can show that eq 1 remains valid for mixtures of flexible chains regardless of their architecture.[5,9,10] Here we shall make use of this observation and assume that the same equation can also be applied to mixtures of cyclic chains. We are interested in homopolymers A and B and copolymers A−B with the topological constraints that both ends of each

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chain are linked and there are no knots, no concatenated rings, no dangling ends and no free linear chains. Normally, the interaction parameter $\chi$ between A and B monomers should also include the effects of these topological constraints. To our knowledge, these effects are not known although some attempts have been made to understand the cases of cyclic homopolymers[11,12] and branched polymers.[13] These effects are under current investigation in our laboratory and will be the subject of a future communication. In the present paper, we shall assume that all the topological constraints and architectural properties are embedded into the form factors entering in the definitions of $S(q)$ and $W(q)$ in eq 1.

Here we are interested in mixtures of homopolymers A and B and block copolymers AB. In this case, the above partial structure factors have contributions from both the homopolymers and the copolymer in the mixture:

$$S_{\mathrm{aa}}(q) = \varphi_{\mathrm{ah}} N_{\mathrm{ah}} P_{\mathrm{ah}}(q) + \varphi_{\mathrm{ac}} N_{\mathrm{ac}} P_{\mathrm{ac}}(q) \tag{4a}$$

$$S_{\mathrm{bb}}(q) = \varphi_{\mathrm{bh}} N_{\mathrm{bh}} P_{\mathrm{bh}}(q) + \varphi_{\mathrm{bc}} N_{\mathrm{bc}} P_{\mathrm{bc}}(q) \tag{4b}$$

$$S_{\mathrm{ab}}(q) = \sqrt{\varphi_{\mathrm{ac}} \varphi_{\mathrm{bc}} N_{\mathrm{ac}} N_{\mathrm{bc}}} P_{\mathrm{abc}}(q) \tag{4c}$$

The subscipts a and b refer to the monomers A and B, whereas the subscripts h and c refer to the homopolymer and the copolymer, respectively. For example $\varphi_{\mathrm{ah}}$ is the volume fraction of monomers A belonging to the homopolymer, $N_{\mathrm{ah}}$ is the degree of polymerization of homopolymer A, and $P_{\mathrm{ah}}(q)$ is its form factor. The same definitions hold for the other quantities changing a to b (monomer B) and h to c (copolymer). Note that $P_{\mathrm{abc}}(q)$ is the contribution to the form factor of the copolymer due to interferences between A and B monomers belonging to the same copolymer (see eqs 5 and 11 below). These formulas apply to both linear and cyclic polymers. The difference between these two architectures is found in the expressions for the form factors $P_{ij}(q)$. Ignoring the effects of excluded volume interactions and assuming Gaussian statistics, one can use the Debye function for the form factor of linear chains while for $P_{\mathrm{abc}}(q)$, one can use

$$P_{\mathrm{abc}}(q) = \frac{(1 - \mathrm{e}^{-u_{\mathrm{ac}}})(1 - \mathrm{e}^{-u_{\mathrm{bc}}})}{u_{\mathrm{ac}} u_{\mathrm{bc}}} \tag{5}$$

with

$$u_{\mathrm{ac}} = f u_{\mathrm{c}} \tag{6a}$$

$$u_{\mathrm{bc}} = (1 - f) u_{\mathrm{c}} \tag{6b}$$

$$u_{\mathrm{c}} = q^2 \frac{N_{\mathrm{c}} a^2}{6} \tag{6c}$$

$$N_{\mathrm{c}} = N_{\mathrm{ac}} + N_{\mathrm{bc}} \tag{6d}$$

$a$ is a statistical length assumed to be the same for both A and B monomers and $f = N_{\mathrm{ac}}/N_{\mathrm{c}}$ is the composition in A monomers of the copolymer.

For cyclic homopolymers, the form factors are given by the Casassa function:[14]

$$P_{\mathrm{ih}}(q) = \frac{2}{\sqrt{u_{\mathrm{ih}}}} \mathrm{e}^{-u_{\mathrm{ih}}/4} \int_0^{\sqrt{u_{\mathrm{ih}}}/2} \mathrm{e}^{t^2}\, \mathrm{d}t \tag{7}$$

with

$$u_{\mathrm{ih}} = q^2 \frac{N_{\mathrm{ih}} a^2}{6} \tag{8}$$

For a cyclic diblock copolymer, we have[10,15]

$$P_{\mathrm{ac}}(q) = \frac{f - \frac{1}{2}}{f^2} C\left(u_{\mathrm{c}}, \left(\frac{1}{2} - f\right)\sqrt{u_{\mathrm{c}}}\right) + \frac{1 - f}{f} H[u_{\mathrm{c}} f(1 - f)] \tag{9}$$

$$P_{\mathrm{bc}}(q) = \frac{\frac{1}{2} - f}{(1 - f)^2} C\left(u_{\mathrm{c}}, \left(f - \frac{1}{2}\right)\sqrt{u_{\mathrm{c}}}\right) + \frac{f}{1 - f} H[u_{\mathrm{c}} f(1 - f)] \tag{10}$$

$$P_{\mathrm{abc}} = \frac{C(u_{\mathrm{c}},0) - f^2 P_{\mathrm{ac}}(q) - (1 - f)^2 P_{\mathrm{bc}}(q)}{2f(1 - f)} \tag{11}$$

with

$$H(x) = \frac{1 - \mathrm{e}^{-x}}{x} \tag{12}$$

and

$$C(x,\nu) = \frac{2}{\sqrt{x}} \mathrm{e}^{-x/4} \int_{\nu}^{\sqrt{x}/2} \mathrm{e}^{t^2}\, \mathrm{d}t \tag{13}$$

The form factors for cyclic homopolymers and copolymers are known and more detailed derivations are given elsewhere.[5,8,10,14−17] However, one should note that they are meaningful only under the condition that the Gaussian approximation is valid. Indeed, the starting assumption which is always used in the derivation of these results is to write the average square distance between two points on a cyclic chain separated by $n$ monomers $\langle r_n^2 \rangle$ in the following form:

$$\langle r_n^2 \rangle = a^2 n \left(1 - \frac{n}{N}\right) \tag{14}$$

where $N$ is the number of monomers of the chain under consideration. This Gaussian approximation leads to the same scaling behavior of the radius of gyration $R_{\mathrm{g}}$ with $N$ as for linear chains, namely

$$R_{\mathrm{g}} \sim N^{0.5} \tag{15}$$

The difference between the radii of gyration of the cyclic chain and its linear counterpart lies only in the prefactor. This description ignores the effects of topological constraints imposed upon rings, as mentioned earlier. It is known, however, that in melts of cyclic chains, topological interactions lead to more compact conformations than predicted by the Gaussian approximation. This immediately leads to a change in the scaling behavior of the radius of gyration which is then intermediate between the collapse state where the exponent $\nu$ in eq 15 is 0.33 and the Gaussian limit where $\nu = 0.5$. Crude models of the Flory type[18] and computer simulations[19] indicate that $\nu$ for melts of cyclic homopolymers should be 0.4. Such refinements of the statistical description of conformation of cyclic chains due to topological interactions are not taken into account in the present investigation. On the other hand, although the RPA is a mean field approximation and fails to describe the physical behavior of systems in the presence of strong fluctuations, one can still use it to

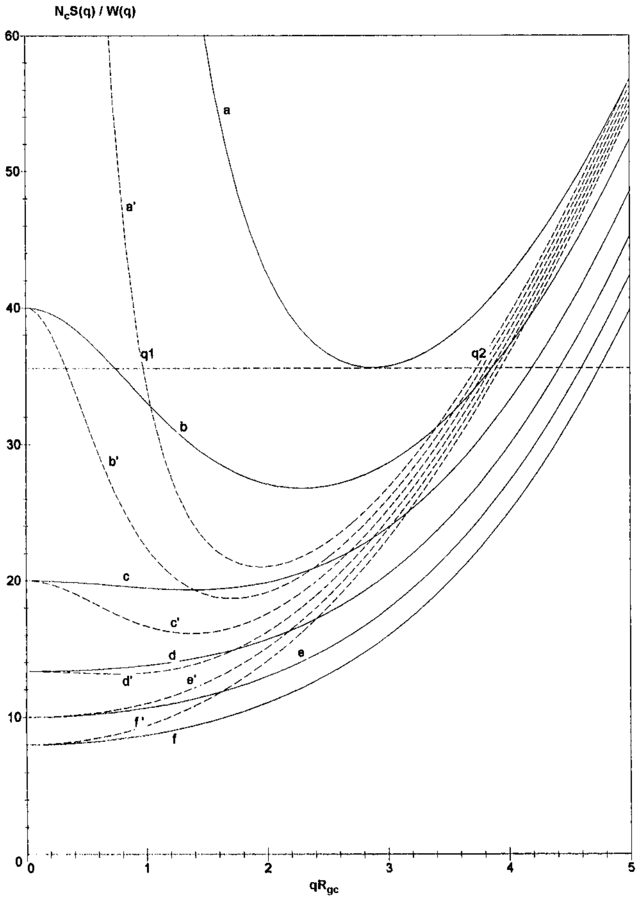

**Figure 1.** Variation of $N_c(S(q)/W(q))$ as a function of $qR_{gc}$ for a mixture of homopolymers A and B and diblock copolymers A−B for several values of $\varphi_c$: (a and a′) $\varphi_c = 1.0$; (b and b′) $\varphi_c = 0.8$; (c and c′) $\varphi_c = 0.6$; (d and d′) $\varphi_c = 0.4$; (e and e′) $\varphi_c = 0.2$; (f and f′) $\varphi_c = 0$. The continuous lines represent the mixtures with cyclic chains, whereas the dashed lines correspond to those with linear chains only. In plotting these curves (see eq 1), we have used $N = N_{ah} = N_{bh} = N_c/2$ and $R_{gah}^2 = R_{gbh}^2 = R_{gc}^2/2$.

make certain predictions. In particular, the spinodal equation is obtained by letting the inverse scattered intensity be zero at $q = q^*$. This equation defines the interaction parameter at the spinodal line $\chi_s$, which is given by

$$2\chi_s = \frac{S(q=q^*)}{W(q=q^*)} \tag{16}$$

The wavelength of the mode which drives the system to criticality is given by $\lambda^* = 2\pi/q^*$, where $q^*$ is obtained by equating the derivative of $(a - b)^2/I(q)$ with respect to $q$ to zero. Depending upon the concentration of copolymer $\varphi_c$, one finds that $q^*$ is different from zero in the case of a microphase separation transition and equal to zero if $\varphi_c$ is small and one deals with a macrophase separation transition (see Figure 3 below).

## Results and Discussion

Figure 1 represents the variations of $N_c(S(q)/W(q))$ as a function of $qR_{gc}$ for mixtures of homopolymers A and B and copolymers A−B at six different volume fractions of the copolymer $\varphi_c$. The continuous curves a−f represent the case of cyclic chains, whereas the dashed curves a′−f′ are the corresponding results for linear polymers in similar conditions. The introduction of the normalization constant factors $N_c$ in the $y$-axis and $R_{gc}$ in the $x$-axis is for convenience only and has no physical meaning. The numerical parameters of the mixtures are the same as those of Hashimoto and co-workers[1,2] and therefore the dashed curves are identical to those reported in refs 1 and 2. The representation in Figure 1 is convenient because it reflects results which can be viewed in various ways and hence could lead to a better insight into their physical meaning. For example, these curves represent the inverse scattering intensity in the ideal limit where the interaction between monomers A and B is zero. This could be the case in neutron scattering experiments where A is an ordinary monomer and B is a deuterated monomer of the same chemical nature as A, assuming that the $\chi$-parameter between these monomers is zero. An example of such experiments was reported by Maschke et al.[7] on mixtures of triblock copolymers made of deuterated and hydrogenated poly(dimethylsiloxane) (PDMS) blocks and deuterated PDMS homopolymers. One can also analyze the results of Figure 1 in the following way. Consider for example curve a obtained for the pure cyclic copolymer A−B when the volume fraction $\varphi_c$ is 1. The dashed horizontal line passes through the minimum of this curve and hence defines the critical parameter at which the microphase separation transition (MST) takes place $\chi_s \approx 18/N_c$. This parameter corresponds to a temperature $T_s$ at which the system undergoes strong composition fluctuations. The critical mode which is most responsible for driving the system out of its stability limit is the one with wavelength $\lambda^* = 2\pi/q^*$. At all temperatures corresponding to the domain $\chi < \chi_s$, the system is stable and shows a one-phase state. The interesting observation with respect to the present investigation is to realize that in the case of the linear copolymer (dashed curve a′), the MST takes place at a much lower interaction parameter $\chi'_s \approx 10/N_c$. The dashed horizontal line shows that all the modes with wavelengths lying between $\lambda_1 = 2\pi/q_1$ and $\lambda_2 = 2\pi/q_2$ are unstable at the temperature corresponding to $\chi = 18/N_c$ and contribute to the microphase separation of the linear copolymer. The latter system shows ordered structures with characteristic lengths from short scales comparable to $\lambda_2$ to long distances comparable to $\lambda_1$. The stability toward MST is substantially enhanced by the simple fact of the existance of a chemical bond between the two ends of the copolymer.

Considering curves b−f and b′−f′, one observes that as the volume fraction of copolymer decreases, the gap between the scattering curves of cyclic and linear chain systems is substantially reduced in the lower $q$ range ($q < q^*$), where the effects of the copolymer are largest. In the upper $q$ range, however ($q > q^*$), the presence of homopolymer is overwhelming and the gap widens as $\varphi_c$ increases. For example, one observes that the second mixture investigated (curves b and b′ corresponding to $\varphi_c = 0.8$ and $\varphi_{ah} + \varphi_{bh} = 0.2$) is deep inside the unstable regime at $\chi_s \approx 18/N_c$. Indeed, the critical parameter for MST of cyclic chains is $\chi_s \approx 13/N_c$ (curve b) and of linear chains is $\chi'_s \approx 9.3/N_c$ (curve b′), which is only slightly lower than the corresponding value for a pure linear diblock ($\chi'_s \approx 10.5/N_c$, as shown by curve a′).

With regard to the nature of the phase transitions, one can see that the pure copolymer cannot undergo macrophase transition at any temperature, a result which is expected since the A and B blocks are linked together. However, with even a trace amount of homopolymer A and/or B, the mixture can also show a macrophase transition which would be observed at $q =$

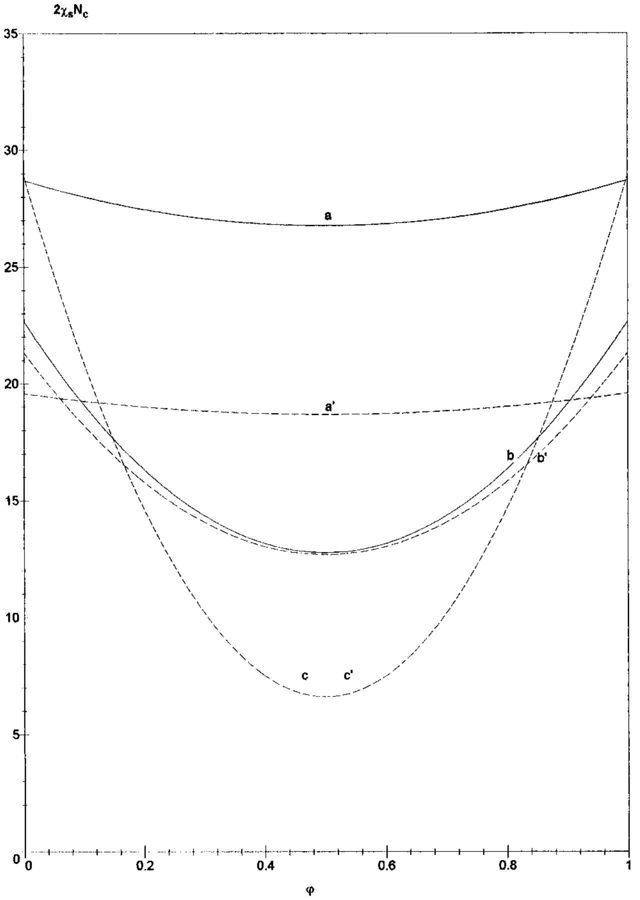

**Figure 2.** Variation of $2\chi_sN_c$ as a function of $\varphi$ for a mixture of homopolymers A and B and diblock copolymers A–B at different values of $\varphi_c$: (a and a′) $\varphi_c = 0.8$; (b and b′) $\varphi_c = 0.4$; (c and c′) $\varphi_c = 0.2$. The continuous lines represent the cyclic chains, whereas the dashed lines correspond to the linear chains.

0 if the temperature is sufficiently high. Consider for example the system with $\varphi_c = 0.8$, if the temperature is lowered in such a way that the interaction parameter reaches $20/N_c$, then the microphase separation (reached for the cyclic chains at $\approx 13/N_c$) would be followed by a macrophase transition. As the volume fraction of homopolymers increases, the critical parameter for the macrophase transition decreases faster than the critical parameter for the MST. They may become equal as for $\varphi_c = 0.6$ and ultimately the MST disappears if the volume fraction of copolymer is low enough as for $\varphi_c \leq 0.2$.

Some of these features are illustrated in the phase diagram of Figure 2 where the critical interaction parameters for macrophase and microphase transitions are shown as a function of $\varphi$, the relative composition of homopolymer A in the blend A/B, i.e. $\varphi = \varphi_{ha}/(\varphi_{ha} + \varphi_{hb})$ for different concentrations of the copolymer $\varphi_c$. Following the same conventions as in Figure 1, the continuous lines represent the results for the cyclic polymers whereas the dashed lines correspond to the case of linear chains. Curves a–c are calculated using $\varphi_c = 0.8$, 0.4, and 0.2, respectively. The two upper curves a and a′ show a wide miscibility gap between cyclic and linear chains. The copolymer in the mixture dominates the phase behavior, and the transition lines plotted here are the generalized spinodals for microphase separation transition. The intermediate curves b and b′ correspond to $\varphi_c = 0.4$. They show a relatively small difference between cyclic and linear chain systems. For symmetrical blends where $\varphi = 0.5$, there is practically no difference between the critical parameters whether one deals with cyclic or linear polymers. The lower curve is the superposition of curves c and c′ for linear and cyclic systems with $\varphi_c = 0.2$. In this case, one has only a macrophase separation transition in which cyclic and linear copolymer systems show the same thermodynamic behavior. This is a direct consequence of the fact that the interaction parameter $\chi(T)$ is assumed to be the same for both open and cyclic chain architecture[11,12] and that all form factors are normalized to 1 at $q = 0$. To monitor the evolution from microphase to macrophase transitions with the concentration of copolymer, one can look at the variation of $q^*$ where the scattering curves of Figure 1 admit a minimum as a function of $\varphi_c$. This representation is shown in Figure 3 and reveals that the MST appears at a much lower concentration of copolymer in the case of cyclic chains. When this concentration is smaller than 0.3, only a macrophase transition can take place in both systems, indicating that the phase behavior is controlled by the blend of homopolymers. When $\varphi_c$ exceeds 0.3, the macrophase transforms into a microphase transition and it is the copolymer component which has the strongest effect on the phase behavior of the mixture with linear chain systems. As $\varphi_c$ increases, the characteristic $q^*$ increases rapidly and tends to approximately $1.9/R_{gc}$ for a pure linear copolymer, as already known.[3,10,20] In the case of the mixture with cyclic chains, the emergence of a microphase transition requires a higher concentration of cyclic copolymers since it appears only at $\varphi_c \approx 0.5$ instead of 0.3 for linear

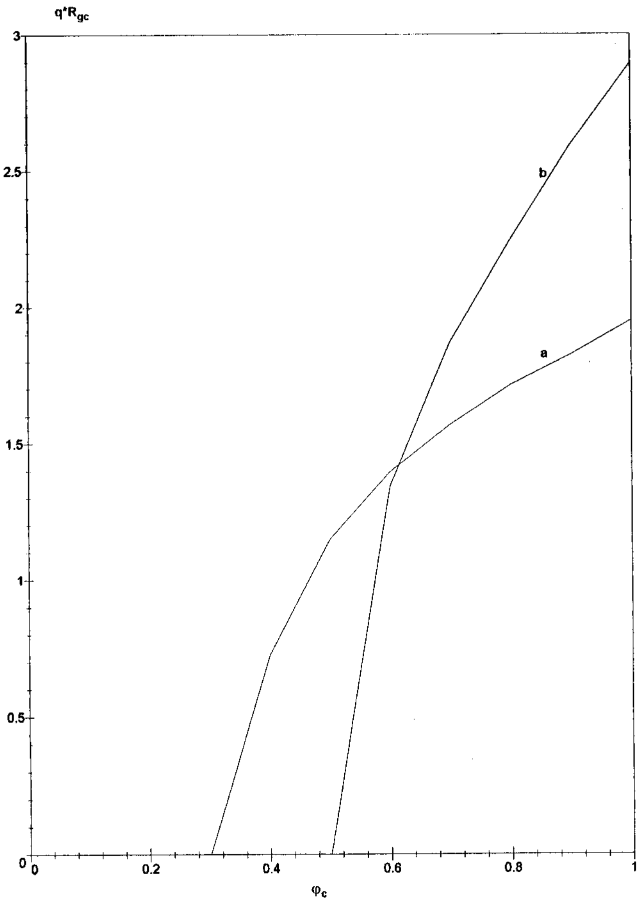

**Figure 3.** Variation of $q^*R_{gc}$ as a function of $\varphi_c$ for a mixture of homopolymers A and B and diblock copolymers A–B. Curve a represents the case where all the chains are linear, whereas curve b corresponds to the case where all chains are cyclic.

copolymers. In the case of mixtures of cyclic homopolymers and copolymers, the composition must exceed 50% in order for the mixture to show a MST. When $\varphi_c$ increases above 50%, the characteristic size $q^*$ also increases much more rapidly than in the case of linear chains. When $\varphi_c$ approaches 1, $q^*$ is of the order of $2.9/R_{gc}$, whereas for linear chains the maximum of $q^*$ is approximately $1.9/R_{gc}$.

## Conclusions

The above observations show that there is a clear distinction between the scattering properties and phase behavior of mixtures of cyclic homopolymers and copolymers as compared to their linear counterparts. The interplay between macrophase and microphase transitions not only depends on the concentration of copolymers but also depends strongly on their architecture. The mixtures containing cyclic copolymers are characterized by a compatibility much higher than that of their linear counterparts. The fact that blends of cyclic homopolymers show compatibility enhancement toward macrophase separation has already been observed experimentally.[21] To our knowledge, however, until now, no experimental observation has been made on cyclic block copolymers to assess the enhancement of compatibility toward MST. The present study is an attempt to present a simple formalism based on the RPA which gives some indication of the scattering and phase behavior of mixtures containing cyclic homopolymers and copolymers. This formalism can also be used to characterize systems with different architectures. Furthermore, other properties such as those accessible by static neutron scattering or X-rays at high $q$'s and quasielastic scattering[22] or neutron spin echo techniques can also be examined within the same framework. In particular, the dynamic properties are the subject of another communication.[23]

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## References and Notes

(1) Ijichi, Y.; Hashimoto, T. *Polym. Commun.* **1988**, *29*, 135.
(2) Tanaka, H.; Hashimoto, T. *Polym. Commun.* **1988**, *29*, 212.
(3) Leibler, L. *Macromolecules* **1980**, *13*, 1602.
(4) de Gennes, P. G. *Scaling concepts in Polymer Physics*; Cornell University Press: Ithaca, NY, 1979; *J. Phys.* (*Paris*) **1970**, *31*, 235.
(5) Higgins, J.; Benoît, H. *Polymers and Neutron Scattering*; Clarendon Press: Oxford, U.K., 1994.
(6) Benoît, H.; Wu, W.; Benmouna, M.; Mozer, B.; Bauer, B.; Lapp, A. *Macromolecules* **1985**, *18*, 986.
(7) Maschke, U.; Ewen, B.; Benmouna, M.; Meier, G.; Benoît, H. *Macromolecules* **1993**, *26*, 6197.
(8) Hammouda, B. *Adv. Polym. Sci.* **1992**, *106*, 89.
(9) Olvera de la Cruz, M.; Sanchez, I. *Macromolecules* **1986**, *19*, 2501.
(10) Benmouna, M.; Borsali, R.; Benoît, H. *J. Phys. II Fr.* **1993**, *3*, 1041.
(11) Léonard, J. *J. Phys. Chem.* **1989**, *93*, 4346.
(12) Léonard, J. *J. Polym. Sci., Polym. Phys. Ed.* **1993**, *31*, 1495.
(13) Fredrickson, G. H. *Macromolecules* **1994**, *27*, 7382.
(14) Casassa, E. F. *J. Polym. Sci., Part A* **1965**, *3*, 604.
(15) Benmouna, M.; Fischer, E. W.; Bensafi, A.; Khaldi, S. *J. Polym. Sci., Polym. Phys. Ed.* **1996**, *34*, 1629.
(16) Burchard, W. In *Cyclic Polymers*; Semlyen J. A., Ed.; Elsevier: London, 1986.
(17) Cosmas, M.; Benoît, H.; Hadziioannou, G. *Colloid Polym. Sci.* **1994**, *9*, 272.
(18) Cates, M. E.; Deutsch, J. M. *J. Phys.* **1986**, *47*, 2121.
(19) Müller, M.; Wittmer, J. P.; Cates, M. E. *Phys. Rev. E* **1996**, *53*, 5063.
(20) Benoît, H.; Hadziioannou, G. *Macromolecules* **1988**, *21*, 1449.
(21) Santore, M. M.; Han, C. C.; McKenna, G. B. *Macromolecules* **1992**, *25*, 3416.
(22) Amis, E. J.; Hodgson, D. F.; Wu, W. *J. Polym. Sci., Polym. Phys. Ed.* **1993**, *31*, 2049.
(23) Benmouna, M.; Benoît, H.; Khaldi, S.; Bensafi, A. *Macromolecules* **1996**, *29*, 8101.

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