# A Self-Consistent Field Theory Study on the Morphologies of Linear ABCBA and H-Shaped (AB)₂C(BA)₂ Block Copolymers

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By using a combinatorial screening method based on the self-consistent field theory, we investigate the equilibrium morphologies of linear ABCBA and H-shaped (AB)₂C(BA)₂ block copolymers in two dimensions. The triangle phase diagrams of both block copolymers are constructed by systematically varying the volume fractions of blocks A, B, and C. In this study, the interaction energies between species A, B, and C are set to be equal. Four different equilibrium morphologies are identified, i.e., the lamellar phase (LAM), the hexagonal lattice phase (HEX), the core−shell hexagonal lattice phase (CSH), and the two interpenetrating tetragonal lattice phase (TET2). For the linear ABCBA block copolymer, the reflection symmetry is observed in the phase diagram except for some special grid points, and most of grid points are occupied by LAM morphology. However, for the H-shaped (AB)₂C(BA)₂ block copolymer, most of the grid points in the triangle phase diagram are occupied by CSH morphology, which is ascribed to the different chain architectures of the two block copolymers. These results may help in the design of block copolymers with different microstructures.

## Introduction

Block copolymers are fascinating soft materials that can self-assemble into a variation of beautiful structures on the nanometer scale. Both theory and experiments have demonstrated that the block copolymer architecture is a controlling factor in generating different nanoscale morphologies. A lot of work [1−12] has been devoted to studying the effects of different block copolymer architectures on the morphological behavior. Lee et al.[1,2] pointed out that for block copolymers with sufficient molecular weight to exist in the strong segregation regime, the morphological behavior of the molecules with multiple junction points can be estimated by imaging all bridge and loop blocks to be cut in half. Hence, block copolymers with simple linear and $A_nB_m$ miktoarm (*n* and *m* denote the number of arm A and arm B, respectively) architectures have been widely investigated.[4−10]

An H-shaped (AB)₂C(BA)₂ block copolymer has four side-arms (AB) that are attached to the two ends of the backbone (C). A schematic representation of the architecture is shown in Figure 1. A linear ABCBA block copolymer is a penta-block copolymer. The difference between the H-shaped (AB)₂C(BA)₂ block copolymers and the ABCBA block copolymers is that the H-shaped (AB)₂C(BA)₂ block copolymers have two AB blocks at both ends of the backbone (C), and the linear ABCBA block copolymers have only one AB block at both ends of the backbone (C). Therefore, studying the morphology differences between the H-shaped (AB)₂C(BA)₂ and the linear ABCBA block copolymer is very helpful in our understanding of the effect of molecular architecture on morphology.
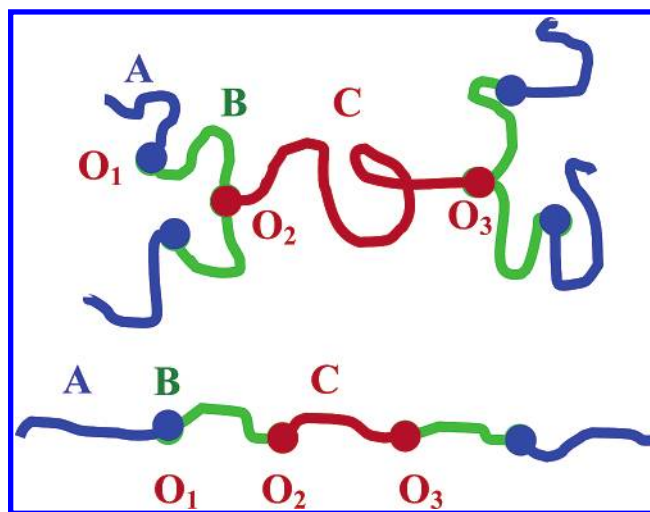
* Corresponding author. E-mail: zysun@ciac.jl.cn (Z.S.); ljan@ciac.jl.cn (L.A.). Tel: 86-431-5262896 (Z.S.); 86-431-5262206 (L.A.). Fax: 86-431-5262969 (Z.S.); 86-431-5685653 (L.A.).
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**Figure 1.** Molecular architectures of the H-shaped (AB)₂C(BA)₂ block copolymer and the linear ABCBA block copolymer.

The self-consistent field theory (SCFT), which is the most accurate mean-field theory for polymers, has been widely used to investigate and screen the microphase structures of block copolymers.[13−21] There are two general classes of techniques for solving the self-consistent field equations for block copolymers. The first is the spectral method, which was suggested by Matsen and Schick in 1994,[13−16] but this method needs a prior assumption of symmetry of the ordered structure. Another one is the real space method, which involves the direct implementation of SCFT for polymers in real space in an adaptive arbitrary cell, which does not require the assumption of the microphase symmetry.[17−19] In this work, we use the combinatorial screening method[17−19] based on the self-consistent field theory to study the equilibrium morphologies of H-shaped (AB)₂C(BA)₂ and

linear ABCBA block copolymers. Moreover, the order−order transitions driven by the architectural changing are investigated.

## Theoretical Method

We consider a melt of $n$ H-shaped $(AB)_2C(BA)_2$ block copolymers and that of $n$ linear ABCBA block copolymers, respectively. Each polymer is composed of $N$ segments, and $f_A N$, $f_B N$, and $f_C N$ form the A, B, and C blocks, respectively. The free energy of the system is given by

$$\frac{F}{nk_BT} = -\ln\left(\frac{Q}{V}\right) + \frac{1}{V}\int d\mathbf{r}[\chi_{AB}N(\phi_A(\mathbf{r}) - f_A)(\phi_B(\mathbf{r}) - f_B) +$$

$$\chi_{BC}N(\phi_B(\mathbf{r}) - f_B)(\phi_C(\mathbf{r}) - f_C) + \chi_{AC}N(\phi_A(\mathbf{r}) - f_A)(\phi_C(\mathbf{r}) - f_C) - W_A(\mathbf{r})(\phi_A(\mathbf{r}) - f_A) - W_B(\mathbf{r})(\phi_B(\mathbf{r}) - f_B) - W_C(\mathbf{r})(\phi_C(\mathbf{r}) - f_C) - P(\mathbf{r})(1 - \phi_A(\mathbf{r}) - \phi_B(\mathbf{r}) - \phi_C(\mathbf{r}))] \quad (1)$$

where $V$ is the volume of the system; $\phi_A$, $\phi_B$, and $\phi_C$ are the monomer density fields normalized by the local volume fractions of A, B, and C, respectively; $\chi_{AB}$, $\chi_{BC}$, and $\chi_{AC}$ are Flory−Huggins interaction parameters between different species; $P(\mathbf{r})$ is the potential field that ensures the incompressibility of the system, $W_\alpha(\mathbf{r})$ is the self-consistent field that conjugates to the density field $\phi_\alpha$ of the block species $\alpha$, and $Q = \int d\mathbf{r}q_{\alpha 0}(\mathbf{r},s)\cdot q_{\alpha 1}(\mathbf{r},f_\alpha N)$ is the partition function of a single chain in the effective field $W_\alpha(\mathbf{r})$. The end-segment distribution function $q_{\alpha\beta}(\mathbf{r},s)$, where $\alpha = $ A, B, C and $\beta = $ 0, 1, gives the probability that a section of a chain, of contour length $s$ and containing a free chain end, has its "connected end" located at $\mathbf{r}$ and satisfies the modified diffusion equation (eq 2). $\beta = 0$ corresponds to $s$ of $q_{\alpha\beta}(\mathbf{r},s)$ increasing from the free end to point $\mathbf{O}_1$, or from point $\mathbf{O}_1$ to point $\mathbf{O}_2$, or from point $\mathbf{O}_2$ to point $\mathbf{O}_3$ (see Figure 1), $\beta = 1$ corresponds to $s$ of $q_{\alpha\beta}(\mathbf{r},s)$ increasing reversely. It should be noted that $q_{c0}(\mathbf{r},s) = q_{c1}(\mathbf{r},s)$ because of the symmetry of the molecular architecture. The modified diffusion equation reads

$$\frac{\partial q_{\alpha\beta}(\mathbf{r},s)}{\partial s} = \nabla^2 q_{\alpha\beta}(\mathbf{r},s) - W_\alpha(\mathbf{r})q_{\alpha\beta}(\mathbf{r},s) \quad (2)$$

Equation 2 is solved by an alternating direction implicit method.[22] The boundary conditions are periodic and the initial conditions of $q_{\alpha\beta}(\mathbf{r},s)$ are as follows: For H-shaped $(AB)_2C(AB)_2$ block copolymers

$$q_{A0}(\mathbf{r},0) = 1$$

$$q_{B0}(\mathbf{r},0) = q_{A0}\left(\mathbf{r},\frac{1}{4}f_A N\right)$$

$$q_{C0}(\mathbf{r},0) = q_{B0}\left(\mathbf{r},\frac{1}{4}f_B N\right)q_{B0}\left(\mathbf{r},\frac{1}{4}f_B N\right)$$

$$q_{B1}(\mathbf{r},0) = q_{B0}\left(\mathbf{r},\frac{1}{4}f_B N\right)q_{C0}(\mathbf{r},f_C N)$$

$$q_{A1}(\mathbf{r},0) = q_{B1}\left(\mathbf{r},\frac{1}{4}f_B N\right)$$

and for linear ABCBA block copolymers

$$q_{A0}(\mathbf{r},0) = 1$$

$$q_{B0}(\mathbf{r},0) = q_{A0}\left(\mathbf{r},\frac{1}{4}f_A N\right)$$

$$q_{C0}(\mathbf{r},0) = q_{B0}\left(\mathbf{r},\frac{1}{4}f_B N\right)$$

$$q_{B1}(\mathbf{r},0) = q_{C0}(\mathbf{r},f_C N)$$

$$q_{A1}(\mathbf{r},0) = q_{B1}\left(\mathbf{r},\frac{1}{4}f_B N\right)$$

which are similar to those in ref 9, 11, and 12.

Expressed in terms of $q_{\alpha\beta}(\mathbf{r},s)$, the densities of the A, B, and C monomers for H-shaped $(AB)_2C(AB)_2$ block copolymers are

$$\phi_A(\mathbf{r}) = 4\frac{V}{NQ}\int_0^{(1/4)f_{A1}N} ds q_{A0}(\mathbf{r},s)q_{A1}(\mathbf{r},f_{A1}N - s) \quad (3)$$

$$\phi_B(\mathbf{r}) = 4\frac{V}{NQ}\int_0^{(1/4)f_BN} ds q_{B0}(\mathbf{r},s)q_{B1}(\mathbf{r},f_BN - s) \quad (4)$$

$$\phi_C(\mathbf{r}) = \frac{V}{NQ}\int_0^{(1/4)f_CN} ds q_{C0}(\mathbf{r},s)q_{C0}(\mathbf{r},f_CN - s) \quad (5)$$

and for linear ABCBA block copolymers, the densities are

$$\phi_A(\mathbf{r}) = 2\frac{V}{NQ}\int_0^{(1/2)f_{A1}N} ds q_{A0}(\mathbf{r},s)q_{A1}(\mathbf{r},f_{A1}N - s) \quad (6)$$

$$\phi_B(\mathbf{r}) = 2\frac{V}{NQ}\int_0^{(1/2)f_BN} ds q_{B0}(\mathbf{r},s)q_{B1}(\mathbf{r},f_BN - s) \quad (7)$$

$$\phi_C(\mathbf{r}) = \frac{V}{NQ}\int_0^{(1/2)f_CN} ds q_{C0}(\mathbf{r},s)q_{C0}(\mathbf{r},f_CN - s) \quad (8)$$

Minimizing the free energy in eq (1) with respect to $W_A$, $W_B$, $W_C$, and $P$ leads to the following self-consistent field equations that describe the equilibrium morphology

$$W_A(\mathbf{r}) = \chi_{AB}N(\phi_B(\mathbf{r}) - f_B) + \chi_{AC}N(\phi_C(\mathbf{r}) - f_C) + P(\mathbf{r}) \quad (9)$$

$$W_B(\mathbf{r}) = \chi_{AB}N(\phi_A(\mathbf{r}) - f_A) + \chi_{BC}N(\phi_C(\mathbf{r}) - f_C) + P(\mathbf{r}) \quad (10)$$

$$W_C(\mathbf{r}) = \chi_{AC}N(\phi_A(\mathbf{r}) - f_A) + \chi_{BC}N(\phi_B(\mathbf{r}) - f_B) + P(\mathbf{r}) \quad (11)$$

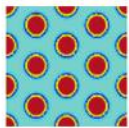$$\phi_A + \phi_B + \phi_C = 1 \quad (12)$$

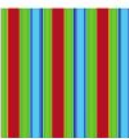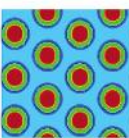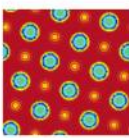We solve the above self-consistent equations directly in real space by using a combinatorial screening algorithm proposed by Drolet and Fredrickson.[17,18] Each iterative procedure continues until the free energy converges to a local minimum, which corresponds to a stable or metastable state. Each minimization is repeated several times using different initial conditions, and we obtain the most stable morphology by comparing the free energies.

For the sake of numerical tractability, the implementation of the self-consistent field equations is carried out in a two-dimensional $L_x \times L_y$ cell with periodic boundary conditions.

## Results and Discussion

To focus on the effect of the block copolymer architectures on the phase behavior, we set the interactions between the different species to be equal, i.e., $\chi_{AB}N = \chi_{BC}N = \chi_{AC}N = 60.0$. By systematically changing the volume fractions of the A, B, and C blocks, we can construct the component triangle phase diagrams in the entire range of the copolymer composition. At

**TABLE 1: Summary of Observed Equilibrium Morphologies**

| Name | Abbreviation | Graphic |
|---|---|---|
| hexagonal lattice phase | HEX | |
| lamellar phase | LAM | |
| core shell hexagonal lattice phase | CSH | |
| two interpenetrating tetragonal lattice | TET2 | |



**Figure 2.** Phase diagram of linear ABCBA block copolymer for $\chi_{AB}N = \chi_{BC}N = \chi_{AC}N = 60.0$.

each grid point, the most stable morphology is obtained with the method described in the above section. Table 1 shows all the observed ordered equilibrium microphase structures that are found for the H-shaped $(AB)_2C(BA)_2$ and the linear ABCBA block copolymers. We can see that for the linear ABC block copolymer,[8] the H-shaped $A_2BC_2$ block copolymer,[12] the H-shaped $(AB)_2C(BA)_2$ block copolymer, and the linear AB-CBA block copolymer, when the interactions between different species are equal, only four stable morphologies are found, i.e., the lamellar phase (LAM), the hexagonal lattice phase (HEX), the core−shell hexagonal lattice phase (CSH), and the two interpenetrating tetragonal lattice phase (TET2).

Figure 2 gives the phase diagram of linear ABCBA block copolymer for $\chi_{AB}N = \chi_{BC}N = \chi_{AC}N = 60.0$. The triangle phase diagram shows no A−C reflection symmetry, which is different from that of linear ABC block copolymer and H-shaped $A_2$-$BC_2$ block copolymer.[8,12] However, by scrutinizing the phase diagram, we can see it actually possesses A−C reflection symmetry except for eight grid points, which are $(f_A, f_B, f_C) \in$ {(0.1, 0.5, 0.4), (0.1, 0.6, 0.3), (0.1, 0.7, 0.2), (0.2,0.3,0.5), (0.2, 0.7, 0.1), (0.3, 0.6, 0.1), (0.4, 0.5, 0.1), (0.5, 0.3, 0.2)}. At the grid points $(f_A, f_B, f_C) \in$ {(0.1, 0.5, 0.4), (0.1, 0.6, 0.3), (0.1, 0.7, 0.2)}, the morphology is HEX, in which the C blocks form the lattices, the B blocks form the matrix, and the A blocks dissolve in the matrix. This phase behavior is the same as that of the H-shaped $A_2BC_2$ block copolymer.[12] In the asymmetric ABA block copolymer, both theory and experiments have shown that when the asymmetry becomes sufficiently large, short A blocks begin to pull out of their domains. Although unfavorable interactions occur when an A block leaves its domain, this is more than compensated for by the fact its B block can relax.[23,24] At the grid points $(f_A, f_B, f_C) \in$ { (0.2, 0.7, 0.1), (0.3, 0.6, 0.1), (0.4, 0.5, 0.1)}, the morphology is TET2, in which A blocks and C blocks form the lattices, respectively, and B blocks form
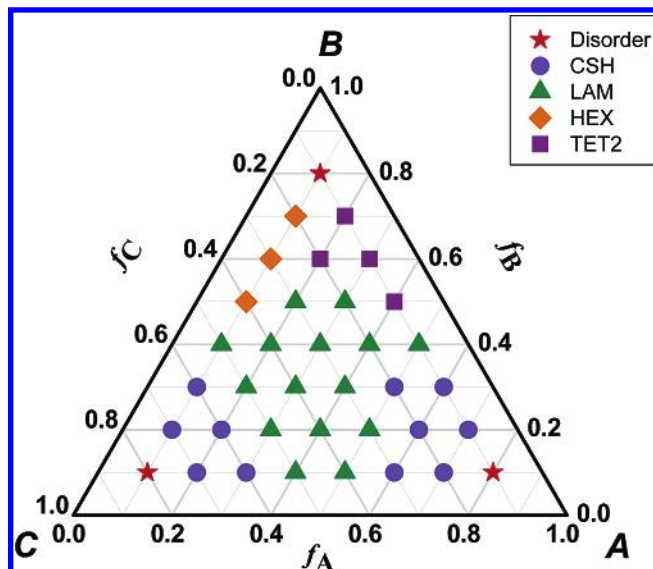
the matrix. At the grid point $(f_A, f_B, f_C) \in$ { (0.2, 0.3, 0.5)}, the morphology is LAM, and at the grid point $(f_A, f_B, f_C) \in$ { (0.5, 0.3, 0.2)}, the morphology is CSH. The asymmetry of linear ABCBA is attributed to the specific chain architecture, i.e., block C joins two B blocks together and there is no free end of block C, which is different from block A. This leads to the different contributions of block C and block A in microphase separation of the linear ABCBA block copolymer. However, when the length of block C is long enough, block C and block A will have similar contributions to the phase behavior of linear ABCBA block copolymer.

Near corner *A* and corner *C* in Figure 2, the CSH morphology is found. However, the core of the CSH is made of block A near corner *C*, whereas it is made of block C near corner *A*. The region of the CSH morphology near corner *C* is smaller than that near corner *A*, which can be attributed to block A having one free end and the length of block A being half that of block C; therefore, block C more easily forms cores of CSH than does block A.

Figure 3 gives the phase diagram of H-shaped $(AB)_2C(BA)_2$ block copolymer. Here, we again choose the interaction parameters between the different species to be equal, i.e., $\chi_{AB}N = \chi_{BC}N = \chi_{AC}N = 60.0$. Apparently, the phase behavior of H-shaped $(AB)_2C(BA)_2$ block copolymer is quite different from that of linear ABCBA block copolymer. For the H-shaped block copolymer, there is no reflection symmetry in the triangle phase diagram. In the center region of Figure 3, the CSH morphology is found. Moreover, most of the grid points are occupied by the CSH morphology. However, we can find in Figure 2 that most of the grid points are occupied by the LAM morphology. This difference in the dominant morphology is attributed to the difference in chain architectures. For the H-shaped $(AB)_2C$-$(BA)_2$ block copolymer, formation of the CSH morphology helps relieve the interfacial crowding,[3,4] as shown in Figure 4. Experimentally, Chen and co-workers found that the change in morphology was due to different interfacial crowding of different chain architecture.[25,26]

Near corner *B* in Figure 3, disorder morphology is found for many grid points. In contrast, the grid points are occupied by the TET2 morphology near corner *B* in Figure 2. This result implies that the H-shaped $(AB)_2C(BA)_2$ block copolymer has little tendency to segregate in these grid points because the
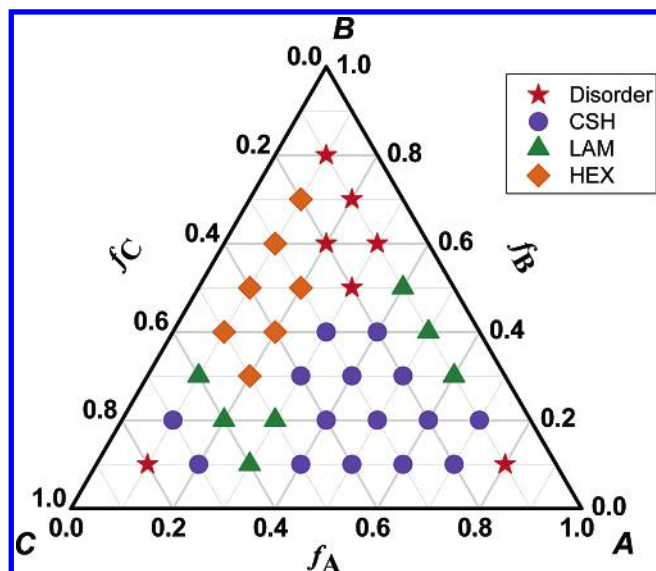
**Figure 3.** Phase diagram of H-shaped $(AB)_2C(BA)_2$ block copolymer for $\chi_{AB}N = \chi_{BC}N = \chi_{AC}N = 60.0$.
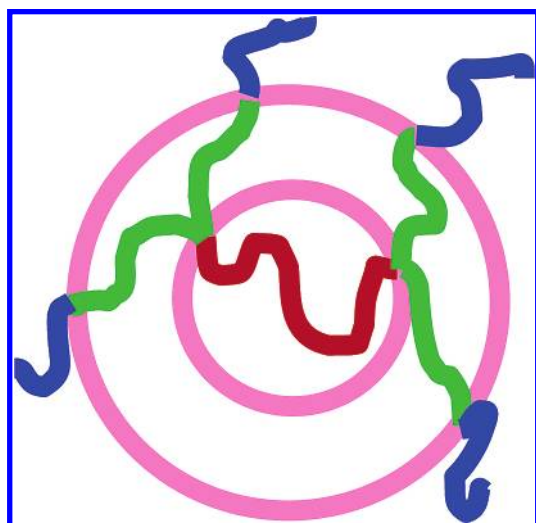


**Figure 4.** Schematic representation of the composition of the core and the shell of CSH morphology for the H-shaped $(AB)_2C(BA)_2$ block copolymer.

disorder state has lower free energy. This is the result of the competition between chain stretching and interfacial crowding.[3,4] If the block C in this region composes the lattice, it will greatly decrease the stretching space of the block C and B, which is disadvantageous to lowering the energy of the system. In other words, this result can be attributed to the lower entropy of H-shaped $(AB)_2C(BA)_2$ because of the junction constraint.[25,26]

Near corner **C** of Figure 3, the CSH and LAM morphologies are found. The CSH morphology in this region is different from that near corner **A**. For the CSH morphology near corner **C**, block A forms the core and block B forms the shell, whereas ear corner **A**, block A forms the matrix, block B forms the shell, and block C forms the core of the CSH morphology. Compared with Figure 2, the region of CSH morphology in Figure 3 is decreased, and parts of the region of CSH morphology in Figure 3 are replaced by the LAM morphology. This can be attributed to the CSH morphology limiting the relaxation of block B in the interface between the C- and B-rich regions. This result can also help us understand that most of the grid points of LAM morphology in Figure 2 are replaced by CSH morphology in Figure 3, in which block C forms the core, block B forms the shell, and block A forms the matrix.

Near edge **BC** of Figure 3, where $f_A = 0.10$ or $0.20$, most of the grid points are occupied by the HEX morphology, and the region of HEX morphology is larger than that in Figure 2. In this HEX morphology, block C forms the lattices, block B forms the matrix, and block A dissolves in the matrix, which is the same as that in Figure 2. At the same grid points, the length of block A of H-shaped $(AB)_2C(BA)_2$ block copolymer is half of that of linear ABCBA block copolymer. Thus, the block A of H-shaped $(AB)_2C(BA)_2$ block copolymer is favored to dissolve in the matrix formed by block B. This chain architecture difference between H-shaped $(AB)_2C(BA)_2$ and linear ABCBA block copolymer can also explain why the LAM morphology is found at higher concentrations of block C for H-shaped $(AB)_2C(BA)_2$ block copolymer.

In other words, H-shaped $(AB)_2C(BA)_2$ block copolymer and linear ABCBA block copolymer have different phase behaviors with equal interaction parameters, because they have different junction constraint and different chain crowding at the interface, which cause the system to adjust the morphologies to lower the overall free energy.[1−4,12,25,26]

## Conclusions

By using a combinatorial screening method based on the self-consistent field theory, we investigate the morphologies of linear ABCBA and H-shaped $(AB)_2C(BA)_2$ block copolymer in two dimensions. Systematically varying the volume fractions of block A, B, and C, we constructed the triangle phase diagrams of linear ABCBA and H-shaped $(AB)_2C(BA)_2$ block copolymer. In our study, we set the interaction energies between the A, B, and C species to be equal, and obtain four different morphologies, LAM, HEX, CSH, and TET2. For the linear ABCBA block copolymer, there is reflection symmetry in the phase diagram except for some special grid points. This implies that block A and block C behave similarly to some extent. For the linear ABCBA block copolymer, most of the grid points of the triangle phase diagram are occupied by LAM morphology, but for the H-shaped $(AB)_2C(BA)_2$ block copolymer, most of the grid points of the triangle phase diagram are occupied by CSH morphology because of different chain architectures. These results may help the design and the synthesis of block copolymers with different microstructures.

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