

SELF-ASSEMBLY OF LINEAR ROD-COIL MULTIBLOCK COPOLYMERS*

Ying-dong Xia, Ji-zhong Chen**, Tong-fei Shi and Li-jia An

State Key Laboratory of Polymer Physics and Chemistry, Changchun Institute of Applied Chemistry,
Chinese Academy of Sciences, Changchun 130022, China

Abstract The self-assembly of the linear rod-coil multiblock copolymers is studied by applying self-consistent-field lattice techniques in a three-dimensional (3D) space. Compared to the copolymer with one rod, the copolymer with more rods ($m_{\text{rod}} \geq 2$) exhibits rich order-order phase transitions with increasing temperature, where the ordered morphology changes from strips to perforated lamellae and finally to lamellae. In addition, taking the copolymer with $m_{\text{rod}} = 2$ as a representative, we further study the effects of the volume fractions of the rods, the spacer coils and the end coils on the phase behaviors respectively, by which the detailed self-assembled mechanism of the linear rod-coil multiblock copolymers is revealed. Our results are expected to provide guidance for the design of the rod-coil materials.

Keywords: Rod-coil multiblock copolymer; Self-assembly; Self-consistent-field lattice.

INTRODUCTION

Rod-coil block copolymers have attracted a great deal of attention due to their ability to self-assemble 10–100 nm length scale structures in the field of semiconducting polymers^[1–4], biological molecules^[5, 6] and high-performance polymer resins^[7, 8]. In contrast to pure rodlike polymers whose solutions and melts exhibit liquid crystalline phase such as nematic and/or layered smectic structures, the covalent linkage of a different class of a coil block to the rodlike polymer results in both liquid crystalline alignment of the rods and the microseparation between rods and coils. In the past decades, many interesting microstructures have been observed for the rod-coil block copolymers, including zigzag, arrowhead, and wave lamellae, honeycombs, and hollow spherical and cylindrical micelles^[9–12], which can be controlled by varying the volume fraction of coil blocks or architecture of rod-coil block copolymer chains.

Theoretical and computational works are approved to be a powerful tool for better understanding the relationship between self-assembled structures and chain architecture. The agreement between theoretical predictions and experiments provides guidance for synthesis of rod-coil block copolymers. For example, Glotzer *et al.*^[13, 14] used Brownian dynamics (BD) to study the T-shaped rod-coil block copolymers in solvent. They found new self-assembled characteristics that stepped strips and bilayer sheets are formed due to the middle-positioned anchor point, and the bilayer sheets can twist into different helical structures dependent on solvent conditions. Another effective way to achieve the rod-coil molecular design is provided by transforming the rod-coil diblock copolymer into a coil-rod-coil triblock copolymer. Van Duijneveldt *et al.*^[15] addressed the

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** Corresponding author: Ji-zhong Chen (陈继忠), E-mail: jzchen@ciac.jl.cn

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effect of steric effects of the coil-rod-coil triblock copolymers arising from flexible terminal chains on smectic stability with Monte Carlo (MC) method, revealing that the addition of the flexible tails stabilizes the smectic-A phase and suppresses the nematic phase. Alsunaidi *et al.*^[16] presented a comparison in the self-assembly of rod-coil diblock and coil-rod-coil triblock copolymers using dissipative particle dynamics (DPD) simulation method and found that the coil-rod-coil triblock copolymer has higher order-disorder transition temperature (T_{ODT}) and the smectic-C is preferred to the smectic-A with respect to rod-coil diblock copolymers. Our group^[17] conducted the self-assembly of coil-rod-coil triblock copolymer melts by self-consistent field theory (SCFT) lattice model, by which the effects of high grafting density at rod and coil interface on self-assembly are demonstrated. A more complicated example of designing the rod-coil molecules can be accessed by linear incorporation of many rod and coil blocks into a polymer chain. Such materials are proved to perform promising materials in area of thermoplastic elastomers, blend compatibilizers and barriers^[18]. However, due to the considerable coupling problems in the equilibrating structures, the simulation on the linear rod-coil multiblock copolymers are less extensively studied^[19–22] and more efforts are needed.

In our previous work^[23], we developed a SCFT lattice model to describe the self-assembly of rod-coil diblock copolymer melts in a three-dimensional space. The main feature of this model is that no liquid crystalline interactions involved and the rods in the self-assembled structures are aligned due to steric requirements, which is facilitate to deal with complex rod-coil block copolymers. With this model, we have investigated the influence of the chain architecture of rod-coil block copolymers on self-assembly, such as coil-rod-coil triblock^[17] and rod-coil-rod triblock^[24] copolymers. Herein we extend our method to the linear rod-coil multiblock copolymers. Besides, the architecture of the linear rod-coil multiblock copolymer is similar to main-chain liquid crystalline polymers (MCLCPs)^[25], and thus our findings are expected to shed some light on the research of the self-assembly of the MCLCPs.

In this paper, the linear rod-coil multiblock copolymer is composed by three parts, *i.e.*, the rods, the spacer coils and the end coil blocks, as shown in Fig. 1. We first concentrate on the effect of the number of the rod of the linear rod-coil multiblock copolymer on self-assembly. To understand the influence of the three parts of the linear rod-coil multiblock copolymer on self-assembly, we then respectively vary the volume fractions of the rods, the spacer coils and the end coils, and the phase diagrams are correspondingly presented in f_{rod} , f_{spacer} and f_{end} versus χN .



Fig. 1 Schematic representation of the linear rod-coil multiblock copolymer

THEORY

We consider a molten system of m linear rod-coil multiblock copolymer chains each of n repeated unit (Fig. 1). The number of segment A containing in each rod is denoted as L_{rod} , and the number of segment B in each spacer coil and each end coil block is denoted as L_{spacer} and L_{end} respectively. We assume the A and B segments have the same size and each segment occupies one lattice site. Thus the total number of lattice sites is $N_L = m[L_{rod}(n+1) + L_{spacer}n + 2L_{end}]$. For convenience, we use the segment size of block copolymer as the length unit. The transfer matrix λ is used to describe the polymer chain, which depends only on the chain model used. For a rod-coil system, we assume that

$$\lambda_{r_{j,s}-r'_{j,s-1}}^{\alpha_{j,s}-\alpha'_{j,s-1}} = \begin{cases} 1, & \alpha_{j,s} = \alpha'_{j,s-1} \\ 0, & otherwise \end{cases} \quad (1)$$

for the rod section and

$$\lambda_{r_{j,s}-r'_{j,s-1}}^{\alpha_{j,s}-\alpha'_{j,s-1}} = \begin{cases} 0, & \alpha_{j,s} = -\alpha'_{j,s-1} \\ 1/(z-1), & \text{otherwise} \end{cases} \quad (2)$$

for the coil section, which represent a self-avoiding chain. Here, the $r_{j,s}$ and $\alpha_{j,s}$ denote the position and bond orientation of the s th segment of the j th copolymer, respectively. The r' denotes the nearest neighboring site of r . The α can be any of the allowed bond orientations depending on the lattice model used. We use the cubic lattice model which has six bond orientations and z is the coordination number of the lattice. Following the scheme of Scheutjens and Leermakers^[26], the end segment distribution function $G^{\alpha_s}(r, s|1)$ that gives the statistical weight of all possible walks starting from segment 1, which may locate anywhere in the lattice, and ending at segment s at site r , is evaluated from the following recursive relation:

$$G^{\alpha_s}(r, s|1) = G(r, s) \sum_{r'_{s-1}} \sum_{\alpha_{s-1}} \lambda_{r_s-r'_{s-1}}^{\alpha_s-\alpha_{s-1}} G^{\alpha_{s-1}}(r', s-1|1). \quad (3)$$

The initial condition is $G^{\alpha_1}(r, 1|1) = G(r, 1)$ for all the values of α_1 . The $G(r, s)$ is the free segment weighting factor, $G(r, s) = \begin{cases} \exp(-\omega_A(r_s)), & s \in A \\ \exp(-\omega_B(r_s)), & s \in B \end{cases}$, where ω_A and ω_B are the chemical potential fields for block species A and B , respectively. Another end segment distribution function $G^{\alpha_s}(r, s|N)$ is evaluated from the following recursive relations:

$$G^{\alpha_s}(r, s|N) = G(r, s) \sum_{r'_{s+1}} \sum_{\alpha_{s+1}} \lambda_{r'_{s+1}-r_s}^{\alpha_{s+1}-\alpha_s} G^{\alpha_{s+1}}(r', s+1|N) \quad (4)$$

with the initial conditions $G^{\alpha_N}(r, N|N) = G(r, N)$ for all the values of α_N . The free energy function of F (in the unit of k_{BT}) can be expressed as^[23]

$$F = \sum_r \left\{ \frac{\chi}{z} \sum_{r'} \phi_A(r) \phi_B(r') - \omega_A(r) \phi_A(r) - \omega_B(r) \phi_B(r) - \xi(r) [1 - \phi_A(r) - \phi_B(r)] \right\} - n \ln Q, \quad (5)$$

where

$$Q = \frac{1}{N_L} \frac{1}{z} \sum_{r_N} \sum_{\alpha_N} G^{\alpha_1}(r, 1|N) \quad (6)$$

is the single-chain partition function. Here, χ is the Flory-Huggins interaction parameter which measures the incompatibility between A and B segments. The $\phi_k(r)$ is the volume fraction field of block species k and conjugated to $\omega_k(r)$, which is independent of the individual polymer configuration. The $\xi(r)$ is the potential field that ensures the incompressibility of the system, also known as a Lagrange multiplier. Minimizing the free energy function F with respect to $\phi_A(r)$, $\phi_B(r)$, $\omega_A(r)$, $\omega_B(r)$ and $\xi(r)$ leads to the following SCFT equations:

$$\omega_A(r) = \frac{\chi}{z} \sum_{r'} \phi_B(r') + \xi(r), \quad (7)$$

$$\omega_B(r) = \frac{\chi}{z} \sum_{r'} \phi_A(r') + \xi(r), \quad (8)$$

$$\phi_A(r) = \frac{1}{N_L} \frac{1}{z} \frac{n}{Q} \sum_{s \in A} \sum_{\alpha_s} \frac{G^{\alpha_s}(r, s|1) G^{\alpha_s}(r, s|N)}{G(r, s)}, \quad (9)$$

$$\phi_B(r) = \frac{1}{N_L} \frac{1}{z} \frac{n}{Q} \sum_{s \in B} \sum_{\alpha_s} \frac{G^{\alpha_s}(r, s|1) G^{\alpha_s}(r, s|N)}{G(r, s)}, \quad (10)$$

$$\phi_A(r) + \phi_B(r) = 1 \quad (11)$$

In our calculations, the real space method is implemented to solve the SCF equations in a cubic lattice with periodic boundary conditions. To prevent bias of the resulting morphologies, our calculations are initiated with different random fields. The calculation stops when the free energy changes within a tolerance of 10^{-8} . The morphologies obtained correspond to either a stable or a metastable state. System sizes range from 40^3 to 80^3 to minimize the effects of simulation box size on the results. By comparing the free energy of the system, the relative stability of the morphologies can be assessed.

RESULTS AND DISCUSSION

There are a wide range of parameters which can be explored in the self-assembly of the linear rod-coil multiblock copolymer system. For this report we confine ourselves to study the effects of the number of the rod in the polymer chain and the component parts of the copolymer, including the rods, the spacer coils and the end coils, on the self-assembly. For convenience, the linear rod-coil multiblock copolymer studied here is symmetric, *i.e.*, the length of the rod, the length of the spacer coil, and the length of the end coils are kept equal respectively. Thus, the ordered morphologies of the linear rod-coil multiblock copolymer depend on four tunable molecular parameters: m_{rod} (the number of the rod in the copolymer), f_{rod} (the volume fraction of the rod in the copolymer), f_{spacer} (the volume fraction of the spacer coil in the copolymer) and χN (the parameter describing the degree of microseparation of the copolymer). Simulation boxes with different sizes are used to make sure that the emergence of the self-assembled structures are not constrained by system size, and the simulations start from both ordered and random configurations to ensure that the observed structure are not kinetically arrested but equilibrium structures. The ordered morphologies observed in our calculation are presented in Fig. 2. Below we proceed with a detailed discussion of the simulation results.

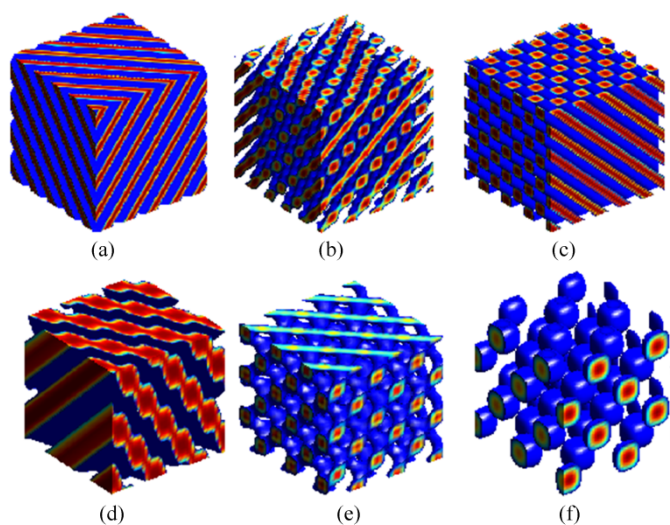


Fig. 2 Ordered morphologies obtained in the present work: (a) lamella, (b) perforated lamella, (c) strip, (d) lamellar strip, (e) gyroid and (f) micelle

We first concentrate on the effect of the number of the rod (m_{rod}) in the linear rod-coil multiblock copolymers on the self-assembly. The copolymers with $m_{\text{rod}} = 1, 2, 3$ and 4 are studied corresponding to the linear rod-coil triblock, pentablock, heptablock and nonablock copolymers. Herein the lengths of the rod, the spacer coil, and the end coil are kept as $L_{\text{rod}} = 6$, $L_{\text{spacer}} = 4$ and $L_{\text{end}} = 4$ for all the copolymers. As shown in Fig. 3, three morphologies are formed, *i.e.*, lamella, perforated lamella and strip (shown in Fig. 2). The notable feature of the phase diagram is that the rod-coil block copolymers with $m_{\text{rod}} = 2, 3$ and 4 exhibit the same phase behavior, which, however, differ from that of the copolymer with only one rod ($m_{\text{rod}} = 1$). When $m_{\text{rod}} = 1$, the

strip structure is stable. Further calculations^[23] prove that the rods in the strip are packed in an interdigitated fashion, which is rationalized to arise from the competition between the rods attempting to maximize their contact with other rods to minimize energy and the coil blocks attempting to maximize their free volume to maximize entropy. When m_{rod} increases, rich order-order phase transitions (OOT) are observed that the ordered morphologies transform from strip to perforated lamella and finally to lamella with increasing temperature. The rods of the copolymer are identified to orient in a common direction in the lamella and the perforated lamella^[23]. Considering the architecture of the rod-coil multiblock copolymer with more rods ($m_{\text{rod}} \geq 2$), besides the end coil blocks (coil blocks tethered to the ends of the rods), the spacer coil also exists (coil blocks between the adjacent rods) which results in jamming between adjacent rods in self-assembly similar to that of rod-coil-rod triblock copolymers^[24, 27, 28]. When the temperature is low, the entropy penalty associated with the stretching of the spacer coils is alleviated for the copolymers with $m_{\text{rod}} \geq 2$, allowing the strip structure stable. As the temperature increased, strong entropy penalty of the spacer coils is suffered and the rods tend to rearrange themselves from the interdigitated fashion to a common direction to lower the system energy, leading to the formation of the lamellar structure. The perforated lamella can be considered as a compromise between the strip and the lamella. For the copolymers with $m_{\text{rod}} \geq 2$ exhibit the same phase behaviors, we choose the one with $m_{\text{rod}} = 2$ as a representative in view of economic computation to conduct the following studies, *i.e.*, the effects of the volume fractions of the rods, the spacer coils and end coils on the phase behaviors respectively. The variations of the length of each part and the corresponding volume fractions are presented in Table 1.

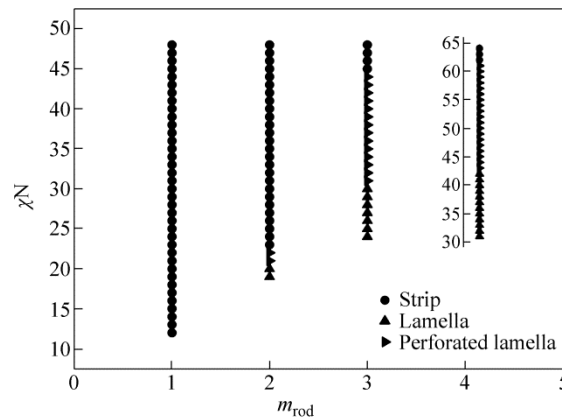


Fig. 3 Phase diagram for the linear rod-coil multiblock copolymer with the number of the rod $1 \leq m_{\text{rod}} \leq 4$

Table 1. The variations of the length of each part of the linear rod-coil pentablock copolymer and the corresponding volume fractions

(a)	L_{rod}	L_{spacer}	L_{end}	f_{rod}	(b)	L_{rod}	L_{spacer}	L_{end}	f_{spacer}	(c)	L_{rod}	L_{spacer}	L_{end}	f_{end}
	4	4	4	0.4		6	2	4	0.09		6	4	2	0.22
	6	4	4	0.5		6	4	4	0.17		6	4	4	0.33
	8	4	4	0.57		6	6	4	0.23		6	4	6	0.43
	10	4	4	0.63		6	8	4	0.33		6	4	8	0.50

To study the impact of the rod volume fraction on the self-assembly, we vary the length of the rods of the rod-coil pentablock copolymer ($m_{\text{rod}} = 2$) from 4 to 10 corresponding to $0.4 \leq f_{\text{rod}} \leq 0.63$, as shown in Table 1(a). Five ordered morphologies are formed, *i.e.*, strip, lamella, perforated lamella, lamellar strip and gyroid (shown in Fig. 2), and their distributions in the phase diagram are presented in Fig. 4. When the rod volume fraction is low, *i.e.*, $0.4 \leq f_{\text{rod}} \leq 0.5$, the stretching of the spacer coil is mitigated and the strip structure is stable. When the rod volume fraction is increased, the spacer coil would suffer entropy penalty if the rods of the copolymer took the way as that in the strip, which induces the rods to rearrange to decrease the system energy. As shown in Fig. 4, the OOT takes place that the ordered morphologies transform from the strip to the perforated lamella and finally

to the lamella at $f_{\text{rod}} = 0.5$, and from the strip to the lamellar strip and finally to the lamella at $f_{\text{rod}} = 0.57$ with increasing temperature, corresponding to the rods packing from the interdigitated fashion to a common direction. Further increasing the rod volume fraction, *i.e.*, $0.57 < f_{\text{rod}} \leq 0.63$, the strip disappears even at low temperatures, where the lamellar strip is stable with the lamella and the gyroid appearing subsequently with the temperature increased.

By changing the length of the spacer coils of the rod-coil pentablock copolymer, we study the influence of the volume fraction of the spacer coils on the self-assembly. The variations are shown in Table 1(b), and the phase diagram with f_{spacer} versus χN is presented in Fig. 5. Five morphologies are formed, *i.e.*, lamella, perforated lamella, lamellar strip, strip and gyroid, as shown in Fig. 2. When the volume fraction of the spacer coil is low, *i.e.*, $0.09 \leq f_{\text{spacer}} \leq 0.17$, the adjacent rods connected by the spacer coils are strongly constrained which results in the entropy penalty of the spacer coil in the self-assembly, and the changed packing fashions of the rods are expected. For example, when $f_{\text{spacer}} = 0.17$, the strip transforms to the perforated lamella and finally to lamella with increasing temperature. When the volume fraction of the spacer coil is further decreased, *i.e.*, $f_{\text{spacer}} < 0.17$, rich OOT behaviors emerge that the perforated lamella evolves to the lamellar strip, to the gyroid and finally to the lamella with increasing temperature. When the volume fraction of the spacer coils is high, *i.e.*, $0.17 < f_{\text{spacer}} \leq 0.33$, no OOT is observed in our calculations and the strip structure is stable, which suggests that the entropy penalty associated with the spacer coil can be neglected.

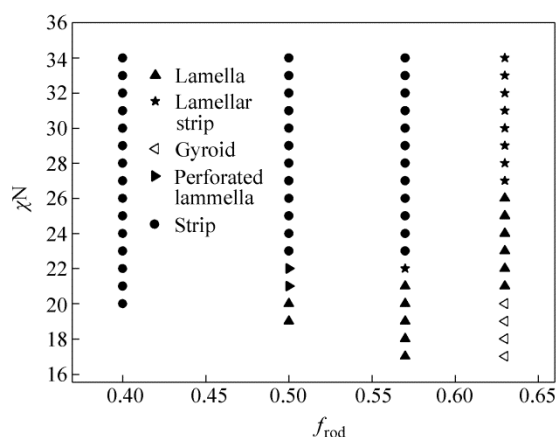


Fig. 4 Phase diagram for the linear rod-coil pentablock copolymer with the volume fraction of the rod $0.4 \leq f_{\text{rod}} \leq 0.63$

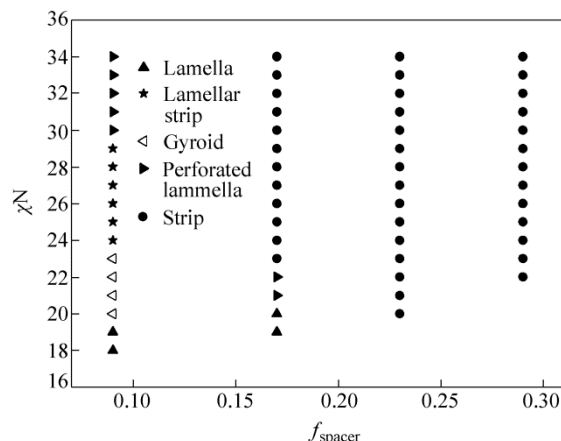


Fig. 5 Phase diagram for the linear rod-coil pentablock copolymer with the volume fraction of the coil spacer $0.09 \leq f_{\text{spacer}} \leq 0.33$

We finally study the influence of the volume fraction of the end coils on the self-assembly by varying the length of the end coils of the rod-coil pentablock copolymer. The variations are shown in Table 1(c). As shown in Fig. 6, the phase diagram is constructed with f_{end} versus χN , and four morphologies are found, *i.e.*, lamella, perforated lamella, strip, and micelle (shown in Fig. 2). To clearly describe the influence of the volume fraction of the end coils on the self-assembly, we also compare the phase behaviors of the rod-coil pentablock copolymers with low and high volume fractions of end coils. When the volume fraction of the end coils is low, *i.e.*, $0.22 \leq f_{\text{end}} \leq 0.33$, the effect of the end coil blocks to maximize their free volume to maximize entropy is trivial, and the system achieves a balance between the rods to maximize their contact to minimize energy and the spacer coils to minimize their elastic strain to maximize entropy. With decreasing temperature, the ordered morphologies transform from the strip to the perforated lamella and finally to the lamella at $f_{\text{end}} = 0.33$, and from the perforated lamella to the lamella at $f_{\text{end}} = 0.22$. When the volume fraction of the end coil is high, *i.e.*, $0.33 < f_{\text{end}} \leq 0.5$, the effect of the end coil becomes dominant, where the rod-coil pentablock copolymers self-assemble into structures with large interface to maximize the free volume to maximize the entropy. As shown in Fig. 6, the strip structure is stable at $f_{\text{end}} = 0.33$, while the micelle is stable at $f_{\text{end}} = 0.5$.

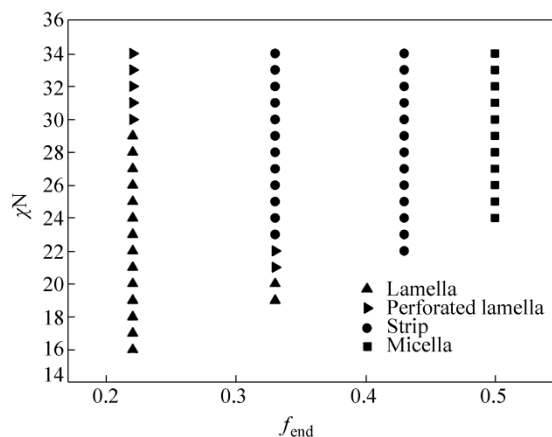


Fig. 6 Phase diagram for the linear rod-coil pentablock copolymer with the volume fraction of the end coil $0.22 \leq f_{\text{end}} \leq 0.5$

CONCLUSIONS

As an extension of our previous work, the self-assembly of the linear rod-coil multiblock copolymer is investigated by applying SCF lattice techniques in 3D space. Six stable morphologies are observed, *i.e.*, lamella, perforated lamella, strip, lamellar strip, gyroid and micelle. Compared to the coil-rod-coil triblock copolymer ($m_{\text{rod}} = 1$), the copolymers with more rods ($m_{\text{rod}} \geq 2$) exhibit the same order-order phase transitions that the ordered morphology changes from the strip to the perforated lamella and finally to the lamella with increasing temperature. Besides, by varying the length of the parts of the linear rod-coil pentablock copolymer ($m_{\text{rod}} = 2$), the effects of the volume fractions of the rods, the spacer coils and the end coils are studied respectively. When the volume fractions are low for the rod blocks or high for the spacer coil, the strip structure is formed with rods packing in the interdigitated fashion, due to the negligible entropy penalty associated with the spacer coil. When the volume fractions are high for the rods or low for the spacer coils, the entropy penalty is strong, leading to the rearrangements of the rods to lower the system energy in the self-assembly. Besides, the influence of the end coil can be ignored when the volume fraction of the end coil is low, while the effects of the end coil become dominant when the volume fraction of the end coil is high which results in the formation of the self-assembled structures with large interfaces. As described above, the effects of many rods and coils imposed on the self-assembly are demonstrated, which are expected to provide guidance for the morphology control of the linear rod-coil multiblock copolymer materials and some insight into the MCLCPs^[25] as well. Other problems of relevance remain to be considered in the subsequent work, such as the polydisperse coil as well as polydisperse rods which may change the aggregation behaviors by the appearance of some defects at the surfaces of the rod aggregates.

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