Self-organization of amphiphilic polymers

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Dedicated to Professor Stanislaw Penczek on the occasion of his 80th birthday

Abstract: Even a cursory analysis of the chemical structure of the large majority of synthetic and natural polymers shows that their monomer units have a dualistic character, that is, they possess a pronounced amphiphilicity. In many cases, this feature has a significant impact on polymer self-assembly. In this mini-review, we discuss some examples illustrating nonconventional forms of self-organization which were recently predicted by computer simulation for simple models of amphiphilic polymers.

Keywords: amphiphilic macromolecules, self-assembly polymers, microphase separation, composites, computer simulation.

Samoorganizacja polimerów amfifilowych

Streszczenie: Analiza struktury chemicznej zarówno syntetycznych, jak i naturalnych polimerów dowodzi, że większość z wchodzących w ich skład jednostek monomerów ma charakter dualistyczny i wykazuje amfifilowe właściwości. W wielu przypadkach cecha ta jest czynnikiem decydującym o samoorganizacji polimeru. W niniejszym miniprzeglądzie literaturowym przedstawiono kilka przykładów ilustrujących niekonwencjonalne formy samoorganizacji przewidziane metodą symulacji komputerowej, przeprowadzonej dla prostych modeli polimerów amfifilowych.

Słowa kluczowe: amfifilowe makrocząsteczki, polimery samoorganizujące się, separacja mikrofazowa, kompozyty, symulacja komputerowa.

Microphase-segregated polymer systems have long garnered significant scientific interest due to their ability to spontaneously form periodic morphologies at controllable length scales [1—3]. Self-assembly is also one of the most universal strategies used in biology for the development of complex and functional (nano)structures: fascinating examples are multimeric proteins and nucleic acid multiplexes, viruses, and biomembranes. Such systems have been extensively studied over years, to allow for a better understanding of their structure and functions. Nowadays, polymer self-assembly is often considered as a basis for the large number of emerging and potential applications, including advanced composite nanomaterials, transformation and production of energy, new information technologies and microelectronics, pharmacology and medicine, food and personal care products.

Polymeric materials are typically characterized by a broad range of length and timescales over which phenomena of both scientific and engineering interest can arise. In this mini-review, we discuss some examples illustrating nonconventional forms of self-organization which were predicted by computer simulation. The focus is on self-assembly of amphiphilic copolymers at mesoscale, *i.e.* at length scales intermediate between atomistic and macroscopic.

The driving force for structure formation in copolymers is competing interactions, *i.e.* sufficiently strong incompatibility of different monomer species, on the one hand, and covalent bonding of units within the same macromolecule, on the other hand. The latter factor prevents the separation of the system into homogeneous macroscopic phases, which can, under specific conditions, stabilize some types of microdomain structures. Usually, such a phenomenon is treated as microphase separation transition (MIST) or order-disorder transition (ODT).

The diblock architecture of the type A_f-*b*-B_{1-f} $(0 < f < 1)$ provides the simplest model for examining polymer self-assembly in a monodisperse copolymer melt. The block copolymers are structures formed by two chemically different polymer chains, A and B, linked together by a covalent bond. Their self-assembly is governed by the chemical composition (the fraction f of segments that belong to, *e.g.* the A block) and the product of Flory–Huggins interaction parameter χ of the copolymer segments and the total number of segments per chain, N. The values of N and χ (or their combination χN) as well as the

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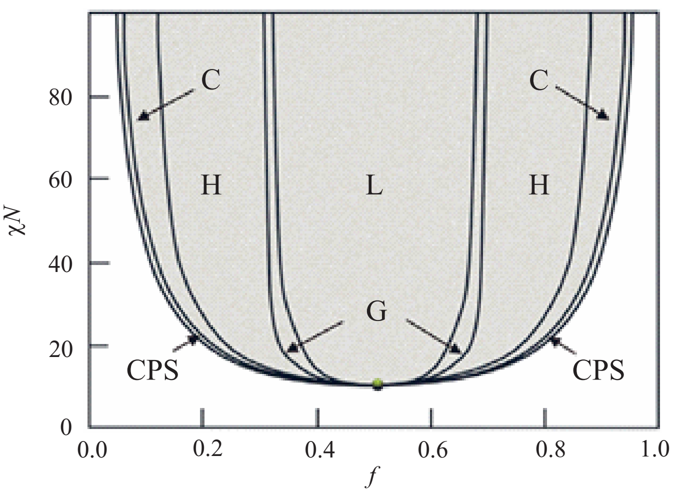


Fig. 1. Phase diagram for a melt of linear diblocks with A and B segments that have equal volumes and Kuhn lengths. The regions of stability for the lamellar (LAM), gyroid (GYR), hexagonal (HEX), body-centered cubic (BCC), and close-packed spherical phases are denoted L, G, H, C, and CPS, respectively. The dot at $f = 1/2$ corresponds to the mean-field critical point, $\chi N = 10.495$

chemical composition of copolymer f are the main parameters in experimental studies and theory.

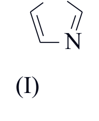
The theory of microphase separation in diblock copolymers has been developed in detail and this subject has been reviewed extensively [1—3]. The phase diagram which defines the regions of stability of morphologies with various symmerty in the space of parameters χN and f, was first predicted by Leibler for the weak segregation of blocks [4]. Later, Matsen [5, 6] and Fredrickson [7, 8] specified this diagram, using the self-consistent field theory (SCFT). Leibler-Fredrickson's classical phase diagram is presented in Fig. 1.

In Fig. 1 one can see the regions of stability of „classical" microdomain phases: A-spheres with close-packed symmetry (region CPS, $f < 1/2$), A-spheres arranged in a body-centered cubic lattice in B-matrix (region C, $f < 1/2$), A-cylinders arranged in a hexagonal lattice in B-matrix (region H, $f < 1/2$), alternating A- and B-lamellae (region L), B-cylinders hexagonally packed in A-matrix (region H, $f > 1/2$), B-spheres arranged in a body-centered cubic lattice in A-matrix (region C, $f > 1/2$), close-packed B-spheres in A-matrix (region CPS, $f > 1/2$), as well as the region of stability of gyroid phase (region G).

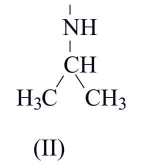
In most of the theoretical studies it is assumed that the polymer blocks are conformationally symmetric; that is, they have equal Kuhn segment length, ℓ, and segmental volume, v. The conformational asymmetry can cause some changes in the phase diagrams. It is known that by introducing a difference in either the statistical segment lengths and/or segmental volumes between the constituents of a diblock, such as in the often studied system of polystyrene and polyisoprene, the microphase boundaries become asymmetric about $f = 1/2$ [9, 10]. Nevertheless, it is commonly believed that the microphase-separated morphology is dictated only by the global features of copolymers: their architecture, chemical composition, and interaction between segments. From this viewpoint, the general and quite expected feature for all mesophases is that the minority component (for example, B) forms domains, while the majority component (A) prevailing in a chain forms a matrix, which surrounds domains. It turns out, however, that such behavior is not universal. Moreover, as we will see, even the types of various morphologies and transitions between them can be counterintuitive at first sight.

It is a commonplace to say that the properties of a copolymer depend not only on its global architecture and chemical composition but also on the local chemical structure of its monomer units. Therefore, an important route in the precise control and theoretical prediction of molecular parameters required to achieve well-defined microphase-separated morphologies is connected with understanding the role of local polymer structure responsible for intra- and intermolecular interaction.

The drawback of the existing theoretical approaches is the representation of each monomeric unit of a real polymer as a point-like interaction site of pure repulsive or pure attractive type. At the same time, even a cursory analysis of the chemical structure of the large majority of real macromolecules shows that each monomeric unit has a dualistic character; that is, repeating polymer unit, which is usually considered as a structureless bead, actually incorporates both repulsive and attractive parts concurrently. Indeed, a large number of synthetic polymers possess a pronounced amphiphilicity in every repeat unit [11]. Typical examples are polymers like poly(1-vinylimidazole) [Formula (I)], poly(N-isopropylacrylamide) (II), poly(2-ethyl acrylic acid) (III), poly(styrene sulfonate), poly(4-vinylpyridine), Nafion (IV), *etc.* In each repeat unit of these polymers there are hydrophilic (polar) and hydrophobic (nonpolar) atomic groups, which have different affinity to the surrounding medium [11]. Many of the amino acids also contain both polar and nonpolar groups simultaneously and, strictly speaking, the interaction between such amino acid residues in proteins cannot be literally reduced to pure hydrophilic or pure hyd-

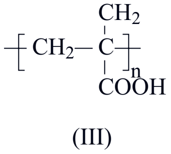


(I)



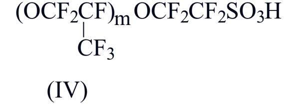
$-\!\!\left[CH_2-\underset{\underset{\underset{\underset{H_3C\ \ CH_3}{CH}}{NH}}{C=O}}{CH}\right]_n\!\!-$

(II)



$-\!\!\left[CH_2-\overset{\overset{CH_3}{CH_2}}{\underset{COOH}{C}}\right]_n\!\!-$

(III)



$-(CF_2CF_2)_nCF_2-\underset{(OCF_2\underset{CF_3}{CF})_m OCF_2CF_2SO_3H}{CF}-$

(IV)

rophobic site-site interactions, as it is presupposed in the standard polymer models. Other important biopolymers — polysaccharides, phospholipids — are also typical amphiphiles. Moreover, among the synthetic polymers, polyamphiphiles are very close to biological macromolecules in nature and behavior. In principle, they may provide useful analogs of proteins and are important for modeling some fundamental properties and sophisticated functions of biopolymers such as protein folding, formation of secondary structures, and enzymatic activity. Therefore, understanding the physics of self-assembly of the copolymers with dualistic monomer-monomer interaction is extremely challenging and important.

The two-letter ("black-and-white") HP model first introduced by Lau and Dill [12] and widely discussed in the literature is the simplest model of hydrophobic/hydrophilic polymers. The model is very computationally efficient, but its principal disadvantage is the representation of each monomer unit of an amphiphilic chain as a point-like interaction site of pure hydrophilic or pure hydrophobic type. At the same time, as noted above, in real amphiphilic polymers, each monomeric unit has a dualistic (hydrophobic/hydrophilic) character, that is, repeating polymer unit actually incorporates both hydrophilic and hydrophobic parts concurrently [11].

In the literature, there are several coarse-grained polymer models in which bead-like monomers are replaced by composite asymmetric objects [13, 14]. Generally, this gets a host of qualitatively new structures. One of the possible extensions of the HP model is the model introduced in Refs. [15—20] (cf. Fig. 2).

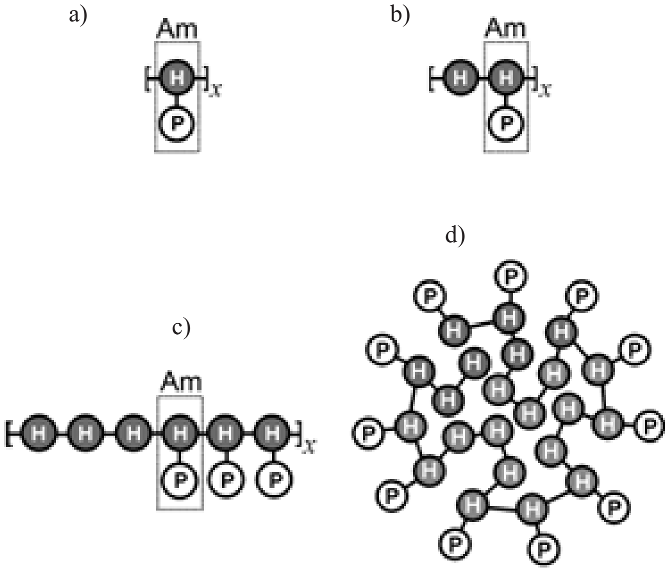


Fig. 2. Models of amphiphilic polymers: (a) amphiphilic homopolymer (poly-Am), (b) regular alternating H-Am copolymer, (c) regular multiblock H-Am copolymer, and (d) protein-like H-Am copolymer in which hydrophilic units are distributed along hydrophobic backbone in a specific (quasi-random) way [21—25]. Each hydrophobic monomer unit (H) is considered as a single interaction site (bead); each amphiphilic group (Am) is modeled by a „dumbbell" consisting of hydrophobic (H) and hydrophilic (P) beads. This composite monomer unit may be considered as a hydrophobic-hydrophilic „HP-dipole"

This is a more realistic coarse-grained model of amphiphilic polymers where the dualistic character of each monomer unit is explicitly represented. Depending on the content of pure hydrophobic and amphiphilic groups, one can simulate amphiphilic homopolymers (poly-Am) and copolymers with the same H/Am composition but with different distribution of H and Am units along the hydrophobic backbone, including regular copolymers comprising H and Am units in alternating sequence, (HAm)x, regular multiblock copolymers (HLAmL)x composed of H and Am blocks of equal lengths L, and random copolymers having different H and Am block lengths (Fig. 2). Generally, a random (quasi-random) amphiphilic copolymer is characterized by its composition, by the average lengths of the hydrophobic and amphiphilic blocks, L_H and L_{Am}, and by the specific distribution of H and Am units along the chain [21—25].

As has been shown in Refs. [14—20], such a relatively trivial modification of the standard HP model can lead to some nontrivial consequences when studying the collapse transition for the single-chain amphiphilic polymers and their aggregation in a selective solvent. In particular, the simulations of solvent-driven conformational transitions under the variation of solvent conditions showed that for the model of amphiphilic homopolymer (poly-Am, Fig. 2a), a variety of novel conformations are possible, depending on the interaction between hydrophobic and hydrophilic sites. Specifically, the thermodynamically stable anisometric globular structures were observed, including disk-like, stretched necklace-like, and cylindrical-shaped conformations [15]. Hydrophobic-amphiphilic (HAm) copolymers with the same composition but with different distribution of H and Am units along the main hydrophobic chain were also simulated [16]. Under poor solvent conditions for hydrophobic segments, all the copolymers form compact conformations, irrespective of the primary structure. However, the morphology of these conformations dramatically depends on copolymer sequence. It was found that single proteinlike polyamphiphiles (Fig. 3d) can readily adopt conformations of compact spherical globules with the hydrophobic backbone clustered at the globular core and the hydrophilic sites forming the envelope of this core and buffering it from polar solvent [21—25]. This morphology closely resembles that of micelles or globular proteins. Similar to natural proteins, the collapsed proteinlike copolymers do not aggregate in solution. The solution properties of charged hydrophobic-hydrophilic proteinlike copolymers have been simulated in the presence of both mono- and multivalent counterions [20, 26—28].

Next, we discuss some recent results from mesoscale simulations of a melt of hydrophobic-amphiphilic copolymers. It can be expected that the microphase separation in the melt of HAm diblocks schematically depicted in Fig. 2c (homopolymer „tail" consisting of hydrophobic units linked to an amphiphilic block of HP-dipoles) will differ significantly from that in the melt of conventional diblocks. Since the two connected sites in each amphiphilic unit are incompatible, it is naturally to expect that this unit will prefer to be at the H/P boundary rather than in H- or P-bulk, the behavior reflecting a surface activity. The sites H and P are assumed to be of equal volume *v*. The length fraction of amphiphilic block (denoted below by letter C) is *f* and the length fraction of sites belonging to the homopolymer tail H is 1-*f*.

Prediction of phase diagrams by any particle-based simulation method is always a very labor-consuming problem. To date, such simulations can be performed only on a supercomputer, and only for fairly simple models. In Refs. [29, 30], the method of dissipative particle dynamics (DPD) and a two-stage simulation strategy was used. At the first stage, a parallel replica DPD code was applied, allowing to effectively select the most ordered structures with the maximum order parameter for a relatively small system. At the second stage, the simulation box of the selected ordered systems was duplicated along three directions and the resultant systems were further simulated for a long time, using another DPD code based on domain decomposition parallelism. Also, large-scale SCFT simulations were carried out for discrete and continuous models of the same copolymers [31]. The predicted phase diagram is shown in Fig. 3.

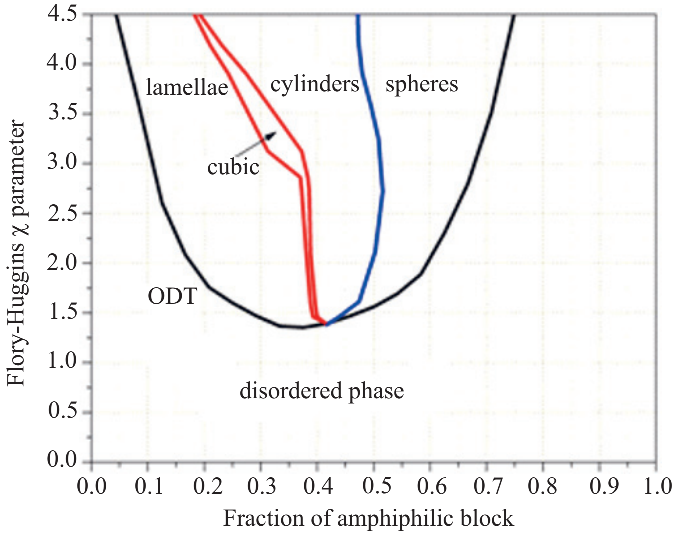


Fig. 3. Phase diagram predicted for a melt of hydrophobic-amphiphilic diblocks [30]

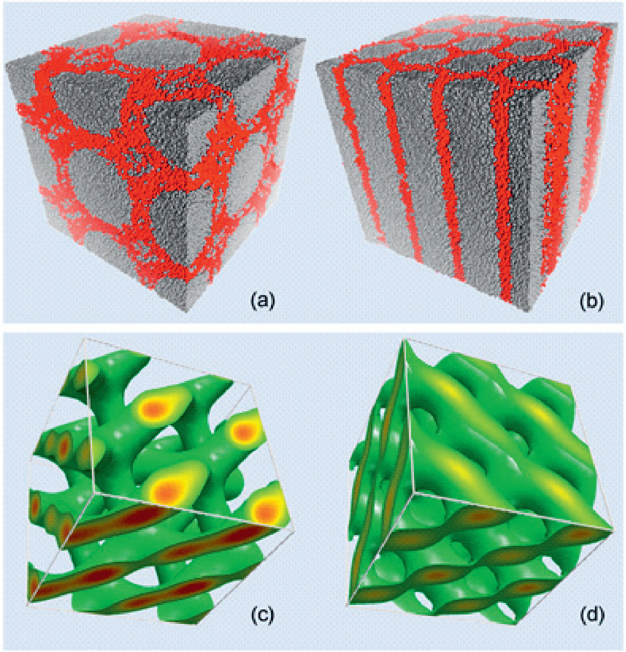


Fig. 4. Typical morphologies found in a melt of hydrophobic-amphiphilic copolymers: (a) BCC, (b) HEX, (c) double diamond (DD), and (d) hexagonally perforated lamellae (HPL). The H-rich domains observed for the BCC and HEX phases look like inverse micelles of spherical and cylindrical shape, respectively. For the DD and HPL phases, the regions filled with P-beads and defined by the isodensity surfaces $\rho_H(\mathbf{r})/\bar{\rho}_H = 1/2$ are shown, where $\rho_H(\mathbf{r})$ and $\bar{\rho}_H$ are local and average density of H-beads, respectively

At purely external similarity of this diagram to Leibler-Fredriksson's classical diagram (Fig. 1) there are a number of fundamental differences. First, the location of various regions here is asymmetric with respect to the critical point (at *f* = 0.5). This is a quite expected result taking into account that hydrophobic-amphiphilic copolymers possess the conformational asymmetry. Second, the regions on this and classical diagrams actually switched places. Indeed, at small *f* we find a lamellar structure instead of spherical microdomains. On the contrary, structures like BCC are observed for copolymers with approximately symmetric composition when one would expect the formation of lamellae. Finally, the matrix (majority H component) and the microdomains (minority P component) also switched places. Two examples of these morphologies are shown in Fig. 4. As seen, for HEX and BCC mesophases, P-beads, which are the part of amphiphilic block, fill space between H-rich domains. These unusual mesophases can be called „inverse mesophases" [32] by analogy with the structures known for surfactants in nonpolar environments.

Besides lamellar (LAM), cylindrical (HEX) and spherical (BCC) structures, the formation of more complex morphologies associated with the bicontinuous cubic phases (BCPs) is also observed (Fig. 3). They include the double-diamond (DD) structure (Fig. 4c) and the Schoen's gyroid. Unlike BCC structure, in which one of the phases is continuous and the other is discrete, these structures are bicontinuous, that is, both phases are continuous. The region of their existence is very narrow (Fig. 3). Interestingly, among BCPs, the DD phase always dominates, while the gyroid is observed very rare. At last, in the region corresponding to the lamellar morphology, the structure whose overall organization is best described by a system of hexagonally perforated lamellae (HPL) can be found. The HPL structure consists of alternating majority and minority component layers in which hexagonally packed domains of majority component extend through the minority component (Fig. 4d). This structural organization can be understood as arising from a competition between the formation of planar interfaces and curved interfaces.

The stabilization of the DD and HPL morphologies deserves a separate discussion. For conventional diblocks, these structures are considered to be metastable, stabilized by compositional fluctuations [7]. However, they are rather typical for triblock copolymers [33] and, especially, for amphiphile/water systems [34, 35]. In diblock copolymers, the GYR phase is stable [7, 8], whereas in binary (or ternary) mixtures of water with surfactants or lipids (and oil) different BCPs (Schwartz's primitive P-structure, double-diamond structure, and Schoen's gyroid) and transitions between them may occur [36, 37]. Generally, hydrophobic-amphiphilic polymers show a pronounced tendency to form structural motifs which are reminiscent of those that are typical for conventional surfactants [38]. In particular, the DD structure observed for compositionally symmetric or nearly symmetric HAm polymers closely resemble the inverse bicontinuous cubic phase well-known for lyotropic mesophases. Similar morphologies have been found in polymeric foams [39] and in assemblies of soap bubbles [40]. The origin of such morphologies in these materials has been discussed for many years and the physical forces causing their formation are well recognized. Essentially „interfacial tension" controls these morphologies, and the way in which these forces are responsible for the structure formation has been comprehensively described in the literature.

It is clear that the discussed nonventional scenarios of polymer self-assembly, both in selective solution and in bulk, are directly connected with details of chemical structure. Thus, one can argue that in the world of synthetic macromolecules, the processes of self-organization, resulting in the formation of mesophases with the characteristic size about tens and hundreds of nanometers, are considerably dictated by local structural details which manifest itself at scales about one nanometer.

The discussion presented above may have important consequences for several related problems in polymer physics. For example, it can shed a new light on the observed morphologies in ionomers [41], including proton-conducting ionomer membranes of fuel cells. For these materials, a percolation of polar channels giving rise to proton conductivity is often observed at surprisingly low volume fraction of a polar component [42—44].

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Prace wybrane przez Komitet Naukowy zostaną opublikowane w czasopiśmie „Polimery".
Opłata konferencyjna do 30 czerwca 2014 r.: 1150 zł netto — uczelnie i jednostki badawcze; 1400 zł netto — firmy. Opłata po terminie — odpowiednio, 1250 zł i 1500 zł.
W przypadku zgłoszenia z firm większej liczby uczestników — każda kolejna osoba 15 % rabatu.
Opłata konferencyjna obejmuje: nocleg w pokoju 2-osobowym, materiały konferencyjne, wyżywienie oraz udział w imprezach towarzyszących, parking. Dopłata do pokoju jednoosobowego — 200 zł/os. (dotyczy całego pobytu).
Prosimy o wniesienie opłaty konferencyjnej na konto:
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Koszt promocji firmy w formie wkładki reklamowej formatu A4 w materiałach konferencyjnych 850 zł netto (w kolorze) lub stoiska promocyjnego na sali konferencyjnej 1800 zł netto.
Informacje: Sekretariat: mgr Olga Motyka, tel.: 34 325 05 09, Instytut Technologii Mechanicznych, Zakład Przetwórstwa Polimerów, 42-201 Częstochowa, al. Armii Krajowej 19c

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