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Review

Block copolymers in electric fields

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ABSTRACT

The structural versatility of block copolymers on the nanometer scale make them highly promising candidates for many applications in soft matter nanotechnology, including optics, electronics, and acoustics. In order to harvest the full potential of nanostructured block copolymer materials and achieve widespread use outside of academia, adaptable strategies are required to control and manipulate their spatial orientation, periodicity, connectivity, and long-range order. Over the past two decades the use of an external electric field has been well established as a viable tool to control a wide variety of structural parameters of nanostructured block copolymers on both mesoscopic and nanoscopic length scales. Covering a wide range of experimental and theoretical work, this review aims to illustrate major scientific advances of the past years, focusing in particular on the underlying physics that governs the fundamental interactions between an external electric field and block copolymer mesophases and its impact on phase behaviour and orientational order in bulk, solution, and thin films.

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Abbreviations: AC, alternating current; AFM, atomic force microscopy; BCC, body centered cubic; BCP, block copolymer; CDS, cell dynamic simulations; CH, cyclohexane; CMC, constant mean curvature; CYL, cylinders; DC, direct current; DMF, dimethylformamide; DSCFT, dynamic self-consistent field theory; GISAXS, grazing incidence small-angle X-ray scattering; GYR, Gyroid; HPL, hexagonally perforated lamellae; LAM, lamellae; LCST, lower critical solution temperature; MWS, Maxwell–Wagner–Sillars; NG, nucleation and growth; ODT, order–disorder transition; OOT, order–order transition; PEO, poly(ethylene oxide); PLA, poly(lactic acid); PMMA, poly(methyl methacrylate); PS, poly(styrene); VDF, poly(vinylidene difluoride); P(*d*-S-*b*-*d*-MMA), poly(*d*-styrene-*b*-*d*-methyl methacrylate); P(*S*-*b*-B), poly(styrene-*b*-butadiene); P(*S*-*b*-I), poly(styrene-*b*-isoprene); P(*S*-*b*-I-*b*-S), poly(styrene-*b*-isoprene-*b*-styrene); P(*S*-*b*-LA), poly(styrene-*b*-lactic acid); P(*S*-*b*-MMA), poly(styrene-*b*-methyl methacrylate); P(*S*-*b*-2VP), poly(styrene-*b*-2-vinylpyridine); RG, reorientation of grains; SANS, Small-Angle Neutron Scattering; SAXS, Small-Angle X-ray Scattering; SCFT, self-consistent field theory; SD, selective disordering; SPH, spheres; SSL, strong segregation limit; TEM, transmission electron microscopy; UCST, upper critical solution temperature; WSL, weak segregation limit.

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1. Introduction

Block copolymers are an intriguing class of macromolecules [1], which consists of two or more chemically distinct and covalently linked homopolymers. The self-assembly of block copolymers [2] on mesoscopic length scales presents a unique way to template the formation of highly ordered, three-dimensional, structural hierarchies with characteristic feature sizes below the wavelength of visible light [3–6]. Directing the orientational and translational long-range order of the underlying block copolymer structure is paramount to achieve all well aspired electromagnetic, mechanical, and acoustic properties envisaged for a wide range of nanostructured soft metamaterials [7].

To harvest the full potential of nanostructured block copolymer materials, adaptable strategies are required to control and manipulate spatial orientation, periodicity, connectivity, and long-range order [8–12]. Several methods have been established, which utilize chemical and physical constraints to direct self-assembly into highly ordered geometries. These techniques include chemically [13] and topologically patterned surfaces [14], confined substrate geometries [15], and external stimuli, such as temperature gradients [16], solvent annealing [17,18], directional epitaxy [19,20], shear [21–24], as well as magnetic [11,25–27], and electric fields.

Considering the latter, the most beneficial properties of an applied electric field from a very practical point of view are three-fold: (i) an electric field can be applied instantly throughout the sample volume, (ii) the applied field strength scales linearly with the inverse distance

between the field-creating electrodes, and (iii) a uniform electric field creates a typical non-contact force, which is gradient-free throughout the sample volume. This is in contrast, for example, to an applied shear field, which constitutes a contact-force field that is associated with a velocity gradient.

As such, an electric field represents a very versatile tool to control both the structure and phase behavior of nanostructured soft matter in confined geometries (e.g., in thin films or in microfluidic devices). In particular, the electric field induced alignment of nanostructured block copolymers can serve as a versatile approach to control both orientational and translational long-range order; both deemed pivotal when block copolymers are to be successfully employed in today’s nano-technology [9,10].

This review will first discuss the underlying physics that controls the alignment of anisotropic block copolymer morphologies in the presence of an electric field. We will then continue to outline how an electric field may further effect the thermodynamic stability of an underlying block copolymer morphology, which may lead to field induced order–disorder or order–order transitions. We shall likewise consider effects in the presence of a competing external field, such as the presence of a surface field or shear.

2. Block copolymer self-assembly

2.1. Polymer miscibility

Contrary to ordinary binary liquids the entropy of mixing is marginal in polymeric systems. Assuming ideal

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