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Orientation and Order in Shear-Aligned Thin Films of Cylinder-Forming Block Copolymers.

RICHARD REGISTER, Princeton University

The regularity and tunability of the nanoscale structure in block copolymers makes their thin films attractive as nanolithographic templates; however, in the absence of a guiding field, self-assembly produces a polygrain structure with no particular orientation and a high density of defects. As demonstrated in the elegant studies of Ed Kramer and coworkers, graphoepitaxy can provide local control over domain orientation, with a dramatic reduction in defect density. Alternatively, cylindrical microdomains lying in the plane of the film can be aligned over macroscopic areas by applying shear stress at the film surface. In non-sheared films of polystyrene-poly(n-hexylmethacrylate) diblocks, PS-PHMA, the PS cylinder axis orientation relative to the surface switches from parallel to perpendicular as a function of film thickness; this oscillation is damped out as the fraction of the PS block increases, away from the sphere-cylinder phase boundary. In aligned films, thicknesses which possess the highest coverage of parallel cylinders prior to shear show the highest quality of alignment post-shear, as measured by the in-plane orientational order parameter. In well-aligned samples of optimal thickness, the quality of alignment is limited by isolated dislocations, whose density is highest at high PS contents, and by undulations in the cylinders' trajectories, whose impact is most severe at low PS contents; consequently, polymers whose compositions lie in the middle of the cylinder-forming region exhibit the highest quality of alignment. The dynamics of the alignment process are also investigated, and fit to a melting-recrystallization model which allows for the determination of two key alignment parameters: the critical stress needed for alignment, and an orientation rate constant. For films containing a monolayer of cylindrical domains, as PS weight fraction or overall molecular weight increases, the critical stress increases moderately, while the rate of alignment drastically decreases. As the number of layers of cylinders in the film increases, the critical stress decreases modestly, while the rate remains unchanged; substrate wetting condition has no measurable influence on alignment response. [Work of Raleigh Davis, in collaboration with Paul Chaikin.]