



ABSTRACT

This thesis investigates instability-driven pattern formation in soft polymeric systems, focusing on the coupling between substrate modification, thin-film deposition, and dewetting dynamics. The work addresses two key aspects: (i) patterning of elastomeric polydimethylsiloxane (PDMS, Sylgard 184) surfaces, and (ii) ordering of ultra-thin polystyrene (PS) films via substrate-directed dewetting under non-equilibrium conditions. Deterministic surface patterning of crosslinked Sylgard 184 is achieved using ultraviolet/ozone (UVO) exposure through transmission electron microscopy (TEM) grids. It is shown that pattern depth is governed not only by exposure time but also strongly by initial film thickness and crosslinker concentration. A non-monotonic dependence on crosslink density is observed, arising from the interplay of oxidation-induced densification, substrate-imposed mechanical resistance, and stress relaxation within the elastomer. This provides a simple route for tunable surface patterning without conventional lithography. Thin PS films deposited on these patterned substrates exhibit two distinct regimes: (i) continuous film formation followed by thermally induced dewetting, and (ii) direct rupture during spin coating (spin dewetting). The results reveal that substrate topography and its orientation relative to the advancing meniscus critically determine rupture location, morphology, and spatial ordering. Under suitable conditions, confinement imposed by the patterned substrate enables a transition from random to ordered droplet assemblies. Hierarchical patterning, achieved by combining soft lithographic imprinting with UVO modification, introduces multi-scale confinement that further enhances ordering and enables complex, multi-level morphology control. In parallel, oxygen plasma treatment of uncured and partially cured Sylgard 184 is shown to induce mechanically driven pattern formation. The formation of a stiff oxidized surface layer over a compliant bulk generates compressive stresses that relax via wrinkling. The wrinkle morphology is strongly influenced by film thickness, crosslinking state, plasma exposure time, and pre-curing conditions, highlighting the role of viscoelasticity and elastic mismatch. Overall, this thesis establishes a unified framework for pattern formation in soft polymer systems by integrating surface modification, thin-film hydrodynamics, and instability-driven self-organization. The findings offer scalable, lithography-free strategies for generating ordered micro- and nano-scale structures with applications in coatings, optics, and soft functional interfaces.



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