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### STUDIES ON THE WRINKLING OF THIN POLYMER FILMS FLOATING ON LIQUID

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Polymer Science and Engineering

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Thomas P. Russell

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#### Abstract

This dissertation aims to broaden our understanding on wrinkling instabilities occurring on floating polymeric sheets, and tries to establish innovative methods that exploit these patterns in studies on material behavior and interfacial phenomena. We will address three major topics in this thesis including, *i*) characterization of the conditions required to buckle an annular disc, *ii*) characterization of wrinkles occurring around a droplet/bubble placed on a membrane that is kept taut at the liquid-air interface, and *iii*) using wrinkling patterns as a probe to understand the interfacial behavior and dynamics of ultrathin films.

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The first project in this thesis is about a thin floating elastic disc wrinkled by a droplet cast at the center. First, we obtain a three dimensional map of the deformed surface using confocal fluorescent microscopy. Then, we study the identical films in an inverted geometry where an air bubble is in contact with the film from its underside. In this configuration, a high-resolution optical profilometer facilitated the topographical analysis, which provided the detailed elasto-capillary measures of the problem. Finally, we study the patterns as a function of boundary conditions and material properties, where physical properties of the sheet, such as molecular confinement and physical aging, also change the size and wavelength of the pattern.

The wrinkle patterns are also characterized in response to the change in surface tension of the bath liquid with the aid of a Langmuir Trough. This experimental set-up also facilitates the pattern formation on annular elastic discs, when variable peripheral forces create contraction within the film. Here, planar capillary forces dictate the controlled boundary conditions and probe a regime of deformation where the wrinkle length and the wavelength are empirically related. We should emphasize that the wrinkling of an annulus does not merely mimic the original droplet experiment, also leading to several remarkable discoveries. First, we observe that, in the studied thickness regime (e.g. the scaled extent and wavelength of wrinkles) observed for a range of thicknesses can be superimposed on another by displacing the data towards the hypothetical threshold required to wrinkle the zero-thickness sheet. We observe another thickness invariant phenomenon when the conditions of fold formation are found similar for films of identical annular aspect ratios.

Finally, we study the time-dependent change in wrinkles forming on viscoelastic membranes. The dynamic behaviors of wrinkles are related to the rheological characteristics of polymer molecules of linear architecture confined into thicknesses comparable to their melt radius of gyration. In this study, we have found that the normalized size of wrinkles could be a measure of the stress and strain exerted by a droplet. We also have demonstrated that confinement significantly influences the dynamics of PS films. We close this chapter demonstrating that the wrinkling dynamics is related to the kinematics of the film under the droplet, and generally compares to the stress relaxation experiments performed on bulk samples.

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