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Volume-Phase Transitions and Swelling Instabilities in Surface-Tethered Responsive Gels

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Responsive polymer hydrogels have opened exciting opportunities for breathable structures that adopt to environmental cues. Such structures can be designed from a variety of chemical motifs that endow specific response behavior at the material level. Moreover, mechanically pinning a responsive gel to a surface presents further opportunities for designing specific shape-volume transitions due to differential swelling that arises in confined structures. We present a simple technique for fabricating responsive polymer networks based on copolymers comprising the photoactive moiety methacroyloxybenzophenone (MaBP). This approach permits the synthesis of photo-cross- linkable polymers that are easily pattered with thicknesses down to 80 Angstroms. Using a combination of neutron reflection, QCM- D, and ATR-FTIR, we have mapped the volume-phase behavior of ultra-thin layers of responsive networks as a function of chemical functionality, cross-link density, and thickness. Interestingly enough, neutron reflection reveals diffuse interfaces at the periphery of surface-attached networks that grow with the degree of swelling, pointing to surface fluctuations that result from biaxial compression. Finally, confocal microscopy has been used to map three-dimensional swelling in patterned structures revealing swelling instabilities from surface undulations to macroscopic buckling depending on the aspect ratio of the pattern.