## **Supplementary Information**

## Kinetic Study of Swelling-Induced Surface Pattern Formation and Ordering in Hydrogel Films with Depth-Wise Crosslinking Gradient

Murat Guvendiren,<sup>a</sup> Jason A. Burdick,<sup>\*a</sup> and Shu Yang<sup>\*b</sup>

<sup>a</sup> Department of Bioengineering, University of Pennsylvania, 210 S 33th Street, Philadelphia, PA 19104, USA. Fax: +1 (215) 573-2071; Tel: +1 (215) 898-8537; E-mail: burdick2@seas.upenn.edu
<sup>b</sup> Department of Materials Science and Engineering, University of Pennsylvania, 3231 Walnut Street, Philadelphia, PA 19104, USA. Fax: +1 (215) 573-2128; Tel: +1 (215) 898-9645; E-mail: shuyang@seas.upenn.edu



**Fig. S1.** Confocal fluorescent depth profiles of PHEMA films photocrosslinked with different concentrations of EGDMA, followed by swollen to equilibrium in water. Films were fabricated by exposing the precursor solution to UV while the surface of the precursor was open to air. Intensity profiles were obtained from confocal xz-scans of swollen films. PolyFluor<sup>TM</sup> 570 (~3 mg/mL) was incorporated into the precursor solution prior to crosslinking for confocal imaging. Solid lines represent the exponential fits. The channel intensity and gain were kept constant during scanning.



**Fig. S2.** Young's modulus and the equilibrium water fraction of the photocrosslinked PHEMA films. Films were fabricated by exposing the precursor solution to UV while the surface of the precursor was covered to inhibit oxygen diffusion. PHEMA film modulus was measured by AFM while the sample was immersed in water. Note that these values are for uniform gels, however, can be used to evaluate the modulus profiles gradient gels. See detailed discussion in previous paper [Ref.20].



**Fig. S3.** Sequential confocal microscopy images (xz-scan) of PHEMA films with 3 wt% (left) and 0.5 wt% (right) EGDMA, and swollen in DI water. The films were placed in a chamber, which was then filled with DI water. Time sequence scans were collected from the same location on the sample. PolyFluor<sup>TM</sup> 570 was incorporated into the precursor solution prior to crosslinking for imaging. Scale bars are 50  $\mu$ m.



**Fig. S4.** Change of (a) equilibrium film thickness  $(h_{eq})$ , (b) equilibrium wavelength  $(\lambda_{eq})$  and (c) equilibrium amplitude  $(A_{eq})$  as a function of the initial film thickness  $(h_0)$ . Data were obtained from confocal microscopy images of hydrogel films equilibrated in DI water for 30 min. Legends indicate wt% EGDMA.



Fig. S5. Change of the critical linear expansion ratio with the initial film thickness.