Supporting Information for:

Enzyme-Triggered Folding of Hydrogels: Towards a Mimic of the Venus Flytrap

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Figure S1. Synthesis of gelatin methacrylate and analysis by ¹**H NMR**. The reaction scheme (as per Ref. 28) is shown on the top. Lysine and arginine residues in gelatin are converted to methacrylate groups by reaction with methacrylic anhydride. NMR spectra are compared for the parent and synthesized polymer. The peaks H^a and H^b at 5.6 ppm confirm the addition of methacrylate groups to gelatin. The integrated intensities due to the aromatic residues, H^a and H^b were used to estimate the degree of functionalization (i.e., the ratio of the number of amino groups modified with methacrylate to the initial number of amino groups), as described in Ref. 27. Here, the degree of functionalization is about 60%.



Figure S2. Folding behavior of bilayers of Gel A (PEGDA) and Gel C (NIPA-LAP). The dimensions of each gel was maintained at 20 mm x 9 mm x 0.15 mm. The NIPA-LAP is identical to that in Figures 2-5. The concentration of PEGDA was decreased from (a) to (c). Note that folding of the flat sheet into a tube is observed only when the PEGDA is reduced to 5 wt%. At higher concentrations of PEGDA, the stiffness of the A layer opposes folding.



Figure S3. Visual observations of hybrid gels folding in cell lysate. Mouse fibroblasts (L929) cells were cultured and the cell lysate was obtained. The hybrid gels were then incubated in the cell lysate. (a) The initial flat sheet folded to form a tube in 6 h due to the presence of MMPs in the lysate. (b) As a control, we added 5 mM EDTA (an inhibitor of MMPs) to the lysate. In this medium, the gel sheet remained flat.