## Shape-Programmed Folding of Stimuli-Responsive Polymer Bilayers

## Abstract

We investigated the folding of rectangular stimuli-responsive hydrogel-based polymer bilayers with different aspect ratios and relative thicknesses placed on a substrate. It was found that long-side rolling dominates at high aspect ratios (ratio of length to width) when the width is comparable to the circumference of the formed tubes, which corresponds to a small actuation strain. Rolling from all sides occurs for higher actuation, namely when the width and length considerably exceed the deformed circumference. In the case of moderate actuation, when both the width and length are comparable to the deformed circumference, diagonal rolling is observed. Short-side rolling was observed very rarely and in combination with diagonal rolling. On the basis of experimental observations, finite-element modeling and energetic considerations, we argued that bilayers placed on a substrate start to roll from corners due to quicker diffusion of water. Rolling from the long-side starts later but dominates at high aspect ratios, in agreement with energetic considerations. We have shown experimentally and by modeling that the main reasons causing a variety of rolling scenarios are (i) non-homogenous swelling due to the presence of the substrate and (ii) adhesion of the polymer to the substrate.



Figure 1. Scheme of rolling of a polymer bilayer according to different scenarios: short-side, long-side, and diagonal rolling.