Supporting Information

Experimental Section

Preparation of Janus discs

The preparation of the Janus discs, their triblock terpolymer precursors, crosslinking and sonication is extensively described in a preceding publication.

Hydrolysis

Hydrolysis of the Janus discs was achieved via treatment of 10 mg/mL solutions with 5 v/v trifluoroacetic acid (p.a. grade, Aldrich) in dichloromethane for 24 – 48 hours. In order to dissolve the particles into water, step-wise dialysis from dioxane into water was performed.

Dynamic Light Scattering (DLS)

Dynamic light scattering was performed on an ALV DLS/SLS-SP 5022F compact goniometer system with an ALV 5000/E cross-correlator and a He–Ne laser (λ_0 = 632.8 nm). Prior to the light scattering measurements the sample solutions were filtered using Millipore or Roth filters (housing: polypropylene, membrane: polytetrafluoroethylene) with a pore size of 5 μm. All samples were analyzed at high dilution. The data evaluation of the dynamic light scattering measurements was performed with the CONTIN algorithm and the values of the hydrodynamic radii were obtained after extrapolating the angle-dependent scattering data to $q^2 \rightarrow 0$.

Scanning Electron Microscopy (SEM)

SEM was performed using a LEO 1530 Gemini instrument equipped with a field emission cathode with a lateral resolution of approximately 2 nm. Prior to imaging, the surfaces were coated with a thin layer of platinum. The acceleration voltage was chosen between 0.5 and 3 kV.

For **cryogenic transmission electron microscopy (cryo-TEM)** studies, a drop of the sample dissolved in water was put on a lacey transmission electron microscopy (TEM) grid, where most of the liquid was removed with blotting paper, leaving a thin film stretched over the lace. The specimens were instantly vitrified by rapid immersion into liquid ethane and cooled to approximately 90 K by liquid nitrogen in a temperature controlled freezing unit (Zeiss Cryobox, Zeiss NTS GmbH, Oberkochen, Germany). The temperature was monitored and kept constant in the chamber during all of the sample preparation steps. After freezing the specimens, the specimen was inserted into a cryo-transfer holder (CT3500, Gatan, München,

Germany) and transferred to a Zeiss EM922 EF-TEM instrument. Examinations were carried out at temperatures around 90 K. The transmission electron microscope was operated at an acceleration voltage of 200 kV. Zero-loss filtered images ($\Delta E = 0$ eV) were taken under reduced dose conditions (100-1000 e/nm^2). All images were registered digitally by a bottom mounted CCD camera system (Ultrascan 1000, Gatan) combined and processed with a digital imaging processing system (Gatan Digital Micrograph 3.9 for GMS 1.4).

Scanning Force Microscopy (SFM)

SFM images were taken on a Veeco Multimode operated in Tapping Mode. Super-sharp probes with finite tip sizes with radii of 2 nm were used. Offline data processing was done using the Nanoscope Software (V6r2.1).

Size distributions (CONTIN plots) of non-hydrolyzed Janus discs

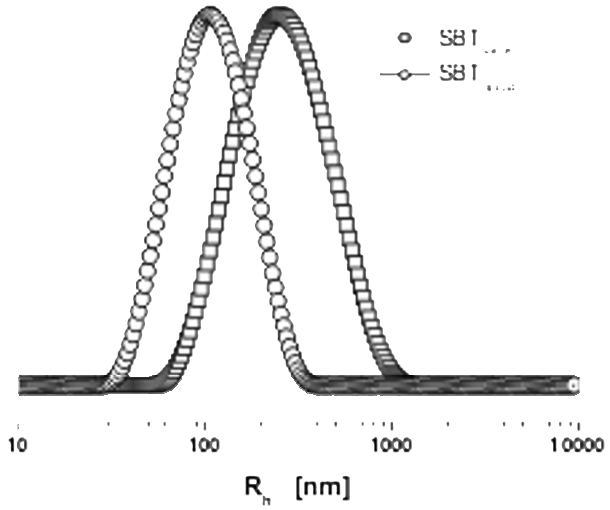

Figure Sup 1. CONTIN plots at 90^0 for the non-hydrolyzed Janus disc precursors leading to the samples SBMA$_{large}$ and SBMA$_{small}$ after acidic hydrolysis (in THF).