A Simple Route to Fluids with Photo-Switchable Viscosities Based on a Reversible Transition between Vesicles and Wormlike Micelles

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Figure S1. Phase behavior and rheology at 25°C of EHAC/ACA mixtures at a fixed [EHAC] of 40 mM and varying [ACA]. The molar ratio of NaOH to ACA is fixed at 1.1. The plot (a) shows the optical density at a wavelength of 700 nm, which quantifies the amount of light scattered from the sample. The plot (b) shows the zero-shear viscosity η_0 obtained from steady-shear rheology. Samples at [ACA] between 10 and 16 mM are transparent and highly viscous, as shown by the photograph of a sample resisting flow in an inverted vial (top left). These samples are also flow-birefringent, as shown by the photograph of a sample viewed under crossed polarizers while being shaken (bottom left). Together, the high viscosity and flow-birefringence point to the presence of wormlike micelles. In contrast, samples with [ACA] > 18 mM have a low viscosity close to water and are turbid, as can be noted from the photograph of a typical sample (top right). The same samples also do not show any flow-birefringence (photograph of a shaken sample under crossed-polarizers in bottom right). Together, the low viscosity and the turbidity point to the presence of vesicles.





Figure S2. (a) SANS data at 25°C from EHAC/ACA mixtures in D_2O at a fixed [EHAC] of 10 mM with various ACA concentrations. The molar ratio of NaOH to ACA is fixed to 1.1. For ACA concentrations of 6 and 6.5 mM, *I* follows a slope of -2 at low *q*, which is indicative of scattering from vesicles. (b), (c) Cryo-TEM images of a sample containing 40 mM EHAC, 20 mM ACA, and 22 mM NaOH show mostly vesicle structures (see Figure 2d also). In some parts of the sample, stretched and interwoven wormlike micelles were also observed in rare cases, as seen in the above images.



Figure S3. UV-Vis spectra of 40 mM EHAC, 20 mM ACA, and 22 mM NaOH before irradiation (cyan line), after UV irradiation for 1 h (blue line), and after subsequent visible light irradiation for 1.5 h (dotted red line). The sample was diluted 20 times with water. The changes at the visible wavelengths are enlarged and shown in the inset. UV irradiation causes a drop in absorbance at UV wavelengths and an increase in absorbance at visible wavelengths, indicating that ACA has been photoisomerized from *trans* to *cis*. Subsequent visible-light irradiation causes an increase in absorbance at the UV wavelengths and decrease at the visible wavelengths, indicating the reverse *cis* to *trans* photoisomerization.