Electronic Supplementary Information

Materials and Methods

DNA-ELP Hybrid Liquid Crystals. Two elastin-like peptides having amidated C-termini, $K_3(VPGVG)_3K_3$-NH_2 (K3ELP) and $K_4(VPGVG)_3K_4$-NH_2 (K4ELP), and synthesized using 9-fluorenylmethyloxycarbonyl (Fmoc) chemistry (80% purity, Coast Scientific) were used as received. Solutions of the ELPs (10 mg/mL) were added dropwise to short fragment (~146 bp) DNA solutions (10 mg/mL), both in 10 mM Tris·HCl (pH 7.3), at room temperature until DNA condensation was visibly observed. Condensation occurred very close to the peptide concentration necessary to achieve charge neutrality. The liquid crystalline phase was collected at the bottom of the solution by centrifugation and then retained in the supernatent solution for all studies.

Circular Dichroism. Far-UV CD spectra of the ELPs (1 mg/mL) in 10 mM sodium phosphate (pH 7.0) were collected on a Jasco J-715 spectropolarimeter equipped with a Peltier temperature controller (Jasco PTC-348WI) using a 0.2 cm quartz cuvette. Wavelength scans were performed from 260 to 190 nm using a step resolution of 0.2 nm, speed of 20 nm/min, 50 mdeg sensitivity, and 4 accumulations. Measurements were made from 5 to 65 ^{o}C in 5 ^{o}C increments. Samples were allowed to equilibrate for about 5 min at each temperature before collecting scans. Ellipticity, θ, was converted to mean

residue ellipticity $[\theta] = \theta/10cl$, where c = concentration in [mol residues/L] and l = pathlength.

X-ray Scattering. Small-angle X-ray scattering (SAXS) measurements were performed on a Rigaku RU-H3R rotating anode X-ray diffractometer equipped with an Osmic multilayer focusing optic ((100 μm)^2 point focus) and an evacuated Statton-type scattering camera. The sample-to-detector distance was 460 mm, which corresponds to a Q range of 0.698 nm^{-1} $\leq Q \leq$ 6.25 nm^{-1} with $Q = (4\pi/\lambda)\sin(\theta/2)$, where θ is twice the Bragg angle. The incident beam wavelength was 0.154 nm, corresponding to 8 keV Cu Kα radiation. Samples retained in their supernatent solution were sealed in glass capillaries to isolate them from vacuum, and temperature was controlled using a Peltier device. The temperature sequence was: 20, 10, 30, 40, 50, 60, 45, 20 ^{o}C. Scattering patterns were acquired with 10 cm × 15 cm Fuji ST-VA image plates in conjunction with a Fuji BAS-2500 image plate scanner, and intensity profiles were obtained. The scattering intensity profiles $I(Q) = |F(Q)|^2 S(Q)$ were fitted with a first-order Bessel function of the first kind for the cylindrical form factor of DNA $F(Q)$ (radius = 1 nm) and a Lorentzian function for the structure factor $S(Q)$. The interaxial spacing was calculated from the Bragg spacing D_{Bragg} as $D_i = (2/\sqrt{3})D_{\text{Bragg}}$, assuming local hexagonal packing.

Polarizing Optical Microscopy. Polarizing optical microscopy was performed with a Zeiss Axiovert S100 TV inverted polarizing microscope (objective: Zeiss Plan-Neofluar

$40 \times / 0.85$ pol) equipped with an LC Pol-Scope retardance imaging system (CRI, Boston, MA), which simultaneously measures the magnitude and direction of birefringence. DNA-ELP hybrid liquid crystals retained in their supernatent solution were prepared on glass slides using vacuum grease to seal the samples under a coverslip and Cytoseal mounting medium around the edge of the coverslip to prevent drying. Since relatively thick samples were necessary to observe the cholesteric fingerprint pattern in these samples, P was calculated from profile plots created using the ImageJ software (*1*) rather than taking the Fourier transform of the birefringence images as described previously for DNA cholesteric droplets (*2, 3*). Variable temperature experiments were performed using a homemade heating block constructed from aluminum with internal copper tubing connected to a water bath. A thermocouple was used to calibrate the setup and monitor the sample temperature. Multiple images were obtained from different regions of a sample at each temperature using the sequence: 10, 20, 30, 35, 40, 50, 20 ^{o}C. Several samples also were measured and the data was averaged.

References

1. NIH Image Program developed at the U.S. National Institutes of Health and available on the Internet at http://rsb.info.nih.gov/nih-image/.

2. M. Leonard, H. Hong, N. Easwar, H. H. Strey, *Polymer* **42**, 5823 (2001).

3. C. B. Stanley, H. Hong, H. H. Strey, *Biophys. J.* **89**, 2552 (2005).