Supplementary Figures

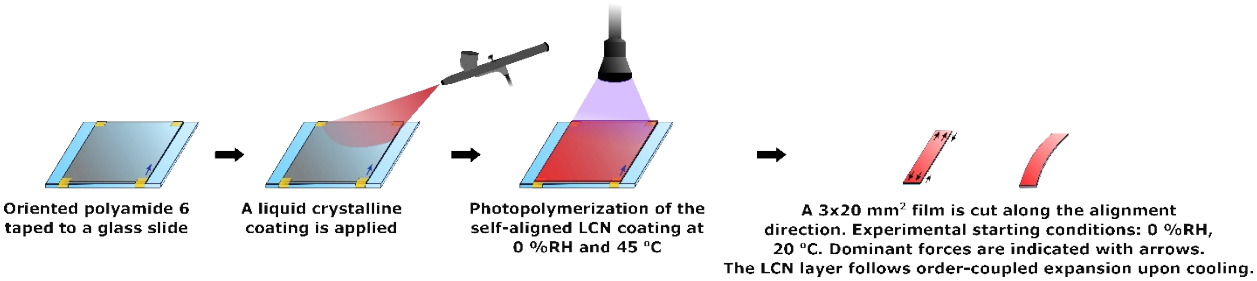

Figure S1. A schematic representation of the LCN/PA6 bilayer fabrication process. From left to right: an oriented PA6 film is cut, cleaned and taped to a 5 x 5 cm^2 glass slide. Liquid crystals are sprayed onto pre-dried PA6 films and UV-cured at 45 °C. Thin strips of 20 x 3 mm^2 were cut from the larger film, and were pre-bent with PA6 side inside the bend at room temperature.

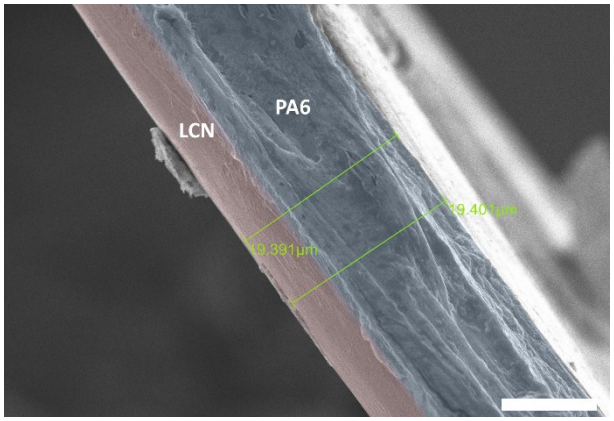

Figure S2. A cross-sectional SEM image of an LCN/PA6 bilayer. The LCN coating (in red) and PA6 substrate (in blue) have been colored to improve contrast. The scale bar equals 10 µm.

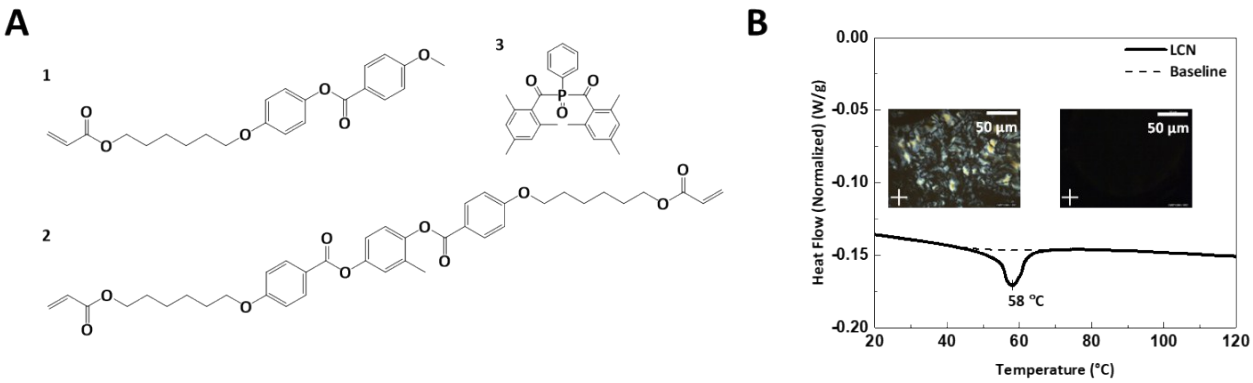

Figure S3. A. Thermo-responsive liquid crystal mixture consisting of 75 wt.% **1**, 24 wt.% **2** and 1 wt.% **3**. **B.** Phase behavior of the liquid crystalline mixture. Temperature scan rate was 5 °C/min; heat flow endo down.

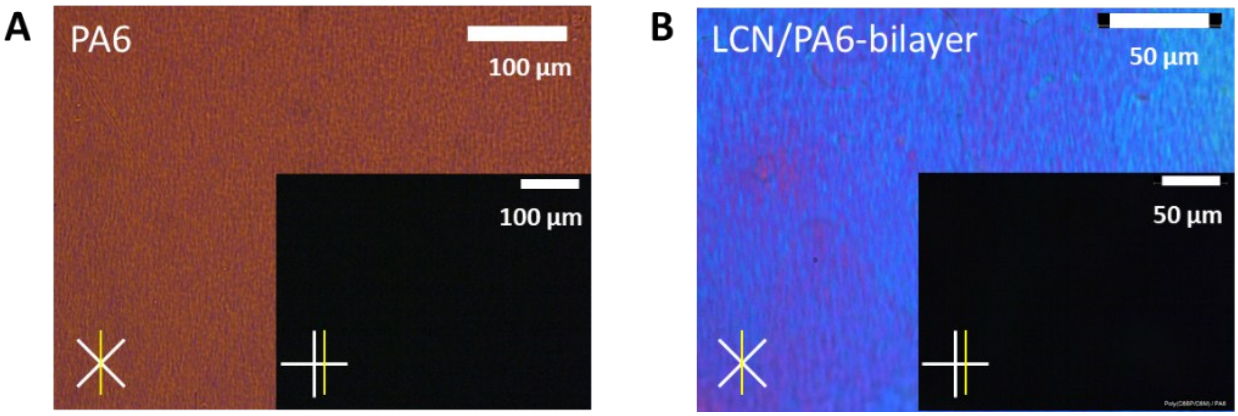

Figure S4. Polarized optical micrographs of: **A.** the oriented 15 µm PA6 substrate. **B.** the well-aligned LCN/PA6-bilayer. The arrows indicate the orientation of the crossed polarizers with respect to the sample orientation in yellow.

The surface alignment of the liquid crystal network was determined using polarized infrared spectroscopy. In total reflection mode the order parameter for the LC surface alignment is approximated, with S is defined as

$$S = \frac{(D-1)}{(D+2)}$$

Where

$$D = \frac{A_{//}}{A_{\perp}}$$

and $A_{//}$ and $A_{\perp}$ are the absorbance of light polarized along and perpendicular the stretching axis of the PA6, respectively. For planar nematic LC materials, the S-value is typically 0.6 – 0.7, while isotropic samples have a value of 0. The ν_{8a} and ν_{19a} vibrational bands at 1606 and 1511 cm^{-1} were used to approximate an order parameter by means of a dichroic ratio in absorbance (see **Figure S4**).[35] Since these bands are associated with the phenyl stretching vibration, one may assume that stretching occurs in the same direction as the long molecular axis of the mesogens. The derived surface order parameters of S_{1606} ~ 0.27 and S_{1511} ~ 0.30 are consistent with a splay-aligned film.[24]

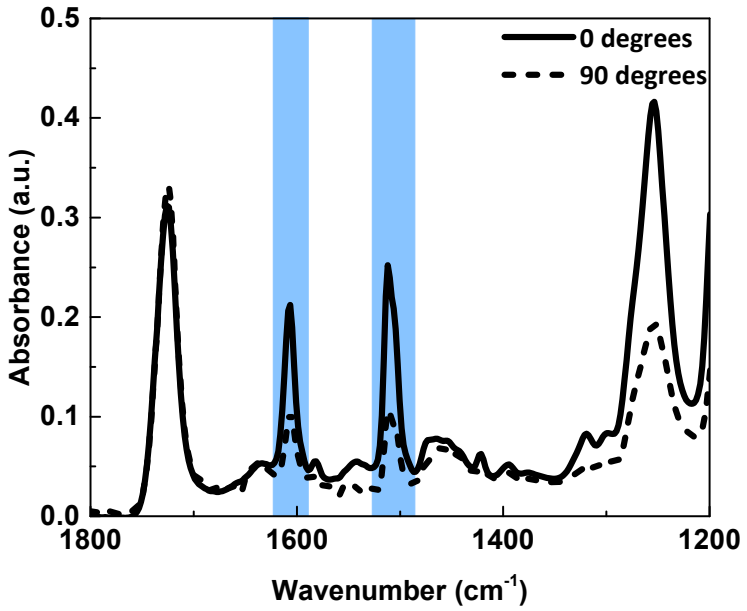

Figure S5. The optical anisotropy in LCN/PA6 bilayers is examined using the vibrational bands ν_{8a} = 1606 cm^{-1} and ν_{19a} = 1511 cm^{-1}. Polarized infrared spectra were taken in reflection mode to estimate an order parameter, S.

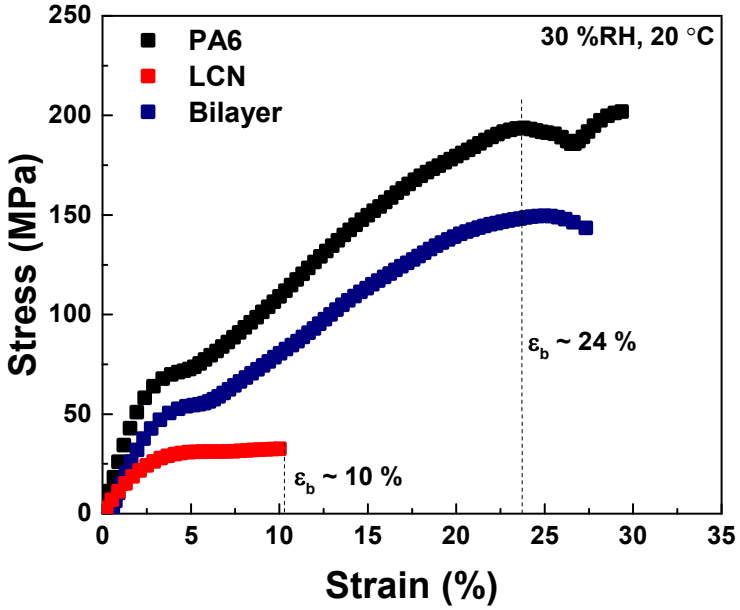

Figure S6. Stress-strain curves of PA6 and LCN/PA6 bilayer actuators, measured in the orientation direction at a strain rate of 1 mm/min (30 %RH, 20 °C). Mechanical failure of the bilayer occurs at an elongation of ~ 24 %.

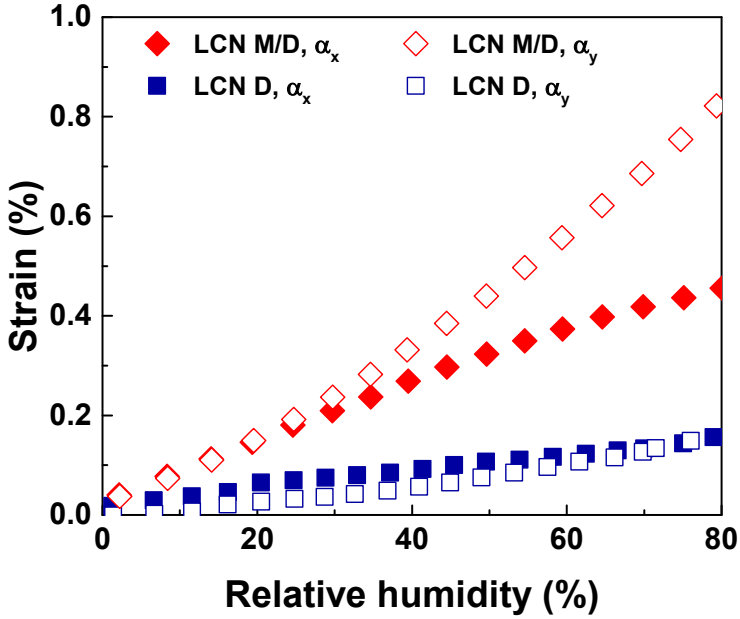

Figure S7. Anisotropic coefficients of humidity expansion given as strain-values (ε) were determined for two different LC compositions. In the legend M and D correspond to mono- and diacrylate, respectively. LCN films composed of mono- and diacrylates significantly swell in the perpendicular direction (red, open symbols). Films that only consist of diacrylates (blue symbols) isotropically swell up to strain values of 0.15 %.