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Confinement-guided self-assembly of ionic superdiscs

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Discotic ionic liquid crystals (DILCs) consist of self-assembled superdiscs of cations and anions that spontaneously stack in linear columns with high one-dimensional ionic and electronic charge mobility, making them prominent model systems for functional soft matter. Compared to classical non-ionic discotic liquid crystals (DLCs), many novel liquid crystalline structures with a unique combination of electronic and ionic conductivity have been reported, which are of interest for separation membranes, artificial ion/proton conducting membranes and optoelectronics. Unfortunately, a homogeneous alignment of the DILCs on the macroscale is often not achievable, which significantly limits the applicability of DILCs. Infiltration into nanoporous solid scaffolds can overcome this drawback. However, little is nknown about the structures of DILCs in nanoscale confinement. Here, we present temperature-dependent high-resolution optical birefringence measurement and 3D reciprocal space mapping based on synchrotron-based X-ray scattering to investigate the thermotropic phase behavior of dopamine-based ionic liquid crystals confined in cylindrical channels of 180 nm diameter in macroscopic anodic aluminum oxide (AAO) membranes. As a function of the membranes hydrophilicity and thus the molecular anchoring to the pore walls (edge-on or face-on) and the variation of the hydrophilic-hydrophobic balance between the aromatic cores and the alkyl side chain motifs of the superdiscs, we find a particularly rich phase behavior, which is not present in the bulk state. It is governed by a complex interplay of liquid crystalline elastic energies (bending and splay deformations), polar interactions and pure geometric confinement, and includes textural transitions between radial and axial alignment of the columns with respect to the long nanochannel axis. Furthermore, confinement-induced continuous order formation is observed in contrast to discontinuous first-order phase transitions, which can be quantitatively described by Landau-de Gennes free energy models for liquid crystalline order transitions in confinement. Our observations suggest that the infiltration of DILCs into nanoporous solids allows tailoring their nanoscale texture and thus their electrical and optical functionalities over an even wider range than in the bulk state, in a homogeneous manner on the centimeter scale as controlled by the monolithic nanoporous scaffolds.

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