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Perfect mixing of immiscible macromolecules at fluid interfaces¹ SERGEI SHEIKO, University of North Carolina at Chapel Hill, KRZYSZTOF MATYJASZEWSKI, Carnegie Mellon University, VLADIMIR TSUKRUK, Georgia Institute of Technology, JAN-MICHAEL CARRILLO, University of Connecticut, MICHAEL RUBINSTEIN, University of North Carolina at Chapel Hill, AN-DREY DOBRYNIN, University of Connecticut, JING ZHOU, University of North Carolina at Chapel Hill — Macromolecules typically phase separate unless their shapes and chemical compositions are tailored to explicitly drive mixing. But now our research has shown that physical constraints can drive spontaneous mixing of chemically different species. We have obtained long-range 2D arrays of perfectly mixed macromolecules having a variety of molecular architectures and chemistries, including linear chains, block-copolymer stars, and bottlebrush copolymers with hydrophobic, hydrophilic, and lipophobic chemical compositions. This is achieved by entropy-driven enhancement of steric repulsion between macromolecules anchored on a substrate. By monitoring the kinetics of mixing, we have proved that molecular intercalation is an equilibrium state. The array spacing is controlled by the length of the brush side chains. This entropic templating strategy opens new ways for generating patterns on sub-100 nm length scales with potential application in lithography, directed self-assembly, and biomedical assays.

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Sergei Sheiko University of North Carolina at Chapel Hill

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