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Crystallization Induced by Electrostatic Correlations in Vesicles of Mixed-Valence Ionic Amphiphiles[1] CHEUK YUI LEUNG, RASTKO SKNEPNEK, LIAM PALMER, GRAZIANO VERNIZZI, MEGAN GREENFIELD, SAMUEL STUPP, MICHAEL BEDZYK, MONICA OLVERA DE LA CRUZ, Northwestern University — Charged amphiphilic molecules, including molecules with biological motifs, have been predicted to organize into elastic membrane or crystalline shells with non-spherical shapes. We demonstrate that pure electrostatic interaction allow (-1) anionic water insoluble amphiphiles and (+3) cationic amphiphiles, which form only micelles in water, to co-assemble into buckled vesicles. The strong interaction between the +3 and -1 head groups increases the cohesive energy of the amphiphiles and favors the formation of crystallized membranes or shells that facet spontaneously into buckled shapes predicted by simulations of vesicles with heterogeneous elastic properties. In situ small-angle and wide-angle X-ray scattering (SAXS-WAXS) experiments conducted at the Advanced Photon Source DND-CAT confirm the presence of crystalline bilayers. Our simulations verify that ionic lateral correlations among the oppositely charged head groups of the co-assembled amphiphiles are responsible for the observed tail crystallization.

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