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Ligand-induced order in Spotted vesicles and Striped micelles

DAVID CHRISTIAN, WOUTER ELLENBROEK, ANDREA LIU, DENNIS DISCHER, MRSEC, University of Pennsylvania — Mixtures of amphiphiles are ubiquitous and assemble into various morphologies, including giant vesicles and cylinder micelles that raise the possibility of mesoscopic segregation within the assemblies – perhaps even in response to binding of small ligands. Here, with anionic and neutral polymer amphiphiles mixed within vesicle and cylinder morphologies, divalent cations are shown to induce meso-scale domains and thus generate ‘*responsive Janus structures*’. Whereas past reports with lipid systems appear conflicted, calcium forms definitive crossbridges between the anionic polymer amphiphiles, rigidifying the charged membranes across a fluid-gel transition and also leading to lateral phase separation without disrupting the assemblies. A systematic phase diagram for these robust assemblies shows that long-lived domains occur in an unexpectedly narrow region near the polyanion’s pK ’s for protonation and cation association. The phase behavior appears well described by a relatively simple model in which – among electrostatic and entropic contributions – counterion entropy outcompetes attractive crossbridging to drive remixing of the highly charged polyacid at high pH, contrary to intuition.

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