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Calcium-induced gel domains in bilayer – more elusive than thought DENNIS DISCHER, DAVID CHRISTIAN, WOUTER ELLENBROEK, ANDREA LIU, MRSEC, University of Pennsylvania — As a highly bioactive divalent cation, calcium can in principle crossbridge anionic groups and induce domain formation and rigidification, but past reports with lipid systems appear conflicted. We mix, as a robust model system, anionic and neutral polymer amphiphiles within vesicle and cylinder micelle morphologies and add calcium. Based on micro- measurements, calcium forms definitive crossbridges between the anionic 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 shows that long-lived domains occur in a narrow region near the polyanion's pK's. 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. Initial observations extend from polymers to a polyanionic lipid involved in cell signaling [phosphatidylinositol (4,5)-bisphosphate], highlighting both the generality and limits of calcium effects.

Dennis Discher
University of Pennsylvania

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