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Title: pH-driven ordering transitions in liquid crystal induced by conformational changes of cardiolipin

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Abstract:

We report an investigation of interfacial phenomena occurring at aqueous-liquid crystal (LC) interfaces that triggers an orientational ordering transition of the LC in the presence of cardiolipin (CL) by varying pH, salt concentration and valence. In particular, the effects of three different conformational isomeric forms of the CL are observed to cause the response of the LC ordering to vary significantly from one to another at those interfaces. An ordering transition of the LC was observed when the CL is mostly in undissociated (at pH 2) and/or in bicyclic (at pH 4) conformation in which LC shows changes in the optical appearance from bright to dark. By contrast, no change in the optical appearance of the LC was observed when the pH of the system increases to 8 or higher in which the CL mostly exists in the open conformation. Fluorescence microscopy measurements further suggest that pH-dependent conformational forms of the CL have different ability to self-assemble (thus different packing efficiency) at aqueous-LC interfaces leading to dissimilar orientational behavior of the LC. Specifically, we found that change in headgroupheadgroup repulsion of the central phosphatidyl groups of the CL plays a key role in tuning the lipid packing efficiency and thus responses to interfacial phenomena. Orientational ordering transition of the LC was also observed as a function of increasing the ionic strength (buffer capacity) and strongly influenced in the presence of mono and divalent cations. Langmuir-Blodgett (LB) and polarization modulation infrared reflection absorption spectroscopy (PM-IRRAS) measurements provide further insight in modulation of the lipid packing efficiency and alkyl chain conformation of the CL at different pH and ionic conditions. Overall, the results presented in this paper establish that LCs offer a promising approach to differentiate different conformations (label free detection) of the CL through ordering transition of the LC at aqueous-LC interfaces.

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