Packing Density Changes of Supported Lipid Bilayers Observed by Fluorescence Microscopy and Quartz Crystal Microbalance-Dissipation

Chiho Kataoka-Hamai, Mahoko Higuchi

International Center for Materials Nanoarchitectonics, National Institute for Materials Science, 1-1 Namiki, Tsukuba, Ibaraki 305-0044, Japan

SUPPLEMENTAL MATERIAL -

Dependence of mobile fraction on pH.

We found that the mobile fraction varied with solution pH (Figure 2). However, the reason remains unclear. In previous studies, electrostatic and van der Waals forces, or an electrostatic force alone, have reasonably explained vesicle fusion.¹⁻⁵ The Debye screening length (~2 nm) estimated for the ionic strength used here (Table 1) was similar to typical bilayer-surface separation,⁶⁻⁸ suggesting that the electrostatic effect was strong. As described in the paper, TR-labeled DOTAP vesicles are positively charged in all of the solutions. In contrast, the glass surface is negatively charged. The overall surface charge decreases with pH due to titratable silanol groups and is at a minimum at pH 3.0;⁹⁻¹⁰ thus, the electrostatic interaction between the vesicles and surface is weaker at lower pH, which may account for the unfavorable fusion at pH 4.0–7.2. However, this interpretation fails to explain the recovery of the mobile fraction in the acidic region. Covalent bond formation will not explain this recovery because the vesicles and surface do not have suitable functional groups available for chemical interactions. Furthermore, no reactions have been reported for similar systems consisting of phosphatidylcholine (PC) vesicles and glass or fused silica at pH 3.^{49,11}

References

1. Anderson, T. H.; Min, Y. J.; Weirich, K. L.; Zeng, H. B.; Fygenson, D.; Israelachvili, J. N. Formation of Supported Bilayers on Silica Substrates. *Langmuir* **2009**, *25*, 6997–7005.

2. Cha, T.; Guo, A.; Zhu, X.-Y. Formation of Supported Phospholipid Bilayers on Molecular Surfaces: Role of Surface Charge Density and Electrostatic Interaction. *Biophys. J.* **2006**, *90*, 1270–1274.

3. Cho, N. J.; Frank, C. W. Fabrication of a Planar Zwitterionic Lipid Bilayer on Titanium Oxide. *Langmuir* **2010**, *26*, 15706–15710.

4. Cremer, P. S.; Boxer, S. G. Formation and Spreading of Lipid Bilayers on Planar Glass Supports. *J. Phys. Chem. B* **1999**, *103*, 2554–2559.

5. Richter, R. P.; Brisson, A. R. Following the Formation of Supported Lipid Bilayers on Mica: A Study Combining AFM, QCM-D, and Ellipsometry. *Biophys. J.* **2005**, *88*, 3422–3433.

6. Bayerl, T. M.; Bloom, M. Physical Properties of Single Phospholipid Bilayers Adsorbed to Micro Glass Beads. A

New Vesicular Model System Studied by ²H-Nuclear Magnetic Resonance. *Biophys. J.* **1990**, *58*, 357–362.

7. Kataoka-Hamai, C.; Miyahara, Y. Label-Free Detection of DNA by Field-Effect Devices. *IEEE Sens. J.* 2011, *11*, 3153–3160.

8. Kiessling, V.; Tamm, L. K. Measuring Distances in Supported Bilayers by Fluorescence Interference-Contrast Microscopy: Polymer Supports and SNARE Proteins. *Biophys. J.* **2003**, *84*, 408–418.

9. Cunliffe, J. M.; Baryla, N. E.; Lucy, C. A. Phospholipid Bilayer Coatings for the Separation of Proteins in Capillary Electrophoresis. *Anal. Chem.* **2002**, *74*, 776–783.

10. Groves, J. T.; Boxer, S. G. Electric Field-Induced Concentration Gradients in Planar Supported Bilayers. *Biophys. J.* **1995**, *69*, 1972–1975.

11. Monson, C. F.; Pace, H. P.; Liu, C. M.; Cremer, P. S. Supported Bilayer Electrophoresis Under Controlled Buffer Conditions. *Anal. Chem.* **2011**, *83*, 2090–2096.