

Formation of charged lipid layers on polyelectrolyte support

R. Krastev^{1,2}, N. Ch. Mishra², T. Gutberlet³

¹ Hahn-Meitner-Institute, Berlin, Glienicke Str. 100, 14109 Berlin, Germany

² Max-Planck Institute of Colloids and Interface, 14424 Potsdam, Germany

³ Laboratory for Neutron Scattering, ETHZ & PSI, CH-5232 Villigen PSI, Switzerland

Adsorption of negatively charged lipid DMPG on PSS or PAH terminated PEMs were studied. Formation of layers of around 30 Å thickness with low SLD typical for phospholipids was observed in both cases. Influence on the presence of electrolyte in the lipid suspension was also observed. Application of complicated fitting models are necessary to fit properly the NR data in the case of adsorption of DMPG onto PAH terminated PEMs.

There is considerable interest to create model systems that maintain the structural and dynamic integrity of native biomembranes, e.g. supports that do not hinder the lateral mobility of membrane components. Supported lipid membranes are being used increasingly as well-defined model systems for fundamental biophysical research and have been proposed as surfaces for the immobilization of proteins. A promising strategy for decoupling the biomembrane from the underlying surface is to rest the biomembrane on a soft cushion composed of a water swellable hydrophilic polymer. The conditions for formation of zwitterionic DMPC lipid bilayers on top of such cushions were obtained in our recent studies [1, 2] and they have shown that cushions prepared on solid support from polyelectrolytes (PE) by alternative cationic/anionic layer-by-layer (LbL) [3] deposition may serve as water swellable support for preparation of lipid membranes.

The present work studied the adsorption behavior of the negatively charged lipid dimyristoyl-phosphatidylglycerol (DMPG) on PE multilayer (PEM) support. The PEM were prepared by LbL technique from the negatively charged deuterated polystyrene sulfonate (dPSS) and the positively charged polyallylamine hydrochloride (PAH). A layer of polyethyleneimine (PEI) was used as a first layer to facilitate the build-up process. PEMs with different termination (Si/PEI/(dPSS/PAH)₆ or Si/PEI/(dPSS/PAH)₆/PSS) were used to study the origin of the interaction between the lipid and the substrate. DMPG was adsorbed from lipid suspensions prepared via extrusion with smallest membrane pore diameter of 50 nm.

Neutron reflectivity (NR) curves from PAH terminated PEM supports are shown on Fig. 1a. Experiments were carried out at three concentrations of electrolyte NaCl – 0, 0.01 and 0.03 M. The electrolyte content in the DMPG suspension influences the dissociation of the lipid molecules [4]. Curve 1 represents a bare sample at 0 M NaCl concentration. The curves at other electrolyte concentrations do not deviate from the one shown. Incorporation of deuterated PE in the PEM structures gives high scattering length density (SLD) of the bare samples when in contact with D₂O. Thus, PEM/D₂O interface is almost invisible for neutrons and only weak oscillations in the reflectivity curves are observed. Curve 2 and 3 were recorded after DMPG adsorption at 0 and 0.03 M NaCl, respectively. The adsorption of a lipid layer with a low SLD at the PEM/D₂O interface causes a local minimum in the SLD profile, and thus this interface becomes visible for the neutrons. This gives rise to a series of pronounced maxima and minima in $R(Q)$. Best fits to the experimental data are shown with lines. There is a certain difference in the curves at 0 M NaCl and that at 0.03 M

NaCl (2 and 3), but the curves at 0.01 (not shown) and 0.03 M NaCl are practically identical. The data were fitted with a two box model which represents the PEM and the lipid layer on top of it. Formation of a layer with low SLD characteristic for the lipid layer with a thickness of around 30 Å onto the PEM is detected in all cases. This most likely represents a formation of a single DMPG layer on the PEM. The presented fits do not fit the experimental curves at higher Q . More complex fitting models have to be used to get reliable agreement which is in progress.

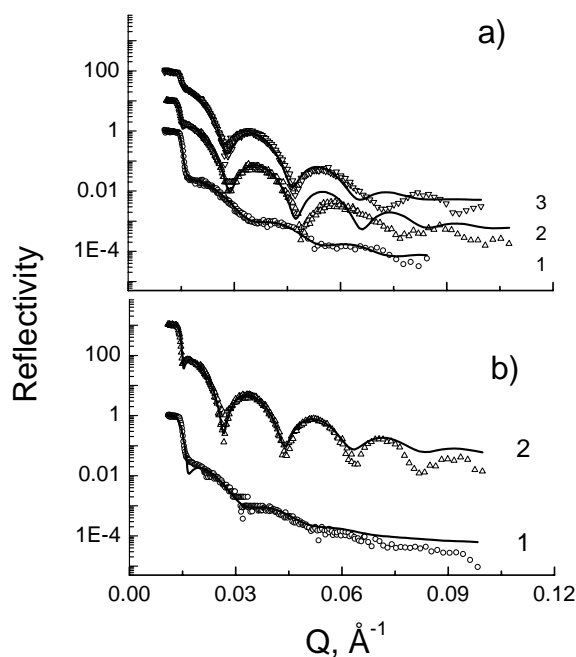


Figure 1: Neutron reflectivity measurements of DMPG layers adsorbed on PEM/D₂O interface.

In Fig. 1b NR curves from PSS terminated PEM supports are shown before and after DMPG (at 0 M NaCl) adsorption. Surprisingly negatively charged DMPG adsorbs also onto negatively charged PEM. A two box model fits the data successfully and shows formation of a 28 Å thick layer with low SLD.

- [1] Delajon, Krastev, Gutberlet, *Langmuir*, in preparation
- [2] Ropers et al., *BENS Exp. Rep.* (2002) 171 and 172
- [3] Decher, *Science* **277** (1997) 1232
- [4] Grigoriev et al, *J. Phys. Chem. B*, **103** (1999) 1013

Work fully performed at SINQ
Proposal-number: II/04 S-13
Instruments: AMOR