Experimental report

14/01/2016

Proposal: **9-13-581**

Council: 10/2014

Title: Tribology of lipid layer: effect of hydration on structural properties

Research area: Soft condensed matter

This proposal is a new proposal

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Samples: D20
Silicon
dspc

Instrument	Requested days	Allocated days	From	To
FIGARO	3	0		
D17	3	3	26/06/2015	29/06/2015

Abstract:

Nature has produced water-based lubricant systems that far outclass the best of most man-made devices. Biolubrification involves complex hydrodynamic flow in the vicinity of soft interfaces as fluid membranes in a wide length scales. A major challenge resides thus in understanding the relations connecting these different scales. This is the main goal of our project which combine the expertise on tribology, biotribology and rheology experiments. We have developed an original set-up which allow us to combine well defined tribology and velocimetry experiments.We have first investigated tribology of lipid monolayer and tri-layer at solid/air interface. A first surprising result is the importance of the humidity rate on the shear plane location and on the tribological properties which exhibit a non-monotonic behavior. To have a better understanding of these results, we need to have hight resolution structural information on monolayer and tri-layer under different humidity rate by using neutron reflectometry. This is the purpose of this proposal.

Experimental Report-Experiment n° 9-13-581

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1 Context

Nature has produced water-based lubricant systems that far outclass the best of most man-made devices [1]. Biological contacts such as the articulating cartilage surfaces in human hips or knees often operate under severe conditions (i.e., high load and low speed), which is related to a boundary lubrication regime [2]. In this regime, actual contact between the surfaces can be prevented by a boundary lubricant that attaches itself to the solid surfaces due to molecular forces, thereby modifying their tribological properties. This boundary regime is characterized by a very low friction coefficient ($\mu = 0.005 - 0.02$, for the human joint) [3]. Briscoe et al [4] found that boundary lubrication under water was far superior to that in air or oil, and mediated by the hydrated surfactant headgroups. These studies emphasize the importance of hydration layers surrounding charges in aqueous media as a basic lubrication element [5]. Self-assembled phospholipid structures are abundant in all organisms and play vital roles in many biological processes. They form bilayers that experience repulsive steric hydration and thermal fluctuations that prevent attraction between adjacent membranes and play an important lubricant role in all biological rubbing surfaces. Stacks of 3 to 7 lipid bilayers separated by aqueous layers have been found in almost all of these biological rubbing surfaces. However, only very few studies have reported quantitative data on the tribological role of phospholipid bilayers [6] and understanding deeply the mechanism of boundary lubrication is still challenging. At the molecular level, recent works have shown that the stacking of lipid bilayers is the most serious candidate for this exceptional biolubrication, but the mechanisms are not understood yet.

In this framework, experiments carried out in Strasbourg (team Physics-Mechanics and Tribology of Polymers and team Membrane and Microforces, Institut Charles Sadron) using an original new set-up *NanoTribo-FRAPP* showed a first interesting results on tribology of lipid monolayer and tri-layer at solid/air interface. A first surprising result is the importance of the humidity rate on the shear plane location and on the tribological properties. To have a better understanding of these results, we need to have high resolution structural information on monolayer and tri-layer under different humidity rate. This is the purpose of the proposed experiments.

2 Experiment

Samples were prepared in ILL using Soft Matter Lab facilities. We made supported monolayer and tri-layer of DSPC (DiStearoylPhosphatidylCholines) on a silicon block by Langmuir-Blodgett technique. The surface pressure was set at 40 mN/m, which correspond to a gel phase of DSPC. The instrument D17 works very well, and thanks to a homemade humidity chamber, we were able to vary the temperature and the humidity of the sample for various contrast of the vapor, and to characterize the evolution of structure with high resolution.

We have done experiments on 5 samples (2 silicon substrates, 1 DSPC monolayer, 2 DSPC tri-layers) for 3 different contrasts (H_2O, 4MW and D_20), both in Gel and Fluid phase for lipids. In total we obtained 28 reflectivity curves. We present below a first preliminary analysis of these experiments.

3 Results

We have firstly checked the bare silicon block under the variation of humidity, and we found that when the relative humidity rises to 90%, there is a very thin layer of water about 0.3 nm formed on the surface in agreement with complete wetting theory (data not shown). Then a supported monolayer and a supported tri-layer were prepared in H_2O. The reflectivity curves shown a reversible change of the structure of tri-layer under the variation of relative humidity from 30% to 90% and then back to 30% (see figure 1). Thanks to the well-designed humidity chamber, we used 3 different water contrasts D_20 / H_20 / SMW (Silicon Match Water: 38%D20+62%H20) in order to facilitate the fitting while keeping the same monolayer or tri-layer.

The first important result we have achieved is that the mono and tri-layer are stable in time even for a large humidity rate ($RH = 90\%$). We also demonstrate the good solvant exchange through the vapor. We observed significant changes in tri-layer structure with the humidity rate.The thickness of the layer of water between the third monolayer and the supported bi-layer was about 0.3 nm at $RH = 30\%$, then rise to about 1.2 nm at $RH = 90\%$. We also tried to increase temperature to that above the gel-fluid lipid phase transition for the supported tri-layer, and no significant changes of structure were observed.

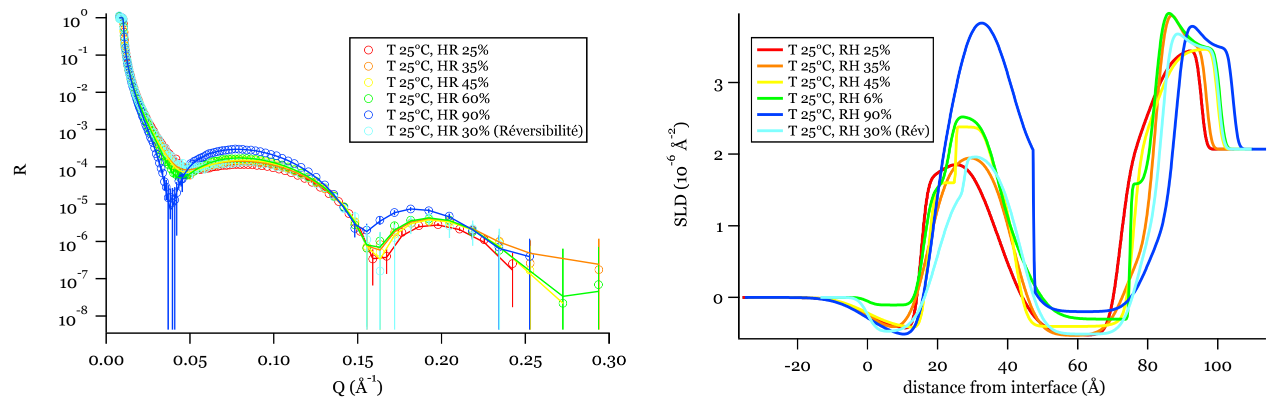

Figure 1: Reflectivity curves of supported DSPC tri-layer (at 25°C) deposited on silicon block with different relative humidity.

4 Conclusion

From tribological experiments, we could expect maybe a structure modification of supported DSPC monolayer and tri-layer by changing the relative humidity of the environment. Thanks to neutron reflectivity, we were able to confirm the effects of humidity on the supported tri-layer. And this structural evolution is reversible in the range of relative humidity from 30% to 90%. These experiments bring us an interest of theoretical study on the effects of humidity for the supported lipid layers and give us indications for future neutron reflectivity experiments. I open also interesting perspectives to investigate interaction of supported bilayer with biomolecules as peptides in proteins.

References

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