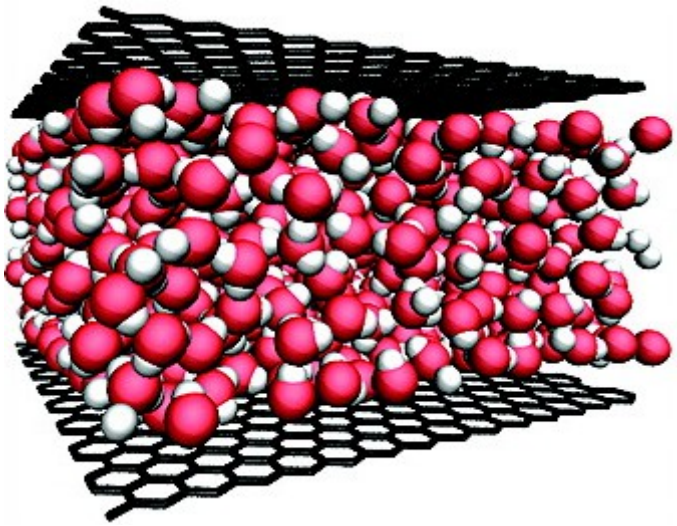
Influence of Interfacial Interactions on Structural and Rheological Properties of Confined Liquids



September 23, 2013

Centre Blaise Pascal, ENS Lyon, France

CBP Centre Blaise Pascal FLMSN ANR Biolub Université de Lyon ENS de Lyon

Influence of Interfacial Interactions on Structural and Rheological Properties of Confined Liquids

September 23, 2013

PROGRAM

08:30-09:00	Breakfast
09:00-09:45	*Molecular biomechanics : Phase Transitions in Lipid Multilayers under Shear and Pressure"* Michael GRUNZE (Heidelberg)
09:45-10:30	*Simple fluids in slit micropore: from local transport properties to shear induced swelling* Guillaume GALLIERO (Pau)
10:30-11:00	*Coffee break*
11:00-11:45	*Vesicles in contact with multiple aqueous phases* Halim KUSUMAATMAJA (Durham)
11:45-12:30	*Observing and understanding membrane interactions* Kheya SENGUPTA (Marseille)
12:30-14:00	Lunch
14:00-14:45	*Adhesion of membranes in confined environments* Thomas LE GOFF (Lyon)
14:45-15:30	*Short range interactions between phospholipid bilayers: disecting the forces* Michael GRUNZE (Heidelberg)
15:30-16:00	Coffee Break
16:00-16:45	*Bottom-up approach of multiscale dynamics in porous media. Application to Gas Shale* Alexandru BOTAN (Boston)
16:45-17:30	*Linear alkanes confined between smooth solid surfaces* Nicolas FILLOT and Daniele SAVIO (Lyon)

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Molecular Biomechanics:
Phase transitions in Lipid Multilayers under shear and pressure

Michael GRUNZE
Angewandte Physikalische Chemie , Universität Heidelberg, and Institute of Functional Interfaces, KIT, Karlsruhe, Germany

In this talk I will discuss the first experiments to study the phase behavior of lipid multilayers in pure water, and in a model synovial fluid under pressure and shear typical in human joints. The data were taken on the new Neutron Reflectometer "Bioref" at the Helmholtz Center Berlin for Materials and Energy (HZB), which was build in a collaboration with the University of Heidelberg. "BioRef" combines neutron reflectivity and IR Reflection Spectroscopy to probe the mechanical and temperature effects on the stability and phase transitions in lipid multilayers. This allows for the first time to correlate molecular conformations in the lipid molecules with mesoscopic phase changes in the multi-lamellar system. The phase behavior of 1,2-Dimyristoyl-sn-Glycero-3-phosphocholine (DMPC) multilayers was studied both for the pure lipids, and with addition of Hyaluronic Acid (HA) to mimic the synovial fluid in a joint. Addition of HA leads to swelling of the multilayers into the micrometer thickness range and stabilizes them against temperature, pressure and shear. We find that the gel-liquid transition in the lipid layers as probed by the frequency of the methylene vibrations in the alkane chains of the DMPC molecules are not effected by the addition of HA. To explain the observation that the swollen multilayers are stable against a appreciable osmotic pressure, we speculate that tethering of HA polymers and mechanically connecting the bilayers leads to the surprising stability. At a shear frequency below 3 Hertz the lamellar system is stable, whereas at higher shear frequency the multilayers are destroyed. Extrapolating our model experiments to the human knee, this means: walk-don´t run.

Simple fluids in slit micropore: from local transport properties to shear induced swelling

Prof. Guillaume GALLIERO
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Using extensive molecular dynamics simulations on simple fluids confined in a slit micropore, we have studied the impact of the confinement on various dynamic properties. In particular we will show how the local variations of the transport properties of the confined fluid (diffusion [1], viscosity [2]), induced by density inhomogeneities, may be described by a simple local average density model [3]. In addition, we will discuss the coupling that may occur between shear and swelling of such highly confined systems [4].

[1] H. Hoang, G. Galliero, J. Chem. Phys. **136**, 184702 (2012)

[2] H. Hoang, G. Galliero, Phys. Rev. E **86**, 021202 (2012)

[3] I. Bitsanis, T.K Vanderlick, M. Tirell and H.T. Davis, J. Chem. Phys. **87**, 1733 (1987)

[4] P. Bordarier, M. Schoen, A.H. Fuchs, Phys Rev E 57, 1621 (1998)

Vesicles in contact with multiple aqueous phases

Halim KUSUMAATMAJA
Department of Physics, Durham University

We study novel soft matter systems consisting of lipid vesicles and phase separating polymer solutions. Our theoretical analysis suggests the existence of an intrinsic contact angle between the membranes and the aqueous phases, reminiscent to Young's angle in standard wetting phenomena. An explicit relation is derived by which the intrinsic angle can be determined from experimental observables. The theory is then used to describe (i) the budding transition, where one of the aqueous phases protrudes from the vesicle body to minimize the interfacial energy between the aqueous phases; (ii) the formation of membrane nanotubes in these systems; and (iii) the competition between these two modes of spending excess membrane area (i.e. budding transition vs. tubular membrane formation).

Observing and understanding membrane interactions

Kheya Sengupta
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We are developing observation techniques to study membranes close to a surface with very high space and time resolution. The experimental results obtained using these techniques, on membrane fluctuations, and dynamics of membrane adhesion driven by generic and specific interactions, will be presented. In addition to exploring how to measure and manipulate the surface interaction potential, we have identified new players, like jamming, as important in driving or restricting adhesion. I will interpret these results in terms of a unified conceptual framework for understanding membrane adhesion under different conditions.

Adhesion of membranes in confined environments

Thomas LE GOFF
Institut Lumière Matière, Université de Lyon

We have studied the dynamics of membranes confined between two walls. This simple model is used in order to grasp the relevance of membrane bending rigidity in cellular adhesion, or in biolubrication.

We obtain effective nonlinear evolution equations for the membrane dynamics in the lubrication limit. The resulting dynamics is very rich and complex.

In the cases relevant to cell adhesion, the dynamics is arrested after a finite time, leading to a frozen configuration. These results suggest a novel scenario for the appearance of finite-size patches during cell adhesion. In addition, we obtain an order-disorder transition when increasing the permeability of the surrounding media. Finally, we show that fluctuations (thermal or resulting from biological activity) restore a coarsening behavior which is similar to that observed in usual phase separation.

Short range interactions between phospholipid bilayers: disecting the forces

M. Grunze I. Fedyanin, A. Pertsin
Applied Physical Chemistry, University of Heidelberg, ,Germany

Short range interactions between soft interfaces are important for the understanding of biolubrication and biofriction. Here the grand canonical Monte Carlo technique is used to reveal the origin of the repulsive pressure operating between supported DLPC bilayers at short separations. By partitioning the interbilayer pressure into physically distinct components, it is shown that the short-range repulsion comes mainly from the direct electrostatic lipid-lipid interaction of the head groups in the opposing leaflets. By contrast, the electrostatic lipid-lipid interaction between DLPE bilayers is strongly attractive, and the short-range repulsion is associated with the hydration (water-lipid) interactions. These findings explain e.g. why DLPC bilayers have a much larger interbilayer (fluid) separation at a given pressure, as compared to that for DLPE.

Bottom-up approach of multiscale dynamics in porous media. Application to Gas Shale

Alexandru BOTAN
Department of Civil and Environmental Engineering, Massachusetts Institute of Technology (MIT), Cambridge MA, USA.

In the context of high fuel prices the reserves of natural gas stored in shales are too large to be ignored. A major difficulty for a proper description of fluid transport in such systems stems from its multiscale texture characterized by a wide pore size distribution. Mass transfer in micropores and mesopores is very different from that in macropores and models for conventional reservoirs, intended to describing transport in macropores, are unsuitable for shale. As a result, any attempt in understanding overall transport requires a multiscale modeling approach. In this work, we built a hierarchical model of kerogen (the carbon-based matrix containing gas or oil in shale reservoir) which includes microporous, mesoporous and macroporous domains as shown in Fig.1. A transmission electronic microscopy (or any imaging technique) image of kerogen obtained from experiments is divided into a grid of equal-size tiles, each of them represents subnanoporous, nanoporous, or macroporous domain.

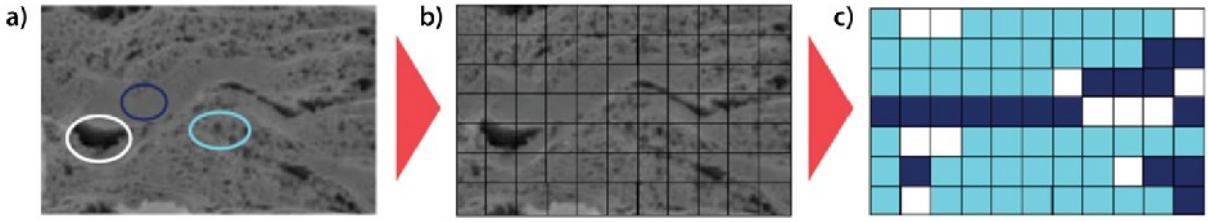

Figure : Hierarchical model of kerogen. A TEM image of kerogen (a) is divided into a grid of equal-size tiles (b), each of them represents subnanoporous (cyan colour), nanoporous (dark blue) or macroporous (white) domain.

Then fluid flow from domain i to domain j (therefore permeability of the whole sample) is described using a novel lattice model originated from the lattice gas automata method.
The foundation of this lattice model will be atom-scale molecular simulations of mass transport processes. The flow behavior will be averaged to allow upscaling of length and time, and the eventual combination with continuum fluid mechanics solvers. An overview of the proposed model and comparison with Lattice Boltzmann Dynamics will be provided.

Linear alkanes confined between smooth solid surfaces

Daniele Savio & Nicolas Fillot

LaMCoS, INSA de Lyon – CNRS, Villeurbanne, France

We focus here on the behavior of confined n-alkane molecules between solid walls of different kind (mostly metals and oxides). Molecular Dynamic simulations are lead to investigate the structure and rheology of the confined fluid, and the major point of interest is the slip at the wall, depending on both solid and fluid materials and the confinement itself.

The first part of the talk concerns the effects of chain length and surface (nature, orientation) on slip, as well as the role of the wall-fluid interactions. Details can be found in [1].

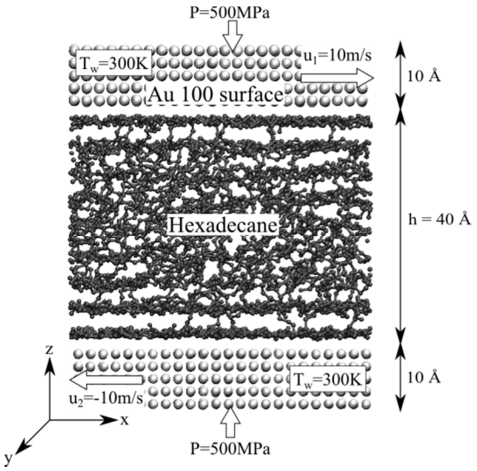

Figure 1 Overview of the molecular dynamics system and the applied operating conditions

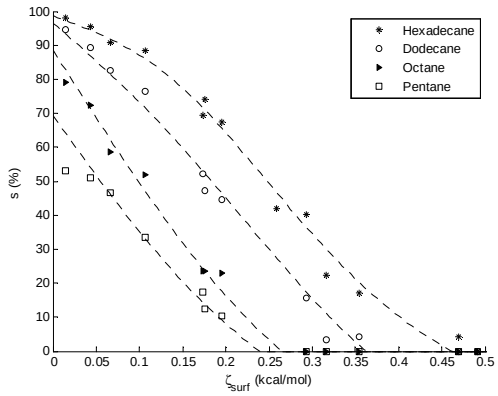

Figure 2 Wall slip values for different linear alkanes as a function of the surface interaction parameter. The points represent results from MD simulations, the dotted lines the predicted values.

A wall slip model based on the competition between these two factors is introduced. A surface parameter accounts for the wall-fluid interaction and commensurability, and is valid for both canonical and complex crystal lattices: this quantity is then linked to the shear stress transferred to the fluid molecules. Finally, a semi-analytical law for wall slip prediction including both the fluid viscosity and the surface characterization parameter is proposed.

The second part of the talk is a highlight on what is happening when the surfaces get closer and nearly touch each other, with a critical amount of fluid in between. Details can be found in [2].

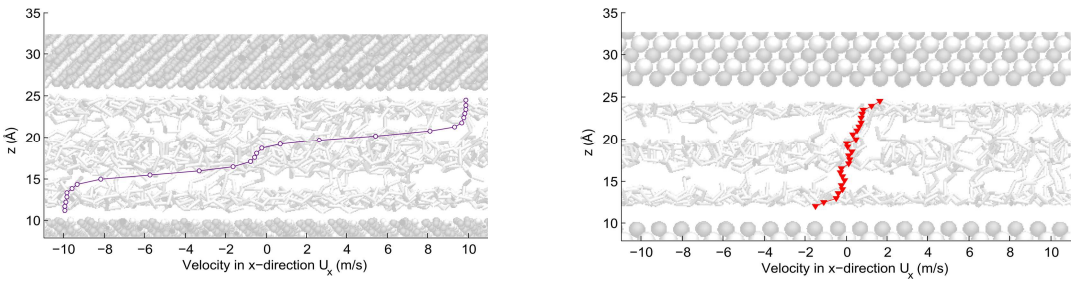

Figure 3 Velocity profiles across the film thickness for different combinations of surface wettability.

[1] Savio et al. Tribol Lett (2012) 46:11–22
[2] Savio et al. Tribol Lett (2013) 50:207–220

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