Experimental report

Proposal: 9-13-758		Council: 4/2018								
Title:	Validation and characterization of a model protomembrane architecture									
Research area: Soft condensed matter This proposal is a new proposal										
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Samples:	C10H20O2									
	С10Н21ОН									
	С30Н62									
Instrument			Requested days	Allocated days	From	То				
IN16B			1	0						
IN5			2	0						
D16			10	2	13/09/2018	20/09/2018				

Abstract:

The aim of this study is to validate a new model membrane structure for a protocell (prebiotic living form) and to perform a first physicochemical characterization of such architecture.

This model consists in a bilayer of mid-chain fatty acid molecules along with alcohols of equal chain length. Between the two layers, perpendicular to the carbon tails, is inserted a hydrocarbon whose size and type effects have to be investigated.

The intercalating agent inside the bilayer is expected to modify its physical characteristics, e.g. the membrane rigidity, permeability and stability. If proven, these effects will have natural impacts on the possible strategies put in place by the first living systems to maintain and protect the biological functions of its boundaries.

Neutron scattering techniques can give unique insights for our investigation, providing information about the molecular arrangement of the hydrocarbon molecules (diffraction with deuterium labelling) and their effects on molecule dynamics at the useful timescales (QENS).

The validation and study of our model can help today's science to understand how the first forms of life resisted to the harsh early Earth's conditions.

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Background

Life origin on our planet is a study matter of many different scientific fields with countless implications. A deep understanding about the physicochemical properties of soft matter is needed: For instance, the behavior of the self assembled structures from amphiphilic molecules can help modeling the first proto-cell membranes.

Fig. 1: Protomembrane model.

A commonly accepted scenario for life suggests it originated within high-temperature, high pressure deep-sea hydrothermal vents. A pivotal issue

is thus to understand how the first proto-cellular form resisted the extreme environment, with wide temperature (2 - 350 °C), pressure (\geq 800 bar) and pH gradients. These harsh conditions impose many constraints: for instance, at T \approx 100 °C, a temperature typically observed near the deep-sea hydrothermal vents [1], a liposome made of a phospholipid bilayer, as in model cellular membranes, would be extremely permeable and fluid, hence incapable to keep useful molecules inside or prevent unwanted ones to enter. Another issue is that phospholipids were probably not abundant on the early Earth at the onset of life. Thus, the first vesicles were most likely composed of simpler single chain fatty acids [2]. Fatty acid vesicles are known to be more sensitive to their surrounding environment [3], making their effectiveness as compartmentalization molecules questionable.

The present work aims at studying a new protomembrane architecture where the bilayer is composed by single chain amphiphiles, with an apolar lipid inserted in its midplane (Figure 1). If proven, this will represent a possible strategy of survival to the extreme early-Earth conditions, making it a strong candidate to model the protomembranes at the onset of life.

Experiment

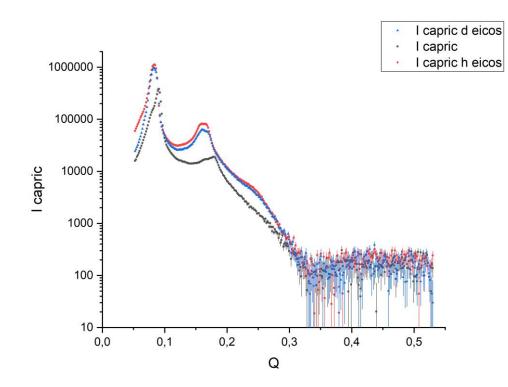


Fig. 2: Scattering intensity curves, integrated over the Omega range $-1^{\circ} - 12^{\circ}$, for the three samples investigated at 70% D₂O/H₂O contrast, 90% RH and T = 40 °C (pure capric acid, in presence of fully protonated and fully deuterated eicosane, heicosane. and d-eicosane respectively).

The experiments were performed using 3 different samples, all prepared as thin films on silicon wafers through vesicle deposition:

- Capric acid + bicine (1:1)
- Capric acid + bicine (1:1) + 5% h-eicosane
- Capric acid + bicine (1:1) + 5% d-eicosane.

For the measurements at different D_2O/H_2O contrasts, each sample was equilibrated for ≈ 1 day inside a BerILL humidity chamber at T = 40 °C and 90% Relative Humidity (RH). The contrasts D_2O/H_2O investigated were 8%, 30%, 50%, 70% and 100% respectively. Some examples of curves, after integration over Omega (sample angle relative to incoming beam) are shown in Figure 2.

To investigate the response of the samples to humidity, another kind of measurements was also performed to follow the swelling kinetics with time: this was done by collecting subsequent Omega scans in the range 1-5 $^{\circ}$ (hence including the Bragg angles corresponding to the first 2 orders). The results for 2 samples are shown in Figure 3 for comparison. A preliminary, qualitative analysis of the two swelling kinetics clearly shows some effects due to the apolar intercalation, which leads to a more ordered (the 2nd reflections better resolved) membrane arrangement. A more detailed and quantitative data analysis is currently ongoing.

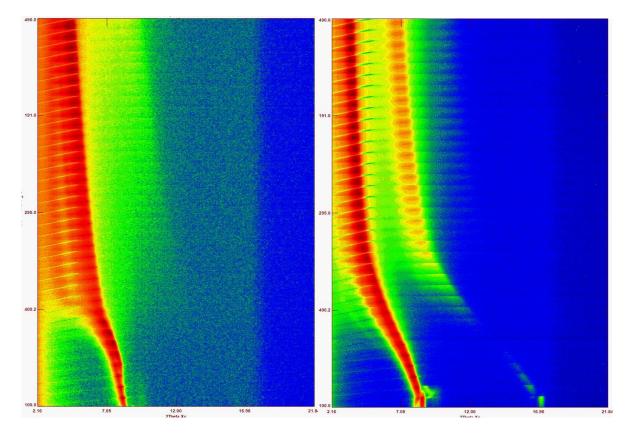


Fig. 3: Swelling kinetics at T = 40 °C, 90 % RH, 70% D₂O/H₂O contrast, for the pure Capric acid sample (left) and the one with the addiction of h-eicosane (right). The x axis is the 2Theta scattering angle (in degrees), while the y axis is proportional to the time.

References

- [1] Dodd et al. Nature (2017) 543, 60-64.
- [2] Monnard et al. Met. Enzym. (2003) 372:133-51.
- [3] Morigaki; et al. Curr. Opin. Coll. Inter. Sci., (2007) 75-80.