SUPPLEMENTARY INFORMATION

Langmuir films at the oil/water interface revisited

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Diffusion at the O/W interface

From the Einstein relation, $D = k_{\rm B}T/\lambda$. For the case of a 2D-diffusion between two liquid phases it can be defined a $D_{O/W,PL}$ which has been related to the viscosities of both bulk phases $\eta_{\rm water}$ (1 cP) and $\eta_{\rm oil}$ (1.9 cP for VAS)¹ through a power law defined by Eq.3²:

$$D_{O/W,PL} \sim (\eta_{\text{water}} + \eta_{\text{oil}})^{-0.85}$$
[3]

Also, a modified hydrodynamic drag term $\lambda = 8(\eta_1 + \eta_2)R$ for the Einstein relation has previously been experimentally verified to be an accurate model for lipid objects with radius *R* in a lipid monolayer between two viscous media with viscosities η_1 and η_2^3 .

The complexity of the phenomenon of lipid diffusion at the liquid-liquid interface resides in the fact that $\Delta[PL]_O$ and R change continuously upon compression as well as upon adsorption, depending on the experiment.

in cP D ~1.7258.10⁻¹¹.T/[$(\eta 1 + \eta 2)R$] m²/s or D ~0.17258.T/[$(\eta 1 + \eta 2)R$] μ m²/s

For $\eta_{\text{water}} = 1 \text{ cP}$ and $\eta_{\text{VAS}} = 1.9 \text{ cP}$ (r.f.¹⁶), T=294K and R=3.99, it results that $D = 4.38.10^{-10} \text{ m}^2/\text{s}$ 438 μ m²/s. This value compares reasonably with data from ².

References

- 1. <u>http://www.engineersedge.com/fluid_flow/fluid_date.thm</u>.
- Negishi, M.; Seto, H.; Hase, M.; Yoshikawa, K., How Does the Mobility of Phospholipid Molecules at a Water/Oil Interface Reflect the Viscosity of the Surrounding Oil? *Langmuir* 2008, 24, 8431-8434.
- 3. Walder, R. B.; Honciuc, A.; Schwartz, D. K., Phospholipid Diffusion at the Oil–Water Interface. *Journal of Physical Chemistry B* **2010**, *114* (35), 11484–11488.



Figure S1. Surface pressure-mean molecular area compression isotherms of EPC at the A/W, A/O and O/W interfaces and EPC/VAS_{0.02} mixture at the A/W interface, normalized by the surface tension of the corresponding free interface (γ_0).



Figure S2. Confocal spectral epifluorescence microscopy of EPC/VAS_{0.02} at the A/W interface Nile red was used a fluorescent probe. VAS was present in a molar fraction 0.2. Samples emission spectra obtained upon excitation at 488 nm are shown. Spectra were built up from the analysis of three different lenses in successive pictures captured at different λ_{em} within the 500-700 nm range.