Abstract ID : **46**

Unidirectional lipid transfer between zwitterionic vesicles under non-equilibrium conditions

Content

Lipid transfer between vesicle systems by monomer diffusion is typically investigated using deterministic kinetic models and fluorescent label-based experiments that yield averaged dynamic information of changes in vesicle properties like size and concentration. Obtaining quantitative information about the distribution of sizes and concentrations at each time remains not straightforward and in many cases, inaccessible.
In this work we have resolved inter-vesicle lipid transfer for asymmetric systems via monomer diffusion under non-equilibrium conditions. Our approach consists of departing from a well-defined non-equilibrium state, where zwitterionic vesicles differing in hydrophobic chain length are incubated, and experimentally monitoring the temporal evolution of size and concentration. For this purpose, we have combined complementary techniques, namely dynamic light scattering and quartz crystal microbalance with dissipation. The size and concentration behavior agree well with the predictions of a kinetic deterministic model and motivate the study of monomer transfer in the framework of stochastic simulations. Furthermore, we have introduced the use of equilibrium Monte Carlo simulations to unravel the quantitative distribution of vesicles with a given concentration present at different times during the transfer process. Results show the temporal evolution of the system from a non-equilibrium state at short incubation times consisting of many coexisting lipid concentrations towards a steady state of a single concentration at long incubation times. The methodology used acts as a reference point to more complex systems, i.e., lipid monomer transfer between confined lipid systems and in the presence of external stimuli.

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Preferred oral

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