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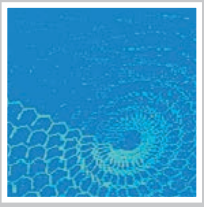

Polymer and Colloid Highlights

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Active Particles to Interrogate Bilayer Mechanics

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In this highlight, we discuss novel ways to deform and sense the mechanical properties of complex fluid–fluid interfaces such as lipid bilayers.

Self-assembly has played a central role in bridging colloid and polymer science with the biological sciences. At fluid–fluid interfaces, self-assembly of surface-active moieties is particularly interesting and relevant. The asymmetry in dielectric properties typically enhances the electrostatic and dispersion forces, and lateral capillary interactions arise due to local deformation of the interface. These structures can display disorder, such as protein aggregate networks at the air water interfaces which sometimes help stabilize beer foams or be highly ordered, such as in 2D colloidal crystals. The lipid bilayer which encloses cells is another fascinating example of such a structured interface. The same forces which drive self-assembly can also lead to mechanical or rheological properties of such interfaces, which oppose the deformations of such interfaces and one needs to go beyond interfacial tension to obtain a good description of the behavior of such systems. The development of experimental and theoretical methods to describe such systems has been an active area of research, see *e.g.* Jaensson and Vermant[1] for a recent review of progress in this area. But most progress has been made understanding how macromolecular or colloidal monolayers[2] or multilayers[3] respond. Deforming lipid bilayers in a controlled manner remains difficult.

Nevertheless, one of the fascinating aspects of biological cells is their ability to actively sense and respond to external stimuli, such as the extension of protrusions to explore their environment.[4] This implies that these bilayers are able to deform strongly, with possible non-linear responses to highly localized forces. Several intracellular of pathogenic bacteria (*e.g.* Listeria) also provide examples of how these can strongly deform cell membranes from inside, leading to the invasion of neighboring healthy mammalian cells.[5]

Minimal artificial soft matter model systems, such as giant unilamellar vesicles, have been successfully used as a minimal model system with which to mimic biological cells, but the realization of a minimal system with localized active internal forces that can strongly deform lipid membranes from within and lead to dramatic shape changes remained challenging. This was addressed by encapsulating self-propelling particles (SPP) in giant vesicles. The SPP were simple Janus particles with H_2O_2 as fuel, which was catalytically decomposed by a Pt layer on one side of the particles. These particles were embedded within vesicles made using dioleoylphosphatidylcholine (DOPC). Experimentally the volume fraction of particles as well as the tension in the bilayer membrane could be varied. The minimalistic system could be modelled quite well using Langevin dynamics simulations of active Brownian particles enclosed in thin membrane shells, described dynamically triangulated surfaces, in collaboration with FZ-Jülich.

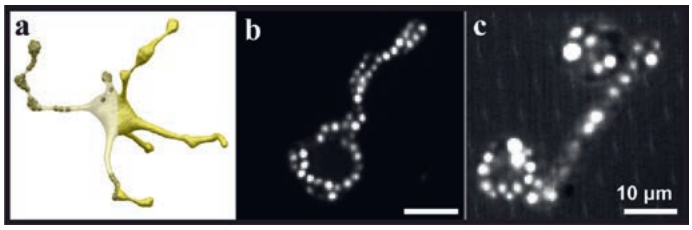

Fig. 1. a) Simulation snapshot shows a dendritic structure of a floppy vesicle to localized forces by SPPs; b,c) Experimental observations of more complex vesicle shapes of active particles at higher particle loadings (particle size in experiments is 1 micrometer).

This combined experimental and simulation study[6] reveals that the most pronounced shape changes are observed at low and moderate particle loadings, with the formation of tether-like protrusions and highly branched, dendritic structures (Fig. 1), with an interesting membrane curvature induced accumulation and feedback mechanism. At high volume fractions only globally deformed vesicle shapes are observed (Fig 1b,c). Moreover, this study demonstrates how the interplay between local active forces, membrane elasticity and viscosity can give rise to a plethora of novel vesicle shapes, which do not exist in equilibrium systems. Analysis of the fluctuation spectra showed a strong departure from the Helfrich curvature-elasticity model when active particles are present, emphasizing the importance of active non-equilibrium processes. In the future, this method could provide a framework to study non-equilibrium thermodynamic and rheological responses of bilayer membranes when the membrane composition becomes more complex.

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[1] N. Jaensson, J. Vermant, *Curr. Op. Coll. Int. Sci.* **2018**, *37*, 136.
[2] L. F Scherz, B. Schroyen, M. Pepicelli, D. A. Schlüter, J. Vermant, D. Vlassopoulos, *ACS Nano* **2019**, *13*, 14217.
[3] M. Pepicelli, *J. Rheology* **2019**, *63*, 815.
[4] D. A. Fletcher, R. D. Mullins, *Nature* **2010,** *463*, 485.
[5] 'The Giant Vesicle Book', Eds. R. Dimova, C. M. Marques, CRC Press, **2019**.
[6] H. R.Vutukuri, M. Hoore, C. Abaurrea-Velasco, L. van Buren, A. Dutto, T. Auth, D. A. Fedosov, G. Gompper, J. Vermant, *Nature* **2020**, *586*, 52.

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