

# ECCo Live 2020

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EARLY CAREER COLLOID  
NETWORK WEBINAR

10<sup>th</sup> September 2020  
13:00-16:00

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# ECCo Live 2020 Programme

Thursday 10<sup>th</sup> September 2020

Time	Event
13.00-13:05	Introduction to ECCo Live
13.05-13:55	<b>Plenary lecture:</b> Cecile Dreiss, Kings College London <i>The colloid science of bile salts and lipid digestion - and other colloid stories</i>
	<b>Break</b>
14.05-14:20	James E. Hallett, University of Oxford <i>Neutron Scattering From (not so) Model Hard Spheres</i>
14.20-14:35	Thomas M. McCoy, University of Cambridge <i>Spontaneous self-assembly of thermoresponsive vesicles using a zwitterionic and an anionic surfactant</i>
14.35-14:50	Katherine Macmillan, Heinrich Heine University <i>Materials from Colloidal Particles using Optical Fields</i>
	<b>Break</b>
15.00-15:15	Guido Bolognesi, Loughborough University <i>Reversible trapping of colloids in microgrooved channels by diffusiophoresis under steady-state solute gradients</i>
15.15-15:30	Timothy J. Murdoch, University of Pennsylvania <i>Sticky but Slick: Reducing Friction using Associative and non-Associative Polymer Lubricant Additives</i>
15:30-15:45	James W. Hindley, Imperial College London <i>Engineering Synthetic Cells That Can Sense and Respond to Their Environment</i>
15.45-16.00	Open Forum
16:00	<b>Close</b>

# The colloid science of bile salts and lipid digestion - and other colloid stories

*Cécile A. Dreiss*

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My research at King's over the last 15 years has explored the structure and properties of soft matter, with a focus on neutron scattering techniques and rheology, in particular: gels [1,2], polymeric micelles [3] and wormlike micelles [2], and supramolecular structures formed by host-guest complexation with cyclodextrins [4]. In this talk I will share my recent ventures in colloid food science and my interest in the unusual biosurfactants that control most of lipid digestion: bile salts.

Bile salts (BS) are biosurfactants produced in the liver and released into the small intestine, which play key roles in lipid digestion: they facilitate enzyme adsorption to fat droplet interfaces (lipase/co-lipase) but also remove insoluble lipolysis products from this interface, carrying them into mixed micelles to the gut mucosa for absorption - all colloid and interface science! Understanding these processes – how bile salts interact with oil/water interfaces, form mixed micelles with products of lipolysis (such as fatty acids and monoglycerides), how they interact with other food compounds (such as the dietary fibres – i.e. polysaccharides - that lower blood cholesterol levels) is paramount to address the current obesity crisis and associated cardiometabolic diseases, and help guide the design of functional foods that can tailor the lipid digestion process and the accessibility of nutrients and bioactive compounds, as well as control the levels of post-prandial glycaemia and lipaemia.

I will discuss how we have characterised bile salts adsorption at the air/water and lipid/water interface by interfacial techniques, including neutron reflectometry [5]; their interactions with oil droplets in emulsions stabilised by a methylcellulose (a dietary fibre and food emulsifier) [6] and finally the morphology of BS micelles and mixed micelles by small-angle neutron scattering and MD simulations, and their interactions with liposomes, as a mimic of the lipid nanostructures found in the GI tract.

[1] Dreiss (2020) *Hydrogel design strategies for drug delivery* [\*\*Curr. Opinion Colloid Interface Sci.\*\* 48, 1-17](#)

[2] Zoratto *et al.* (2019) *Supramolecular gels of cholesterol-modified gellan gum with disc-like and wormlike micelles*, [\*\*J. Colloid Interface Sci.\*\*, 556, 301-312](#)

[3] Puig-Rigall *et al.* (2019) *Pseudo-polyrotaxanes of cyclodextrins with direct and reverse X-shaped block-copolymers: a kinetic and structural study*, [\*\*Macromolecules\*\*, 52, 1458-1468](#)

[4] Serres-Gómez *et al.* (2018) *Supramolecular hybrid structures and gels from host-guest interactions between  $\beta$ -cyclodextrin and PEGylated organosilica nanoparticles*, [\*\*Langmuir\*\*, 34, 10591-10602](#)

[5] Pabois *et al.* (2019) *Molecular insights into the behaviour of bile salts at interfaces: a key to their role in lipid digestion* [\*\*J. Colloid Interface Sci.\*\*, 556, 266-277](#).

[6] Pabois *et al.*, (2020) *Interactions of bile salts with a dietary fibre, methylcellulose and impact on lipolysis*. [\*\*Carbohydrate Polymers\*\*, 115741](#)

## Neutron Scattering From (not so) Model Hard Spheres

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Keywords: Neutron Scattering, Hard Spheres, Nanoparticles

PMMA particles are long established as the model particle system of choice to study hard-sphere behaviour in nonpolar solvents. Although a well-established synthetic route to produce highly stable latexes with surface layers of poly(12-hydroxystearic acid) comb stabilizer (PHSA) exists, this polymer is not commercially available and can prove challenging to produce. As such, while their utility in fundamental studies of colloidal phase behaviour is beyond doubt, variations in the quality of the stabiliser layer can result in some inconsistencies between the behaviour of PHSA-PMMA produced by different research groups. A promising alternative is to use polydimethylsiloxane-based stabilisers. Crucially, well-defined methacryloxypropyl-terminated PDMS stabilisers are commercially available, dramatically improving consistency between different reported PMMA syntheses and lowering the barrier of entry for new researchers to perform fundamental investigations with PMMA. Despite this promise, uptake of this alternative route to stabilisation in nonpolar solvents has been relatively slow, with many investigations still favouring the PHSA route.

We have studied the structure of PDMS-stabilised PMMA using small-angle neutron scattering, to better inform future investigations making use of this stabiliser. We achieved a reduction in the size of particles, to make ones suitable for neutron scattering, and we enhanced their stability using a cross-linking agent. By using deuterated methyl methacrylate monomer, we were able to highlight the distribution of the stabiliser and cross-linking agent. We found that the interfacial stabiliser layer is complex, with PDMS shells that are thicker than those typically used to stabilise PMMA latexes and, for the cross-linked particles, with PDMS stabiliser incorporated into the PMMA core [1].

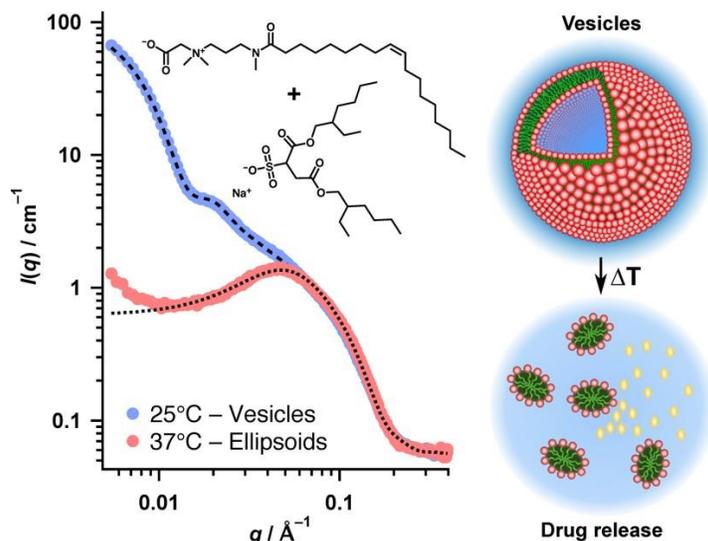
[1] Hallett, J. E., Grillo, I., & Smith, G. N. (2020). A neutron scattering study of the structure of poly (dimethylsiloxane)-stabilized poly (methyl methacrylate)(PDMS-PMMA) latexes in dodecane. *Langmuir* **36**(8), 2071-2081.

# Spontaneous self-assembly of thermoresponsive vesicles using a zwitterionic and an anionic surfactant

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Keywords: Surfactants, self-assembly, small-angle scattering.

The work I hope to present at the ECCo Live 2020 online webinar rigorously demonstrates factors driving vesicle self-assembly in an aqueous, dual surfactant system comprising Aerosol-AT (AOT) and oleyl amidopropyl betaine (OAPB); see Fig. 1 for structures. Small-angle neutron and X-ray scattering are used to gain structural information whilst the effects of surfactant ratio, salt concentration and temperature are examined to probe physical phenomena and mechanisms for disassembly. Surfactant ratio and salt concentration were found to dictate micellar geometry by altering packing constraints and intermolecular interactions. Heating vesicle forming solutions to physiological temperature (37°C) can cause them to collapse into smaller ellipsoidal micelles (2-3~nm), with higher salt concentrations (>10 mM) inhibiting this transition. These aggregates could serve as responsive carriers for loading or unloading of aqueous cargoes such as drugs and pharmaceuticals, with temperature changes serving as a simple release/uptake mechanism.



**Fig 1:** Small-angle neutron scattering data from a 10 and 20 mM aqueous mixture of AOT and OAPB (structures inset) and the accompanying structural transition from vesicles to ellipsoids upon heating from 25 to 37°C respectively.

[REF]: Thomas M. McCoy, Joshua B. Marlow, Alexander J. Armstrong, Andrew J. Clulow, Christopher J. Garvey, Madhura Manohar, Tamim A. Darwish, Ben J. Boyd, Alexander F. Routh and Rico F. Tabor, Spontaneous self-assembly of vesicles using a zwitterionic and anionic surfactant as thermo-responsive carriers. *Biomacromolecules*, 2020.

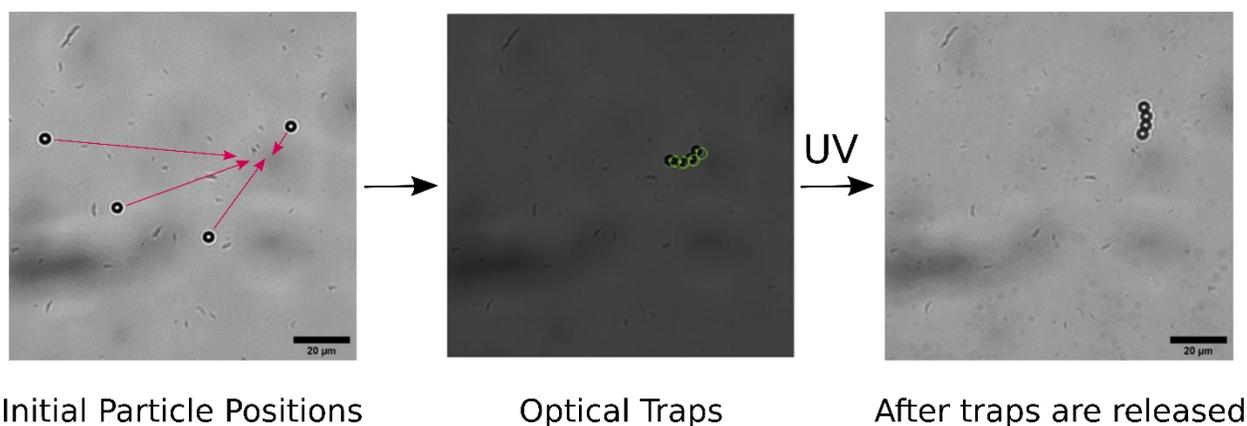
# Materials from Colloidal Particles using Optical Fields

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Keywords: optical fields, self-assembly, UV click

Optical tweezers exploit the interaction of colloidal particles with highly-focussed light to manipulate and manoeuvre these particles [1]. In addition to single optical traps, multiple traps or extended potential energy landscapes (optical fields) have been applied using periodic interference patterns, speckle patterns created using ground glass and freely configurable patterns created using spatial light modulators [1, 2]. Despite this, the capacity of these optical potential energy landscapes to trap multiple colloidal particles into a designed structure has yet to be fully explored. In order to pursue this goal, here we investigate a thiol-yne UV click reaction to attach colloidal particles together. The particles are arranged into defined clusters using a spatial light modulator then UV light is applied to attach them together (see Figure 1). We have successfully produced a variety of particle clusters using this method and we have investigated the reaction parameters in order to improve the efficiency of the particle sticking. In the future, we aim to use optically-created potential energy landscapes to imprint a structure on a dispersion of colloidal particles that, once organised, can be fixed together by using UV light.



**Figure 1** A series of images showing the successful attachment of four colloidal particles.

- [1] R. F. Capellmann, J. Beverunge, F. Platten and S. U. Egelhaaf, *Rev. Sci. Instrum.*, **2017**, 88, 056102  
[2] F. Evers, R.D.L. Hanes, C. Zunke, R.F. Capellmann, J. Beverunge, C. Dalle-Ferrier, M.C. Jenkins1, I. Ladadwa, A. Heuer, R. Castañeda-Priego and S.U. Egelhaaf, *European Physical Journal Special Topics*, **2012**, 222, 2995–3009

# Reversible trapping of colloids in microgrooved channels by diffusiophoresis under steady-state solute gradients

*Guido Bolognesi,<sup>1</sup> Naval Singh,<sup>1</sup> Goran Vladislavljević,<sup>1</sup> François Nadal,<sup>2</sup> Cecile Cottin-Bizonne<sup>3</sup> and Christophe Pirat<sup>3</sup>*

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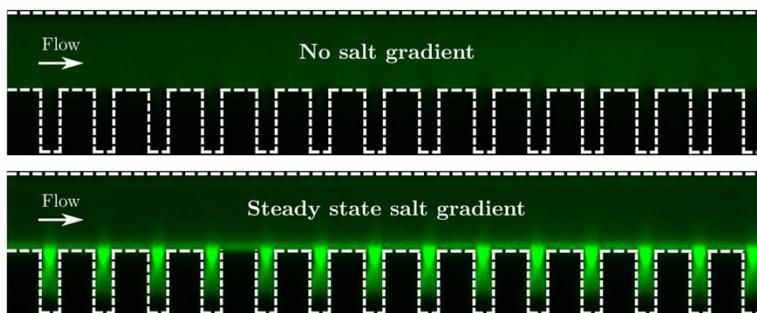
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Keywords: Microfluidics, Diffusiophoresis, Diffusioosmosis, Solute Gradient

The ability to manipulate particles by diffusiophoresis – a phoretic phenomenon leading to particle motion along a solute concentration gradient without the use of an external field – has gained an increasing attention in recent years [1]. For instance, transient salt concentration gradients have been successfully used to achieve enhanced particle transport into dead-end structures by DP and diffusioosmosis (DO) effects [2], whereas steady-state solute gradients have been used for the trapping and characterisation of liposomes and exosomes in open microchannels [3].

In this study [4], we report a novel mechanism for reversible trapping of particles in dead-end microstructures, namely microgrooves, via steady-state solute gradients in a continuous flow setting. As shown in the figure, the charged fluorescent colloids are accumulated within the micro-grooves by pumping parallel electrolyte streams within a microchannel fitted with a microgrooved wall.

The trapping mechanism was investigated via confocal microscopy and finite element simulations. The phenomenon is fully reversible and particles can be cyclically trapped into and released from the grooves by controlling the salt concentration of the parallel streams via a flow switching valve. The proposed method offers great potential for microfluidic bio-analytical testing applications, including bio-particle pre-concentration and signal amplification.



## References

- [1] D. Velegol et al., *Soft Matter*, 2016, 12, 21; [2] S. Shin et al., *PNAS*, 2016, 113, 2; [3] M. K. Rasmussen et al. *Nat. Commun.*, 2020, 11, 2337; [4] N. Singh et al., 2020, arXiv preprint arXiv:2007.11114.

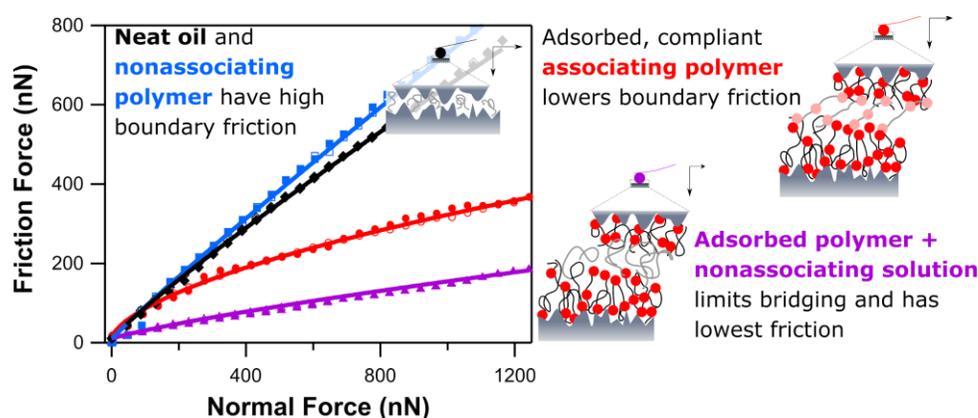
# Sticky but Slick: Reducing Friction using Associative and non-Associative Polymer Lubricant Additives

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Keywords: atomic force microscopy, nanotribology, extensional rheology, lubrication

Low viscosity lubricants require an increased loading of additives to compensate for an increased prevalence of solid-solid contact. One strategy to sustain the current trend towards lower viscosity lubricants is to develop multi-functional additives. Oil-soluble olefin copolymers (OCP) additives are commonly used as viscosity modifiers to improve the temperature dependent rheology of engine oils. However, functionalizing OCP with a random distribution of a small amount (<5 mol%) of polar-aromatic groups along the polymer provides additional potential for enhanced boundary lubrication. Understanding the boundary lubrication mechanism of FOCP requires a multi-scale approach.<sup>1</sup> Extensional relaxation time measurements of bulk polymer solutions show that the functionalized olefin copolymer (FOCP) can form transient, associative bonds between polymer chains, while quartz crystal microbalance with dissipation monitoring (QCM-D) demonstrates that the amphiphilic nature of FOCP drives adsorption at the solid-liquid interface. Colloid probe atomic force microscope (AFM) measurements show that the adsorbed layer forms a steric barrier that reduces friction in the boundary regime (Figure 1). AFM also shows that FOCP in a bulk oil interacts with the adsorbed polymer, resulting in an increased interfacial viscosity, important for promoting hydrodynamic lubrication, and the formation of adhesive bridges. Unexpectedly, boundary lubrication is further enhanced by immersing adsorbed FOCP in solutions lacking additives that form associations. Mechanistic insight from this study can inform the development of high-performance, multi-functional lubricant additives.



1 - T.J. Murdoch et al, *ACS Appl. Polym Mater.*, 2020 DOI: 10.1021/acsapm.0c00687

# Engineering Synthetic Cells That Can Sense and Respond to Their Environment

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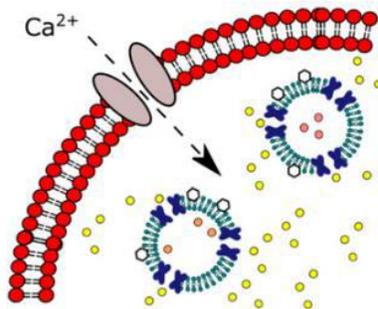
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**Keywords:** Self-assembly, synthetic cell, synthetic biology, stimuli-responsive, lipid vesicle

The development of soft matter devices that are responsive to specific stimuli is essential for engineering the next generation of drug delivery systems, as well as the construction of synthetic cells and tissues. This research focuses on the development of three responsive lipid vesicle designs, utilising mechanosensitive channel reconstitution, photopolymerization and lipid domain patterning to create nano- and micro-scale synthetic cells responsive to ionic/phospholipase<sup>[1]</sup>, light<sup>[2]</sup> and temperature<sup>[3]</sup> stimuli respectively.

Combining functional vesicle modules across different length scales enables the creation of synthetic cells capable of responding to a variety of local and applied stimuli. Such systems can be engineered to function in sense-and-response applications including drug delivery, diagnostics and bioremediation.



*Figure 1. Activation of a synthetic signalling pathway in a multicompartiment artificial cell*

## References

- [1] J. W. Hindley, D. G. Zheleva, Y. Elani, K. Charalambous, L. M. C. Barter, P. J. Booth, C. L. Bevan, R. V Law, O. Ces, *Proc. Natl. Acad. Sci. U. S. A.* **2019**, *116*, 16711–16716.
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