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Phase Behaviour in Charged Colloidal Systems

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The subject of this research proposal is the properties of concentrated colloidal suspensions. Colloidal suspensions are ideal model systems for understanding crystallisation and the glass transition. The colloidal suspensions of interest are mixtures of a dispersant (water or organic solvent) and spherical micro particles made of silica, PMMA or other polymers, with diameters in the range of hundreds of nanometres [1].

Colloidal systems serve as model systems for many problems in condensed matter physics. As the inter-particle distance is hundreds of nanometres, light scattering methods can be used rather than X-ray scattering [2]. Further, the motions are Brownian in nature rather than ballistic, so dynamics can be studied in real time. In addition the colloidal interaction is adjustable by changing the effective charge, which can be done by changing the ionic concentration in the solvent [3]. Due to these advantages, colloidal system studies can help test physical theories and provide insights into the properties of atomic and molecular materials.

Although systems with different interactions (eg hard sphere, charged systems etc) have been studied for many years, there has been little attempt to unify these to determine if there are any universal behaviours. This will be explored in this project, by comparing previous results on hard spheres with new measurements for systems with repulsive interactions of different strengths.

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Simulation of Brownian motion of a colloidal spherical particle near a flat surface

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The study of colloidal interactions mediated by steric repulsions, van der Waals forces, or structural and depletion forces form a major research area in understanding the behaviour of colloidal particles undergoing Brownian motion in a dispersion or near an interface. The energy scale of the forces affecting a Brownian particle are of the order of thermal energy (5-10 kT), but play a crucial role in determining colloidal stability. Several techniques have been developed to measure these colloidal forces where *total internal reflection microscopy* (TIRM) is one of the techniques that can quantify colloidal interactions by noninvasively probing the potential energy (PE) of a single Brownian particle levitated above a surface. This is achieved via tracking the instantaneous separation distance between a single Brownian colloidal particle and a flat surface and then constructing the PE function. Even though TIRM is highly sensitive to very weak interactions, its accuracy is highly dependent on various factors such as experiment duration, sampling rate and experimental noise.

Here, our aim is to simulate Brownian motion of a single colloidal spherical particle near a flat plate in a colloidal dispersion containing non-adsorbing polyelectrolyte molecules based on the earlier methodology developed by Sholl *et al.*¹ for particles in an electrical double layer (EDL) force field in a TIRM measurement. In this work, we extend the Brownian dynamics algorithm developed by Ermak and McCammon² and utilised by Sholl *et al.*¹, to the simulation of a particle in the presence of both van der Waals and depletion forces as well as EDL forces. In order to investigate the effect of various experimental parameters on the accuracy of TIRM experiment data, simulation conditions are kept similar to those of the experiments. This study will enable us to effectively design and optimize TIRM experiments resulting in the accurate measurement of colloidal forces

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Engineering Red Blood Cells and Polyethylene Glycol on Diagnostic Materials to Detect Fibrinogen

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Currently, there is a clinical demand for a rapid, sensitive, low-cost and easy-to-use diagnostic that can measure fibrinogen concentrations in the blood.

Through the use of red blood cells (RBCs), polyethylene-glycol 400 (PEG 400) and fibrinogen solutions, we have identified two PEG 400 concentration-dependant mechanisms that can be exploited for detection purposes. At low concentrations, we have identified that the hypertonic environment PEG 400 provided could enhance fibrinogen-induced RBC aggregation. Conversely, at high concentrations, PEG 400 could precipitate fibrinogen into a sticky substance, entrapping the RBCs in the process.

In this study, we evaluated these mechanisms towards producing positive (ie. Fibrinogenic) and negative (Ie. Afibrinogenic) results on several diagnostic materials. We compared the efficacy and fidelity between both types of results through differences in visual characteristics such as colour.

Blood Conditions	Blood Solution	Additive Solutions	Tube Conditions	Results		
				-ve	+ve	
Type: AB+ Age: 0 weeks	- 0.6% (v/v) RBCs - 10% (v/v) PEG 400 - Balance PBS	+ve: 10 g/L Fibrinogen in PBS -ve: PBS	- Added 50 uL Blood Solution - Added 50 uL Additive Solution - Incubated for >1 hour at Room Temperature. -Centrifuged for 1 min at 4000 rpm -Shook gently for 5 seconds	Tube Supernatant		
				Tube Bottom		
	Tube Supernatant					
	Tube Bottom					

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Ellipsoidal droplet production and arrested coalescence: shape transformation and evolution

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Ellipsoidal-shaped droplets can be created using microfluidic flows and tuning of droplet rheology. Study of non-spherical shapes provides further insight into formation of larger networks, for example in dairy product, but also enables synthesis of more complex colloids and materials. Arrested coalescence¹ has been shown to cause numerous complex shape transitions in emulsion systems, but previously only spherical droplets were studied². Here the dynamics of such shape transitions are studied using micromanipulation and droplets with varying aspect ratio and elasticity. A theoretical model of the final shape of pairs and triplets of ellipsoidal droplets is developed and used to compare with experimental results. We find good overall agreement but show that additional constraints affect the final packing.

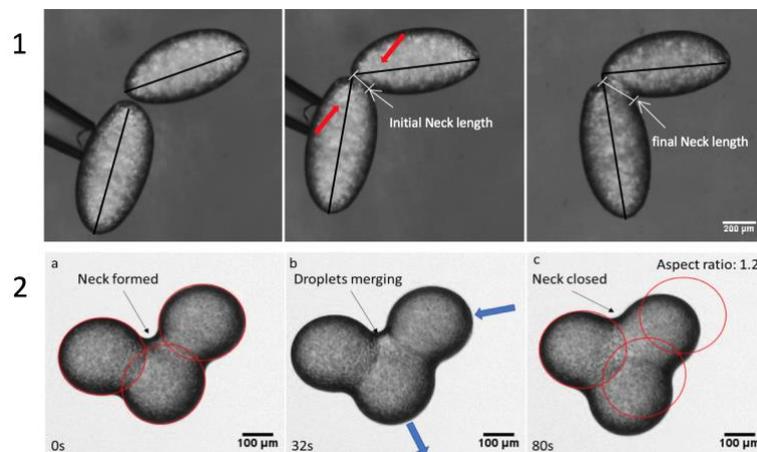


Figure 1. (1) ellipsoid doublets coalescing and restructuring into a more compact shape via rotation of one of the ellipsoids. (2) an ellipsoidal triplet restructuring as a result of fluid meniscus-induced deformation.

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Aligned droplet patterns by dewetting of polymer bilayers

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The dewetting of thin polymer films on solid substrates has been extensively studied in the past two decades.¹⁻³ Here we report a novel phenomenon whereby aligned polymer droplets spontaneously form by dewetting on a substrate, and the order extends over a large scale (millimetres). The patterns are formed by dewetting a poly (4-vinylpyridine) (P4VP) film on a polystyrene (PS) film, spin-coated from their ethanol and toluene solutions, respectively. The mechanism of ordered droplet formation was investigated and the final ordered pattern was found to result from a series of aligned holes in the P4VP film formed during spin-coating. These holes are in turn likely triggered by polymer concentration fluctuations induced by shear force during spin coating.⁴ The experimental conditions that contribute to the appearance and the order of the resulting aligned droplets were identified. The potential of these ordered patterns to be used in applications is being explored.

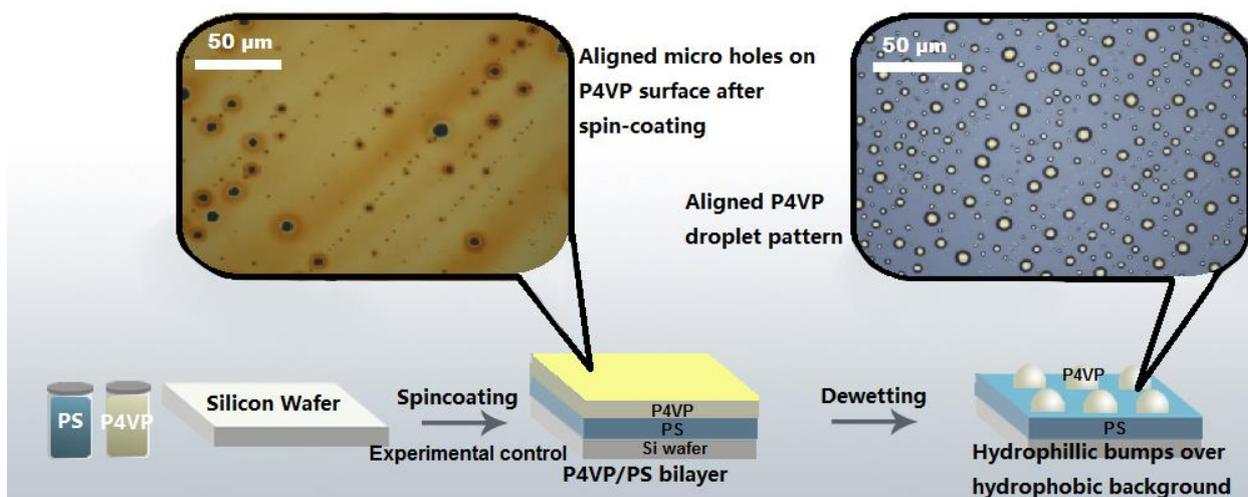


Figure 1 Schematic representation of the process of formation of spontaneously aligned droplets via dewetting of P4VP/PS bilayer. Insets: Optical micrographs of film surface before and after dewetting.

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Nanocellulose Gel for Biomedical Applications

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Red blood cells (RBC) present different antigens on their surfaces, and therefore are classified into a specific blood type¹. Amongst the known blood group (BG) systems, the ABO and Rh systems present a considerable relevance for preliminary blood typing. Currently, the conventional blood typing can be performed through a variety of techniques, such as the gel card method. This approach relies on the reaction between RBCs and antibodies and their elution through a gel column by centrifugation². Agglutinated RBCs constitute positive results and are trapped on the gel column, whereas negative results are formed by individual RBCs pellet on the bottom. The obtainment of a sustainable, renewable and low cost material is desired for a new generation of a high quality and sensitive blood typing test. We introduce TEMPO-oxidized nanocellulose gels as a suitable matrix with a wide potential for the gel card method for blood grouping. Indeed, the gel presented well-defined and visually clear results from agglutination and non-agglutination of RBCs. To increase RBC viability without affecting the gel colloidal stability, the evaluation of additives was necessary. Carbohydrates and synthetic polymers were able to balance the gel tonicity, however haemolysis rates did not decrease. In contrast, the macrobinder-containing gel regulated the osmotic pressure to levels found in the human blood plasma (Figure 1). Indeed, this molecule showed the ability to increase cell viability without affecting gel properties. In conclusion, this study highlights the immense potential of nanocellulose gel for biomedical applications.

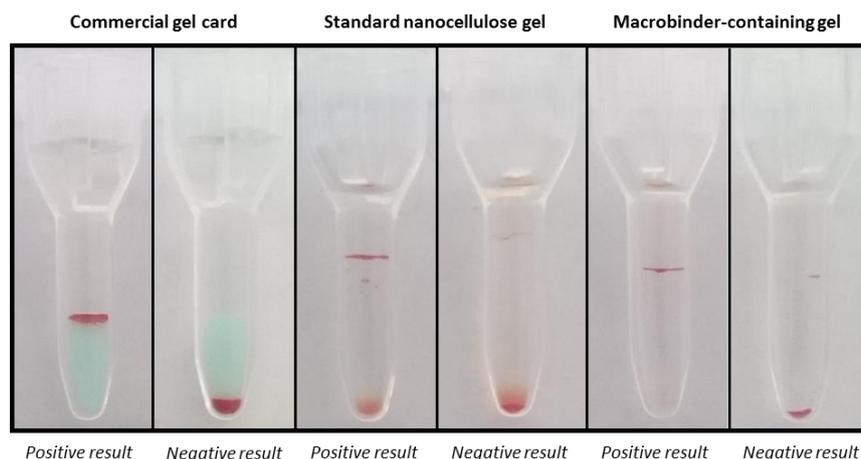


Figure 1. Positive and negative RBC agglutination for standard and macrobinder-containing nanocellulose gel.

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A Scalable Method to make Anisotropic Nanoparticles by Nanolithography

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Particles of nanoscale size are immensely fascinating to the researchers for their exceptional properties. Often the anisotropy in the structures (shape or composition) of these nanoparticles add some phenomenal degree to their various applications in real life. Among the range of applications, one of the less explored application of the anisotropic nanoparticles are in the films formation. This leads one to probe the interaction of the anisotropic particles with surfaces in presence of various DLVO and non-DLVO forces.

The aim of this work is to establish a scalable method for making metallic anisotropic nanoparticles, by nano-lithographic method and to understand the interaction of these particles with the surface. The process includes formation of a template allowing the metal deposition of the shape of interest. The template can be made up of any shape which is possible by combination of thermal scanning probe lithography¹, namely NanoFrazor and Reactive Ion Etching (RIE). Circular holes written on the polymer by this instrument enable the formation of rod-like structures through quite a few intermediate steps. The template produced by this method is free of drawback of the conventional Anodic Aluminium Oxide (AAO) template² which does have several disadvantages such as toxicity, time consuming procedure etc. The template reported in this article is capable of high yield, quite liberal about choosing the material of particles and also doesn't restrict the nanoparticles in any particular application. Anisotropic TIRM experiments allow us to understand the interaction forces of the particles (100 – 200 nm diameter and 500 nm length) with the surface.

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Charge reversal and aggregation of humic substances induced by hydrophobic monovalent counter-ions

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Abstract

Humic substances (HSs) are considered as natural colloidal particles or polyelectrolytes and represent a big portion of recalcitrant organic carbon in the terrestrial environment. HSs have more than 50% of elemental carbon with hydrophobic aliphatic and aromatic moieties as well as hydrophilic polar and charged groups. In soil, sediments and water bodies, HSs interact with inorganic and/or organic pollutants and control their fate depending on the charging and aggregation of HSs. Considering the importance of HSs in the natural environmental condition, we focused to investigate the charging and aggregation behavior of humic substances (HSs) in the presence of hydrophobic monovalent cations, namely, tetraphenylphosphonium TPP⁺. Three types of HSs standard Suwannee river fulvic acid (SRFA), Suwannee river humic acid (SRHA), and Leonardite humic acid (LHA) with different aromaticity were used in this investigation. The hydrophobic counter ion TPP⁺ induced the charge reversal of the HSs and also formed the large visible aggregates of all the HSs at lower pH. The iso-electric point (IEP) or charge reversal pH of LHA was higher than that of SRFA and SRHA in every concentrations of TPPCl (tetraphenylphosphonium chloride), demonstrating the strong hydrophobic interaction between HSs, especially LHA, and TPP⁺. The large remarkable aggregation was observed in the case of LHA with TPP⁺, indicating the existence of stronger hydrophobic attraction among LHAs and TPP⁺. Microscopic observation and the size determination by dynamic light scattering were carried out to confirm the aggregation. The HS aggregates showed fractal structure with values of fractal dimension D_f between 2-2.2 in unstirred conditions, revealing that the HS aggregates with TPP⁺ were formed via cluster-cluster aggregation with restructuring. The fractal dimension D_f increased to 2.8-2.9 in stirring conditions, and the aggregates deemed to be formed by breakage and the regrowth between smaller aggregates and larger aggregates.

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Shrinkage Behavior of Microbubble

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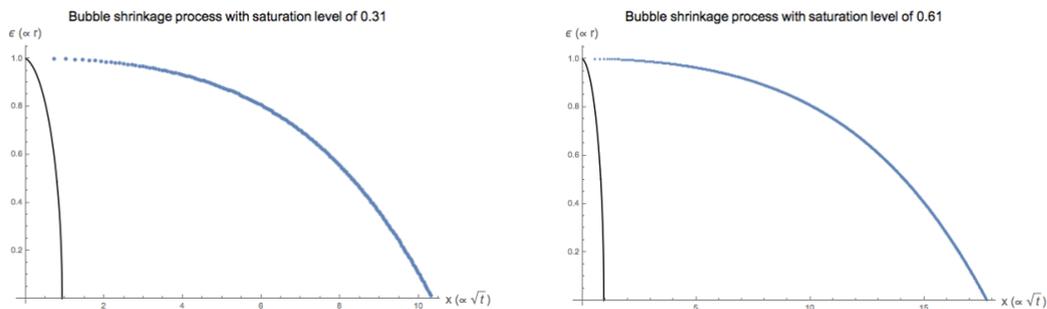
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In the 1950s, P.S. Epstein and M.S. Plesset mathematically described the growth and shrinkage process of bubbles in solution, which is driven by the Laplace pressure within the bubble and the saturation level of dissolved gas in the surrounding solution. The gas concentration immediately adjacent to the bubble is described by Henry's law where the pressure is the external pressure plus the Laplace pressure and the rate of growth or shrinkage is described by the diffusion equation. However, the initial conditions that the bubble is isolated, stationary and far from a wall are not easy to achieve experimentally. As we know, bubbles are effected by buoyancy in solution, making it difficult to test the theory.

My experiment overcomes the buoyancy of bubbles by using a micro-pipette probe, which affects the shrinkage rate by less than ten percent when the radius of the bubble is larger than 0.05mm. We find that a bubble takes about 100 times longer to dissolve completely into solution than expected from EP's theory, as shown in the figures below [1].

Our results indicate that there is something missing in their theory. I am investigating if the missing physics is an energy barrier to the transfer of a gas molecule from inside the bubble to the solution.



[1]: The black curves are theoretical curves from EP's theory and the blue ones are from my experiment results.

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Fully printed fullerene solar cells with high performance molecular nematic liquid crystalline

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Solar cells are one possible source of alternative energies. In addition to the conventional solar cells based on inorganic materials, cells made using organic oligomers or polymers – so called organic solar cells (OSCs) are emerging. They are attractive because (i) they can be deposited onto flexible plastic substrates and (ii) this deposition can happen out of solution, e.g. in a printing process, thus allowing for high throughput, high reproducibility and low cost.

At the University of Melbourne, the group of Dr David Jones has recently synthesized an oligomer, BQR (Fig. 1) that can be produced in large quantities and gives power conversion efficiencies of 10% in OSCs when blended with a fullerene acceptor such as PCBM.¹ This blend would therefore be ideal for a larger scale production of OSCs. At the research centre CSIRO, initial tests were carried out to make such OSCs by slot-die coating. It turned out that the resulting films were too brittle for flexible OSCs.

In this project, I will discuss whether it is possible to make less brittle films of BQR blended with fullerenes such as P3HT that still give high efficiencies when deposited by slot-die coating onto flexible substrates.

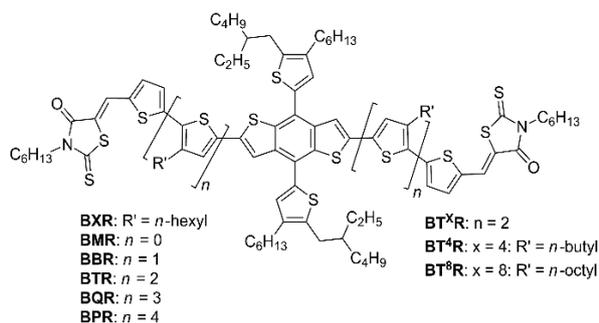


Fig 1: The chemical structure of BXR series.¹

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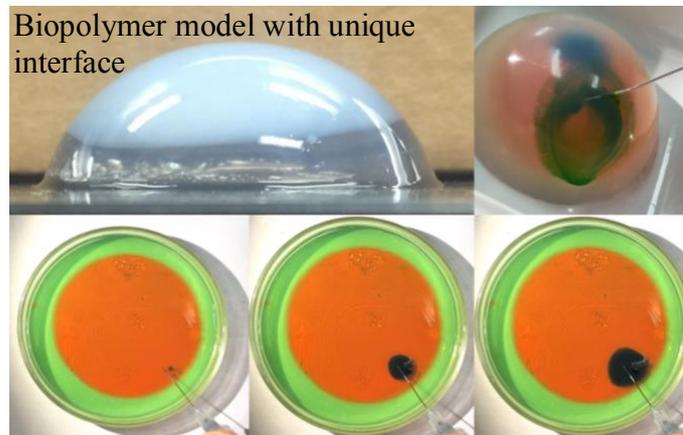
Biocompatible fluid for biological tissue separation

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Adhesion of biological tissues affects many natural and surgical processes and is a critical element of wound healing and medical treatments¹. Because biological tissue can vary widely, new means of measuring adhesion are needed, as are new ways of separating adhering tissue in a safe and reproducible manner. We have developed a microfluidic approach to study the adhesion of model and biological surfaces and tissues and to evaluate the effect of fluid rheology on the performance of separation processes. Novel combinations of biopolymers are used to mimic biological tissues and enable study of new therapeutic approaches to tissue cleaving and separation. Separation efficiency using fluids with different rheological properties was studied and compared to hydrodynamic models of pressure distribution in flow^{2,3,4}. Experimental variables such as flow rate, viscosity, and normal stress, as well as imposed pressure are used to determine the limits of adhesion and other tissue interactions.



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Ion Migration in Methylammonium Lead Halide Perovskite Films

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Ion migration plays a critical role in the hysteresis and stability of perovskite solar cells. My research topic focuses on the iodide ion migration phenomenon in Methylammonium Lead Halide Perovskite Films under external electrical field, including both 2D and 3D structure. The history of ion migration will be illustrated, as well as the following influence on the solar cell efficiency. The mechanism of ion migration will also be presented. The resulting pictures and videos obtained from our experiment are also showed in my presentation.

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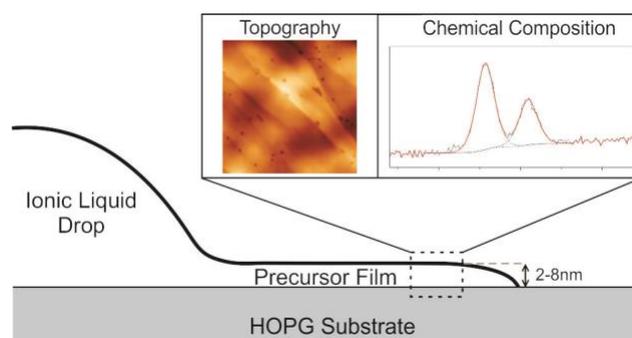
Precursor Film Formation of an Ionic Liquid on Highly Oriented Pyrolytic Graphite

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This study has determined the topographical and chemical characterisation of a precursor film formed by the ionic liquid (IL), hexyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide, [HMIM] [NTf₂] on a highly orientated pyrolytic graphite (HOPG) surface. Contact angle measurements and tapping mode atomic force microscopy (TM-AFM) were performed to examine the wetting properties and the morphology of the precursor film formed from [HMIM] [NTf₂]. This was then accompanied by synchrotron based X-ray photoelectron spectroscopy (XPS) and near edge x-ray adsorption fine structure (NEXAFS) spectroscopy to determine the chemical composition of the precursor film, and the orientation of the IL at the graphite surface respectively. AFM images revealed that precursor film formation was a two-stage process. First, very thin rigid IL patches were formed. These rigid patches expanded laterally with time with no detectable change in thickness. Second, a soft continuous film of IL was formed, of a few nm thickness. The thick film spread over the already established rigid film of IL, after which began a dewetting phenomenon revealing the thin rigid layer of [HMIM] [NTf₂] beneath. This autophobic behaviour was manifested by hole formation which expanded over time. XPS spectra collected from the precursor film region showed the presence of both the cation and anion in the film. The spectra did not indicate the presence of any chemical interactions between the IL components and the HOPG surface (confirmed by the absence of any change in the positions of the N 1s peaks arising from the anion and the cation). NEXAFS spectra revealed a preferential flat orientation of the imidazole ring relative to the HOPG surface. The study shows the utility and effectiveness of combining force microscopy and molecular spectroscopy for full characterisation of the behavior of precursor films formed during the spontaneous spreading of ionic liquid droplets.



Versatile and robust encapsulation systems using hybrid polymer-inorganic microgels

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Encapsulation is used to deliver reagents to specific reaction sites in a controlled manner. It has seen widespread implementation in fields as diverse as catalysis and cosmetics, most notably within the pharmaceuticals industry. The complexity and cost involved in encapsulation inhibits its use in the broader chemical industries. Due to the current use of toxic and expensive reagents, there is a growing need for a simple yet efficient encapsulation system – a system that not only incorporates cost effective reagents but is robust and provides enhanced selectivity and sustainability in a range of applications.

Hybrid polymer-inorganic microgels have been found to be promising encapsulating materials, with the potential for controlled and tuneable surface chemistry and sustained cargo release. Through entrapment within the gelled structure or electrostatic adhesion, a variety of materials, from macroscopic particles to small ions can be encapsulated. This phenomenon can be exploited for either sorption or release purposes. Coating the microgel with a silica shell provides improved chemical and mechanical properties. Such reinforced particles have increased mechanical strength, vital to their long-term performance, and the silica surface offers diverse opportunities for chemical functionalisation. This polymer-silica core-shell chemistry results in a facile method for encapsulation and delivery, and eliminates the need to modify the underlying polymer surface.

The versatile nature of the hybrid material allows its adaptation toward many applications, from minerals processing to soil conditioning. Utilising elementary reactions, mild conditions and ‘one-pot’ reactions provides an efficient and easily tailored system.

Quantification of long-range forces for underwater superoleophobic surfaces

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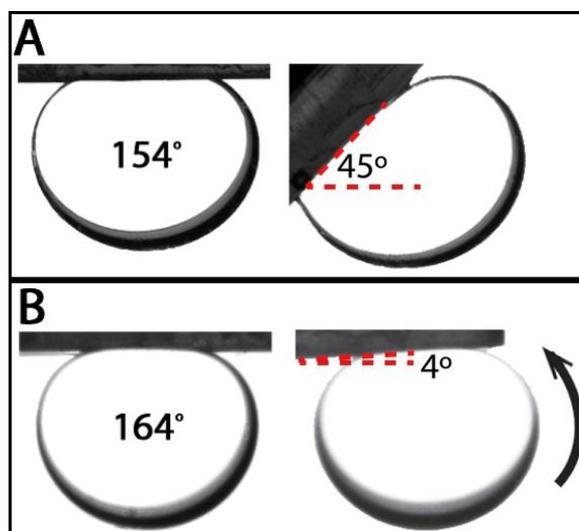
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Lubricant-infused surfaces that have slippery and anti-fouling properties have attracted great attention due to their promising impact on many medical, environmental, and industrial applications.¹ However, as their function relies on a lubricant layer being trapped in the surface roughness, it is crucial to reach a thorough understanding of the factors that determine the lubricant layer stability. In this work, the long-range van der Waals interactions that are responsible for the stability of thin lubricant films were quantified and the conclusions qualitatively tested against experimental results. Here, the system studied was a structured surface that has underwater superoleophobic properties: a wrinkled layer of hydrophilic poly(4-vinylpyridine) (P4VP), prepared on a shrinkable substrate². Lifshitz theory³ was used to estimate the Hamaker constant for the system, as a function of P4VP layer thickness. In addition, the capillary effect of trapping water in the specific surface micro- and nanostructure developed through the spontaneous wrinkling of the P4VP surface was estimated. Wrinkled P4VP surfaces with micro-scale wrinkles showed low adhesion to different oils with water droplet roll-off angle of $6^\circ \pm 1^\circ$, however the theoretical calculations showed that the P4VP is not an ideal polymer for such an application. Other polymers are being identified, for which adhesion of oil can be decreased by minimizing the contact area between the oil and the surface, and by stabilizing the infused water layer.



Underwater contact angle of an oil droplet (identify the oil type) and roll-off angle of the same droplet on (A) wrinkled P4VP film with wrinkle width xx nm, and (B) 36 wrinkled P4VP film with wrinkle width xx nm.

Protocols for Chemical Removal in Water Treatment

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This study aims to establish protocols for chemical removal in water treatment, and validate CCPs (Critical Control Points) and QCPs (Quality Control Points) for these chemicals at full scale water treatment and water recycling sites. The main chemicals of interest and removal barriers are PFAS (Per- and poly-fluoroalkyl substances) compounds removed by GAC (Granular Activated Carbon), CoCs (Chemical of Concerns) removed by RO (Reverse Osmosis) and other molecules such as taste and odour (TO) compounds removed by Ozone. The proposed research will include:

- 1) Establishing protocols and methods for PFAS removal by a GAC barrier and validating QCPs and CCPs at a full scale site.
- 2) Development of bioassay screening tests and fluorescence measurements as alternatives to chemical analytics for PFAS removal performance;
- 3) Use of an established protocol for CoCs removal by RO to understand a NatVal (National Validation framework for water recycling schemes) status and validate CCPs at a full scale site.
- 4) Use of an established protocol for taste and odour compounds removal by an Ozone barrier and establish CCP validation at full scale.
- 5) Establish and validate the performance of the different barriers for removing chemicals in an operational plant for water recycle in Antarctica using bioassay to screen plant performance as an alternative to chemical analytics.

The bioassay and fluorescence methods have been established to measure PFAS compounds across a wide range of concentrations (from mg/L to ng/L). A EC₅₀ (Half maximal effective concentration) has been established as the benchmark to measure the toxicity of PFAS and Fluorescence Excitation-Emission Matrix and Regional Integration has been used to quantify spectra for on-line detection of PFAS compounds at very low concentrations.

Effect of Methyl Isobutyl Carbinol on bubble rise velocity and stability of foam film formed under dynamic conditions.

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Methyl Isobutyl Carbinol (MIBC) is one of the most commonly used frothers in processing of Australian copper ores. The main reasons for the frother addition is to: (i) provide a narrow distribution of fine bubble size, (ii) prolong the bubble residence time in the flotation tank (by lowering the bubble's terminal velocity), and (iii) extend the lifetime of the froth to ensure efficient separation of floated value mineral particles. Hence, it is important to understand how frother concentration, as well as presence of electrolyte would affect these processes.

We have measured the terminal velocity of an isolated single bubble rising in solutions of MIBC of various concentration in the absence and presence of KCl. We also have studied the evolution and the lifetime of foam films formed upon bubble collision with the solution-air interface, i.e. under dynamic conditions. We attempt to correlate these experimental data with the properties (interfacial tension, interfacial rheology) of solution-air interface.

Transparent lubricant-infused surfaces for anti-biofouling applications

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The ecological and economic impact of marine fouling is devastating, affecting wild fisheries, aquaculture, marine sensors, and shipping.¹ We have developed an approach to stopping marine fouling based on lubricant-infused nanostructured surfaces, inspired by the *Nepenthes* Pitcher Plant². Our group has previously developed Teflon nanostructured wrinkles, which are robust to wear and have superhydrophobic properties.³ In this work,⁴ the nanostructured wrinkles were infused with silicone oil and their ability to inhibit the growth of marine bacteria reached 99% compared to the non-infused system. The wrinkles were optimised to retain the inert silicone oil and the amount of oil present on the surface was quantified for the first time using fluorescence. The amount of oil was then related to the performance of the surfaces both in terms of wettability and bacterial inhibition. The surfaces were also tested for their performance in a real-world field test conducted over seven weeks at Watson's Bay in Sydney's eastern suburbs where they showed resistance to marine fouling, compared to the uninfused control samples. The nanoscale wrinkles are spontaneously formed, tuneable, and very easy to produce. In addition, their polymeric nature makes them easily moulded and the infused surfaces display very good underwater transmittance, making them ideal for use with underwater sensors or cameras.

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Development of 3D printed microfluidic perfusion devices to measure cryoprotectant permeability

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We need new cryopreservation protocols with novel cryoprotectants since many endangered Australian plant species are resistant to existing cryopreservation protocols. Although over 40 molecules have been identified as having some cryoprotective ability, most current cryopreservation media are based on the cryoprotectants such as Me₂SO and Glycerol, and the toxicity of these molecules remains a serious issue. So, it is important to study the biophysical response of the cells to different potential cryoprotectants in order to develop “cell-type specific” novel cryopreservation protocols. One of the key parameters in a molecule’s cryobiological potential is cell permeability¹.

Various devices have been developed using conventional photolithography techniques to measure the permeability of different cryoprotectants. However, this technique has a number of limitations for applications related to biological systems. The development of soft lithography based on PolyDiMethylSiloxane(PDMS) by the Whitesides group has complemented conventional photolithography and enabled more microdevices to be fabricated for biological applications. These devices have been employed in the study of cell osmotic behavior for many years.

One disadvantage of soft lithography is that it requires photomasks to create the initial pattern, which are comparatively expensive for pattern features <25µm. In the past decade, the technological advances in 3D printing have enabled the easy fabrication of inexpensive microfluidic devices. Using 3D printing a master mould can be made which can be repeatedly used for creating PDMS replicas with soft lithography².

Here we report the development of 3D printed microfluidic perfusion devices for the investigation of biophysical response of individual cells to various cryoprotectants. Using these devices, we could successfully trap individual cells and analyse the cell shrink-swell kinetics with improved accuracy.

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Patterning of Photo-responsive Smart Materials

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Smart materials exhibit properties that react to changes in their environment. By changing an external condition, such as pressure, temperature, electric or magnetic field, stress, light, pH, their properties can be altered. This can manifest as a change in colour, volume, viscosity, shape, size or structure. In the best circumstances, this change is reversible and repeatable in nature, which allows some useful and fascinating applications in areas as diverse as electronics, construction, aviation, medicine, transport, etc.

Smart materials can be put into two categories:

- Active Smart Materials: Stimuli such as magnetic, thermal or electrical fields induce changes in their material and geometric properties, thereby enabling them to transduce energy. Example: Piezo-electric materials, magneto-strictive materials, ER fluids, etc.
- Passive Smart Materials: Capability to transduce energy is absent. They cannot act as transducers or actuators but as sensors. Example: Fibre optic cable.

Light is an appealing stimulus as it is clean, abundant and low-energy. A common example of a photo-responsive smart material is photo-chromic lenses which are coated with a UV-responsive material effecting a change in their colour when exposed to sunlight.

In my work at IITB and Monash, I plan to create patterned structures of photo-responsive materials, in particular graphene. At IITB, the plan involves femtosecond laser assisted patterning of graphene for various potential applications. At Monash, the focus will be on synthesising various graphene dispersions utilising photosensitive stabilisers and a range of organic and inorganic particles. Extensive studies on the patterning process, photochemistry and surface chemistry will be performed to evaluate the change in the properties of these photo-responsive materials after patterning. Structural and optical characterization will be used to substantiate these findings and to determine the changes in surface properties on photo-switching of the light sensitive materials. Once these characterisations have been completed, the materials will be tested in proof of concept devices such as sensors and optoelectronic arrays.

Uptake of Cubosomes into the Cellular Environment

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Therapeutic peptides and proteins represent the fastest growing class of new drugs with application as therapeutics, diagnostics and in vaccines. These complex, fragile macromolecules are, however, associated with significant drawbacks including long-term stability and degradation by the human immune system. In addition, many of these therapeutics have significant hydrophobic character, which makes encapsulation and storage an issue.¹ Encapsulation of these therapeutics in a carrier particle can both protect the protein against degradation, deliver the protein to the desired site of action, and offer controlled release properties.¹ Lipid based particles offer a range of advantages including potentially retaining the protein activity, and controlled release. Cubosomes are sub-micron sized (approx. 100-200nm) dispersed lipid cubic phase particles that have been shown to successfully encapsulate hydrophobic proteins and peptides in the lipid bilayer. Uptake of Cubosomes into a cellular environment has been found to be a slow process as seen in figure 1, with diffusion of therapeutic drugs across the cellular membrane over a prolonged timescale, this was tested using confocal microscopy.² We have formulated Cubosomes based on a mix of lipids to more successfully mimic the complexity of the native cell membrane. Peptide and protein uptake into these nanoparticles was shown to depend on the lipid composition, the cubic nanostructure, and the geometrical and charge characteristics of the encapsulated proteins.³ Uptake of these protein-loaded Cubosomes into a range of cell-lines was monitored using confocal microscopy and toxicity of these nanoparticles was determined again using confocal and FACS analysis.

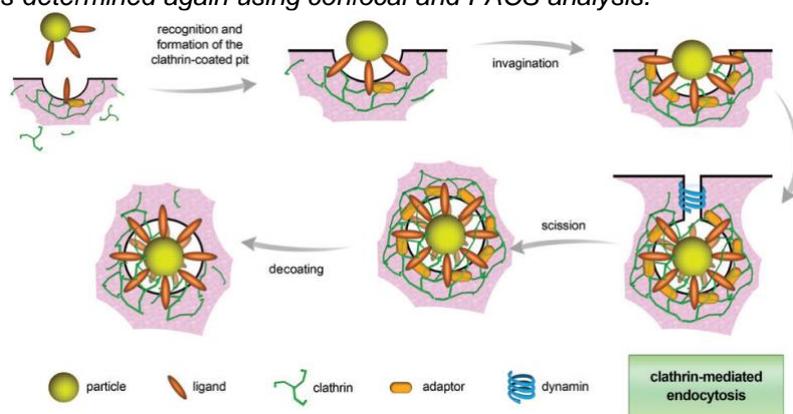


Figure 1 Clathrin Mediated Endocytosis diagram showing uptake of a particle.⁴

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Phytantriol modified with DSPE-PEG2000: a Temperature-Dependent Lipid-based Nanomaterials for On-demand Drug Delivery

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Purpose: Self-assembled lipid-based liquid crystalline (LC) systems, which can take advantage of the amphiphilic property of lipids to encapsulate drugs with different water solubility, have been obtaining attention as a potential drug delivery system¹. The LC nanostructures can be tuned via an external stimulus to achieve different release profile. In particular, thermo-responsive LC systems undergo nanostructure transition with increasing temperature, which can be exploited for on-demand therapeutics. This study aims to determine the temperature-dependent phase transition of LC systems consisting of phytantriol (PHYT) and DSPE-PEG2000.

Methods: PHYT was mixed with 5% molar ratio of DSPE-PEG2000 in chloroform. After removal of the chloroform, the mixture of lipid was then hydrated in excess PBS buffer of pH 7.4 and then sonicated to form a dispersion. The LC phase changes were determined on a temperature ramp (at 5 °C increment from 25 °C to 80 °C, 13keV) by using small angle X-ray scattering (SAXS) at the Australian Synchrotron.

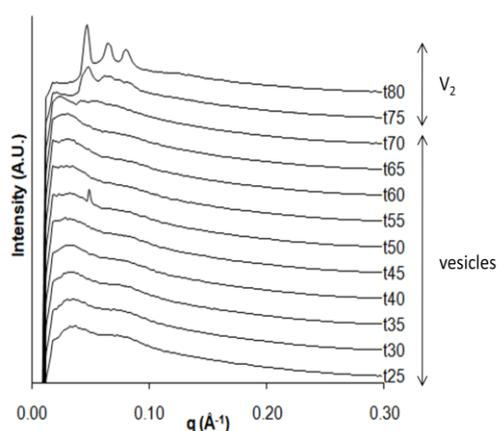


Figure 1. Phase diagram of PHYT

Results and discussion: The phase transition of hydrated PHYT is from bicontinuous cubic (V_2) to reversed hexagonal (H_2) with increasing temperature. When modified with DSPE-PEG2000, the phase transition profile is altered, from vesicle to V_2 (Figure 1).

Such identity changes stem from the alteration of the lipid critical packing parameter (CPP)². The large hydrophilic head group of DSPE reduces the CPP of the system, thereby switching the initial phase from V_2 to enclosed vesicles. With increasing temperature, the hydrophobic tail component in the system become bulkier. This inherently increases the CPP, resulting in the phase transition to V_2 .

Conclusion: The phase profile of PHYT (V_2/H_2) was successfully modified in the presence of DSPE-PEG2000 (vesicles/ V_2) via a reduction in the lipid critical packing parameters.

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CFD study of buoyancy driven droplet coalescence

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Liquid-liquid extraction is one of the critical mass transfer processing operations used in the mining and petroleum industries. High performance extraction columns achieve high mass transfer rates while processing large material throughputs. Droplet coalescence plays a vital role in determining the mass transfer rate in an extraction column, due primarily to its role in determining droplet size distributions. However models for determining droplet coalescence under realistic operating conditions are presently not available, or poorly validated. The present study forms one step in the development of a numerical tool able to predict droplet coalescence in industrially relevant scenarios.

Within the present study we use simulations to predict the impact and possible coalescence between two toluene droplets within a water continuous phase. The upper droplet is held stationary on a needle, while the lower droplet rises via buoyancy. We use a previously developed MSIC (Multi-Scale Interface Capturing) Multiphysics algorithm to predict the collision^{1,2,3}. The technique employs a novel sub-grid scale method to calculate the film pressure between the colliding droplets, and the volume of fluid method (VOF) for capturing droplet interface deformation. We obtain a phase map showing collision outcome as a function of diameter ratio and Weber number (We , interpreted as a non-dimensional impact velocity). Similar to the better understood gas-liquid system, results show a non-monotonic transition of coalescence-bouncing-coalescence as We increases, for equal diameter droplets, however the transition We numbers between the regimes are quite different. For different droplet diameters the results are more complex.

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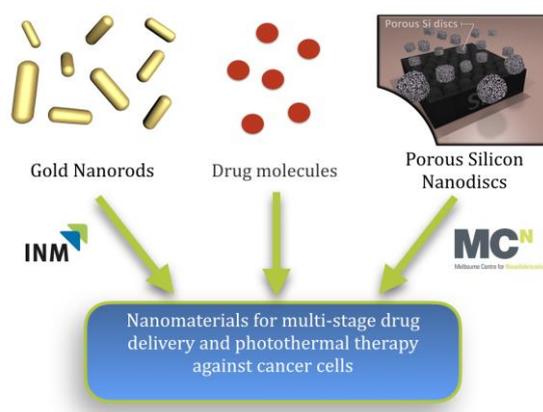
Design of multistage platforms for drug delivery and photothermal therapy against cancer cells

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Gold nanorods (GNRs) have emerged as promising candidates for photothermal therapy where they are stimulated by electromagnetic radiation and heat their surrounding¹. Porous silicon nanoparticles (pSi NPs) hold attractive prospects as nanovectors for drug delivery systems due to their biocompatibility, biodegradability and large surface-to-volume ratios². We integrate small GNRs into larger pSi NPs to design an advanced strategy for cancer therapy. GNRs are self-assembled within the pSi NP pores via DNA interactions, preventing undesired release. Double-stranded oligonucleotides are used to dehybridize above a certain temperature and release a commonly used chemotherapy medication, doxorubicin. Electromagnetic stimulation heats the GNR causing the dehybridization of the DNA releasing the drug and the GNRs from the porous silicon carrier. Once delivered to tumor tissue, hyperthermia is induced by the GNRs through laser excitation for a combined chemophotothermal therapy.



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Gas-induced flocculation of magnetic graphene oxide adsorbents

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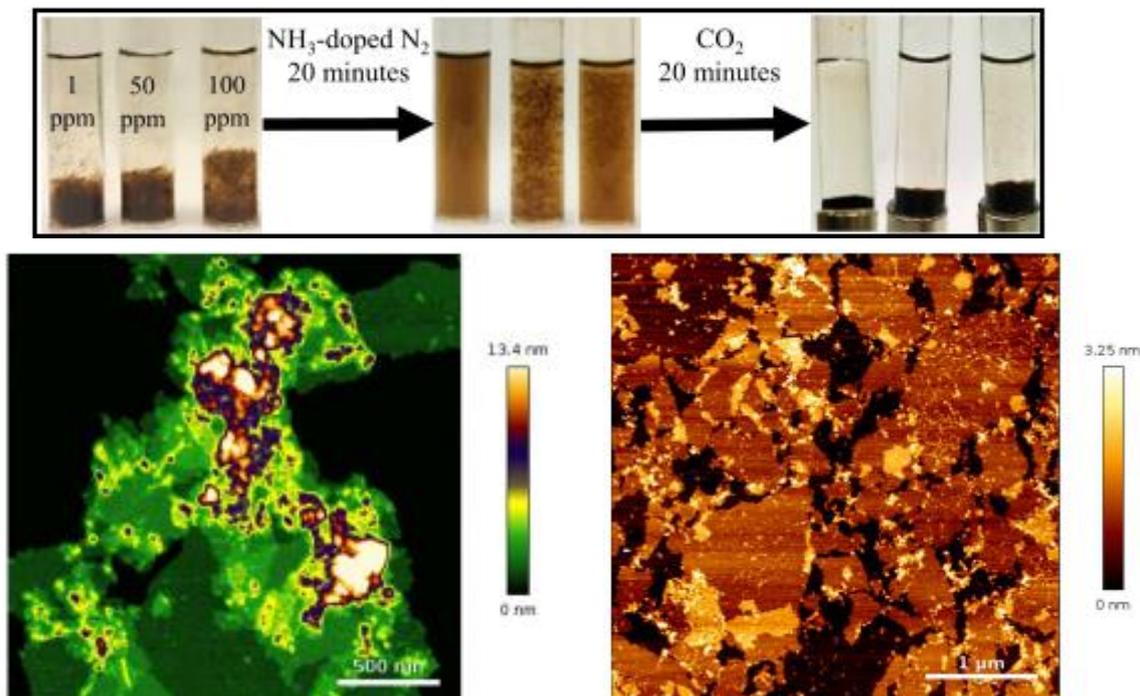
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It has previously been shown that graphene oxide can be noncovalently magnetized through adsorption with magnetite nanoparticles (mGO).¹ This magnetic composite was also shown for the first time to be efficient at capturing Eu³⁺, a popular model ion for nuclear waste, while forming easily removable magnetic aggregates. It has also been shown for the first time that the aggregation of these nanoparticle self-assemblies could be controlled through sparging with NH₃-doped N₂ and CO₂, which appeared to work through a combination of pH-driven surface charge stabilization and an increase in ionic strength due to salt formation.

Atomic force microscopy was applied to demonstrate that the unprecedented capture efficiency of mGO is likely due to europium locally precipitating on the surface of the mGO. The captured europium was shown to be strippable by 0.1 M EDTA and 1 M HNO₃, indicating the presence of two or more europium speciations or binding modes. Attempts to further characterize these species via powder XRD, ATR-IR, and thermogravimetric analysis were only partially successful. Future use of techniques such as X-ray photoelectron spectroscopy may provide further insight into how Eu is captured with such unprecedented efficiency, which is motivated as the system shows great promise for environmental and economic applications that require metal capture, making it worthy of further investigation.



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Mechanical and friction properties of cellulose-hemicellulose hybrid hydrogels

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The remarkable load bearing capacity and lubricating properties of natural biphasic materials such as articular cartilage and plant cell walls inspire us to study mechanics and tribology of biphasic hydrogels. These systems share a similar set of structural and mechanical characteristics, which include biphasic network structure, formation of surface coatings, as well as ability to incorporate non-Newtonian and viscoelastic fluids confined within pores. In this work we apply plant cell wall biomimetic principles to create regenerated cellulose-based hydrogels via dissolution in ionic liquids and subsequent solvent exchange into water to produce gels with controlled content of hemicelluloses.

The regenerated cellulose hydrogels mostly consisted of amorphous cellulose as confirmed by nuclear magnetic resonance spectroscopy and X-ray diffraction. By utilizing a rheometer-based technique that uniquely incorporates in situ mechanical characterization (compression–relaxation and oscillatory shear), we found that mechanical response of poroviscoelastic hydrogels is governed by the aggregation and rearrangement of cellulose matrix, as well as by fluid pressurization inside micro-pores. We compare the influence of plant cell wall polysaccharides, such as xyloglucan and arabinoxylan, on the mechanical properties of cellulose hydrogels and report a three time increase in mechanical toughness of Cellulose-Xyloglucan (CXG) and Cellulose-Wheat Arabinoxylan (CWAX) hybrid hydrogels. In addition, we quantify how poroelastic effects contribute to the formation of a fluid lubricating film between hydrogels upon compression via a mechanism similar to the ‘weeping’ lubrication of articular cartilage.

Evaluation of dynamic mechanical and micro-hydrodynamic behavior provides insights into the modes of tribo-rheological and interfacial behavior of biphasic gels, thus mimicking key aspects of articular cartilage lubrication under compression and sliding conditions.

Measurement of Polyelectrolyte Multilayer Mechanical Properties using Biaxial Tensile Stress Measurements

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The interfacial mechanical properties of emulsifiers at the oil-water interface are important for the stability and properties of emulsions. Deformability of emulsion droplets will influence the hydrodynamics and surface forces of oil droplets when undergoing collisions. This will then influence rates of coalescence and also the bulk rheology and tribology of an emulsion (important for textural aspects of emulsion use, such as foods and creams). Traditionally, emulsifier interfacial mechanical properties have been studied using either shear or dilational interfacial rheology, which work well for emulsifier systems such as surfactants and proteins. However, some more modern emulsifier systems, such as multilayer emulsifiers, cannot be so easily studied using these methodologies, as the construction of the interfacial layer is a multi-step process, and the layer cannot self-heal once disrupted (which will occur repeatedly when measured with conventional interfacial rheology).

Polyelectrolyte multilayers are one such potential emulsifier system for which it would be useful to obtain mechanical properties. Although complex to produce, emulsions stabilised with polyelectrolyte multilayers are becoming more viable due to the development of droplet microfluidic platforms as a means to generate emulsions with complex stabilizing layers. We have chosen to study the mechanical properties of polyelectrolyte multilayers using a technique more commonly used in tissue mechanics studies: a BioTester5000 Biaxial Test System (CellScale division of Waterloo Instruments Inc., Waterloo, Ontario, Canada)¹. The use of this technique relies on the formation of the interfacial layer on a supporting surface, in this case PDMS. PDMS was selected to allow measurements to be made for the multilayer on a negatively charged hydrophilic interface, and a non-charged hydrophobic interface. The latter interface acts as a mimic of an oil-water interface for the purpose of this investigation.¹ The multilayer system studied was Polyacrylic Acid (PAA) and Polydiallyldimethylammonium chloride (PDADMAC). In addition to studies of the effect of different terminated layer on the mechanical properties, studies were performed with cross-linked multilayers, to determine the extent to which mechanical properties may be enhanced post-formation.

* Measurements were performed for a partially dehydrated multilayer, to allow tracking measurements of film deformation to be detected.

Investigation of Targeted Delivery of Drug Nanocrystal Loaded Liposomes by Azide-cyclooctyne Functionalisation to Cancer Cells

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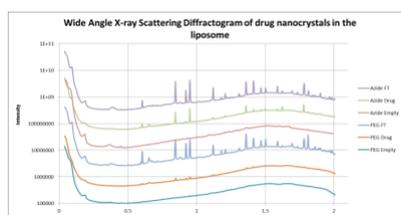
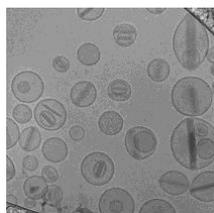
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Liposomes, as drug delivery carriers constructed by phospholipid bilayers, can encapsulate high dose of drug and achieve targeted delivery by surface modification. Copper-free click chemistry is a type of fast and high-yielding chemistry reaction between azide and cyclooctyne¹. The crystallisation of drugs in functionalised liposomes can decrease the administration frequency and minimise side effects dramatically.

Liposomes were prepared with the thin film hydration method. Drug was loaded using pH-gradient loading method and crystallised with the freeze-thaw method². Liposomes were characterised by DLS for particle size, cryo-TEM for nanocrystal morphology, WAXS for nanocrystal polymorphism, reverse dialysis for surface azide quantitation, and SANS for shape and sizes of the drug nanocrystals. Then lung cancer cells were then metabolically labelled³ using acetylated dibenzylcyclooctyne (DBCO) modified unnatural mannose. The azide functionalised liposomes were then incubated with the cells for targeting and the binding efficiency was determined quantitatively with TIRF.

Liposomes with a particle size of 120-150 nm, achieved a drug encapsulation efficiency of 95%. Drug nanocrystals were clearly visualised in cryo-TEM (Fig 1) and its polymorphism was determined by WAXS in Australian Synchrotron (Fig 2). The amount of available azide group on the surface of liposomes was determined to be 10%. Currently, we are investigating the click chemistry binding efficiency of liposome nanoparticles to A549 lung cancer cells quantitatively.



Liposomes with encapsulated drug nanocrystals and click chemistry modifications possess a great potential in cancer drug targeted delivery. Drug nanocrystals were successfully made in the liposome and its polymorphism was determined. The amount of available surface modification was quantified, and cell targeting in-vitro is currently under investigation.

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Shaped and Stimuli-responsive Droplets for Improved Drug Delivery

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Abstract: A shaped non-spherical endoskeleton droplet can be prepared to match the shape of a target surface with a larger surface area than a spherical droplet¹ and has been found to be responsive to a number of stimuli². Although useful for consumer product applications, current shaped droplets are too large for other uses like aerosol delivery of medicines³. Here we investigate production of shaped and stimulus-responsive droplets, at much smaller length scales than previously achieved, in order to accomplish enhanced drug delivery to the respiratory tract. Instead of direct molding of the elongated droplets we apply a novel approach developed earlier⁴ to produce micron-scale rod-shaped droplets via careful control of surfactant adsorption. High aspect ratio rod-like lipid droplets have been prepared (Fig 1) and are tested for improved aerosolization and delivery using standard medical protocols.

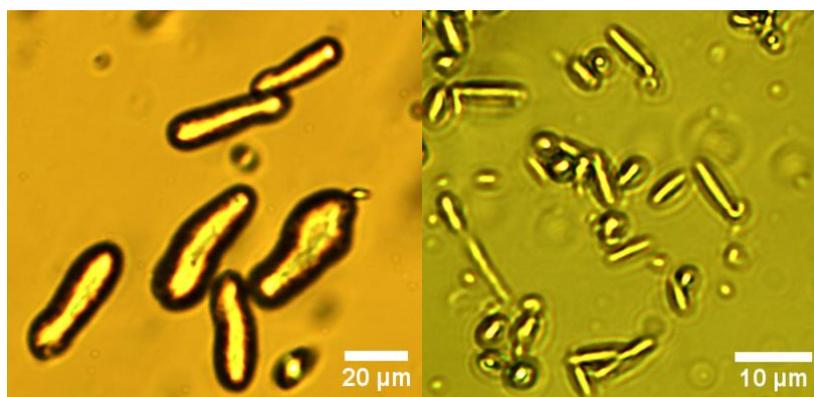


Fig 1: Micrographs of lipid rods prepared by emulsification method, demonstrating the ability to produce micron-scale rods without direct molding.

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The synthesis and determination of Nano scale graphene oxide (Nano-GO) and its physical properties as a function of lateral size.

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My project seeks to detail the properties of Nano-GO, thus determining at what lateral size GO acts like a particle and not a molecule. Using a modified version of the Hummers method for Graphene Oxide synthesis, we will produce a range of Nano-GO fragment sizes that are <100 nm. To achieve this, portions of the reaction mixture are removed at predetermined times, thus, limiting the degree of oxidation of the starting reactant (activated carbon nanoparticles) and creating Nano-GO of different sizes. If this synthesis fails to produce the required scale in size, a controlled successive oxidation of GO will be used to overcome this problem. An elemental analysis of each sample will determine whether the degree of oxidation is a factor in the synthesis of specifically sized fragments. Once successfully produced, the Nano-GO will be tested for its behaviour in several ways including: self-assembly, adsorption, interfacial properties, and dispersion stability. In addition, optical properties will be considered in the form of quantum dot fluorescence. The comparison of each fragment size and their properties will hopefully show us that there is a size at which GO behaves as a molecule rather than a particle.

Probing the surface characteristics of polymeric nanoparticles decorated with cell-penetrating peptides

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Cell penetrating peptides (CPPs) are cationic amino acids sequences that can be conjugated to the surface of polymeric nanoparticles to enhance interactions with the nano-bio interface.¹ Our aim was to investigate the distribution of CPPs with three different architectures (short, long linear and branched) on the surface of PLGA nanoparticles produced using microfluidics.

The nanoformulations were prepared by mixing an organic polymer solution (PLGA, 10 mg/mL in acetonitrile) and an aqueous PVA solution (2% w/v) using the NanoAssemblr™ Benchtop Device. The CPPs RRH, TAT and a branched TAT were conjugated to the PLGA nanoparticles with a post-microfluidics approach or in an *in situ* microfluidics approach by adding the CPP solution to the aqueous solution before mixing. Afterwards, the CPP-tagged nanoformulations were incubated with gold nanoparticles for 2 h and characterised. Size and zeta-potential of the nanoparticles were determined using dynamic light scattering and laser doppler microelectrophoresis. The surface distribution of CPPs was investigated using TEM.

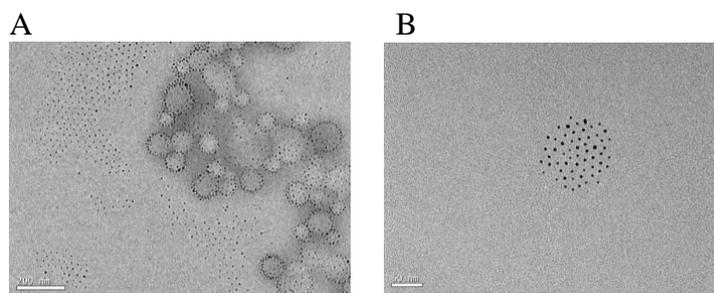


Figure 1. Micrographs of gold-labelled PLGA nanoparticles (A) RRH-tagged nanoparticles prepared using a post-microfluidic approach showing a surface distribution of gold nanoparticles. (B) TAT-tagged PLGA nanoparticles prepared using an *in situ* microfluidics with a distribution throughout the nanoparticle.

The size of the nanoparticles did not change with the addition of CPPs, however the zeta-potential became less negative. The distribution of CPPs on the surface of the nanoparticles depends on the preparation method used.

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Lift forces on a sphere in bounded and wall-bounded flows

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Particle lift forces act perpendicular to the direction of flow. In a flowing suspension lift forces can move particles away from or towards walls, or cluster particles at certain radial positions within a pipe. In the biological context, lift forces contribute to the separation between platelets and red blood cells that results in a ‘Cell Free Layer’ forming adjacent to blood vessel walls, as well as providing a separation mechanism that is the basis of novel cancer-detecting cell sorting techniques¹.

Models that can predict the lift forces on rigid spherical particles under low inertia (Particle Reynolds number $Re_p \lesssim 1$) conditions, particularly in wall-bounded parabolic flows, are not available or adequately validated. As part of continuing project to develop such a model, this 3D numerical study examines the shear induced lift forces on a small rigid spherical particle within a linear shear flow, both within an unbounded domain and also within a domain bounded on one side by a rigid wall. We set the slip velocity of the particle (u_{slip}) and calculate the resulting lift force. The numerical solutions are validated against analytical solutions from Saffman (1965)², McLaughlin (1991)³ and McLaughlin (1993)⁴. The lift forces obtained for both domain types agree well with the above models, provided that $Re_p \ll 1$. In the unbounded simulations, the influence of domain size is tested and it is found that a domain size of at least 50 times the particle radius is required to accurately predict the lift at $Re_p = 0.1$. For the wall bounded flows the lift force can either increase or decrease with increasing wall particle separation, dependent upon Re_p .

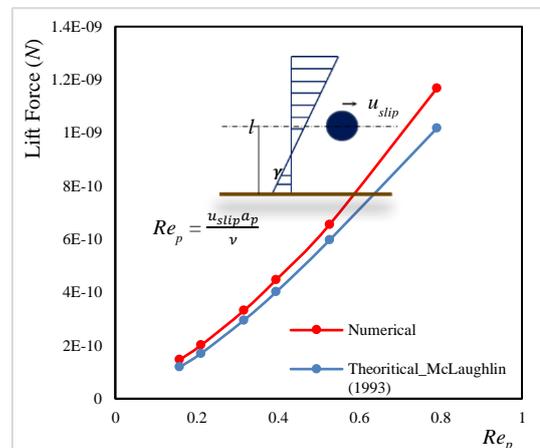


Figure 1. Lift force comparison for $a_p = 100\mu\text{m}$, $\gamma = 10\text{s}^{-1}$ in water at $l = 200\mu\text{m}$.

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