17th Australia-Japan Colloids Symposium

Soft Matter

AUSTRALASIAN COLLOID AND INTERFACE SOCIETY

Version 1.0

Welcome Message

On behalf of the organising committee I am very pleased to welcome your participation in the 17th Australia-Japan Colloids Symposium. These meetings have a history stretching back to 1992 and have provided a great opportunity for Japanese and Australian Colloid and Interface Scientists to engage both intellectually and socially. Many fruitful collaborations and deep friendships have resulted from these meetings.

The COVID-19 pandemic has had a profound influence on the lives of people all over the world and this event is not immune from its influence. The pandemic means that we are unable to meet in person and hence this event is being held virtually. Whilst this brings many challenges it also has provided us with a number of opportunities. With the support of the Australasian Colloids and Interface Society we have been able to make the event free and this has enabled us to bring together a record number of participants (over 180 at this point). Additionally, we are trialling a number of new approaches in how our scientific presentations are being delivered including pre-recorded talks, breakout virtual rooms for poster presentations, graphical abstracts for social media promotion and 1 minute pre-recorded lightning talks for poster presenters giving them the opportunity to get you excited about their presentation.

I very much hope that in the near future we are able to resume meeting in person I think that this event will also show us a new way that we can augment our traditional meetings.

I must acknowledge the outstanding work of the organising committee, Saffron Bryant, George Franks, Stuart Prescott, Anna Wang and Catherine Whitby in getting this event off the ground in a very short period of time. You all have been wonderful to work with. Thank you.

I must acknowledge the valuable guidance of Professor Takanori Takiue as our international adviser and the strong support of Shigeru Deguchi and Naoyuki Ishida when the idea of this meeting was first raised. This event would never have taken place if it wasn't for the initial efforts of Greg Warr and Ben Boyd, who first championed the idea.

Thank you for your attendance. I think the exciting program we have assembled will reward your participation well.

I wish you all a safe and healthy 2020. Sincerely,

Vince Craig

The Organising Committee

George Franks, University of Melbourne

Stuart Prescott, UNSW

Anna Wang, UNSW

Catherine Whitby, Massey University

Vince Craig, ANU

Saffron Bryant, RMIT

Adviser to the local organising committee
Takanori Takiue, Kyushu University

Important Information

- For the latest updates and information, visit: https://complexfluids.net/aj/
- If you're having technical difficulties and need to contact the organizing committee, please email: vince.craig@anu.edu.au, or saffron.bryant@rmit.edu.au

Courtesy on Zoom

- Please keep yourself muted unless participating directly in the discussion.
- Please turn off your video camera when you're not talking.
- Please ask questions via the text chat and the session chair will collate them for the speaker. During the question session, the raise hand feature can also be used, but priority will go to the questions in the chat session.
- If presenters are sharing video as well as their slides, side-by-side mode can be a nice way of seeing both more easily.
- Zoom can be accessed via the desktop client, mobile client or web browser.

Time Zones for Reference

Japan	Eastern Australia	Western Australia	New Zealand	United Kingdom	USA (New York)
GMT+9	GMT+10	GMT+8	GMT+12	GMT+1	GMT-4

Keynote Speakers

Tam Greaves is an Associate Professor within Physics at RMIT University, and has been at RMIT since 2014. She completed her Ph.D. in Experimental Physics in 2004 at Monash University, Australia. In 2005 she joined CSIRO as a Postdoctoral Fellow, and worked there for nearly 10 years. She is a frequent user of the Australian Synchrotron SAXS/WAXS beamline, and member of the advisory committee for the BioSAXS beamline which is being built over the next 3 years. Tam conducts research into understanding the fundamental physicochemical and thermal properties of ionic liquids, their mixtures, liquid nanostructure and the solvophobic effect. She is currently developing ionic liquid solvents for use with biological molecules in a broad range of applications, including protein solubility, stability and crystallization. A key focus of her research is the development of high throughput methodologies for the experimental design, sample synthesis, characterisation and data analysis.

Kiyoshi Kanie is a Professor in the Institute of Multidisciplinary Research for Advanced Materials at Tohoku University in Sendai. He received his PhD from Kyoto University in 2000. He held research associate positions at the University of Tokyo and Tohokhu University before being appointed an Associate Professor at Tohokhu University in 2008 and a Full Professor in 2019. His research interests encompass, the design and synthesis of functional materials, liquid phase synthesis of functional inorganic nanoparticles with controlled size and shape and the development of organic-inorganic hybrid materials with dynamic functions. He was the recipient of the Science Award of the division of colloid and surface chemistry of the Chemical Society of Japan in 2010 and received the Japan Institute of Metals and Materials meritorious award in 2014.

Naoyuki Ishida is Associate Professor in the Graduate School of Natural Science and Technology at Okayama University. He received his Ph.D. degree form Kyoto University in 2000. From 2000 to 2012, he worked at the National Institute of Advanced Industrial Science and Technology (AIST) as a research scientist and moved to Okayama University in 2012. He also spent one year at University of Leeds in 2006 and eight months at the Australian National University in 2017 and 2018 as Visiting Fellow. His research is aimed at understanding of surface forces by direct measurement and he has made significant contributions to our understanding of the forces between hydrophobic surfaces. His research also focuses on interaction forces in non-aqueous liquids, interactions of biomolecules and surface functionalisation with stimuli-responsive polymers.

Catherine Whitby was awarded her PhD from the University of Melbourne in 2001. She held research associate positions at the University of Hull (United Kingdom) and the University of Sydney (Aust.) before being appointed as an ARC Future Fellow and Senior Research Fellow in the Ian Wark Research Institute at the University of South Australia. In 2014 she was appointed as a Senior Lecturer in Chemistry in the School of Fundamental Sciences at Massey University in New Zealand. She is a physical chemist with expertise in colloid and surface chemistry. She uses nanomaterials to modify the chemistry of drop and bubble surfaces. This strategy enables her to control the structure, stability and flow of soft materials. Her findings have been applied in food and pharmaceutical products and in drilling fluids.

Program

17th September – DAY ONE
Morning

Tokyo time	Sydney time	Speaker	Title	Page
Chair: George Franks				
8.45 – 9 am	9.45 – 10 am	Vince Craig	Welcome	
9 – 9.15 am	10 – 10.15 am	Syuji Fuji	Shape-designable liquid marble	1
9.15 – 9.30 am	10.15 – 10.30 am	Raymond Dagastine	Mass transfer across nanometre thin films between microbubbles	2
9.30 – 9.40 am	10.30 – 10.40 am	Joe Berry	Mechanics of Soft Materials	3
9.40 – 9.50 am	10.40 – 10.50 am	Neha Sharma	Surface coating of Liposomal Membranes with Gum Ghatti	4
9.50 – 10 am	10.50 – 11 am	Grant Webber	Specific-ion Effects in Complex Polymer Brush Systems	5
10 – 10.30 am	*11 – 11.30 am*	*Morning tea break*		
Chair: Saffron Bryant				
10.30 to 11 am	11.30 am - 12 pm	Keynote: Prof. Kiyoshi Kanie	Size- and shape-controlled liquid phase synthesis of inorganic nanoparticles and application to organic-inorganic hybrid	6
11 to 11.15 am	12 – 12.15 pm	Hideya Kawasaki	Ultrasonically activation of colloidal gold nanoclusters toward sono-catalysis	7
11.15 to 11.30 am	12.15 – 12.30 pm	Cathy McNamee	Change in the inter-surface forces between two charged surfaces containing salts by the presence of a liquid flow	8
11.30 to 11.45 am	12.30 – 12.45 pm	Srinivas Mettu	“Soft tip” Atomic Force Microscopy in Probing the Interactions of Different Oil Components with Calcite Surface	9
11.45 to 12 pm	12.45 – 1 pm	Michael Higgins	Hydration Layer Structure of Biofouling Resistant Nanoparticles	10
12 to 1 pm	*1 to 2 pm*	*Lunch break*		

Afternoon

Tokyo time	Sydney time	Speaker	Title	Page
12 to 1 pm	*1 to 2 pm*	*Lunch break*		
Chair: Catherine Whitby				
1 to 1.30 pm	2 to 2.30 pm	Keynote A/Prof. Tamar Greaves	Solvent properties of protic ionic liquid-water mixtures, and their application to biological molecules	11
1.30 to 1.45 pm	2.30 – 2.45 pm	Chiho Watanabe	Delayed diffusion and unique phase behaviour in polymer crowding micro-emulsions	12
1.45 to 2 pm	2.45 – 3 pm	Stephen Holt	The adsorption of glucose oxidase on mesoporous aluminium	13
2 to 2.10 pm	3 – 3.10 pm	Saffron Bryant	Selective Ion Transport across a Lipid Bilayer in a Protic Ionic Liquid	14
2.10 to 2.20 pm	3.10 – 3.20 pm	Hank (Qi) Han	Conformational Changes of Protein with Ionic Liquids: A Multi-technique Approach	15
2.20 to 2.30 pm	3.20 – 3.30 pm	Durga Dharmadana	Human neuropeptide substance P self-assembles into semi-flexible nanotubes that can be manipulated for nanotechnology	16
2.30 to 3 pm	*3.30 to 4 pm*	*Afternoon tea break*		
Chair: Ben Boyd				
3 – 3.15 pm	4 – 4.15 pm	Anna Wang	Aggregation of highly negatively charged vesicles	17
3.15 – 3.30 pm	4.15 – 4.30 pm	Rico Tabor	Precise molecular packing from designed surfactants for vesicles and wormlike micelles	18
3.30 – 3.45 pm	4.30 – 10.45 pm	Hiroki Matsubara	Common black film stability and synergetic adsorption in ionic−nonionic mixed surfactant systems	19
3.45 – 4 pm	4.45 – 5 pm	Thomas McCoy	Spontaneous self-assembly of thermoresponsive vesicles using a zwitterionic and an anionic surfactant	20
4:10 pm onwards	5:10 pm onwards	Poster Session 1		

18th September – DAY TWO
Morning

Tokyo time	Sydney time	Speaker	Title	Page
Chair: Ray Dagastine				
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8.50 – 9 am	9.50 – 10 am	Ramzi Kutteh	Microhydrodynamics of arbitrary-shape passive and active particles in linear flow: daily life at the small scale	23
9 – 9.10 am	10 – 10.10 am	George Franks	Elastic plastic fracture mechanics investigation of toughness of wet colloidal particulate materials: influence	24
9.10 – 9.20 am	10.10 – 10.20 am	Ishihara Shingo	Modelling of interaction force between particles and simulation of breakage behaviour	25
9.20 – 9.35 am	10.20 – 10.35 am	YK Leong	Thixotropic clay gels: microstructure and role of colloidal forces	26
9.35 – 10 am	*10.35 – 11 am*	*Morning tea break*		
Chair: Rico Tabor				
10 - 10.30 am	11 – 11.30 am	Keynote: Prof. Naoyuki Ishida	Evaluation of Non-DLVO Forces between Surfaces in Liquids by Atomic Force Microscopy	27
10.30 - 10.45 am	11.30 - 11.45 am	Yuki Uematsu	Is the origin of the negative surface charge of hydrophobic water surface OH ion adsorption?	28
10.45 – 11.10 am	11.45 – 12.10 pm	Duc Nguyen and Liwen Zhu	Soft-hard Janus nanoparticles for polymer encapsulation	29
11.10 - 11.20 am	12.10 - 12.20 pm	Lee Shi Ting	LSPR-mediated high-resolution imaging for the study on cell adhesion and spreading	30
11.20 - 11.30 am	12.20 - 12.30 pm	Aaron Elbourne	Antibacterial liquid metals: biofilm treatment via magnetic activation	31
11.30 - 12.30 pm	12.30 - 1.30 pm	Poster Session 2		
12.30 -1.30 pm	*1.30 – 2.30 pm*	*Lunch break*		

Afternoon

Tokyo time	Sydney time	Speaker	Title	Page
12.30 - 1.30 pm	*1.30 - 2.30 pm*	*Lunch break*		
Chair: Arlene McDowell				
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2 - 2.15 pm	3 - 3.15 pm	Leonie van 't Hag	SANS studies with lipid cubic phase to investigate protein encapsulation and conformation	33
2.15 - 2.30 pm	3.15 - 3.30 pm	Masumi Villeneuve	Hydrophobic seed proteins on starch granule surface enables gluten-free rice bread making	34
2.30 - 2.45 pm	3.30 - 3.45 pm	David Beattie	An ATR FTIR study of the interaction of tio2 nanoparticle films with β-lactoglobulin and bile salts	35
2.45 - 3 pm	3.45 - 4 pm	Andrew Clulow	Mimicking lipid self-assembly in digesting milks	36
3 – 3.30 pm	*4 – 4.30 pm*	*Afternoon tea break*		
Chair: Anna Wang				
3.30 – 3.45 pm	4.30 – 4.45 pm	Iijima Kazutoshi	Preparation of thin films and hydrogels of polysaccharides via polyion complex nanoparticles	37
3.45 – 4 pm	4.45 – 5 pm	Nick Reynolds	Self-healing hydrogels formed from nanofibrillar assemblies of the amino acid phenylalanine	38
4 – 4.15 pm	5 – 5.15 pm	Takuya Yanagimachi	Voltage driven characteristic instability of nematic liquid crystal on chemically patterned substrate	39
4.15 – 4.30 pm	5.15 – 5.30 pm	Nigel Kirby and Susanne Seibt	1. Small angle scattering at the Australian Synchrotron. a fantastic tool for colloid and interface science 2. The dynamics of gold nanorod growth – a SAXS study	40 41
4.40 – 4.50 pm	5.40 – 5.50 pm	Vince Craig	Closing	

Thursday 17th September

Shape-Designable Liquid Marble

Syuji Fujii[1,2], Junya Fujiwara[3], Florian Geyer[4], Doris Vollmer[4], Hans-Jürgen Butt[4], Tomoyasu Hirai[1] and Yoshinobu Nakamura[1,2]
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Liquid marbles (LMs) are liquid droplets coated with solid particles adsorbed at the liquid-gas interface and behave as non-wetting soft, elastic objects [1,2]. A requirement for the particles to work as an effective LM stabilizer is that they are hydrophobic (intrinsic water contact angle close to or larger than 90°). LMs can be prepared using solid particles with various chemical compositions including organic, inorganic and composite particles. Most of the literature is concerned with (near) spherical particles and their flocs. However, there are no studies on LMs stabilized with well-defined non-spherical particles or stabilizers. Thus, it is crucial to reveal and understand the relationship between solid particle shape and LM structure formation to utilize the LMs.

Fig. 1 Initials of the term polyhedral LM: PHLM, formed using transparent 2.35 mm-sized plates and dyed water. LMs with different polyhedral morphologies. SEM image of a dried cyanoacrylate-treated 3 µL polyhedral LM using 0.34 mm-sized plates.

In this study, a new type of armored droplets, so-called polyhedral LMs are introduced (Figure 1) [3]. These LMs consist of liquid droplets stabilized by hexagonal plates made of poly(ethylene terephthalate), which adsorb to the liquid-air interface. Depending on the specific combination of plate size and droplet diameter, the plates self-assemble into highly ordered hexagonally arranged domains. Even tetrahedral-, pentahedral-, and cube-shaped LMs composed of only 4 to 6 plates are demonstrated. During evaporation of the internal liquid, due to the high adsorption energy of the plates at the liquid-air interface, the overall surface area stayed constant resulting in strongly deformed polyhedral LMs. In line with this, highly asymmetric and super-long polyhedral LMs and letters are obtained due to the strong interfacial jamming exerted by the rigid hexagonal plates. This is particularly pronounced for larger plate sizes leading to LMs with unusually sharp edges.

References

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[3] F. Geyer, Y. Asaumi, D. Vollmer, H.-J. Butt, Y. Nakamura, S. Fujii, Adv. Funct. Mater. **29** (2019) 1808826.

Mass transfer across nanometre thin films between microbubbles

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The role of interfacial coatings in gas transport dynamics in foam coarsening is often difficult to quantify. The complexity of foam coarsening measurements or gas transport measurements between bubbles requires assumptions about the liquid thin film thickness profile in order to explore the effects of interfacial coatings on gas transport. It should be possible to independently quantify the effects from changes in film thickness and interfacial permeability by using both atomic force microscopy (AFM) and optical microscopy to obtain time snapshots of this dynamic process. Further, it is expected that the surfactant and polymer interfacial coatings will affect the mass transfer differently. We measure the mass transfer between the same nitrogen microbubbles pairs in an aqueous solution using two methods simultaneously. First, we quantify the bubble volume changes with time via microscopy and second, we use AFM to measure the film thickness and mass transfer resistances using a model for the gas transport. Modelling of the interface deformation, surface forces and mass transfer across the thin film agrees with independent measurements of changes in bubble size. We demonstrate that an anionic surfactant does not provide a barrier to mass transfer, but does enhance mass transfer above the critical micelle concentration. In contrast, a polymer monolayer at the interface does restrict mass transfer. This approach may also have potential in providing a novel method to rapidly screen interfacial species with potential utility in the areas of ultrasound contrast imaging agents, oxygen carriers and therapeutic gases [1].

[1] Y. Q. Yang, M. D. Biviano, J. X. Guo, J. D. Berry and R. R. Dagastine, *Journal of Colloid and Interface Science*, 2020, **571**, 253-259.

Mechanics of Soft Materials

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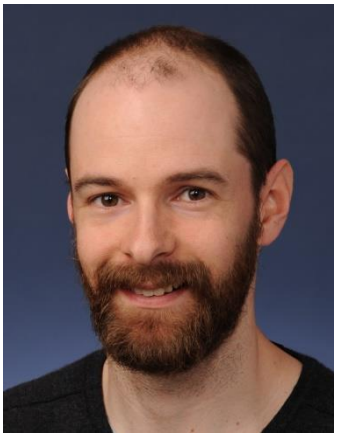
Soft materials have critical application in paints, food processing, biology and medicine. The function of these materials depends upon responses to external stimuli that may be orders of magnitude different in length and time-scales. A change in something as simple as size or material stiffness of a drug delivery particle can mean the difference between successful transport & release of a drug to a specific disease site in the body or expulsion; a change in porosity or viscoelastic response of a hydrogel used for cell culture can significantly impact biological functions such as cell proliferation and differentiation; and a change in elastic modulus can determine if capsules containing flavour, fragrance or paint survive processing & handling. These widely varying but poorly understood characteristics are fundamental to the future design of advanced materials.

There is currently a lack of appropriate models required to determine the material properties of micro-capsules and micro-particles. Instead, measurements are typically carried out on larger samples under the assumption that materials made from the same components have the same properties regardless of size and shape. For example, interfacial rheology of complex interfaces is commonly carried out on flat interfaces, and material property measurements of hydrogels are undertaken on large, thick films. Here we show that size and shape do matter, and present new models required to interpret data resulting from experiments directly undertaken with micro-capsules and micro-particles. The new models open up the possibility of testing the rheological and/or mechanical properties of capsules and particles directly at the length-scale and geometry relevant to the required application.



Surface coating of Liposomal Membranes with Gum Ghatti

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Liposomal membranes are self-assembled lipid vesicles which are extensively studied for drug delivery processes. Polysaccharides such as amylopectin, dextran and pullulan were shown to exhibit prominent interaction with liposomal membranes1. *Gum Ghatti* (GG) is a proteoglycan derived from *Anogeissus Latifolia*, widely available in the Indian subcontinent. It is widely used emulsifier and thickening agent in pharmaceuticals, paper industries and food items. In this study, we investigated GG-induced morphological changes in the nano-sized liposomal membranes made up of neutral phospholipids; 1,2-dioleoyl-sn-glycero-3-phosphocholine (DOPC), 1-palmitoyl-2-oleoyl-sn-glycero-3-phosphocholine (POPC) and positively charged lipid; 1,2-dioleoyl-3-trimethylammonium-propane (DOTAP). We aimed to coat GG on the surface of different liposomal membranes via the aid of electrostatic interactions between lipid molecules and negatively charged GG. The changes were evaluated by difference in size and surface charge of the membranes before and after addition of GG. Further, the morphological changes in the membranes were observed with optical microscopy and transmission electron microscopy (TEM). Rise in size of the membranes was found to vary with concentration of GG. Figure 1 represents TEM images of POPC and POPC_GG liposomal membranes which show a coating of GG on the surface of POPC membranes. Interestingly, introduction of GG in cationic liposomal suspension caused aggregation which can be seen with turbidity in the solution, possibly attributed to fusion of the individual membranes. It is in agreement with a previous study where polysaccharides exhibit coating or bridging of lipid vesicles depending on the concentration2. Our study demonstrates change in activity of polysaccharide (GG) with the composition of lipids, and contributes to the present outlook of GG.

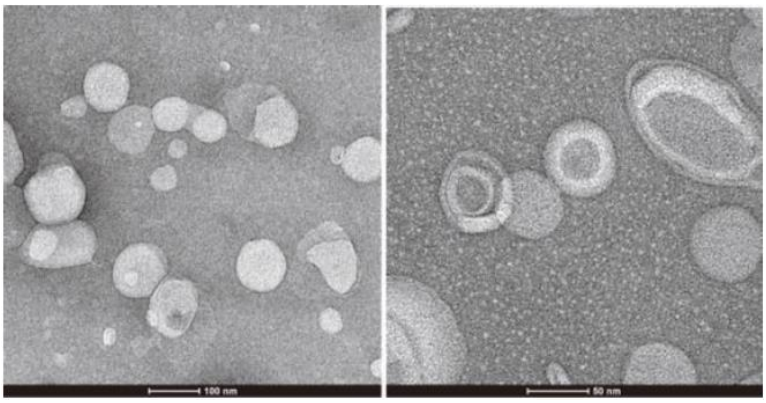

Figure 1. TEM images of POPC (left) and POPC_GG lipid vesicles (right).

References

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[2] Sumamoto et al., *J. Biochem.* 1980, **88**, 1219.

Specific-ion Effects in Complex Polymer Brush Systems

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Physicochemical behaviour of soft-matter and colloidal systems can be impacted by the identity of any electrolyte present.[1] These specific-ion effects go beyond simplistic models based on ion valency and concentration and have be known for well over a hundred years. In spite of years of research there is currently no widely accepted theory capable of explaining or, importantly, predicting specific-ion effects universally. The concept of an underlying series that can be used to order the magnitude of effects for a subset of ions has recently gained wider acceptance, with myriad experiments indicating other ions can alter their position in this series. Series reversal is even possible depending on the solvent. While most experimental and theoretical studies to-date have focused on single electrolytes in a pure solvent, real systems that exhibit specific-ion effects are often more complex. In this presentation we will discuss recent research on two complex systems; thermoresponsive polymer brushes in aqueous solutions of mixed electrolytes and specific-ion effects on multi-responsive polymer brushes.

Electrolytes can act to either "salt-in" (stabilise) or "salt-out" (destabilise) a polymer brush immersed in a solvent. We have used brushes of poly(ethylene glycol) methyl ether methacrylate (POEGMA) copolymer to study the effect of mixtures of two salting-in, two salting-out and a salting-in plus salting-out electrolyte. The impact of two salts of the same type is concentration dependent, where the net effect can be cooperative or antagonistic. For mixtures of different salt type the overall behaviour is more temperature sensitive, with the salting-out ion dominating at low temperature (below the polymer lower critical solution temperature) and the salting-in ion dominating at high temperature.

A poly(2-(2-methoxyethoxy) ethyl methacrylate-*co*-2-(diethylamino)ethyl methacrylate) [P(MEO_2MA-*co*-DEA)] copolymer brush of 90:10 mol% composition was used to study specific-ion effects on a multi-responsive brush; PMEO_2MA is thermoresponsive, PDEA pH responsive.[3] Here the effect of a particular salt was pH dependent; for example, KCl salted-in the brush at pH 4 but acted to destabilise the brush at pH 9. In this case, the order of the specific-ion effect could be reversed by changing pH and thus the charge state of the PDEA.

[1] T.J. Murdoch; B.A. Humphreys; E.C. Johnson; G.B. Webber; E.J. Wanless, *J. Colloid Interface Sci.*, **2018,** 526, 429-450 DOI: 10.1016/j.jcis.2018.04.086.
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KEYNOTE: Size- and Shape-controlled Liquid Phase Synthesis of Inorganic Nanoparticles and Application to Organic-inorganic Hybrid Materials

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Recent remarkable progress in inorganic nanoparticles (NPs) synthesis enables us to obtain various types of monodisperse NPs with uniform size and shape.[1]-[6] Thus, new breakthroughs could be expected in NP-based novel functional materials. Especially, NP-based periodic structure formation has attracted a considerable attention in material science.[7]-[8] Among organic soft materials, liquid-crystalline (LC) organic dendron is one of the most representatives to form self-organized periodical structures. Thus, we focused our attention on introduction of such self-organization ability into inorganic NPs. As dendrons, we synthesized phenetyl ether-type dendrons with an amino-group at the apex. These dendrons themselves show thermotropic LC phases. The dendrons are attached as the outer corona, through amidation, to the carboxylic groups at the surface of the inner aliphatic corona encapsulating the NP. CO_2H-modified monodisperse Au, CdS, and Fe_3O_4 NPs were synthesized for the purpose of the study. The dendron-modified gold NP showed an LC hexagonal columnar phase at 130 °C and formed a simple cubic (SC) LC phase at 150 °C. The result indicated that the dendron-modified gold NPs can be regarded as organic-inorganic hybrid dendrimers with thermotropic LC behaviour.[9]-[10] The dendron-modified CdS NPs also formed a thermotropic cubic LC phase with a novel, low-symmetry structure, space group $P2_13$ by annealing at 150 °C for 15 h. In contrast, unannealed dendron-modified CdS NPs formed an amorphous structure. In such a state, it showed strong photoluminescence (PL) when UV irradiated at 365 nm. However, PL was quenched when the dendron-modified CdS NPs formed the cubic phase. Such PL quenching behavior was totally reversible, and appears to be associated with the periodic structure.[11] Such unusual PL behavior might be a powerful tool to develop future functional devices.

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Ultrasonically Activation of Colloidal Gold Nanoclusters Toward Sono-Catalysis

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Thiolate-protected Au NCs (Au_nSR_m), where *n* and *m* denote the numbers of gold atoms and thiolate ligands (SR), have ultrasmall sizes (typically less than 2 nm in the core) and contain up to hundreds of gold atoms. Au NCs have discrete energy levels because of the quantum size effect, which is significantly different from the larger plasmonic Au nanoparticles (>3 nm) with continuous energy levels. Our groups have focused on photo-functionality of Au NCs, such as photoluminescence, photocatalytic activity, and photosensitizing ability [1-5]. Apart from light, ultrasound has several advantages, such as operational simplicity, safety, and being environmentally benign. Recently, ultrasound excited nanomaterials (e.g., TiO_2) have been drawing intense research because of the potential applications such as ultrasound-activated catalysis (sonocatalysis), ultrasound-activated sensitizers (sonodynamic therapy), and ultrasound-induced hydrogen production from water (Sono-Hydro-Gen process)[6,7]. However, the ultrasound-activated mechanism remains unresolved, and there is no evidence on ultrasound activation of nanomaterials. As a result, the strategy for high-efficient sono-catalytic nanomaterials becomes obscure.

Herein, we demonstrate the ultrasound (1 MHz) can excite thiolate-protected Au NCs by observation of sonoluminescence and excited oxygen (i.e., singlet oxygen) from Au NCs[8]. The acoustic wavelength of ultrasound in water ranges from about 75 to 0.15 mm for a frequency between 20 kHz and 10 MHz. As this wavelength is significantly above the dimensions of Au NCs, the direct coupling of the ultrasonic acoustic field with the Au NCs is unlikely. On the other hand, microbubble cavitation (the growth and rapid collapse of microbubbles) occurs in water under ultrasonication to emit light (sonoluminescence). It is proposed that the sonoluminescence from the water could excite Au NCs when the sonoluminescence spectrum overlaps with the absorption spectrum of the Au NCs, as like resonance energy transfer process.

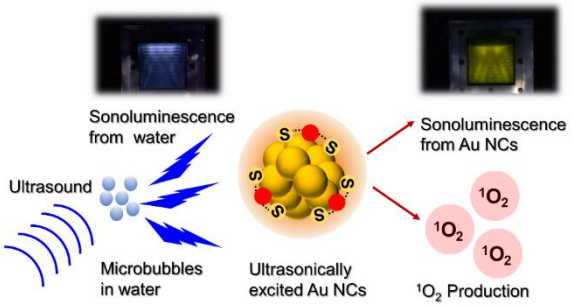

Fig. Ultrasonic activation of Au NCs to produce $^1O_2$ and sonoluminescence

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Change in the Inter-Surface Forces Between Two Charged Surfaces Containing Salts by the Presence of a Liquid Flow

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The forces acting between particles in an aqueous solution determine whether those particles will disperse or aggregate. The ability to control these forces allows the physical properties of these systems to be controlled and therefore their applications to be improved. In real systems of particles in aqueous solutions, the liquid often contains a flow. The effect of a flow on the forces acting in such systems, however, is still not clear.

In this study, we used a combined Atomic Force Microscope-peristaltic pump system to determine the effect of a flow in aqueous salt (NaCl or $MgCl_2 \cdot 6H_2O$) solutions between a negatively charged silica particle and a negatively charged silicon wafer on the forces acting in the system [1]. A flow decreased the inter-surface repulsive forces, if the water contained ions that showed specificity to the surfaces, i.e., Na^+ or Mg^{2+} ions. The difference in the repulsive forces measured in the absence and presence of a liquid flow became greater, when the concentration of these ions were increased by increasing the bulk salt concentration. Fitting of the force curves with the DLVO theory showed that an increased flow rate caused the surface potentials and Debye lengths of the systems to decrease. The difference between the values measured in the absence and presence of a liquid flow became larger for higher bulk salt concentrations. The Debye length decrease was explained by a shrinkage of the electrical double layer due to (1) the deformation of the electric double layer by the liquid flow, and (2) an increase in the number of ions near the charged surfaces and the subsequent electrostatic screening increase, due to ions being introduced into the electrical double layer by the flowing liquid. The decreased surface potential was explained by (1) the increased electrostatic screening resulting from the decreased electrical double layer thickness, and (2) an increase in the number of specifically adsorbed ions to the silica or silicon wafer surfaces.

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“Soft tip” Atomic Force Microscopy in Probing the Interactions of Different Oil Components with Calcite Surface

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The interactions of oil with rocks are complex, which involve van der Waals, electrostatic, hydration, cation-π, steric interactions, etc. These interactions have been the forefront of diverse fields of chemistry, biology, and material science, especially they control the adsorption and transport of oil molecules on rock surface in petroleum industry. [1] [2] On the other hand, in recent years, there has been an emerging interest on the impacts of ionic chemistry on calcite-oil interactions. Scholars have found that Ca^{2+}, Mg^{2+}, and SO_4^{2-} ions changed the binding strength of oil molecules on calcite surface by altering the surface properties, e.g., lattice structure, charges, and wettability of calcite surface. [3] [4] A deep understanding of calcite-ion-oil interactions help the oil industry smartly develop the oil reserves. The force spectroscopy technique that measures the attractive and repulsive forces between oil droplet and solid surface are crucial in developing this understanding.

We studied the ionic effects on calcite-oil interactions by immobilizing a droplet of mineral oil or crude oil to a tipless cantilever and used this "soft tip" to probe the calcite surface in different salt solutions (NaCl, Na_2SO_4, $CaCl_2$, and $MgCl_2$) with an atomic force microscopy (AFM). It was observed that for mineral oil that contains only hydrocarbons and aromatics, the total forces were repulsive in NaCl and $MgCl_2$ solutions but were attractive in Na_2SO_4 and $CaCl_2$ solutions, and the adhesion results followed $CaCl_2$ >Na_2SO_4 >NaCl (reference)>$MgCl_2$. While for crude oil that comprises of hydrocarbons, aromatics, resin, and asphaltenes, it was observed that the total forces were repulsive only in Na_2SO_4 solution but were attractive in NaCl, $CaCl_2$, and $MgCl_2$ solutions, and the adhesion results followed $MgCl_2$ >>NaCl (reference)>Na_2SO_4 >$CaCl_2$. The force results firstly suggested that the interactions of oil with calcite surface could be extremely different due to the differences in oil chemical components, i.e., cation-π interactions [5] and cation bridging interactions [6] for mineral oil and crude oil, respectively. Secondly, these two kinds of interactions responded differently to Ca^{2+}, Mg^{2+}, and SO_4^{2-} ions, that is, the cation-π interactions were mitigated by Mg^{2+} but were enhanced by Ca^{2+} and SO_4^{2-}, whereas the cation bridging interactions were mitigated by Ca^{2+} and SO_4^{2-} but were enhanced by Mg^{2+}.

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Hydration Layer Structure of Biofouling Resistant Nanoparticles

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Hydrophilic surface chemistries can strongly bind water to produce surfaces that are highly resistant to protein adsorption and fouling. The interfacial bound water and its unique properties have intrigued researchers for decades yet the relationship between the water three-dimensional structure and function in antifouling coatings remains elusive. Here, we use hydrophilic, epoxy organosilane modified silica nanoparticles to demonstrate cheap, robust and practically applied coatings that we discover have broad-ranging, ultra-low antifouling properties when challenged by various proteins, bacteria and fungal spores. To understand their remarkable antifouling properties, Frequency Modulation-Atomic Force Microscopy is used to directly observe the interfacial water structure on single nanoparticles at sub-atomic resolution (Figure 1), which we validate using all-atom MD simulations that strikingly predict similar structures of water layers on the original and ultra-low fouling surfaces [1]. Unprecedented convergence of experimental and modelling data reveal that suitably spaced, flexible chains with hydrophilic groups interact with water molecules to produce a confluent, *quasi-stable* layer, consisting of dynamic interfacial water, provides an effective basis for antifouling performance of ultrathin, hydrophilic surface chemistries

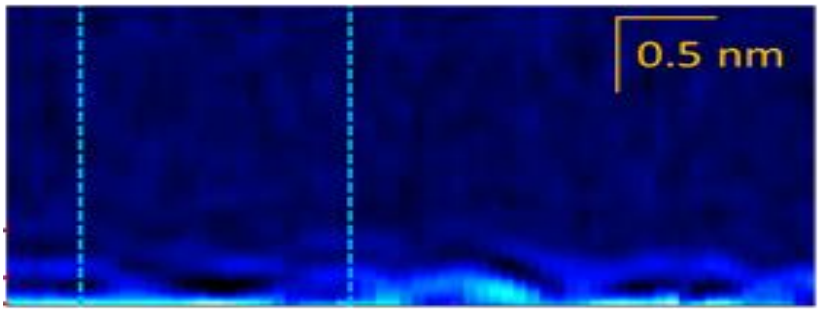

Figure 1: 3D FM-AFM cross-sectional image of interfacial water structure on silica nanoparticle.

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KEYNOTE: Solvent properties of protic ionic liquid-water mixtures, and their application to biological molecules

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Protic ionic liquids (PILs) are cost efficient "designer" solvents which can be tailored to have properties suitable for a broad range of applications. PILs are also being combined with molecular solvents to enable more control over the solvent environment, driven by a need to reduce their cost and viscosity. However, there are relatively few structure-property studies which look at these more complex mixtures. We have explored the solvation properties of common PIL-molecular solvents using various techniques,[1] including Kamlet-Aboud-Taft parameters determined from solvatochromic dyes, and MD simulations.[2-3] These have identified many interesting solvent properties of these solutions and gives insight into their interactions with solutes.

In the second part of this presentation I will discuss how we are using our understanding of PIL-water solvent properties to design and characterise solvents for biological molecules. In particular, we are targeting being able to control protein solubility and stability, which are critical for applications in bioprocessing, biocatalysis, protein crystallography and cryopreservation. We have explored lysozyme as a model protein in various PIL-water systems, predominantly using spectroscopic techniques and small angle x-ray scattering (SAXS).[4-5] From this we have been able to identify which PILs are more biocompatible, and to identify specific conformational changes of lysozyme due to the presence of PILs. More recently, protein crystallography has been used to identify specific binding sites of the PIL ions and water to lysozyme.

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Delayed diffusion and unique phase behaviour in polymer crowding micro-emulsions

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Molecular behaviour in milliliter-scale bulk solutions and small picoliter-scale microemulsions is often different[1–3]. This difference is attributed to as “size effect”. Although the factors of the size effect are considered to be the small volume and the membrane interface, few attempts have been made to separate and quantify these factors. In this study, we evaluated the size effect by measuring the molecular diffusion in highly concentrated polymer crowding solutions confined in micro-emulsions. First, molecular diffusion in various sized spherical oil-in-water micro-emulsions of polymer crowding solution covered by a lipid monolayer[4] was investigated by fluorescence correlation spectroscopy (FCS) (Fig.1). The results show that the molecular diffusion of the globular protein bovine serum albumin (BSA) and the linear polymer poly(ethylene glycol) (PEG) slowed down as the radius R of the spherical emulsion decreased below 20 μm at polymer concentrations are less than one molecule of the inter-protein distance and above the overlap concentration c^*, respectively. Furthermore, the molecular diffusion in the disk-shaped emulsions was measured in the same way and the diffusion coefficient D was found to be delayed than that in the spherical emulsions under the same volume conditions. On the other hand, the diffusion coefficient D was independent of the distance from the membrane interface. These results suggest that the slow molecular diffusion is not due to the distance from the membrane nor the small volume effect but is due to the increase in the membrane area/volume ratio due to micrometric confinement. In addition to these results, we will illustrate the liquid-liquid phase separation (LLPS) in such micro-emulsions and discuss the origin of the size effect caused synergistically by polymer crowding and confinement in microscopic spaces[5,6].

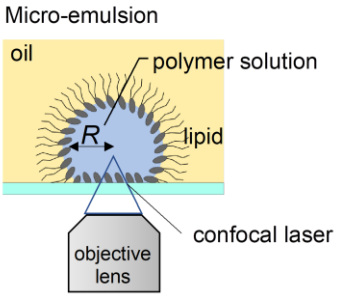

Fig.1 FCS measurement in a micro-emulsion.

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The Adsorption of Glucose Oxidase on Mesoporous Aluminium Oxide Films

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Protein adsorption at an inorganic nanoporous material is a fundamental process in the chromatographic separation of proteins and synthesis of an artificial biocatalytic system.[1-3] The nanoporous material possesses both an outer surface and internal pore surfaces, and both can be regarded as adsorption sites for protein molecules. The distribution of protein molecules at both adsorption sites affects the chromatographic separation and performance of the biocatalytic system. The purpose of this study is to clarify whether negatively-charged glucose oxidase (GOD) can penetrate into pores in mesoporous aluminum oxide (MAO) films. The MAO film used in this study was prepared by a surfactant-templated method,[4] and contained ordered pores with diameter of *ca*. 10 nm, much smaller than that of a conventional nanoporous aluminum oxide.

The MAO film was deposited on a microscope cover glass slip for *ex situ* structural characterization of the by X-ray reflectometry (XRR), neutron reflectometry (NR), and scanning electron microscopy. The same deposition 'recipe' was then used for films deposited on thick silicon wafers for *in situ* experiments, where the GOD containing solution is in contact with the MAO film, using NR. The NR profiles before/after adsorption GOD were continuously measured in a flow cell with varying H_2O/D_2O ratios applied to 'highlight' different aspects of the system. The results indicated almost exclusive outer surface adsorption with the negatively charged GOD molecules unable to enter the positively charged channels.

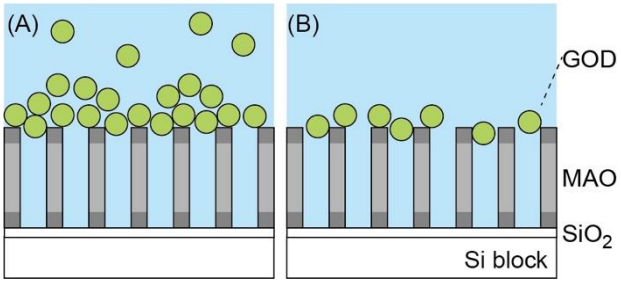

Fig. 1 Schematic illustrations of the distribution of GOD molecules at the MAO film in 10 wt% GOD solution and in water after removal of excess amount of GOD adsorbed.

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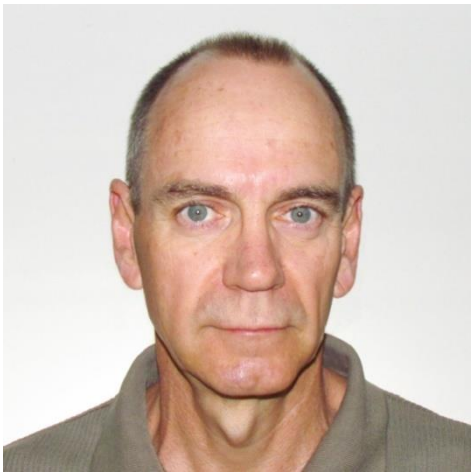

Selective Ion Transport across a Lipid Bilayer in a Protic Ionic Liquid

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Spontaneous bilayer formation and amphiphilic self-assembly in ionic liquids is well established.[1, 2] However, to date, this has been a largely uncontrolled process. Using a *tethaPod*™ system (Figure 1), we demonstrate the formation of a tethered lipid bilayer in a purely ionic liquid environment.

Furthermore, using electrical impedance spectroscopy, we show for the first time the function of a membrane transport protein in a pure ionic liquid. The transporter in question; valinomycin, not only retains full functionality in an ionic liquid, it also retains its selectivity for potassium over sodium ions.

This paves the way for the development of new, non-aqueous systems such as microreactors, and biosensors. Furthermore, stability in an ionic liquid could be used for long-term storage of tethered membranes and related devices, or use at temperatures not accessible with an aqueous environment.

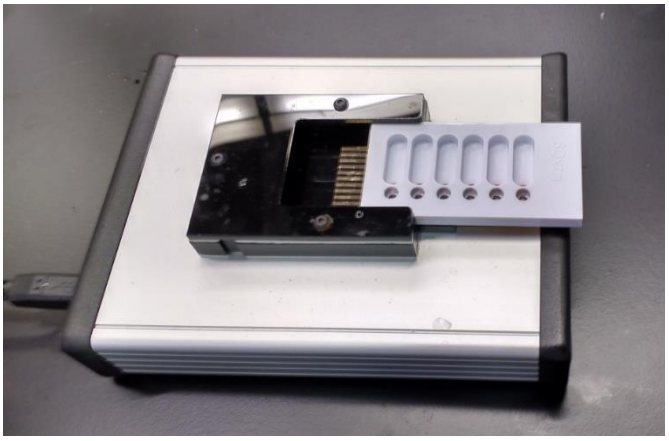

Figure 1. Photograph of a *tethaPod*™ system from *SDx Tethered Membranes Pty Ltd* (Sydney, Australia).

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Conformational Changes of Protein with Ionic Liquids: A Multi-technique Approach

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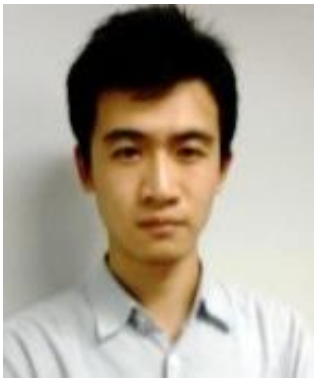
Ionic liquids (ILs) are liquids that are comprised entirely of ions. They are designer solvents which can help stabilise some proteins. However, it is challenging to understand protein structure in ILs. In this talk, we show a wide range of ILs used for lysozyme and GFP as model proteins. We introduce a multi-technique approach including spectroscopies, small angle x-ray scattering and crystallography to explore the conformational changes of proteins and such approaches can improve our understanding on structure-property relationships for future solvent design for proteins.



Human neuropeptide substance P self-assembles into semi-flexible nanotubes that can be manipulated for nanotechnology

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Tailoring peptide self-assembled nanostructures for specific applications has been reported in diverse areas, from nanoelectronics to water purification, to drug delivery [1]. Unlike proteins, short peptides are accessible synthetically, with sequences that can be easily modified. However, the majority of peptide nanostructures reported to date form polymorphic, irreversible, insoluble "amyloid-like" nanofibrils [2] . Expanding the space of peptide nanostructure morphologies and properties is hence desirable. Specifically, few sequences were reported to form nanotubes, and these only include unnatural peptides, chimeric molecules or truncated natural sequences [3]. Other than a few exceptions [4], these sequences formed poly-disperse assemblies. Here we report, for the first time, the intrinsic, reversible self-assembly of the human neuropeptide Substance P into high aspect-ratio, semi-flexible, monodisperse nanotubes that can associate into tapes or films with nematic properties. We show that substance P nanotubes are versatile materials for nanotechnology, as they can be precipitated or mineralised while conserving their core-shell morphology and dimensions. Our discovery will open new biomimetic approaches for innovative bio-nanotechnology applications, beyond direct relevance for neuropeptide functional assemblies.

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Precise molecular packing from designed surfactants for vesicles and wormlike micelles

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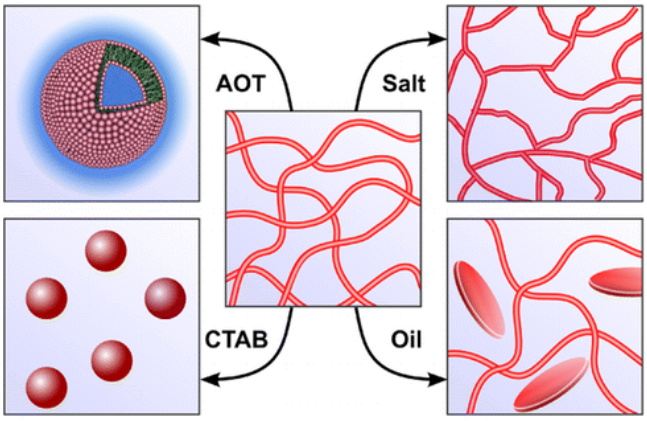
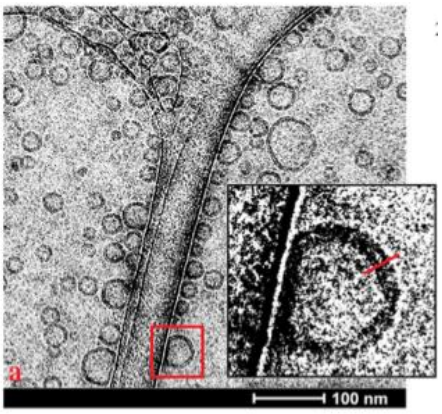
Controlling molecular packing of surfactants results in their diverse bulk aggregation, from small micelles to elongated worms, vesicles and liquid crystalline phases. We explore the utility of two common approaches to achieving desired self-assembly structures: (a) additives that locate between surfactant molecules, adjusting their interactions and packing; and (b) using molecular design to achieve specific packing arrangements. The former is more easily achieved, but is naturally more sensitive to other components in the system and suffers limitations for formulation design. Nonetheless, we use betaines as a model class of surfactant that favour formation of wormlike micelles, and demonstrate that by addition of various different components, self-assembly can be directed towards formation of nano-discs, microemulsion droplets, vesicles or networked worm structures [1].





Conversely, through molecular design of highly tailored carbohydrate surfactants, we demonstrate that similar control over self-assembly can be achieved in a much more robust fashion, rendering molecules that form non-ionic worms and vesicles [2]. This class of molecules shows not only diverse self-assembly in dilute micellar systems, but also forms a rich panel of liquid crystal structures at higher concentrations. Moreover, these molecules can be synthesized from sustainably derived building blocks, giving them potential as 'green' molecules for industrial formulation. The design rules that we develop may aid formulation of future products, as well as contributing to our fundamental understanding of how surfactant architecture and interactions control self-assembly in bulk solution.

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Aggregation of highly negatively charged vesicles

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The adhesion of lipid bilayers is a critical process in membrane trafficking, multicellularity, creation of tissues, and for fusion-based processes. However, while oppositely-charged lipid bilayer membranes adhere and fuse readily, like-charged membranes typically only adhere in the presence of divalent cations, fusogenic peptides, or depletants.

We use a recently reported method to generate solutions of giant vesicles that can relieve membrane tension by elastic and extensive means [1]. We find that having a highly dynamic bilayer system enables like-charged lipid vesicles to form assemblies ranging from pairs to three-dimensional foams (shown below), without the addition of fusogens or adhesives.

Further analysis of the shape of the hemifused pairs reveals that they appear much like bubbles. Indeed, with only monovalent salts and fatty acids in solution, our system draws strong parallels with bubbles. Force balances along the junction between vesicles shows that the hemifused pairs are at equilibrium.

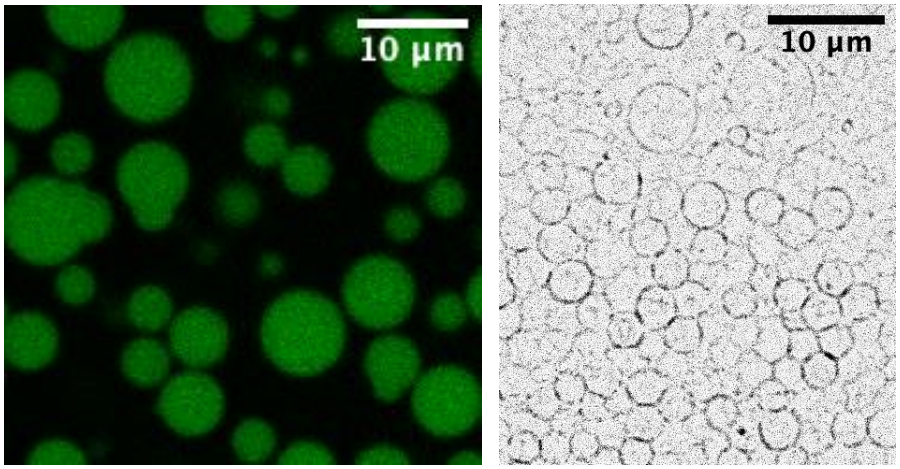

Figure 1 Microscopy of vesicles reveal hemifused pairs (left) and foams (right)

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Common Black Film Stability and Synergetic Adsorption in Ionic−Nonionic Mixed Surfactant Systems

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In dry foams, the interaction between surfactant-adsorbed films generates disjoining pressure as the drainage of the foam film proceeds. Common black films (CBFs), typically thinner than 100 nm, are stabilized by the long-range electrostatic and van der Waals forces. The classical DLVO theory explains the physical property of foam films in this region well. Foams stabilized by surfactant mixtures are common in technical applications. However, the most studies in the literature were focused on foam films stabilized by a single surfactant, and only a few reports are available for understanding foam films stabilized by surfactant mixtures.

As reported in many literatures, the thickness of CBFs is determined mostly by bulk electrolyte concentration through the shielding of the surface charge of the adsorbed films. Therefore, in the absence of inorganic salt, the increase in the nonionic surfactant ratio in the bulk solution simply leads to film thickening due to the reduction of the ionic surfactant (electrolyte) concentration in the solution. However, in real foam systems, it is widely accepted that mixing of nonionic and ionic surfactants in the adsorbed film has more positive influence on the foam film stabilization. For example, the synergism in the mixed adsorbed films leads to more elastic and kinetically stable foams due to slower coarsening, drainage, or coalescence. Furthermore, the synergetic adsorption of surfactants gives rise to better foaming by the reduction of surface tension as compared to the foaming of solutions of the individual surfactants.

In this talk, we will propose an important static aspect of the synergetic adsorption, which has a greatly influence on the foam film stability, by using phase diagram of the adsorbed film (PDA). For this purpose, the thickness and stability of CBFs in two ionic–nonionic mixed surfactant systems were compared. When the attraction between the two surfactants in the adsorbed film was relatively weak, the foam film collapsed as the bulk composition of the nonionic surfactant, $\hat{X}_2$, became 0.09 as observed for cetyltrimethylammonium chloride (CTAC) – *n*-dodecyl-β-D-maltoside (β-$C_{12}G_2$) system. However, the CBF of lithium perfluorooctanesulfonate (LiFOS) – tetraethyleneglycolmonooctyl ether (C_8E_4) system was stable up to $\hat{X}_2$ > 0.96 due to the strong synergism between the ionic and nonionic surfactants maintaining a high ionic surfactant composition in the adsorbed film, thus leading to a high surface charge density that stabilize the CBF.

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

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The work I hope to present at Australia Japan Colloids Symposium 2020 is a comprehensive study of 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. Smallangle 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.

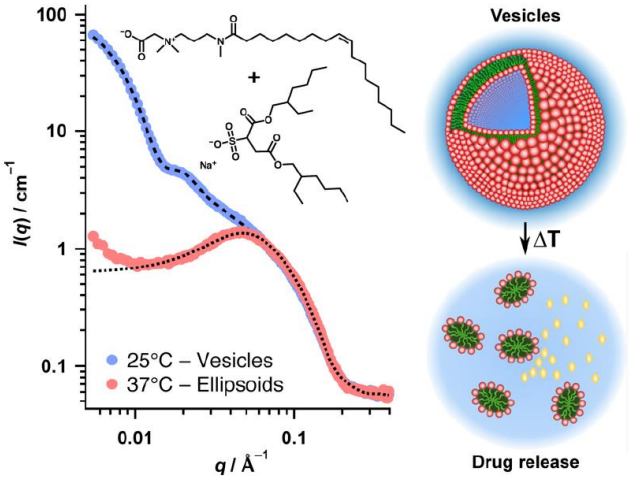

Fig 1: Small-angle neutron scattering data from a 10 and 20 mM aqueous mixture of AOT and OAPB (structures inset) and the accompanying structural transition from vesicles to ellipsoids upon heating from 25 to 37°C respectively.

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Friday 18th September

A Model System for Experimental Studies of Gas Generation in Compressible Sediments

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Anaerobic lagoons and ponds are used in many industries to biologically digest large volumes of wastewater, with biogas a useful by-product. However, lagoon design is inadequate due to limited capacity to model the complex multi-phase hydrodynamic and biological behaviour. A part of improving our understanding of lagoons lies with the transport of gas through the system. Gas is generated in the sludge layer at the bottom of the lagoon. The sludge has a yield stress such that bubble growth and movement is limited[1] (see Figure (a)). Instead, the gas diffuses through the liquid in the sludge. Near the top of the sludge, bubbles can grow such that buoyancy overcomes the bed strength and weight, and gas is released. This can lift and fluidise solids from the bed. Solids and gas can form a scum or foam layer at the top of the lagoon that can inhibit biogas capture and damage the lagoon covers. Understanding the phenomena present in such a complex three-phase system requires laboratory experiments with known rates of gas generation and known material properties. Unfortunately, these hard to control in biological systems due to varying rates of degradation and changing material properties with degradation.

This work details a well-controlled experimental model system enabling detailed investigations of the mechanisms for gas transport in compressible sediments. Coagulated micronised calcium carbonate is used as an inert compressible sediment such that the solids concentration and bed strength vary as a function of depth. This is doped with 1% magnesium hydroxide, which is oxidised by hydrogen peroxide dosed in the liquid to create oxygen gas (see Figure (b)). This system allows systematic coverage of the parameter space, including sedimentation rate, gas generation rate, sediment depth (and therefore concentration and compressibility) and bed area. These can be varied simply by changing the initial concentration (and height) of the suspension, the initial concentration of reactant and the sample area. This simple model system identifies a rich tapestry of phenomena, ranging from bubbles trapped in thick sediments, single bubbles escaping thin sediments, local collapse of non-cohesive sediments, bulk density inversion due to sufficient gas production, bed fluidisation, particle flotation due to both bubble adhesion and density inversion, scum formation and foaming.

Figure: (a) X-ray tomograph of large bubbles within a thick magnesium hydroxide sediment after 6 h of gas generation[1]; (b) Gas bubbles in settled calcium carbonate spiked with magnesium hydroxide and peroxide, showing trapped oxygen bubbles in thick sediment and escaping bubbles in thin sediment (image courtesy Bharath Krishna, Masters Research Project student, The University of Melbourne); (c) Presenting Author

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Microhydrodynamics of Arbitary-Shape Passive and Active Particles in Linear Flow: Daily Life at the Small Scale

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Passive and active small particles present in both natural and technological flow-driven particulate processes are typically nonspherical in shape. The physical properties of such processes depend ultimately on the particle dynamics, which in turn is greatly influenced by particle shape through hydrodynamic interactions (HI). We have recently extended to the presence of linear flow two complementary approaches for Stokesian Dynamics (SD) simulations of arbitrary shape particles in quiescent fluid, namely the constraint SD approach and the rigid body SD approach . Either approach models arbitrary shape particles as groups of primary spheres whose HI are furnished by any desirable mobility algorithm for spheres , but while the constraint approach is practically effective for any size flexible particles and rigid particles with only small numbers of primary spheres, the rigid body approach is useful for any size rigid particles. We show results from SD simulations of some interesting systems involving passive and active nonspherical particles in various settings, performed with both foregoing approaches. They highlight the surprising and often counterintuitive aspects of physics at the microscale.

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Elastic plastic fracture mechanics investigation of toughness of wet colloidal particulate materials: influence of saturation

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Previous use of linear elastic fracture mechanics to estimate toughness of wet particulate materials underestimates the toughness because it does not account for plastic deformation as a dissipation mechanism. Elastic plastic fracture mechanics using the J-integral approach was used for the first time to measure the fracture toughness (J_{IC}) of wet micron sized alumina powder bodies as a function of saturation. The samples were prepared by slip casting. The saturation was controlled by treatment in a humidity chamber. The elastic modulus (E) and the energy dissipated by plastic flow (A_{pl}) were measured in uniaxial compression. The critical stress intensity factor (K_{IC}) was measured using a diametral compression sample with a flaw of known size. The fracture toughness (J_{IC}) was calculated from these measured quantities and the geometry of the specimen. Elastic plastic fracture mechanics was used for the first time to quantitively account for plastic deformation of wet particulate materials. [1] Plastic deformation was responsible for the majority of energy dissipated during the fracture of wet colloidal particulate materials. The linear elastic fracture mechanics approach previously used accounted for less than 1% of the total energy dissipated in fracture. Plastic deformation around the crack tip increases with saturation of the particulate body. Toughness (J_{IC}) was found to increase with increasing saturation due to plastic deformation that increased with saturation level. The improved understanding of toughness as a function of saturation will aid in providing quantitative analysis of cracking in drying colloidal films and bodies.

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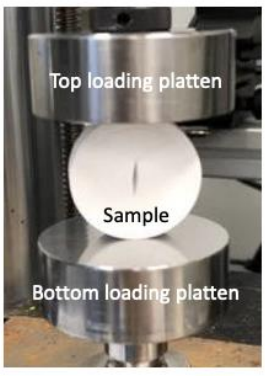

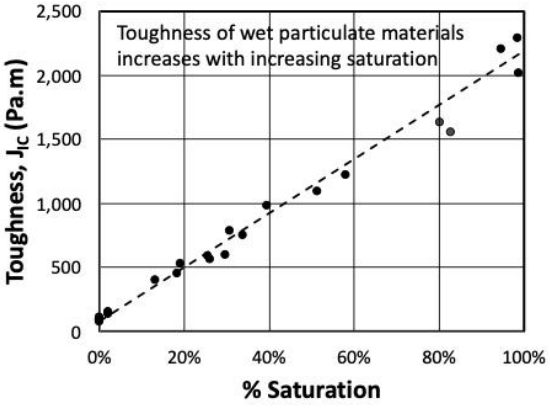

Modelling of interaction force between particles and simulation of breakage behaviour

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Grinding is one of the most effective operations to produce fine particles, and the significance of grinding grows with each passing year due to the development of nanotechnology. Grinding has been used in many industry fields, for example, foods, cosmetics, medicines, mining and so on. However, in general, the efficiency of grinding is quite low because the mechanism of grinding and breakage have not elucidated. For efficient grinding, it is necessary to understand the breakage mechanism of the materials. In this work, it has been attempted to analyse the breakage phenomena using computer simulation. ADEM (Advanced Distinct Element Method) [1] has been developed and it can represent both non-spherical particles motion and particle breakage behaviour. While conventional DEM only represent discrete bodies, the ADEM enables the representation of a continuum by considering the interaction force between primary particles using joint spring. The interaction force by the joint spring needs to be determined so that the macroscopic deformation and breakage behaviour is consistent with the actual behaviour, which is dependent on the size and configuration of the primary particles used. If the relationship between known forces such as van der Waals forces and the interaction forces of the joint springs can be clarified, it will be possible to predict the breakage behaviour of particles theoretically. Here, we attempted to reproduce the compressive deformation and breakage behaviour of brittle and plastic materials and investigated the modelling of inter particle forces.

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Thixotropic Clay Gels: Microstructure and Role of Colloidal Forces

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Gels of NaMt and hectorite displayed prominent time-dependent or ageing behaviour. This behaviour may last for days or even months. The microstructure showed flexible nano-sized platelets interacting attractively to form the network junctions and repulsively opening up the structure (Fig 1a,b,c,d). These junctions are formed by heterogeneous charged and vdW attraction in the jagged and curled edges with faces of nearest platelets. The EDL repulsive force governs the development of the 3-D structure as it moves to attain a minimum free energy state and this force is responsible for the time-dependent behaviour. At high ionic strength, 0.3M KCl, the bentonite gels became time independent behaviour (Fig. 1e) - vdW dominates the interaction force. Leong model described the ageing behaviour well for the low salt the gel (Fig 1e). For the nanodiscotic Laponite gels, the 3-D structure is formed from sheets (Fig. 1d) made from primarily disks interacting in the overlapping face-face interaction at the edge. The sheet charge property is the same as the disk; face negative and edge positive.

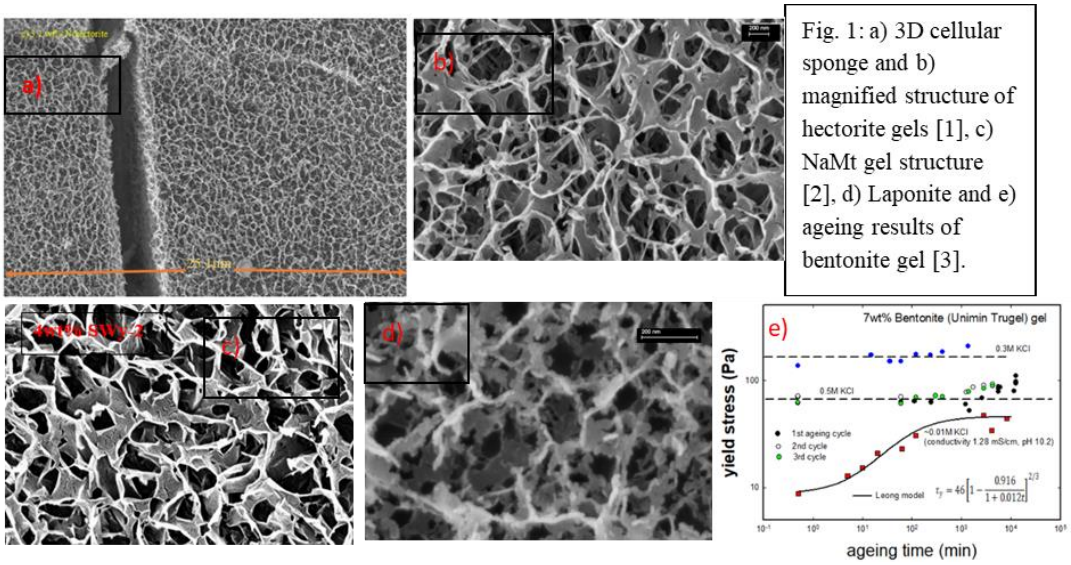

Fig. 1: a) 3D cellular sponge and b) magnified structure of hectorite gels [1], c) NaMt gel structure [2], d) Laponite and e) ageing results of bentonite gel [3].

With this microstructure knowledge, a new model may be needed to explain behaviour like layer stacking and large dried platelets, low permeability, intercalation property. Overlapping face-face attraction at the edge enlarged the platelet size in the dried powder. Capillary force and layered cations mediating face-face repulsion allowed layer stacking to occur upon drying. In the wet state, the open cellular structure can accommodate any additive molecules in the relatively large pore spaces negating their need for interaction with the platelet surfaces prior to platelet stacking and intercalation upon drying.The stacking tendency of the platelets intercalates the trapped molecules in-between. The complete interpenetration of the structure with nano-pores and channels (Fig. 1a) trapped the water molecules by strong capillary forces imparting low permeability to the clay gels.

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KEYNOTE: Evaluation of Non-DLVO Forces between Surfaces in Liquids by Atomic Force Microscopy

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Understanding of the interaction forces between surfaces in liquids is fundamental in many fields of science and engineering because the stability of colloidal dispersions (including two phase fluids such as emulsions and bubbles) is generally dominated by the forces between two opposing particles. The theoretical basis for analyzing surface forces between charged particles in aqueous solutions was provided by Derjaguin-Landau-Verwey-Overbeek (DLVO) theory[1] in the 1940s and is still used widely today. However, developments in direct force measurements with the surface force apparatus (SFA) and the atomic force microscope (AFM) have extended our understanding of the forces that lie outside of the DLVO paradigm, often called non-DLVO forces, which have also significant effect on the stability of colloidal dispersions.

This talk will cover our latest studies on the non-DLVO forces using AFM. The hydrophobic attraction has been the subject of numerous studies using SFA and AFM since it was measured directly for the first time[2] and had been known to be much greater in range than the van der Waals (vdW) force. Recent studies, however, on the "pure" hydrophobic attraction is not unusually long-ranged, generally having a range of less than 10–15 nm.[3] In this talk, I will present the recent results of investigations on the characteristics of this "short-range" hydrophobic attraction, such as the measurements of between dissimilar surfaces[4] and effect of surfactants.

Interaction forces in organic solvents is also significant for handling various kinds of colloidal dispersions in organic solvents used in industries including paints, cosmetics, and electronics. However, the forces in non-aqueous solvents have been much less understood than those in aqueous solutions, because the DLVO theory often cannot describe the forces in non-aqueous solvents.

Recently, we have investigated in different organic solvents the dispersibility of the silica particles modified with silane coupling reagents with various terminal groups, along with the measurements of the interactions between the modified particle and substrate by AFM. I will present the ideas on how the forces in the organic solvents dominate the dispersibility of the particles.

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Is the origin of the negative surface charge of hydrophobic water surface OH ion adsorption?

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The electrification of hydrophobic surfaces is an intensely debated subject in physical chemistry. We theoretically study the zeta potential of hydrophobic surfaces for varying pH and salt concentration by solving the Poisson-Boltzmann and Stokes equations with individual ionic adsorption affinities [1]. Using the ionic surface affinities extracted from the surface tension of the air-electrolyte interface, we first show that the adsorption and repulsion of small inorganic ions such as H_3O^+, OH^-, HCO_3^-, and CO_3^{2-} are irrelevant for the zeta potential observed in experiments because the surface affinities of these ions are too small. Even if we take hydrodynamic slip into account, the characteristic dependence of the zeta potential on pH and salt concentration cannot be reproduced. To explain the zeta potential of hydrophobic surfaces, instead, we introduce a minute amount of impurities in the water and consider their acidic and basic reactions with water. We find good agreement between our calculations and the reported experimental data of Teflon surfaces. Our theory suggests that the impurities are weak acids ($pK_a = 7$) and weak bases ($pK_b = 12$) at a concentration of the order of 10^{-7} M.

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Soft-Hard Janus Nanoparticles for Polymer Encapsulation

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We demonstrate a scalable continuous feed method for the synthesis of polymer Janus nanoparticles with different composition and aspect ratios between lobes, including with one hard cross-linked polystyrene lobe and one soft film-forming poly(methyl methacrylate-co-butyl acrylate) (P(MMA-co-BA)) lobe, through emulsion polymerization. The Janus nanoparticles with the soft P(MMA-co-BA) lobes can spontaneously self-assemble around solid particles in an aqueous phase, forming a thin polymer film. The process is achieved at room temperature and under mild conditions. The Janus nanoparticles show excellent encapsulation ability for a variety of solid particles, such as organic and inorganic pigments, carbon nanotubes and calcite, without requiring polymerization. The role of the surface charge of the particles, wettability, and morphology are investigated to explain fundamental questions on the encapsulation mechanism.

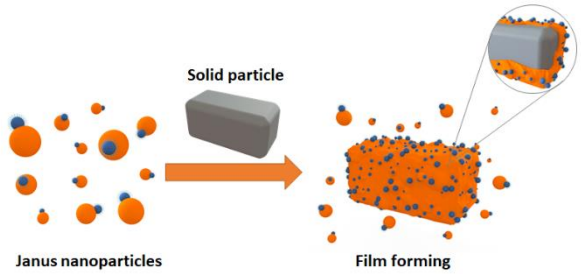

Figure 1. Schematic of encapsulation of particles in aqueous phase by film forming Janus nanoparticles

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Duc Nguyen


Liwen Zhu

LSPR-mediated high-resolution imaging for the study on cell adhesion and spreading

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LSPR-mediated microscopy imaging through the fabrication of 2D sheet from self-assembled gold nanoparticles (AuNPs) has successfully demonstrated a non-scanning high-resolution imaging method for live cell imaging. This nanomaterial-based imaging method can produce image with high axial and lateral resolution even under a regular epifluorescence microscope.[1] In this work, we further applied this technique to monitor the dynamic behaviour of focal adhesion throughout the process of cell adhesion and spreading. It is aimed to provide better understanding on the role on focal adhesion during the cell adhesion dynamics.[2] NIH3T3 fibroblast cell that stably expressing Venus-paxillin is used to image focal adhesion at cell–substrate nanointerfaces under total internal reflection fluorescence (TIRF) microscope. Preliminary result has shown the possibility to track the dynamic movement of paxillin cluster along the fibrillar structure in real-time.

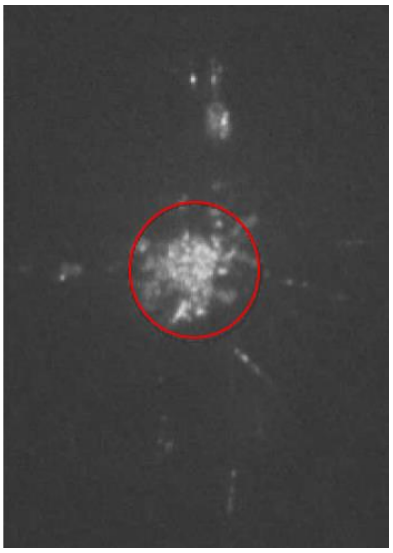

Fig. 1 Early stage of cell-attached nanointerfacial image on AuNP sheet with 20 nm SiO_2 spacer layer under TIRF microscope (incident angle: 75°, exposure time: 500 msec)

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Antibacterial Liquid Metals: Biofilm Treatment via Magnetic Activation

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The rise of antibacterial resistance has made the treatment of biofilm-related diseases an on-going challenge. This situation has made the occurrence of post-operative, biofilm-based infections much more prevalent and increasingly difficult to treat. As such, the quest for next-generation antimicrobial technologies must pivot towards targeted therapies for which pathogenic species cannot develop resistance. Recently, liquid metal (LM) nanoparticles have emerged as a new class of biocompatible nanomaterial that simultaneously possess both metallic and fluidic properties. In essence, the nanoparticles are hard spheres which remain internally fluidic. Importantly, gallium-based LM particles, such as Galinstan (a Gallium-Indium-Tin alloy), can be magnetically functionalised via the incorporation of internalised magnetic iron nanoparticles. In this study, the use of magneto-responsive Galinstan based liquid metal (LM) nanoparticles is assessed as a new-class of antibacterial materials. Importantly, when exposed to an oscillatory magnetic field the LM nanoparticles are physically activated, meaning that they both spin and undulate within the solution. When place in-contact with a bacterial biofilm the movement of the particles is capable of physically inactivating the bacterial cells while disrupting the surrounding dense biofilm matrix. The antibacterial efficacy of the LM nanoparticles was assessed against both Gram-positive, *Staphylococcus aureus*, and Gram-negative, *Pseudomonas aeruginosa* bacterial biofilms.

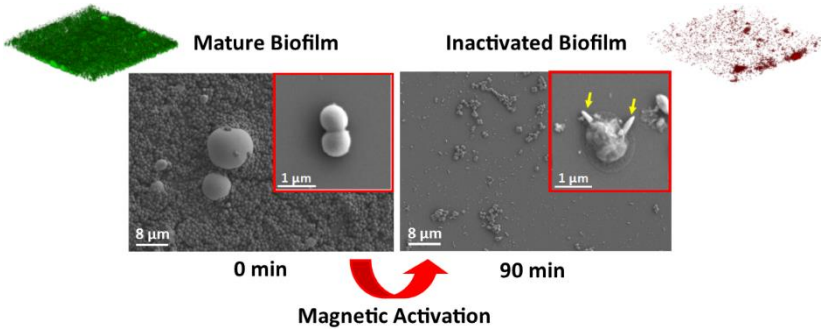

Figure 1. SEM and confocal images of a *S. aureus* biofilm in the presence of Galinstan nanoparticles before and after exposure to a magnetic field.

KEYNOTE: Protein adsorption at the oil-water interface: role of protein self-association

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Many food emulsions are stabilised against drop flocculation and coalescence by thick films of adsorbed milk proteins at the drop surfaces.[1] The major protein in milk serum is *b*-lactoglobulin, a small globular protein. Depending on the pH, temperature, and salt concentration, *b*-lactoglobulin exists in aqueous solutions as monomers, dimers, or higher oligomers.[2] *b*-lactoglobulin diffuses rapidly from the aqueous phase to the oil-water interface. There it unfolds irreversibly to form a viscoelastic layer.[3] The surface charge of *b*-lactoglobulin and the hydrophobicity of the oil phase are known to affect the adsorption process.[4-6] In this talk, I will present results from a study done under conditions designed to tease out the differences in interfacial activity between the monomer and dimer species of *b*-lactoglobulin. We used drop profile tensiometry and interfacial shear rheology to probe the dynamics of the protein adsorption at the oil-water interface from aqueous salt solutions. We explored the influence of the salt concentration and the type of salt present on the elasticity of the protein network. Our key finding is that the formation of the viscoelastic layer is directly linked to the proportion of dimer present in solution. These results suggest simple routes for tailoring the properties of *b*-lactoglobulin networks at immiscible interfaces.

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SANS studies with lipid cubic phase to investigate protein encapsulation and conformation

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Understanding protein-lipid interactions is crucial for evolving food technology, biological and biomedical applications of nanomaterials. Knowledge regarding the effect of the multiple components in the system on the nanostructure, within the context of the application, is needed. Lyotropic liquid crystal design rules were developed and the effect of protein encapsulation on lipid self-assembly materials was extensively studied by us in recent years.[1]

Recently, significant progress in this field was made by developing a small-angle neutron scattering method with perfectly contrast-matched lipid phases in D_2O to study biomolecule encapsulation.[2] In this presentation, I will show how we used this to obtain a protein-eye view of the in meso crystallisation method of integral membrane proteins from the bicontinuous cubic phase over time.[3] Studies to date have, by necessity, focused on structural transitions occurring in the lipid mesophase. This opens up the possibility of studying the structure (and function) of proteins, therapeutic peptides as well as other biomolecules in a lipid environment.

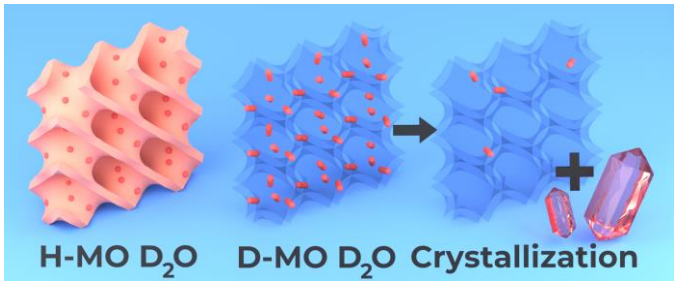

Figure 1. Illustration of a "Protein-Eye View of the in Meso Crystallization Mechanism" [3] using small-angle neutron scattering and deuteration of the cubic phase forming lipid monoolein (MO).

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Hydrophobic Seed Proteins on Starch Granule Surface Enables Gluten-Free Rice Bread Making

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Prevalence of wheat allergy and celiac disease has led to a growing demand for gluten-free foods. Rice flour is considered as a promising substitute for wheat flour, as rice does not contain gluten. Rice flour is made from the ground endosperm of paddy rice seeds, and the endosperm contains many organelles called amyloplasts, which are collections of dozens of crystalline starch grains. When rice flour is dispersed in water, starch grains in the amyloplast spontaneously scatter, and the conditions of the starch grains is thought to determine the characteristics of the rice flour products. An indicator of starch grain quality is the degree of starch damage, which is determined by using enzymes that hydrolyze only damaged starch. On the other hand, storage proteins are accumulated around starch grains, but their properties and relevant reactions are rarely used to evaluate the quality of rice flour.

In baking bread with rice flour, the selection of rice flour with respect to the starch-damage is important. Rice flour of low starch-damage (L) results in a bulky bread comparable to wheat flour bread without the addition of thickening agents. Our previous study has shown that the surface tension of water with L dispersed in it is lower than that of water with high starch-damaged rice flour (H) dispersed in it. Furthermore, L was less water absorbent than H, suggesting that the surface hydrophilicity of starch grains is different[1]. However, it was not clear what the biochemical indicator, the extent of starch damage, means in terms of interfacial chemistry. The formation of surface film at the dispersed aqueous solution/air interface comprising starch and protein was reported previously[2]. In this study, we focused on the surface condition of starch granules and hydrophilicity of the storage proteins.

Although having same chemical compositions, L, which is suitable for bead making and H, which is unsuitable for bread baking were assessed. Organized surface films of dispersed solution of L and H were transferred on polymer coated Cu grids for transmission electron microscopy (TEM) observation. Most of the L starch granules had clearer contours, whereas most of the H starch granules had less clear contours. The same experiment was performed on friction-applied L, and the supernatant of the dispersed aqueous solution became gluey and the contours of the starch granules were blurred in TEM observation. Proteins on the surface of starch granules were eluted and recovered for L and H, and fractions with different hydrophilicity were obtained by simplified stepwise liquid chromatography. Most of the proteins eluted from L were hydrophobic, whereas most of the proteins eluted from H were hydrophilic. Based on these findings, the relationship between the proteins and starch granule surface gelatinization and their effects on the film formation by starch granules will be discussed.

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An ATR FTIR Study of the Interaction of TiO2 Nanoparticle Films with β-Lactoglobulin and Bile salts

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The technique of in situ particle film attenuated total reflection Fourier transform infrared spectroscopy (ATR FTIR) has been used to probe the adsorption and co-adsorption (sequential) of a common food protein (*Beta*-lactoglobulin – BLG) and two representative bile salts (taurocholic acid – TCA, and sodium glycocholate – GCA) onto the surface of titanium dioxide (TiO_2) nanoparticles. Evaluating binding interactions between commonly used (historically now, in some countries) food additives and food components, as well as the body's own digestion chemicals is a critical step in understanding the role of colloid chemical phenomena in digestion and bioavailability. TCA is seen to adsorb significantly to TiO_2 but without any significant ability to be retained when it is not present in the aqueous phase. GCA is seen to also adsorb significantly, and via two distinct binding mechanisms, with one being resistant to removal. BLG, as expected, adsorbs significantly, is irreversibly bound, and has altered conformation when adsorbed at pH 2 (stomach conditions) as opposed to pH 6.5 (small intestine conditions). This altered conformation is not interface dependent, and is mirrored in solution spectra of the BLG. Sequential co-adsorption studies indicate that TCA and GCA adsorb to TiO_2 nanoparticle surfaces and display similar degrees of reversibility and binding in the presence of pre-adsorbed BLG as compared to in the absence of pre-adsorbed BLG.

Mimicking Lipid Self-assembly in Digesting Milks

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Introduction: Milk is nature's emulsion for delivering fats and fat-soluble nutrients to infants and remains a mainstay of the adult diet thereafter for many. The milk fat globules that deliver these nutrients comprise 98% triglycerides with hundred of unique acyl chains esterified onto the glycerol backbones, resulting in thousands of possible unique triglyceride structures.[1] Digestion of the apolar triglycerides into monoglycerides and fatty acids by endogenous lipases breaks down the milk fat globules and allows the absorption of fat-soluble nutrients entrained within them in the intestines.

Methods: Small angle X-ray scattering (SAXS) with *in situ* lipolysis has revealed the spontaneous assembly of the amphiphilic milk fat digestion products into a progression of liquid crystalline structures over time during *in vitro* lipolysis. The milks of different species yield different self-assembled structures that are robust to standard milk processing and torage techniques.[2-3] SAXS has also shown that lipolysis can enhance the solubility of lipophilic drugs in digesting milk and infant formula, enhancing their bioavailability.[4-5]

Results & Discussion: We hypothesise that controlling the liquid crystalline structures that form during milk lipid digestion is advantageous for nutrient absorption by each individual species and is thereby key to nutrient delivery. A key issue in testing this hypothesis is the chemical complexity of the milk fats themselves, making the analysis of the individual lipid components within the fat globules challenging. To that end, this presentation will discuss the lipid liquid crystalline structures formed in a variety of milks and milk-like emulsions during digestion and how they can be mimicked with simplified emulsions of homotriglycerides.[6] These simplified emulsions provide representative digestive colloid structures through which to analyse the impact of lipid digestion on bioactive delivery. Lipid self-assembly in digesting milk-like emulsions measured using SAXS will be discussed and compared with mammalian milks and off-the-shelf milk substitutes.[7]

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Preparation of Thin Films and Hydrogels of Polysaccharides via Polyion Complex Nanoparticles

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Introduction

Polyion complexes (PICs) formed with cationic and anionic natural polysaccharides are attractive materials as cell scaffolds due to their high biocompatibility, biodegradability and specific biological functions based on their chemical structures. However, PICs of polysaccharides have poor moldability. We have developed fabrication techniques of films and hollow fibers of polysaccharide PICs [1, 2]. In this study, thin films and hydrogels of polysaccharides were prepared via PIC nanoparticles.

Materials and Methods

Anionic polysaccharides such as heparin (HEP), chondroitin sulfate C (CS), hyaluronic acid (HYA), and carboxymethyl cellulose (CMC), and cationic chitosan (CHI) were used. At first, polysaccharide composite nanoparticles were prepared by mixing diluted solution of anionic polysaccharide and CHI. Polysaccharide composite films were prepared by casting these polysaccharide nanoparticles. Polysaccharide composite hydrogels were fabricated by mixing nanoparticle dispersion and PBS solution. The resultant films and hydrogels were characterized by using FT-IR and a scanning electron microscope. Culture of cells on the thin films and in the hydrogels were also examined.

Results and Discussion

Free-standing, transparent films of HEP/CHI, CS/CHI, HYA/CHI, and CMC/CHI with a thickness of several micrometers were obtained by casting polysaccharide nanoparticles of anionic polysaccharides and CHI [3]. By mixing CS/CHI nanoparticle dispersion with PBS, PIC hydrogels were successfully obtained.

Then, cells were cultured on the thin films and in the hydrogels. Adhesion of cells depended on the kinds of anionic polysaccharides. Cells were also successfully incorporated in CS/CHI hydrogels by mixing cell dispersion with PIC nanoparticle dispersion and PBS.

These results indicated that PIC nanoparticles can be used as components of cell scaffolds made of PIC of polysaccharides.

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Self-healing hydrogels formed from nanofibrillar assemblies of the amino acid phenylalanine

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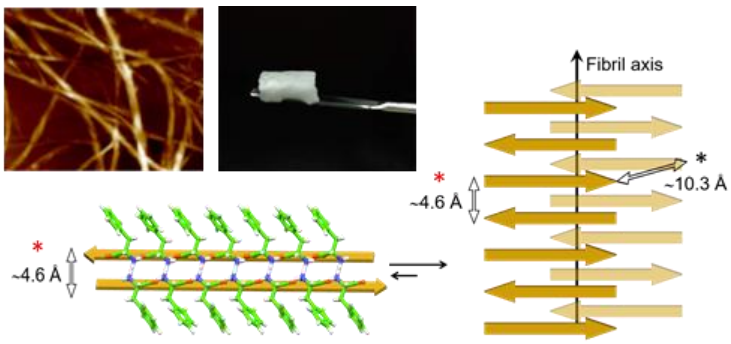

Self-assembling proteins and peptides have long been of technological interest due to their ability to form nanoscale nanofibrillar networks and hydrogels with desirable physiochemical properties. These materials have many applications in diverse fields including as regenerative medicine, drug delivery, environmental science and biosensing.[1] Simplistic building blocks are appealing as they provide easily accessible, synthetically simple and versatile scaffolds onto which bespoke functionalities can be added. This has driven researchers to find the minimalistic self-assembling peptide motif, with numerous examples of self-assembling materials made from ultrashort peptides with as little as two amino acids.[1] Here we present a further simplification of this minimalistic sequence and show that the single amino acid phenylalanine can self-assemble into β-sheet rich helical nanoribbons formed from intertwined protofilaments with exceptionally high Young's modulus. Furthermore, at high concentrations and under the correct conditions these nanofibrillar assemblies coalesce into highly homogenous, non-crystalline, self-healing hydrogels that display storage and loss moduli significantly higher than similar non-covalently cross-linked biomolecular nanofibrillar scaffolds. These remarkably stiff nanofibrillar hydrogels properties can be harnessed for various technological and biomedical applications, such as self-healing, printable, structural, load-bearing 3D scaffolds.[2]

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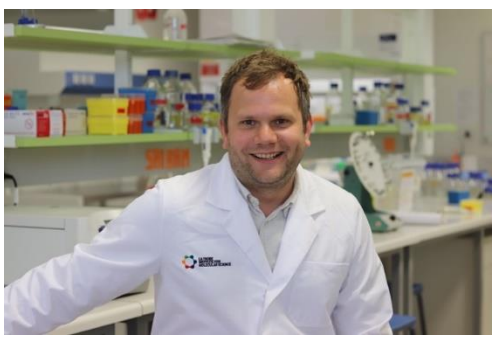

Voltage Driven Characteristic Instability of Nematic Liquid Crystal on Chemically Patterned Substrate

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Surface anchoring plays a fundamental role for controlling the molecular alignment of a nematic liquid crystal (NLC). To achieve a suitable alignment, the solid substrate of LC cell is coated by polymer film or grafted polymer brush. Recently, nano scale patterns were examined to achieve precise control of the alignment of NLC [1]. The patterns were prepared by partially removing the grafted poly(6-(4-methoxy-azobenzene-4'-oxy) hexyl methacrylate) (PMMAZO) on the silicon substrate. The NLC aligned perpendicular to the region of polymer brush. On the other hand, they aligned parallel to the region of bare silicon. However, the dynamics of NLC under the applied voltage was not studied for the LC cell of the patterned surface. In this work, the dynamics of NLC on the stripe patterns of bare silicon was studied.

The grafted PMMAZO on the silicon substrate was removed by using the electron-beam lithography followed by oxygen plasma etching, and the stripe patterns of bare silicon surrounded by grafted PMMAZO were prepared. The width and length of the stripes were 300 nm and 100 □m, respectively. The LC cell was prepared using the silicon substrate and glass plate coated by indium thin oxide (ITO). The NLC 4-cyano4'-pentylbiphenyl (5CB) was injected into the cell, and which was observed by using a polarized light microscope. A DC voltage was applied between the silicon and ITO surfaces.

As in Figure 1, the patterned region became brighter than that of 0 voltage, when the positive DC voltage (3.6 V) was applied. Then the brightness I gradually decreased, and became 0 at around $t \approx 70$ s. Considering that the 5CB molecules become stable when they align parallel to the applied electric field, the final state after $t \approx 70$ s is the most stable. The increase in I is an unusual behaviour which is due to the instability of the molecular orientation of 5CB. When a negative voltage was applied, I was not larger than that of 0 voltage, and finally became 0. The instability of the molecular orientation did not appear in this case. Similar time dependence of I was observed when the voltage was changed from 1.0 to 4.6 V.

The change in I is explained by considering the interaction between molecules and bare silicon surface. We propose an effective model to explain the characteristic instability.

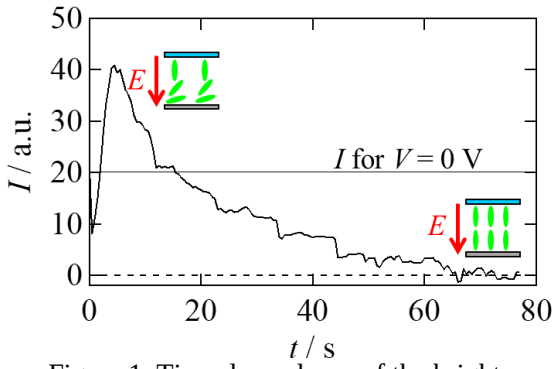

Figure 1. Time dependence of the brightness I.

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Small Angle Scattering at the Australian Synchrotron. A Fantastic Tool for Colloid and Interface Science

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The Australian Synchrotron has been operating a world class small and wide angle X-ray scattering (SAXS/WAXS) beamline [1] since 2009, which is ideally suited to colloid and interface applications. The beamline was designed for flexibility and performance, featuring :

- A diverse range of simple ex-situ sample environments suited to soft matter systems, such as temperature stages for determining chemical and temperature dependent phase diagrams, high throughput analysis such as in wellplate formats (in some cases many thousands of samples per day), moisture gradients, and User-supplied sample environments
- Flexible in-situ experiment capability, allowing you to mount your chemical reaction system on the beamline to study reactions, dissolutions, phase transformations, precipitations in real time
- grazing incidence scattering capability, for dry solid thin films (solid/air interface), as well as flow-cells for wet samples (solid liquid interfaces)
- a high degree of automation, and an easy to use interface that lets you see and understand your results during the beamtime.
- A high performance endstation with automated camera length adjustment, to allow fast changes in detector setup to study structure over length scales between 0.25 and 150 nanometres.
- World class signal:noise and sensitivity for dilute systems, and time resolution up to 25 or 500 frames/second (detector dependent) to study fast changes.

Soft matter and interface science are the most popular and productive disciplines on the beamline. This fantastic instrument is publically available through an open, competitive proposal process to prospective Users worldwide. Beamtime is at no cost to Users for open IP research, and travel support is provided to interstate Australian and New Zealand researchers. Beamtime is regularly available for purchase for commercial Users.

This presentation will outline the diverse range of capabilities and opportunities for colloid and interface science, illustrated by interesting experiments performed on the beamline over recent years.

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The Dynamics of Gold Nanorod Growth – A SAXS Study.

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The applications of gold nanorods reach over a broad range in biology, plasmonics and sensing. Recent progress in understanding of the growth mechanism at different stages, including the symmetry breaking step, renewed the interest in wet-chemical nanoparticle growth processes. Despite several studies of the crystallisation dynamics of ascorbic acid-catalysed nanorod syntheses, the growth kinetics of gold nanorods in a hydroquinone-based synthesis are not well-understood. A more detailed insight into the growth of nanocrystals can be provided by *in situ* observations. Time-dependent optical properties of growing gold nanorods can be analysed using absorption spectroscopy, whereas *in situ* investigations of structural evolution are more challenging, making the use of strong X-ray scattering sources such as the synchrotron necessary. This provides sufficient time resolution to detect single steps of nanoparticle growth to study the growth kinetics.

We studied the growth process of hydroquinone-based seeded-growth of gold nanorods *in situ*. The synthesis in aqueous CTAB solution showed a novel double-sigmoidal growth, which can be linked to the reaction speed of single gold crystal facets. We find this optimised synthesis route as highly reproducible, resulting in monodisperse gold nanorods without significant side products. From the spectral and scattering data, we can explain for the first time the growth mechanism of the hydroquinone-based synthesis from cuboctahedral seeds to an ellipsoidal state to the final spherical capped cylinder. Furthermore, the influence of changing the gold acid and hydroquinone precursor concentrations in the initial solution was investigated, giving further insights into the nanoparticle growth mechanism. The high quality of the synthesis and X-ray scattering experiments at the SAXS beamline of the Australian Synchrotron made it possible to actually determine the size of the CTAB layer around the growing nanorod during the synthesis.

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POSTERS

Poster Sessions

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Efficient transfer of *in-situ* polyethoxylated alkyl amine modified gold nanoparticles from aqueous to organic media

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Key words: gold nanoparticles, phase transfer, surface charge, pH, colloidal stability

Phase transfer of nanoparticles, specially from polar to nonpolar solvent is necessary because the preparations of the desired particle in nonpolar solvents require more complicated and meticulous steps which make the production process laborious. In this study, the gold nanoparticles (AuNPs) were directly prepared in aqueous media using polyethoxylated alkyl amine (AMIET), a nonionic surfactant that acts as reducing and stabilizing agent in the synthesis process. However, the critical issue of the phase transfer of colloidal nanoparticles is their colloidal stability[1]. It is often challenging to transfer the nanoparticle from a stable colloid system to another phase, where the particle are at first not able to disperse. Although different strategies like ligand exchange, surface modification with chemical functional group, additional coating layers or common solvents are discussed for phase transfer, the particles might partially lose their surface properties to be stably dispersed in the desired nonpolar phase. Beyond these traditional methods, herein we demonstrate the efficient phase transfer of laboratory synthesized AMIET-AuNP without addition of any specific chemical. The objective of this work is to establish a robust and repeatable phase transfer methodology of AMIET-AuNP. Therefore, this study is an attempt to destabilize the aqueous colloidal system by changing the pH of the system to obtain nearly zero surface charged particles that should be stable in nonaqueous phase. Consequently, almost complete phase transfer was promptly achieved at the state so called isoelectric pH where the AuNPs tends to coagulate or flocculate. Moreover, it is noticeable that at this unstable state, the AuNPs transfer its phase spontaneously regardless of the density of nonaqueous solvents (Fig-1). It is suggested that the surface bound ligand shell can undergo conformational changes depending on the surrounding solvents that favors the particles for phase transfer[2].The complete phase transfer, preserved morphology and ligand behavior has been evident by UV-visible and NMR spectra. Our current strategy may confer multiple possibilities for phase transfer of other nonionic surfactant coated metal nanoparticles.

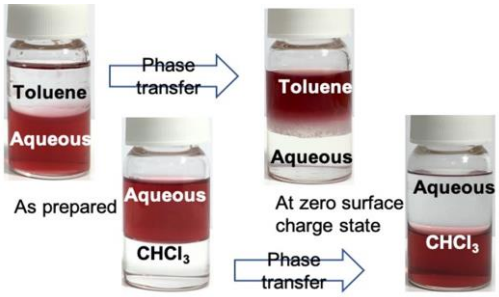

Fig-1: Appearance after the addition of organic solvents to AMIET-AuNP as prepared and after adjusting to the isoelectric pH.

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Contact angle as a powerful tool in anisotropic colloid synthesis

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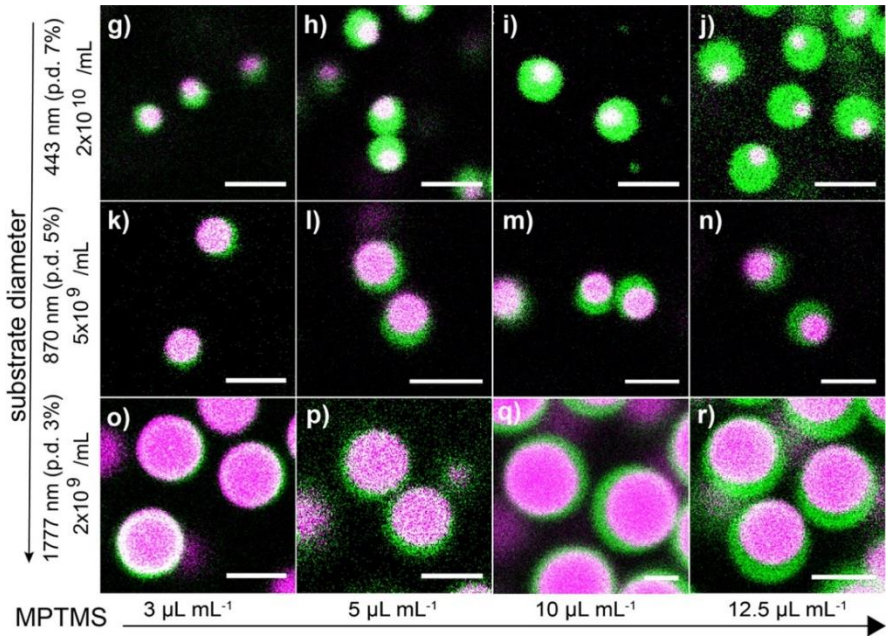
Nucleation and growth is a technique widely used to prepare colloids, in which droplets are adsorbed onto substrate particles. Changing the contact angle of the substrates can greatly alter the morphology of the product particles. Here [1], we investigate the nucleation and growth of 3-methacryloxypropyltrimethoxysilane (MPTMS) both onto Stöber spheres and onto (cross-linked) MPTMS* spheres. The former results in 'snowman' particles with a cap-shaped MPTMS* compartment, and we show that their morphology is highly controllable via the MPTMS content in the reaction mixture. The contact angle of the MPTMS* compartment decreases with droplet diameter, suggesting that this wetting process is affected not only by surface tension but also by line tension. In contrast to Stöber spheres, MPTMS* substrate particles yield highly reproducible and tuneable 'engulfed-sphere' colloids with an internal reference axis (see Figure) but with a homogeneous mass distribution. These engulfed-sphere particles can be fully index-matched for confocal microscopy on account of their homogeneous refractive index. Suitable index-matching mixtures of polar and of low-polar media are presented, where cyclohexyl iodide (CHI) is introduced as a new medium for colloids of high refractive index. Finally, the index-matched engulfed-sphere colloids are self-assembled into (close-packed and long-range) plastic phases, and the particles' rotational diffusion inside the crystal phases is tracked via confocal microscopy.



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Stratification In Drying Films: Diffusion And Diffusiophoresis

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Stratification in drying films – how a mixture of differently-sized particles arranges itself upon drying – is examined. It is seen experimentally that smaller particles preferentially accumulate at the top surface, but it is not understood why.[1] Understanding this could allow the design of formulations that self-assemble during drying to give a desired structure. Potential applications are across a wide range of industries, from a self-layering paint for cars, to a biocidal coating in which the biocide stratifies to the top surface, where it is required.

As solvent evaporates from a film, the top surface, initially at height H, descends at velocity $\dot{E}$. Particles which are unable to diffuse away from the top are collected by the top surface. Whether or not the particles can diffuse sufficiently quickly away from the surface is characterised by the dimensionless group called the Péclet number, $Pe = 6\pi\eta R\dot{E}H/kT$, where R is the particle radius, η is the solvent viscosity, and kT is the thermal energy. Hence it is expected that drying dispersions with different size particles will lead to different particle arrangements in the dried film. On the basis of diffusional arguments alone, it would be expected that larger particles stratify to the top surface. However, other physical processes, including diffusiophoresis, may also be important.

As will be explored in this presentation, by deriving transport equations, the magnitude of different contributions can be compared, and numerical solutions for the film profile are produced. Adding a diffusiophoresis term to the diffusional model predicts that the top surface is formed of small particles. This demonstrates that diffusiophoresis is a feasible explanation for the experimental observations.

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Effect of surfactant ionicity on critical micelle concentration in aqueous ionic liquid mixtures

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Protic ionic liquids are the largest known solvent class capable of promoting surfactant self-assembly. However, ILs are increasingly used as mixtures with molecular solvents, such as water, to reduce their cost, viscosity and melting point, and the self-assembly promoting properties of these mixtures are largely unknown. Here we investigated the critical micelle concentration (CMC) of ionic and non-ionic amphiphiles in ethylammonium nitrate (EAN)-water mixtures to gain insight into the role of solvent species, and effect of solvent ionicity on the self-assembly process. The amphiphiles used were the cationic cetyltrimethylammonium bromide (CTAB), anionic sodium octanoate sulfate (SOS), and the non-ionic surfactant tetraethylene glycol monododecyl ether (C12E4). Surface tensiometry was used to obtain the CMCs and free energy parameters of micelle formation, and Small angle x-ray scattering (SAXS) was used to characterise the micelle shape and size.

The EAN-water solvents displayed self-assembly results consistent with a salt in water for EAN proportions below 5 mol% across all three surfactants, leading to CMC values lower than the CMC observed in water. A steep incline in the CMC was observed for concentrations between 5 mol% to 50 mol% of EAN for SOS and C12E4. However, CTAB displayed more complex behaviour where the CMC remained below the CMC of water until 33 mol% EAN. Across all surfactants, a plateau in CMC values were observed at very high EAN concentrations, which could indicate that there is a shift in the dominant solvent beyond EAN concentrations of 50 mol%.

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Specific-ion effects are ion specific

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Specific-ion effects (SIEs) induce or influence physicochemical phenomena in a way that is determined by the identity of the ions present, and not merely by charge or concentration. Such effects have been known for 130 years since the seminal work of Hofmeister and are often categorised according to the well-known Hofmeister series.[1] Examples of SIEs are ubiquitous throughout the chemical, biological, environmental and material sciences, and are traditionally explained in terms of the influence ions have on the structure of water. However, this explanation is unsatisfactory as it is unable to adequately explain and predict frequently-observed series reversals and anomalies, so a predictive theory remains elusive.[2–5] Utilising a DDEC6 charge distribution analysis,[6] in conjunction with an adjusted electrostatic potential energy calculation, derived from the well-founded Coulomb's Law, a new ab initio parameter (þ) has been developed. This provides a physical basis to recent work,[7] both predicting and post-hoc explaining SIEs in a multitude of systems, such as polymer cononsolvency, enzyme activity and the Gibbs Energy of transfer from aqueous to nonaqueous systems, laying the foundations for a general predictive theory of SIEs.

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Examining specific ion effects in glycerol from concentration depth profile using NICISS

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Adsorption or desorption of ions at solvent surface influences various properties of the solvent and ions present in the solution. This nature of an ion plays very important role in unraveling specific ion effects with applications extending towards physical, chemical, biological, and atmospherically relevant processes[1].To provide detailed information about ionic behavior at solvent surface, obtaining direct depth profile information of various cations and anions in variety of vapor-solvents interface is important[2].

In the present research work, Neutral impact collision ion scattering spectroscopy (NICISS) is used to obtain concentration depth profile of ions at vapor-glycerol interface [3]. Specific ion trends are investigated in salty glycerol solution of 11 salts (NaCl, NaBr, NaI, CsCl, CsBr, CsF, KF, KBr, KI, LiCl and LiBr) to provide a comparison between depth profile onsets of various anions and cations . Glycerol is a polar solvent with high dipole moment (2.56 D), dielectric constant and very low vapor pressure[4]. As NICISS measurement is done under UHV condition, low vapor pressure & polar solvent nature is the one of the reason for selecting glycerol with further scope in investigating similar effects with other polar & non-polar solvents such as formamide, propylene carbonate and water.

Keywords: Specific ion effects, Concentration depth profile, Glycerol, Vapor-solvent interface

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Quantitative Determination of Protein Solubility in Ionic Liquids

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Proteins are often utilised for a range of applications in the pharmaceutical, biological, chemical and food industries[1-2]. The ideal solvent for hydrophilic proteins is usually buffered water due to its minimal cost, and ability to mimic the native environment of proteins, however many proteins are hydrophobic and have poor solubility in water. Because of this, organic solvents have been investigated as an alternative solvent for biocatalysis[3] and protein extraction[4], but often have detrimental effects on the protein stability and structure. We propose to use ionic liquids (ILs) as an alternative solvent, or as an additive in aqueous solutions, to quantify the solubility and stability of proteins. Initially the model protein lysozyme will be tested in ILs from highly dilute to neat. A novel, high throughput method has been developed to quantitatively determine the solubility of lysozyme. The aim is to explore specific-ion effects and how these differ for concentrated IL solutions compared to conventional dilute salts. A variety of techniques including UV/vis spectroscopy, Fourier-transformation infrared spectroscopy, circular dichroism and small angle x-ray scattering will be used to describe the stability and structure of the protein, and to gain insight into its interactions with ILs. It is hoped that any solubility trends present for lysozyme or specific ions can then be extrapolated to other proteins. Further studies will be done to compare any variations in the specific ion effects on different proteins and to begin building a database of quantified protein solubility and stability in ILs.

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The effect of salt and particle concentration on the dynamic aggregation of detonation nanodiamonds in water

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Nanodiamonds have received significant attention from the scientific and engineering communities due to their unique physiochemical properties. These properties are exploited in many applications from drug delivery and bio-sensing to composite materials and abrasives [1, 2]. Most detonation nanodiamond particles are colloidally stable in water and many organic solvents without the need for stabilizing ligand molecules. However, their colloidal properties and aggregation behavior in simple and controlled aqueous environments remain largely unexplored and poorly understood.

Previously, we have shown that detonation nanodiamond (DND) particles can self-assemble into a lace-like network in water [3]. The current study aims to systematically understand this phenomenon. We use dynamic light scattering and synchrotron-based small-angle X-ray scattering to examine the effects of both particle and salt concentration on the particles dynamic aggregation behavior. The complex structures that are dynamically and reversibly formed by nanodiamonds in water will be discussed in the context of the particles surface chemistry and compared to well-known colloids like silica nanoparticles.

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Pickering Emulsions for 3D Printing of Hierarchical Porous Ceramic Architectures

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Good control of the rheological (flow) properties of colloidal pastes is essential for their success as feedstocks for 3D printing, namely the Direct Ink Writing (DIW) technique. In DIW, feedstock inks are extruded through a nozzle of the 3D printer directly onto the platform. Therefore, a feedstock with a high storage modulus is required for its particle network to behave in a solid-like manner in retaining its shape before and after printing. Additionally, it should be a shear thinning fluid that is able to flow well under high shear stresses are applied during printing. We present an approach of introducing a hierarchical organisation of porosity into ceramic products.1 By controlling the gaps between printed scaffold filaments, we create millimetre-scale porosity. Furthermore, via soft templating of oil droplets, stabilised by the colloidal ceramic particles (known as Pickering emulsions), 20 micron-scale porosity can be developed. The printed objects are strengthened by sintering at high temperature. This creates complex-shaped, crack-free components with a controllable porous architecture and remarkably low density, which are advantageous in biomedical, catalytic, and aerospace applications. The effects of formulation (surfactant and oil concentrations, solids particle size, and mixing speed) on rheology and pore size and morphology have been investigated. The rheological properties (storage modulus, yield stress and recovery of storage modulus) of the emulsions have been found to delineate the samples with good shape retention from those that slump. Additionally, the internal features of the sintered structures have been analysed via X-ray tomography and SEM.

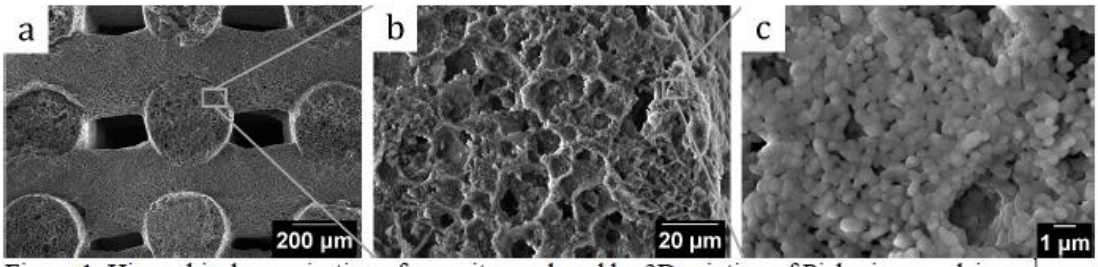

Figure 1. Hierarchical organisation of porosity produced by 3D printing of Pickering emulsions.[1]

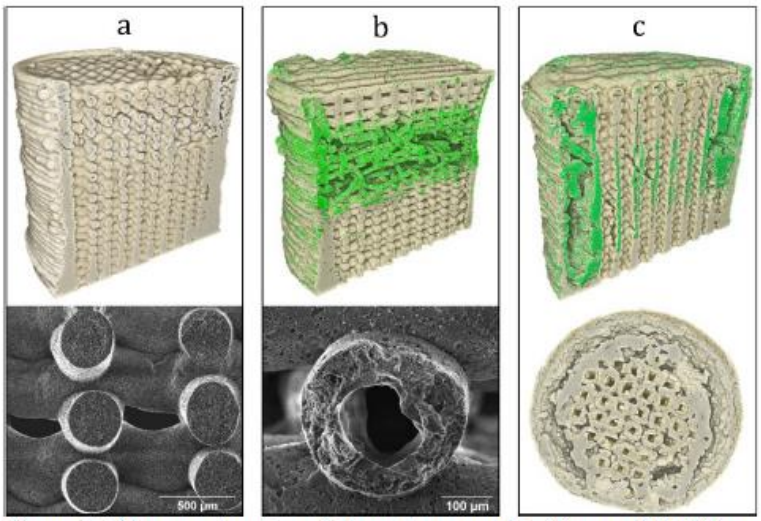

Figure 2. 3D reconstruction of X-ray tomographic slices and SEM images for the characterisation of internal structures.[1]

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Polar-Nonpolar Interface Of Bicontinuous Cubic Phase In Nonionic Surfactant/Water Systems

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Several substances are known to form triply periodic minimal surface(TPMS)-like structures, including the bicontinuous cubic phases (Q phases) of lyotropic liquid crystals. In the type II Q phase, the midplane in the non-polar region of the bilayer is located on a TPMS, and the bilayer is periodically connected in three dimensions. On the other hand, in the type I, the middle of the polar region is located on a TPMS.

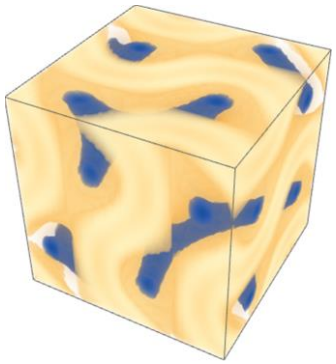

Fig. Electron density of $C_{12}EO_6$/water Q_I phase.

We have succeeded in single-crystal X-ray structural analysis of the type II Q-phase, a lipid monoolein/water-based Q-phase[1] and a phytanthiol/water-based Q-phase [2], respectively, and revealed the electron density distributions.

We report here on the single-crystal X-ray structure analysis of the type I Q phase in non-ionic surfactant/water systems using $C_{12}EO_6$, $C_{12}EO_7$ and $C_{12}EO_8$, respectively [3]. We used single crystals of type I Q-phase and measured X-ray diffraction at Spring-8 BL40B2 using a rotating crystal method. The amplitudes of the structure factors were converted from the X-ray diffraction data. Two model were used to optimize the experimental amplitudes of structure factors. One model was the PS model, in which the polar-nonpolar interface is parallel to an TPMS, and the other was the CMCS model, in which the interface is a constant mean curvature surface. After the optimization, the PS model agreed well with the experimental amplitudes and R-factor of about 0.05, while the CMCS model does not agree with the experimental amplitudes and R-factor of about 0.2. Next, we used the experimental amplitudes and model phases to calculate the electron density (Fig.). It showed that the polar region is located on the TPMS and the polar-nonpolar interface is parallel to the TPMS.

Two layers of $C_{12}EO_{6\text{-}8}$ polar chains face each other on the TPMS, sandwiching a thin water layer. It is suggested that the constant length of the polar chains, regardless of their location, is entropically advantageous and allows the interface to shift away from the CMCS. The other findings are discussed in this presentation.

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Drag reduction and boundary slip at silicone oil-water interfaces

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The interaction between liquid flows and solid surfaces has become an important issue for the manipulation of fluids at small scales[1]. The friction resulting from this interaction has a significant impact on fluid transport and energy dissipation which result in high pumping pressures and high shear rates in the fluid. Lubricant-infused surfaces (LIS)[2], in which a lubricant is trapped within the surface micro- or nano-structure, minimize the contact of the flowing liquid with the solid substrate and, therefore, offer a low friction surface which reduces the hydrodynamic drag significantly. This drag reduction is normally explained as an apparent slip of the fluid due to the mobility of the lubricant layer. Theoretical models predict that this apparent slippage increases with the lubricant height h_o and the ratio of the viscosity of the flowing liquid over that of the lubricant μ_w/μ_o (see Fig.1) [3]. Here, using highly accurate microfluidic measurements, we perform experimental observations and numerical simulations on a nanostructured wrinkled surface infused with a thin silicone oil film[4]. Our findings suggest that the apparent slip model is not sufficient to describe the large reduction in drag observed on LIS (up to 28 %, close to the values reported for superhydrophobic surfaces). We propose that the nucleation of gas at the liquid-lubricant interface, facilitated by flow, is an additional mechanism that could explain the large drag reduction observed in LIS.

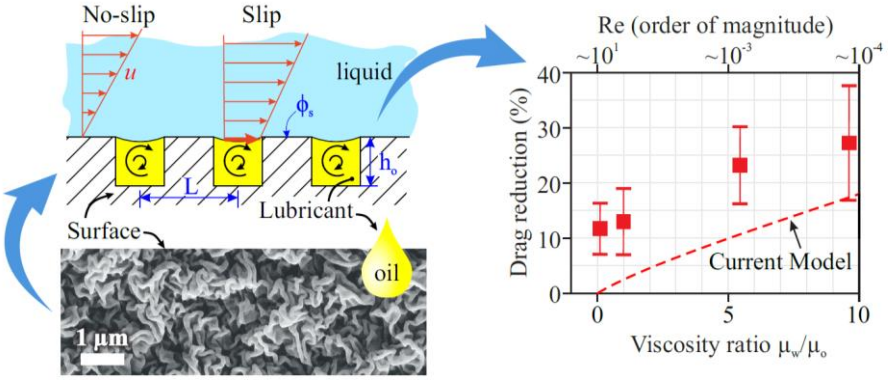

Figure 1: (left) Wrinkled Teflon surfaces are infused with silicone oil, leading to microscale pools of oil between the wrinkles. (right) Discrepancy in the drag reduction estimation between our experiments and the apparent slip model (current model).

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Hetero-coagulation of nanoparticles in emulsion droplets: A green approach for producing functional structured materials

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Microstructured materials, such as microcapsules or microbeads, have been widely used for applications in drug delivery, protein separation, heterogeneous catalysis and water treatment. However, preparation of such systems can be time-consuming and require use of specific lab equipment, toxic chemicals or high temperatures.[1]

This talk presents a green approach to produce functional silica microbeads and microcapsules at room temperature via salt driven assembly of nanoparticles in emulsion droplets.[2,3,4] Such an approach is fast, safe and exhibits a high level of tuneability. The use of an emulsion as template enables us to control both diameter and structure of the final systems, with single emulsions leading to microbeads and double emulsions leading to microcapsules. Furthermore, metal oxides such as TiO_2 and Fe_3O_4 can easily be incorporated in the structure, conferring additional photocatalytic and magnetic properties to the systems.[2,3,4] Such a method is a green and versatile approach for preparing functional structured materials using readily available nanoparticles and easily accessible lab equipment.

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Microfluidic-assisted Formulation of PLGA Nanoparticles for Intracellular Delivery of Anti-Chlamydial Protease Inhibitor JO146

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The obligate intracellular bacterium *Chlamydia trachomatis* is a major global health burden, being a leading cause of sexually transmitted diseases and trachoma. Drug delivery to chlamydia is challenging due to their unique biphasic developmental cycle and ability to enter into an antibiotic-refractory persistent state[1]. Previously, we identified a small-molecule peptide inhibitor JO146 [Boc-Val-Pro-Val^{P}$(OPh)_2$] that specifically targets the chlamydial serine protease HtrA[2]. JO146 is highly efficacious in attenuating infectivity of chlamydia in various host cell types[2,3]; however, its efficacy was not observed during persistence *in vitro*[4].

In this study, PLGA (poly(lactic-*co*-glycolic)acid) nanoparticles were explored as a delivery system for JO146 to improve delivery to the target site and antibacterial potency against *C. trachomatis* infections. JO146-PLGA nanoparticles with three different diameters were formulated to investigate the optimal particle size to reach the target protease. To determine the effect of formulation parameters on average particle size, JO146-PLGA nanoparticles were prepared by microfluidics and optimized using a Design of Experiments approach. Three different sized formulations (95, 150, 220 nm) were designed and had drug loadings of 5.7%, 7.3% and 6.2%, respectively. In simulated physiological conditions (PBS, pH 7.4, 37°C, 96 h), the 95 nm nanoparticles increased in size by 20% over 96 h, whilst the others remained unchanged. The smaller nanoparticles (95 nm and 150 nm) had an initial rapid drug release of approximately 80% within 1 h. In contrast, the 220 nm nanoparticles displayed sustained release up to 80% of the drug loading after 96 h, while 100% was released from the other formulations over the same period.

Microfluidics enabled the controlled production of nanoparticles of different sizes. JO146-loaded PLGA nanoparticles and size influenced particle stability and drug release kinetics. Future testing of the PLGA nanoparticle formulations against *C. trachomatis* cells will add further insight to formulation optimization for targeting chlamydial infections.

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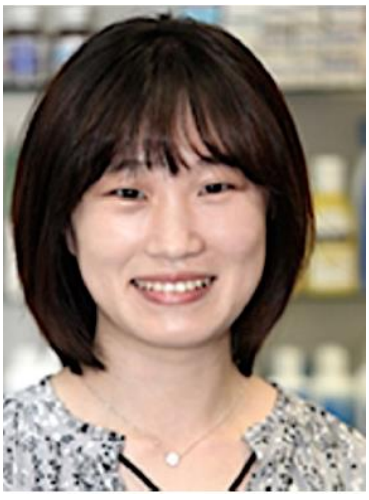

Battacin-inspired ultrashort antimicrobial peptides: structure-activity relationship

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Reports from the World Health Organisation (WHO) and Australia Government described the antimicrobial resistance as one of the key health issues facing our generation, recognizing it as health priority. These data indicate the urgent need for research on novel antimicrobial strategies. Antimicrobial peptides (AMPs) are produced by various organisms as part of their normal immune response and a growing number of these peptides have been shown to self-assemble into nanostructures that are thought to play a role in their antimicrobial activity. While most of studies to date have focused on long peptide sequences and proteins, recent works had demonstrated that short peptide sequences also self-assemble and display antimicrobial activity. Pioneering works in the domain have established that two nanotube-forming short synthetic sequences – the diphenylalanine peptide and cyclic D,L-α alanine peptide - interact with bacterial membranes and display antimicrobial activity. Thus, we hypothesise that novel families of antimicrobial agents can be designed from small self-assembling minimal building blocks and that their antimicrobial spectrum of activity can be tuned with minimal sequence variation (1-3). This project aims to determine the correlation between the self-assembly properties of short designed AMPs and their membrane-targeting antimicrobial activity, via a thorough characterisation of their self-assembling properties and interactions with model lipid membranes, which will allow us to optimise the peptide sequences and enable the design of drug formulations and biomaterials.

Here, we are focusing on battacin-inspired ultrashort peptides containing from 3 to 5 amino-acids residues (4). Antimicrobial studies showed activity of the peptides against gram positive and negative bacteria as well as fungi species. A set of biophysical techniques, including Fourier transform infrared spectroscopy, small angle X-ray scattering, and electron microscopy showed that 7 out of 8 of these designed antimicrobial peptides self-assembled into liquid crystalline beta-sheet nanofibrillar aggregates in aqueous media, with different kinetics depending on sequence, resulting in the formation of hydrogels with a range of viscosities. The versatility of the formed nanostructures is well suitable for nanotechnology applications and those peptides could be used in the design of biomaterials such as nanostructured surfaces and topologies for antimicrobial purposes such as filters.
The peptide HG 2.77 (diphenylacetyl-Dab-Dab-1-Nal-NH2), that didn't assemble, presented a lower antimicrobial activity compared to the peptide HG 2.41(Fmoc-D-Lys-Lys-1-Nal-1-Nal-Lys-NH2) that self-assembled and had the highest activity from the designed peptides, indicating that there is a correlation between the self-assembly, amphiphilicity, positive net charge and peptide length with the antimicrobial activity. Moreover, in order to investigate the mechanism of action of HG 2.41 and HG 2.77 we performed Small angle X-ray scattering, X-ray reflectometry and neutron scattering experiments. The results showed the selective interaction of those peptide with different model membranes.

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Engineering Polymeric Nanocapsules with High Aspect Ratio as a High Drug-Payload and Long-circulating Drug Delivery System

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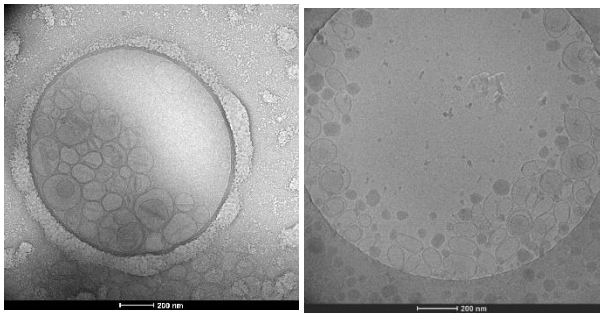
INTRODUCTION

Most nanoparticles used for medical applications suffer from low treatment efficiency and serious side effects, due to their short circulation time in the body and immediate-release mechanism. Elongated shape was found to be able to minimize the internalization of nanoparticles by macrophages and hence increase the circulation time[1]. Polymeric material provides a less permeable membrane so a smaller amount of drug can be released over a longer period of time. This project thus aims to make use of both features to engineer elongated nanocapsules to increase circulation time and improve controlled release.

METHODS

Vesicle templating method[2] was adapted, in which elongated liposomes[3] were used as the template and polymers were directed to grow via RAFT polymerization on the surface of the liposomes. The shape and aspect ratio of the nanocapsules prepared after polymerization were characterized by cryo-TEM and small angle neutron scattering (SANS) with BILBY[4]. Lastly, cellular interactions between macrophages and elongated nanocapsules will be compared to their spherical counterparts and their drug release profile will be constructed via ultrafiltration.

RESULTS



a b

Figure 1 a) Elongated liposomes were successfully prepared with encapsulated drug nanocrystals; b) Elongated shape was maintained in the final polymeric nanocapsules

Cryo-TEM images showed that elongated liposomes were successfully prepared with encapsulated drug nanocrystals (Figure 1a). With these templates for adsorption of RAFT oligomer, the RAFT polymerization was driven onto the surface of the liposomes. There was no destruction of the interior space for drug encapsulation and the elongated shape of the liposome templates were kept relatively intact (Figure 1b) comparing to some nanocapsules made with spherical templates in the literature[5]. The disappearance of the drug nanocrystal might be due to the high reaction temperature resulting In higher solubility and further investigation is needed.

CONCLUSIONS

Elongated nanocapsules were prepared with elongated liposome templates. Further investigation will focus on keeping the drug in nanocrystal form. Cellular interactions and drug release will be investigated to decrease dosage frequency and the side effects.

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Metal Organic Framework: A Safe Gene Delivery Vehicle

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Metal-organic framework (MOF) is a hybrid nanomaterial with excellent loading capacity. Among different classes of MOFs, the zeolitic imidazole framework-8 (ZIF-8) is highly investigated for delivering biomolecules. The ZIF-8 crystal is organized by divalent zinc ions and 2-methylimidazole linkers. Distinctive features of ZIF-8 makes it an advanced material to load different biomolecules (*i.e.*, proteins, enzymes, carbohydrates). Moreover, nucleic acid (NA) can also be encapsulated by ZIF-8 by biomimetic mineralization to prepare NA@ZIF-8 composite.[1] Pure ZIF-8 itself is not non-toxic to healthy cells and can act as a neutral delivery vehicle. It is a challenge to determine how the crystal structures impact cellular uptake and hence gene delivery. On the other hand, cancer cells are continuous, self-propagating cells that have an acidic microenvironment. ZIF-8 disintegrates and releases the loaded NA in acidic pH of cancer cells. Moreover, it can be an inexpensive way to deliver genetic material compared to other conventional gene delivery. In this project, we will consider the unique properties of ZIF-8 to deliver genetic material to minimise the effect of dysregulated genes in cancers.

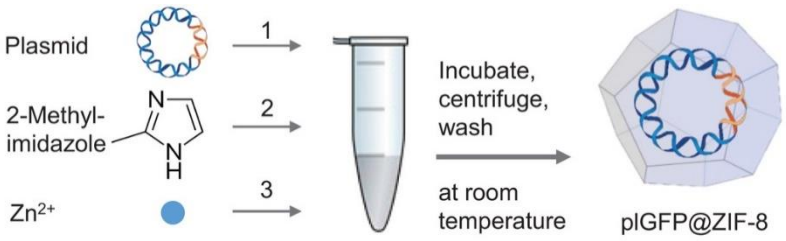

Figure. General approach to encapsulate nucleic acid in ZIF-8 crystal.

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Understanding structural networks within peptide hydrogels by transmission electron microscopy and cryogenic scanning electron microscopy

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Peptide hydrogels are one of the most promising nano-biomaterials used in a wide range of applications such as scaffolds for tissue engineering, wound healing, sustained drug delivery systems, nanocomposite for fabrication and removal of toxic materials from polluted water[1]. These wide applicability is as a result of excellent biocompatibility, biodegradability, environmental responsiveness and bio functionality[2]. Understanding peptide nanostructures is critical to tailoring their applications for efficient use[3].

Transmission electron microscopy (TEM) and cryogenic scanning electron microscopy (cryo-SEM) are common characterization tools used for structural elucidation[1]. TEM examines primarily nanoscale structures which form networks in the matrix of hydrogel systems. The peptide nanostructures commonly exhibit high-aspect ratio morphologies as fibers, tubes, ribbons, or tapes. These structures interact to form network by a process of entangling, branching or cross-linkages in a 3-dimensional architecture which is best examined by cryo-SEM[4].

Here we compare the characterisation by TEM and cryoSEM of the self-assembled hydrogels formed by substance P, a neuropeptide that we discovered to form semi-flexible nanotubes[5]. We analyzed how these nanotubes interact to form a network matrix by cryo-SEM. The nanotubes as observed in TEM are consistent with other biophysical characterization[5], while 3-D structural networks can be characterized by cryoSEM. Pores due to cryogenic artefacts[3, 4] could be distinguished from structural networks in cryoSEM images. The occurrence of these cryogenic artefact were systematically characterized to support improvement of the cryoSEM protocols for peptide hydrogels.

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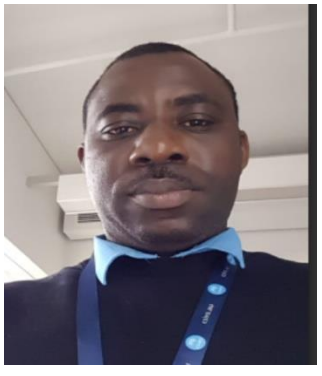

QCM-D and Neutron Reflectometry Investigation of Cellulose-Mucin Interactions Under Plasma Treatment Towards ETSA

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Epidemic thunderstorm asthma (ETSA)[1] is associated with inhalation of airborne pollen grains and aerosolized pollen fragments, causing hypersensitive immune reactions[2] that might lead to an asthma attack. The wall of pollen grains (intine) contains cellulose which is hypothesized to initially interact with the nasal and tracheal mucous layer[3] when inhaled. The air-way mucous layer is comprised of mucin (a major glycosylated proteinaceous element) and water, which serves as a first-line-of-defence against inhaled pollen particles. Although immunological and meteorological studies have been conducted in this regard, the fundamental cause and mechanism of ETSA are under-investigated. This study is focused on unraveling inherent cellulose-mucin interactions employing quartz crystal microbalance with dissipation (QCM-D) and neutron reflectometry (NR) examining the adsorption of mucin on cellulose while mimicking a thunderstorm environment, such as the affect of plasma treatment on cellulose-mucin interactions. Here, we generate air-plasma and plasma-activated water to treat our model cellulose surfaces[4], simulating the ionized surface chemistry of thunderstorm-borne pollen particles and examine subsequent interactions.

In this poster, we describe the use of QCM-D and NR to investigate cellulose-mucin interactions and the effect of plasma treatment on these biointerfacial interactions. The advanced molecular data obtained from this study, coupled with immunological and meteorological investigations, will enable the mechanistic understanding, treatment, and prevention of ETSA.

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Site-selective biasing of galvanic replacement in silver nanoprisms by photoexcitation of LSPR modes

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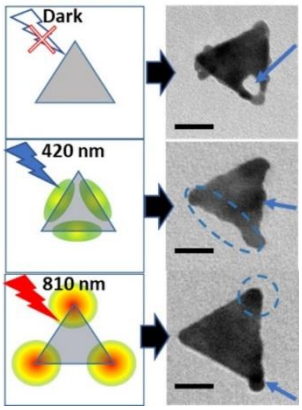

Surface plasmons are collective oscillations of free electrons by an incident electromagnetic field and are prominently exhibited by noble metals such as Ag, Au, and Cu at nanoscale dimensions. The localised surface plasmon resonance (LSPR) resulting from this light-matter interaction leads to remarkable opto-electronic effects such as optical near field enhancement close to nanoparticle surfaces, generation of hot carriers and finally thermalization (photo-thermal effect) of the plasmon excitation. These phenomena have been of significant research interests, particularly to harness them for driving chemical reactions. Anisotropic nanoparticles such as silver nanoprisms exhibit multiple LSPR modes, like the in-plane and out-of-plane dipolar and quadrupolar modes. These modes can be selectively photo-excited using light of narrow bandwidth which leads to localised hot spots within the nanoparticle depending on the mode excited.[1] These plasmonic hotspots also act as chemical hotspots and can be used to drive and control chemical reactions on nanoparticle surfaces. Galvanic replacement is a spontaneous reaction that occurs due to the difference in the standard electrode potential of two components participating in the reaction. The GR reaction leads to the corrosion (oxidation) of the component having the lower reduction potential, while simultaneously reduction of the second component leading to interesting nanoparticle morphologies such as nanoshells, nanocages, nanoframes .[2, 3] We report selective photoexcitation of LSPR modes as a means to control the chemical reaction sites in silver nanoprisms by taking galvanic replacement between silver nanoprisms and chloroaurate ions as a probe for the site-selectivity. [4] When the reaction between silver nanoprisms and chloroaurate ions is carried out under excitation of the in-plane dipole mode, the reaction occurred at the vertices of the nanoprisms; whereas it occurred at the nanoprism edges when carried out under excitation of in-plane quadrupole mode. No such site-selectivity was observed when the reaction was performed in dark conditions (no photoexcitation) We attribute this effect to hot-electron injection process at the sub-particle sites (plasmonic 'hot-spots') where localised plasmon eigenmodes drive selective reactivity. Our approach allows mapping of plasmon-driven sub-particle reaction sites with a high spatial resolution. We also probe the changes in the LSPR of Ag-Au alloy nanoprisms obtained under different photoexcitation conditions using Electron Energy Loss Spectroscopy (EELS), which reveals peculiar changes in the optical properties of the resultant bimetallic particles. The selective photoexcitation of different LSPR modes of anisotropic metal nanoparticles during a chemical reaction presents an exciting opportunity to control and spatially bias chemical reactions on the nanoparticle surface.

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Cascaded nanooptics to probe microsecond atomic-scale phenomena

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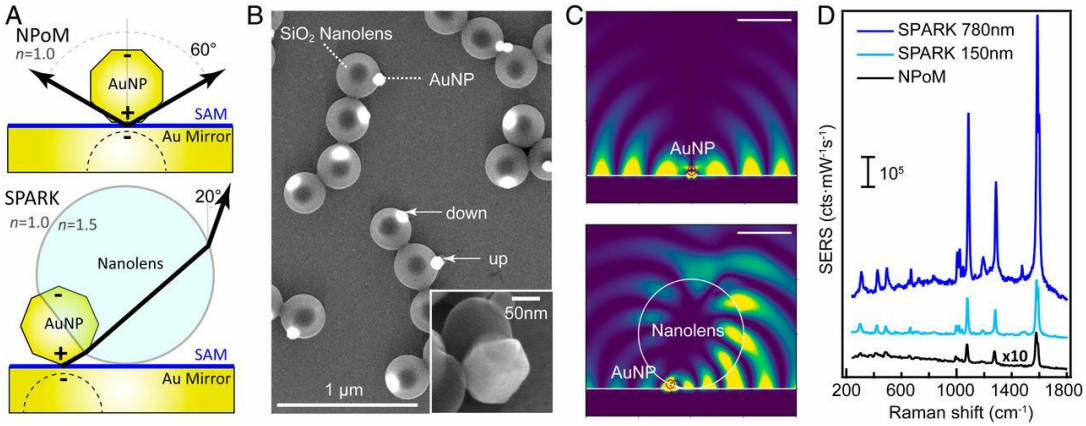
Plasmonic nanostructures can focus light far below the diffraction limit, and the nearly thousand-fold field enhancements obtained routinely enable few- and single-molecule detection. However, for processes happening on the molecular scale to be tracked with any relevant time resolution, the emission strengths need to be well beyond what current plasmonic devices provide. Here [1], we develop hybrid nanostructures incorporating both refractive and plasmonic optics, by creating organosilica colloids fused to plasmonic nanojunctions. Drastic improvements in Raman efficiencies are consistently achieved, with (single-wavelength) emissions reaching 10^7 counts·mW^{-1}·s^{-1} and 5×10^5 counts mW^{-1} s^{-1} molecule^{-1}, for enhancement factors >10^{11}. We demonstrate that such high efficiencies indeed enable tracking of single gold atoms and molecules with 17 µs time resolution, more than a thousandfold improvement over conventional high-performance plasmonic devices. Moreover, the obtained (integrated) megahertz count rates rival those of luminescent sources such as single-dye molecules and quantum dots, without bleaching or blinking.



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Probing the local 3D structure of disordered colloids with fluctuation x-ray scattering

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Fluctuation x-ray scattering studies how the x-ray diffraction pattern changes as a small x-ray beam is scanned relative to the sample. The ensemble of diffraction patterns from different sample positions can reveal information about the local 3D structure in disordered materials. We have developed a fluctuation scattering technique called the pair-angle distribution function (PADF) method that recovers three- and four-body correlations in the sample, including local angular structure[1]. This is a natural generalisation of the pair-distribution function obtained from small-angle x-ray scattering (SAXS). The PADF technique has been recently applied to disordered carbon materials[2] and self-assembled lipids[3]. Here we present simulation and experimental results applying the PADF technique to colloids[4]. We show that the angular peaks in the PADF can distinguish favoured local structures in disordered colloids, such as icosahedral clusters, face-centred cubic clusters, body-centred cubic clusters and hexagaonal close packing.

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Evaluation of electrocatalytic performance of Co_3O_4 nanoparticles with different ratios between Co^{2+} and Co^{3+} on oxygen evolution reaction (OER)

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Development of new efficient technologies for the generation of hydrogen fuel is utmost important as the detrimental environmental consequences of fossil fuels are continuously increasing. Electrochemical water splitting has been identified as one of the sophisticated and efficient approaches to tackle this challenge.[1] The hydrogen evolution reaction (HER) and the oxygen evolution reaction (OER) are the cathodic and anodic reactions involved in water splitting, where their sluggish reaction kinetics limits the practical applicability.[2] Among them, OER is the most difficult reaction to undergo as it has a four step reaction mechanism.[3] Therefore it is important to discover efficient electrocatalysts that can facilitate this process. Cobalt oxide with a spinel structure containing Co^{2+} and Co^{3+} oxidation states has been identified as an efficient electrocatalyst for OER in electrochemical water splitting in alkaline media.[4] However, the role of the oxidation state of cobalt in catalytic activity is still not clear. We synthesised a range of cobalt oxide nanoparticles with controllable stoichiometry by controlling the reaction parameters of hydrothermal synthesis and calcination temperatures. UV-Vis absorption spectroscopic data suggests that different materials have different ratios between Co^{2+} and Co^{3+}. Among these, oxides synthesized using cobalt hydroxide particles of homogeneous morphology displayed the best catalytic activity with a low overpotential of 232 mV at η=10 mA•cm^{-2}. Further, these materials showed low Tafel slope values, long term stability, low charge transfer resistance and considerably high electrochemical surface area that resulted in an outstanding catalytic activity for OER in alkaline solutions. Overall, the work suggests that in addition to the dependence on particle size and morphology, the ratio of Co^{2+} and Co^{3+} remarkably influences the catalytic activity of the spinel Co_3O_4 particles for OER.

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Synthesis and Application of Silica Hollow Particles

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Hollow particles are gas/solid composite particles having cavities inside. In this study, we adopted the inorganic particles template method to synthesis hollow silica particles. This method facilitates control of the size and shape of synthetic particles. Also, by changing the precursor of synthetic particles, it becomes possible to synthesize various particles and organic/inorganic hybrid particles.

In this study, at first, we have tried to synthesize silica hollow particles, second, synthesize organic/inorganic hybrid hollow particles by using different precursors, third, synthesize silica hollow particles aggregation using agglomerated particles as template. In the synthesis of hollow silica particles, calcite was used as a template and tetraethoxysilane (TEOS) was used as a precursor to synthesize. The hybrid hollow particles were synthesized using a mixed solution of methyltriethoxysilane (MTES) and TEOS as a precursor. In the case of silica hollow particle aggregates, synthesis was performed using vaterite as a template. I would like to report the result on the day.

Part of this research was carried out under the Aichi Research Center for Knowledge Base (Phase III; 2019-2021) "Mass production in hollow particles utilizing MI and synchrotron radiation and accelerated development of functional materials". I would like to thank you for writing here.

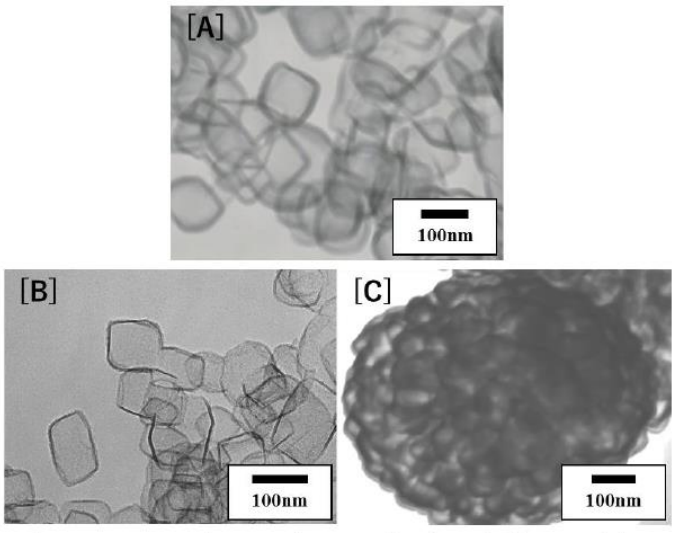

Figure.1 TEM and STEM images of various hollow particles
A) Particles using calcite as a template
B) Organic/Inorganic hybrid particles using MTES as a precursor
C) Particle aggregates using vaterite as a template

Hydrogen-sulfide responsive bicontinous nanospheres

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Polymeric nanoparticles are a potential strategy to re-vitalize traditional chemotherapeutic drugs; reducing systemic toxicity and increasing local concentrations in tumours. Polymer nanostructures are advantageous over lipid particles due to greater physical resilience and reduced non-specific leakage of drug, which necessitates a triggering mechanism to allow drug to escape.

Hydrogen sulfide (H_2S) is a gaseous transmitter that is reported to have increased expression in some cancers[1]. Utilisation of the specific reduction of azide (N_3) moieties by H_2S is a recent avenue for stimulated release of drug from nanocarriers[2]. This study investigates formulation of bicontinuous nanospheres (BCN) from aryl-azide containing block-co-polymers for H_2S response and encapsulation and release of model compound.

Methods

Polymer synthesis was achieved via ARGET ATRP using a methyl-PEG (mPEG) macroinitiator and synthesized 2-azidobenzyl carbamate monomer (ABOC). Polymers were analysed via NMR, FTIR and GPC. Polymers were formulated into particles via nanoprecipitation before characterization via DLS, cryo-EM and EM-tomography.

Particle response to H_2S was determined via exposure of nanoparticles to hydrogen sulfide generated *in-situ* from sodium hydrosulfide in PBS (5 mM), with response confirmed via FTIR, ^{1}H NMR and DLS.

Particle loading was attained by incorporating Nile red into the organic solvent prior to nanoprecipitation, with unencapsulated drug removed via size exclusion chromatography. Release of Nile red was examined in response to H_2S via fluorescence.

Results

A series of mPEG-pABOC polymers were synthesized with Mn (^{1}H NMR) 5476 Da to 10013 Da and a narrow distribution, PD 1.16 – 1.41.

Self-assembly of these polymers resulted in spherical particles of 155 – 180 nm with a narrow PDI. Cryo-EM imaging showed a complex morphology with apparent irregular internal bilayers surrounding internal pores. These particles appear morphologically similar to reported BCNs[3], with SAXS analysis showing disordered internal structure.

When exposed to H_2S, particles show a reduction in DLS particle count of over 90% coupled with loss of the strong azide peak at 2100 cm^{-1} on FTIR while ^{1}H NMR showed loss of aromatic and benzylic protons from the polymer backbone. This suggests reduction of the azide and subsequent self-immolation leading to generation of a free amine changing the nature of the bilayer, leading to disaggregation of the structure.

Hydrophobic compound was encapsulated, with an encapsulation efficiency of 68% for Nile red. No encapsulation of hydrophilic drug was seen. Release of hydrophobic drug was rapid in both hydrogen sulfide and control environments, with 50% of the encapsulated drug released by 3 hrs. In PBS, particles released 90 % of drug by 24hrs, while in the presence of H_2S 100% was released by 12 hrs.

Conclusions

A series of aryl azide containing polymers were synthesized that self-assemble into bicontinuous nanospheres. These particles respond rapidly to hydrogen sulfide and disassemble.

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APPENDIX

Previous Australia-Japan Colloids Symposia

No.	Location	Dates	Delegates	Umbrella Meeting
16	Asahikawa, Japan	22nd-24th September 2016	25 Japanese 10 Australians (28 others)	67th Divisional meeting on Colloid and Surface Chemistry (Chem. Soc. Japan)
14	Siem Reap Cambodia	15th-19th November 2015	30 Japanese 19 Australians (14 others)	Western Pacific Colloids WPC2015
13	Nagoya Japan	18th-20th September 2013		
12	Cairns, Australia	21 to 23 November 2011		
11	Chiba, Japan	19-22 September 2010		63rd Divisional meeting on Colloid and Surface Chemistry, the Chemical Society of Japan
10	Adelaide, Australia	1-3 February 2009		Australian Colloid and Interface Symposium 2009 (ACIS2009)
9	Matsumoto, Japan	19-23 September 2007	35 Japanese 16 Australian	60th Divisional meeting on Colloid and Surface Chemistry, the Chemical Society of Japan.
8	Terrigal, Australia	27-30 November 2005	30 Japanese 34 Australian 2 UK delegates	
7	Ube, Japan	8-11 September 2004	110 Japanese 15 Australian	57th Divisional meeting on Colloid and Surface Chemistry (Chem. Soc. Japan)
6	Sydney, Australia	16-17 February 2003	15 Japanese 75 Australian	Australian Colloid and Interface Symposium 2003 (RACI)
5	Kyushu University, Japan	13-15 July 1998	110 Japanese 22 Australian	
4	Sorrento (near Melbourne), Australia	April 1996		
3	Kyushu University, Japan	October 1994	50 Japanese 20 Australian	
2	Sorrento (near Melbourne), Australia	December 1992	12 Japanese 40 Australian 8 from USA&Europe	
1	Kyushu University, Japan	May 1992	60 Japanese 10 Australian	

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