



2024 ANNUAL MEETING OF THE GDR SOPHY

A MEETING @SORBONNE UNIV. ON DEC. 11TH – 12TH 2024

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Location of the meeting: Sorbonne University, in the *Durand amphitheater*, place Jussieu (Paris)

December 11 th	
10:00am-10:15am	<u>Forewords</u> by Sylvain Deville
10:15am-11:15am	Keynote lecture #1: Architected polymeric foams generated from liquid templates - Aurélie Hourlier-Fargette (ICS, Strasbourg)
11:15am-11:35am	Biomimetic Fiber-Reinforced Foam: A High-Strength, Low-Density and Humidity-Responsive Material for Scalable Applications Jordan Danae Dornak (Aato Univ.)
11:35am-11:55am	Study of biomimetic membranes composed of glycosylated lipids Ramanujam Ramanujam
11:55am-1:20pm	Lunch provided on site
1:20pm-1:40pm	Colloidal assembly under electrodiffusiophoresis - Sophie Marbach (Phenix, CNRS)
1:40pm-2:00pm	Towards a new family of ionic colloidal crystals based on long-chain polyelectrolytes and small spherical nanoparticles Florent CARN (MSC Paris 7, CNRS)
2:00pm-2:20pm	Out-of-equilibrium assembly: from cooperative motion to percolated 2D networks - Laura Alvarez (CRPP, Pessac)
2:20pm-2:40pm	Mechanical tuning of residual stress to control memory and aging in soft glassy materials - <i>Paolo Edera</i> (ESPCI, Paris)
2:40pm-3:00pm	Structure and mechanics of a hydrogel/nanoparticle double network Gonzalo Sánchez Vera (C3M, ESPCI, Paris)
3:00pm-3:20pm	Soft Solid Marangoni Stresses - Nicolas Bain (ILM, Lyon)
3:20pm-3:50pm	Coffee Break provided on site & poster session
3:50pm-4:10pm	Fluid Mechanics for Functional Materials - Claas Willem Visser (U. of Twente)
4:10pm-4:30pm	Millimetric marble gliding in a soap film - Youna Louyer (IPR, Rennes)
4:30pm-4:50pm	Multi-scale simulation of geopolymers formation starting from the atomic scale – Romain Dupuis (LMGC, CNRS, U. Montpellier)
4:50pm-5:10pm	From soft to hard in Saint-Gobain materials: scientific questions Laurence Talini (SVI, Saint-Gobain Recherche / CNRS)
5:10pm-5:40pm	Future actions by GDR SoPhy. Panel discussion with SoPhy's board

7:30pm-9:30pm: Cocktail on the 24th floor of the Zamanski tower

December 12th

9:00am-10:00am	Keynote lecture: From amorphous calcium carbonate precursors to mineralized biological materials: the case study of sea urchin biominerals Marie Albéric (LCMCP, Sorbonne-Univ. Paris)
10:00am-10:20am	Confinement-induced phase separation: Evaporation-driven colloidal skin formation - Raphael Saiseau (U. of Twente)
10:20am-10:40am	Coffee break provided on site & poster session
10:40am-11:00am	Freezing of gas bubbles in a liquid - Bastien Isabella (ILM, Lyon)
11:00am-11:20am	The shape of a frozen hydrogel - Axel Huerre (MSC, Paris 7, CNRS)
11:20am-11:40am	Designing amine-rich double hydrophilic block copolymer, a novel route to functionalize mesoporous materials Julien Schmitt (Institut Charles Gerhardt, Montpellier)
11:40am-12:00pm	Lyotropic behavior of PICs micelles – Martin In (CNRS / L2C, Montpellier)
12:00pm-12:15pm	<u>Closing remarks</u> by Sylvain Deville
12:15pm-2:00pm	Lunch provided on site

Posters

- P1 Orange Carotenoid Protein encapsulated in mesoporous materials: a possible new way of OCP production Elizaveta Grishaeva (LRS, Sorbonne U., UPMC, CNRS)
- P2 From liquid to solid foams: towards new mechanically auto-assembled architected polyurethane foams with fibers Sébastien Vergne (ICS, Strasbourg)
- P3 Soft matter & hard matter composite to model a membrane-less organelle for biomineralization Yeseul Park (CRPP, Pessac)



Study of biomimetic membranes composed of glycosylated lipids

Ramanujam Ramanujam^{*1}

¹PhD student – India

Abstract

The research activity focuses on studying biomimetic membranes. Compared to other classical amphiphiles, the self-assembly behavior of microbial glycolipids has not yet been studied in depth. Some microbial glycolipids (MGs), i.e., rhamnolipids or single-glucose-containing lipids, can assemble themselves into membranes, which could modify the structure and properties of biological membranes upon interaction. The physical properties of biomimetic membranes solely composed of MGs have not been studied thoroughly, yet. Some physical properties i.e., bending rigidity, the thickness of the membrane, area/molecule, etc., still need to be studied, along with the impact of the MGs on phospholipid-based biomimetic membranes.

^{*}Speaker

Millimetric marble gliding in a soap film

Youna Louyer^{*1}, Benjamin Dollet, Isabelle Cantat, and Anaïs Gauthier

¹Institut de Physique de Rennes – Université de Rennes, Centre National de la Recherche Scientifique – France

Abstract

Foam based materials are a promising tool in the building industry for their thermal and acoustical performance, or simply because they are lighter. Such materials are usually made from a suspension of solid particles, which produces a solid foam when it is dried. To understand how particles interact and organize in the foam, we need to understand the interaction between the liquid films and the solid marbles.

In our experiment, we study the interaction between a horizontal soap film and one or several millimeter-sized marbles. We focus on both the static position (figure1.a) of the beads and their dynamics, and characterize the forces at the origin of their motion. In particular, we demonstrate that the gravitational distortion of the film due to its own weight - a small deformation, generally neglected when considering soap film shapes - is the cornerstone to explain the dynamics of a single particle. Counter-intuitively, we expect the relative impact of the film weight to gain importance as the soap film and particle size are reduced.

We finally focus on the interaction between two particles. We show that the interaction force between two marbles can be deduced from the orbits that spontaneously appear when they approach each other (Figure 1.b). At large distances, the attractive force arises from the film deformation (due to its own weight or from the particles weight). At small distances, the interaction between the two menisci around the marbles causes a much stronger attraction.

^{*}Speaker

Soft Solid Marangoni Stresses

Bain Nicolas^{*1}

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Abstract

Despite its importance in any adhesion and wetting phenomena, there is a fundamental property that is not yet understood in soft solids: surface elasticity. Also called the Shuttleworth effect, surface elasticity is intimately linked to the solid physico-chemistry and can be boiled down to one question. Does stretching the surface of a soft solid change its surface tension? In 2021, we demonstrated that the mechanical response of a textured silicone gel could only be explained by an elastic surface(1). It is, however, still unclear whether the measured surface elasticity is a true material property or a mere consequence of the surface preparation. This presentation will focus on a novel experimental setup that exploits Marangoni stresses and local TFM techniques to characterize the surface mechanics of pristine surfaces.

<u>Références</u> :

(1) Nicolas Bain, Anand Jagota, Katrina Smith-Mannschott, Stefanie Heyden, Robert W. Style, and Eric R. Dufresne, Phys. Rev. Lett. 127, 208001 – Published 8 November 2021.

Fluid Mechanics for Functional Materials

Claas Willem Visser^{*1} and Jieke Jiang

¹University of Twente – Netherlands

Abstract

Multi-scale materials exhibit length scales between the molecular size and the part size. These are ubiquitous in nature, because diverse functionalities, efficient material use, and high performance can be integrated integrated within a single material. However, fabricating these is challenging.

I will present an overview of our research on in-air microfluidics, in which droplets and bubbles are ejected into the air and subsequently converted into functional 3D multiscale materials. The first part will address design and fabrication of micro-particles with controlled shape, size, morphology and composition. These particles are for example used in cell encapsulation, controlled release, and CO2 capture. The second part will address how ejected bubbles are 3D-printed into functional multiscale materials. Here, the focus is on polymer foams, for which subtle mechanisms (oxygen inhibition) were exploited to control the open-to-closed cell transition.

^{*}Speaker

Out-of-equilibrium assembly: from cooperative motion to percolated 2D networks

Laura Alvarez *1,2

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Abstract

The fascinating out-of-equilibrium self-organization processes found in nature are driven by complex interactions. These intricate behaviors can be mimicked using synthetic soft matter systems to design new adaptive and smart materials, harnessing the fundamental properties found in their biological analogues. Here, we explore the out-of-equilibrium assembly of two types of colloidal model systems: rod-like colloidal particles and Janus active colloids. By tuning particle interactions with AC electric fields, we induce a diverse range of microstructures and dynamic behaviors.

In particular, we elucidate the formation of 2D porous percolated networks of rod-like particles, leveraging the inherent polydispersity of the colloidal suspension (1). This work experimentally demonstrates that polydispersity is essential for percolation in systems of anisotropic particles, validating previous theoretical predictions (2). Additionally, we explore a new form of collective behavior in Janus colloids, leading to the formation of polar clusters, particularly when Janus particles with two distinct swimming speeds are present. Experimental observations, supported by a Viseck-like ABP model, reveal how interactions between fast and slow particles drive cooperative dynamics and species segregation (3).

These works illustrate how the fine-tuning of particle interactions and careful material design can direct self-assembly, enabling the creation of both static structures and dynamic, coordinated systems, and offering pathways for designing advanced materials with programmable collective properties.

(1) A. V. Kyrylyuk, P. van der Schoot. PNAS ,105, 8221 (2008)

(2) L. Alvarez^{*}, D. Levis^{*}, E. Sesé, I. Pagonarraba, L. Isa (in preparation)

(3) J. Fojo, L. Rodríguez-Arco, M. Fernández, C. Fernández-Rico^{*}, L. Alvarez^{*} (in preparation)

Freezing of gas bubbles in a liquid.

Bastien Isabella^{*1}, Sylvain Deville, and Cécile Monteux

¹ILM – Université Claude Bernard - Lyon I – France

Abstract

In many different industrial or scientific fields, such as environmental science, metallurgy, or even food preservation, some gas bubbles can be trapped in materials during the solidification process.

In this talk, I will present our work about the study of the dynamics of nucleation of gas bubbles during freezing and especially near the water/ice solidification front. To achieve this, we use confocal fluorescence cryomicroscopy to observe in situ the behavior of air or gas bubbles during all stages of the freezing process. More particularly, we are studying the nucleation phenomena induced by the freezing and the interaction of these nucleated bubbles with the solidification front. In addition, the impact of gas diffusion and freezing parameters on bubble nucleation and engulfment will also be discussed.

These observations will help us to better understand the physical and chemical mechanisms involved in freezing liquids containing dissolved gases, or bubbles, and to determine how to control or restrain the engulfment of these bubbles during solidification.

From soft to hard in Saint-Gobain materials: scientific questions.

Laurence Talini^{*1}

 $^1 {\rm Surface}$ du Verre et Interfaces – SAINT-GOBAIN, Centre National de la Recherche Scientifique – France

Abstract

 $^{^*}Speaker$

Multi-scale simulation of geopolymers formation starting from the atomic scale

Romain Dupuis $^{\ast 1},$ Anthony Saliou , Katerina Ioannidou , Roland Pellenq , and Arnaud Poulesquen

¹Laboratoire de Mécanique et Génie Civil – Centre National de la Recherche Scientifique, Université de Montpellier – France

Abstract

Geopolymers are a type of alkali aluminosilicate polymer that are synthesized from a liquid precursor. Over the past several years, we have been conducting a joint theoretical and experimental investigation into the formation of geopolymers at various length and time scales. This work presents significant challenges due to the complex nature of geopolymer formation.

Our first objective was to separate the high-pH effect, caused by the alkali-based solutions used to dissolve the metakaolin, from the aluminum effect. We found that these two effects counteract each other. Through simulations of aluminosilicate formation at high pH, using parallel tempering molecular dynamics, we observed the formation of grains with a size of approximately 1 nm, which is consistent with values measured using small-angle X-ray scattering (SAXS).

In addition, we have been examining the interaction between two aluminosilicate grains in solution from a fully atomistic perspective using metadynamics. This has allowed us to reconstruct a potential of mean force for the interaction between two grains at the scale of a few nanometers. For example, we have observed that the size and aggregation of grains is controlled by pH in a consistent manner with respect to the experiments and simulations at the molecular level.

Our multi-scale simulation approach provides new insights into the mechanisms governing the formation and properties of geopolymers. This understanding can guide the design and optimization of new geopolymer materials and processes.

From liquid to solid foams : towards new mechanically auto-assembled architected polyurethane foams with fibers

Sébastien Vergne^{*1}, Guillaume Cotte-Carluer¹, Antoine Egele¹, Leandro Jacomine¹, Wiebke Drenckhan¹, and Aurélie Hourlier-Fargette¹

¹Institut Charles Sadron, Strasbourg – CNRS – France

Abstract

Polyurethane (PU) is one of the most widely used polymer materials, valued for its thermal and mechanical properties, with applications in numerous fields including biomedical engineering and construction. Traditionally, PU foams are obtained by chemical foaming, but this technique does not allow control of the foam morphology. Liquid foam templating with physical foaming is an alternative method which consists in producing cellular materials by introducing gas bubbles into a polymer matrix, which then solidifies to achieve desired structural characteristics. Here, we develop a system to generate homogeneous PU foams by liquid foam templating, with a special focus on the mixing of the precursors, as this mixing determines the starting time of the solidification process. We study quantitatively the efficiency of the millifluidic mixing between the two polyurethane precursors with static mixers and show that increasing the number of mixing elements makes the emulsion more homogeneous. We optimize the bubbling at the outlet of the mixing part. Additionally, we show an example of a foam with a single fiber, as a first step towards the creation of new architected materials made of foams and fibers.

^{*}Speaker

Colloidal assembly under electrodiffusiophoresis

Carlos Silvera Batista , Kun Wang , Hannah Blake , Vivian Ndosu-Madueke , and Sophie $\mathrm{Marbach}^{*1}$

¹PHysicochimie des Electrolytes et Nanosystèmes InterfaciauX – CNRS – France

Abstract

Electric fields can tune colloidal interactions and forces leading to diverse static and dynamical structures. So far, however, field-driven interactions have been limited to dipoledipole and hydrodynamic contributions. Here, we propose that under the right conditions, electric fields can also induce interactions based on local chemical fields and diffusiophoretic flows (1). We use a simple approach where faradaic reactions at electrodes induce 3D chemical gradients of pH, that we can measure via fluorescent probes.

Global pH gradients lead to 2D focusing of a particle suspension away from the electrodes through electrodiffusiophoresis. Simultaneously, the electric field induces *local* pH gradients by driving the double layer of particles far from equilibrium. Such local pH gradients induce diffusiophoretic transport, leading to aggregation in the 2D plane. By adjusting the voltage and frequency of the electric field, we can tune the effective interactions and observe either very dynamic aggregates or a collective chemotactic-like collapse of particles. Remarkably, such collapse occurs without reactions at a particle's surface.

This diversity of behavior unlocks a broad set of possibilities to engineer diffusiophoresis and colloidal assembly by externally-controlled electric fields.

(1) Batista, C. A. S., Wang, K., Blake, H., Nwosu-Madueke, V., & Marbach, S. (2025). Artificial chemotaxis under electrodiffusiophoresis. *Journal of Colloid and Interface Science*, 677, 171-180.

Towards a new family of ionic colloidal crystals based on long-chain polyelectrolytes and small spherical nanoparticles.

Florent $Carn^{*1}$

¹Matière et Systèmes Complexes – Université de Paris, CNRS : UMR7057 – France

Abstract

The mixing of polyelectrolyte chains and oppositely charged nanoparticles in aqueous solution results in the formation of electrostatic assemblies. The size, shape and compactness of these assemblies can vary greatly with the Debye length, the concentration ratio of the partners and the characteristics of each partner (i.e. charge density, size, shape, persistence length of the chains).1 It is also known that the nature of the phase separation observed around the charge stoichiometry can vary from one system to another (i.e. liquid/liquid or liquid/solid). However, to our knowledge, this type of 'asymmetric association' between long flexible linear chains and small spherical particles has not been known until now, either theoretically or experimentally, to allow the formation of assemblies with an ordered structure on large spatial scales (i.e. a colloidal crystal). The aim of this talk is to present a recent result showing that, contrary to what is generally observed, it is possible to form colloidal crystals by the electrostatic coupling of a semi-flexible polyanion, hyaluronic acid, and gold nanoparticles coated with a self-assembled layer of cationic ligands.2 This result is all the more remarkable given that the size distribution of the two partners is not particularly narrow and the polyelectrolyte contour length (L ≈ 230 nm) is large compared to the particle size ($R \approx 4$ nm). On the basis of small-angle X-ray scattering (SAXS) measurements, I will show in which region of the state diagram these crystalline structures were formed and highlight the role played by the flexibility of the PEL chains in the formation of these ordered structures.

For instance: Skepo, M. et al. Complexation, Phase Separation, and Redissolution in Polyelectrolyte-Macroion Solutions, Macromolecules 2003, 36, 508-519

• Shi, L.; Carn, F.; Boue, F.; Buhler, E. Gold Nanoparticle–Polyelectrolyte Complexes with Tunable Structure Probed by Synchrotron Small-Angle X-ray Scattering, ACS Applied Nano Materials 2023, 6, 3990

^{*}Speaker

The shape of a frozen hydrogel

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¹MSC – Université de Paris, CNRS : UMR7057 – France

²Institut Jean le Rond d'Alembert – CNRS UMR 7190, Institut Jean Le Rond d'Alembert, Sorbonne Université – France

³Institut Jean Le Rond d'Alembert – Sorbonne Universite, Centre National de la Recherche Scientifique, Centre National de la Recherche Scientifique : UMR7190 – France

Abstract

Hydrogel is an elastic material composed of a polymer matrix with the ability to absorb a large amount of water, which can make up 99% of the gel mass in the case of superabsorbent hydrogels. Repeated cycles of freezing and thawing on hydrogel pieces lead to the formation of a very specific fibrous structure allowing them to mimic properties of many human tissues such as muscles. They are therefore a useful tool in medical science for tissue engineering.

However, the physics behind the freezing of a hydrogel remains to be understood. Here, we present experimental results on the freezing of hydrogel structures under various conditions. We mainly study agar gels with an agar concentration between 7 and 50 g/L. We cool a copper substrate to a temperature ranging from -80 to $0 \circ C$ on which we place the gel. We then measure the freezing front and study the deformation of the material.

We will discuss how the geometry can influence the freezing process as hydrogels can be molded into various geometries. We explore the deformation of this elastic material under internal expansion due to the change of water in ice by characterizing the typical modification of the overall shape. We further develop a model taking into account the expansion of the ice and the elastic shape of the material. This model is able to predict the frozen shape with a great accuracy. Differences with the pure water case are highlighted.

Orange Carotenoid Protein encapsulated in mesoporous materials: a possible new way of OCP production

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Abstract

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Orange Carotenoid Protein (OCP) is a two-domain carotenoprotein that acts as a photoprotecting agent in cyanobacteria. It is made up of a single, 300 amino acid polypeptide and a ketocarotenoid that works as chromophore. The photoprotective mechanism of OCP is based on photo-induced protein structural rearrangement and carotenoid internal translocation during which the stable, resting orange form turns into light-activated red form. The OCP red form binds to the phycobilisome (cyanobacterial light-harvesting system) and help dissipation of excess energy under strong illumination conditions. The relative structural simplicity and water-solubility make OCP a peculiar light-sensitive system in several fields, ranging from sensors to optogenetics.

The mechanism of OCP photoactivation is still under debate, and recent works suggest that it might change with the nature of the bound ketocarotenoid. In this framework, the availability of OCP binding different ketocarotenoids is particularly important.

The usual way to produce OCP is by expression in *E.coli*, but this approach has limitation on the kind of carotenoid that the formed OCP can bind. On the other hand, OCP extraction from cyanobacteria require extensive purification steps and gives limited amounts of purified OCP.

An alternative, "synthetic" procedure to build OCP complexes was studied in this work. The two building blocks of OCP – Apo-OCP (which is relatively easy to express from bacteria) and a carotenoid (canthaxanthin in this work) – were introduced in two types of

^{*}Speaker

mesoporous material: Mesoporous Siliceous Foam (MSF) and Metal Organic Frameworks (MOFs).

OCP was found to self-assemble inside MSF, and current efforts are aimed to develop an extraction procedure from the silica matrix. Studies on MOFs has shown the possibility to encapsulate OCP; furthermore, carotenoid exchange upon exposure to organic solvent was observed.

Confinement induced phase separation: Evaporation-driven colloidal skin formation

Raphael Saiseau^{*1}, Lorenzo Botto, and Alvaro Marin

¹University of Twente – Netherlands

Abstract

When a droplet containing a suspension evaporates on a super-hydrophobic substrate, a 3D colloidal cluster of varying porosity and geometry depending on the initial concentration, evaporation rate or inter-particles interaction (1,2,3). Using a bidisperse suspension also offers supplementary degrees of control on the resulting cluster shape allowing energy-efficient methods to make new materials through particles' self-assembly (4,5).

We present an analytical approach of the advection-diffusion problems in a confining environment which is then applied to evaporation driven colloidal particles' self-assembly. The resulting colloidal distribution will be compared to Brownian particles dynamics simulations as a test case scenario. The transport of the colloids in an evaporating droplet is then considered by adding particles hydrodynamic interactions, resulting in non-trivial dependance of the concentration distribution on the initial packing fraction.

Lastly, by applying these results to the formation of a glassy layer, we will present cases leading to delayed shell formation scenario which is of utmost importance to correctly predict the skin accumulated mechanical stresses during the droplet evaporation.

(1): Seyfert, Carola, et al. "Evaporation-driven colloidal cluster assembly using droplets on superhydrophobic fractal-like structures." *Soft Matter* 17.3 (2021): 506-515.

(2): Lintingre, E., et al. "Control of particle morphology in the spray drying of colloidal suspensions." *Soft Matter* 12.36 (2016): 7435-7444.

(3): Sekido, Takafumi, et al. "Controlling the structure of supraballs by pH-responsive particle assembly." *Langmuir* 33.8 (2017): 1995-2002.

(4): Liu, Wendong, et al. "Segregation in drying binary colloidal droplets." $ACS\ nano\ 13.5$ (2019): 4972-4979.

(5): Liu, Wendong, Michael Kappl, and Hans-Jurgen Butt. "Tuning the porosity of supraparticles." ACS nano 13.12 (2019): 13949-13956.

Biomimetic Fiber-Reinforced Foam: A High-Strength, Low-Density and Humidity-Responsive Material for Scalable Applications

Jordan Danae Dornak^{*1}, Juha Koivisto², Karen Charlot-Wauquier^{*}, Ammar Hassan, Eelis Lehtinen , and Ali Aaraf Gillani

> ¹Department of Media [Aalto] – Finland ²Department of Applied Physics [Aalto] – Finland

Abstract

Biomimetic design, inspired by natural structures, allows us to develop advanced materials. This research draws on the anisotropic closed-cell structure of wood to create a dynamic foam that exhibits material properties characterized by high strength and low density. Current observations present in foams up to 40 cm in size are capable of withstanding curvatures up to 30° in bending. Existing research aims to scale this technology to produce samples spanning several meters. The bubbles of the fiber reinforced foam are elongated using temperature dependent viscosity of methylcellulose and constricted drying. This method induces anisotropic alignment, improving the foam's yield strength by up to 64 times along the primary axis compared to the cross direction. Observations concluded that the oriented fiber-reinforced structures exhibited load-bearing capacity and resilience in bending. Additionally, the foam undergoes repeatable, humidity-driven dynamics: it bends up over approximately 4 hours in response to rising humidity and returns to its original position in around 15 minutes as humidity decreases. This manufacturing approach yields a foam material with high strength and low density capable of withstanding significant curvatures. The foam's closed cell structure composition has resulted in excellent thermal insulation. Recent experiments also confirm the foam's reactivity under humidity cycles. The proposed novel foam manufacturing process can be easily scaled up from laboratory trials to industrial production volumes. Its adaptability to curved shapes make it a promising option for applications that require lightweight, durable components in extended lengths and flexible design specifications.

References

Reichler, M., Rabensteiner, S., Törnblom, L., Coffeng, S., Viitanen, L., Jannuzzi, L., Mäkinen, T., Mac Intyre, J. R., Koivisto, J., Puisto, A., & Alava, M. J. (2021). Scalable method for biobased solid foams that mimic wood. Scientific Reports, 11(1). https://doi.org/10.1038/s41598-021-03764-0

Miranda-Valdez, I. Y., Coffeng, S., Zhou, Y., Viitanen, L., Hu, X., Jannuzzi, L., Puisto, A., Kostiainen, M. A., Mäkinen, T., Koivisto, J., & Alava, M. J. (2023). Foam-formed biocomposites based on cellulose products and lignin. Cellulose, 30(4), 2253–2266. https://doi.org/10.1007/s10570-022-05041-3

Structure and mechanics of a hydrogel/nanoparticle double network

Gonzalo Sánchez Vera^{*1}

¹Chimie Moléculaire, Macromoléculaire et Matériaux (UMR7167) – ESPCI Paris, PSL Research University – France

Abstract

When polymer chains or colloidal particles dispersed in a solvent connect with each other to form a system-spanning network, the sample changes its macroscopic appearance, turning from a liquid to a gel with predominantly solid-like properties.

The rheology of thus-formed polymer or colloidal gels depend strongly on the interaction between elementary constituents, as well as on their concentration. When polymers are crosslinked by strong covalent interactions to form diluted hydrogels, they result in high stretchable elastic solids that can withstand quite large deformations before fracturing. On the other hand, colloidal gels are typically characterized by more complex microstructures and high sensitivity to the imposed deformation, and yield under a moderate shear strain.

The synergetic effect arising from mixing polymers and colloidal particles to make composite material is well-known. However, its microscopic origin and its effect on weakly non-linear rheology of composite materials is still poorly understood. This is partly due to the experimental challenge associated to the structural complexity resulting from mixing two species with such different behavior.

In this work, we develop a model double network system, composed of a colloidal gel made of silica nanoparticles interpenetrated with a poly ethylene glycol (PEG) hydrogel. The two networks are formed in controlled conditions to give reproducible structures and mechanical properties across a wide range of compositions. We probe the time-dependent formation of the multiscale structure through a combination of X-ray, neutron and light scattering, and we relate it to the evolution of the linear viscoelastic moduli probed by rheology.

This work paves the way for a comprehensive study of structure-function relationship in composite materials deformed in the linear and weakly non-linear regime, as I will show in my preliminary experimental results.

^{*}Speaker

Mechanical tuning of residual stress to control memory and aging in soft glassy materials

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²Chimie Moléculaire, Macromoléculaire et Matériaux (UMR7167) – Ecole Supérieure de Physique et de Chimie Industrielles de la Ville de Paris, Institut de Chimie - CNRS Chimie, Centre National de la Recherche Scientifique – France

Abstract

Soft Particle Glasses are out of equilibrium systems. This makes their nonlinear mechanical properties evolving in time (ageing) and directional dependent (Bauschinger effect). What is the origin of these phenomena? Is there a way to control the mechanical state of the material ?

Combining systematic rheological experiments and particle dynamics simulations we introduce a conceptual framework to rationalise these phenomena in terms of internal stress distribution.

Based on this understanding we develop a protocol to control both the directional bias of mechanical properties and aging.

The protocol, introduced for SPG (simple yield stress fluids), is generalised to be applied also to thixotropic materials.

Lyotropic behavior of PICs micelles

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Abstract

Polyelectrolyte complexes micelles (PICs) have been used as structuring agents for more environment friendly synthesis of silica-based mesoporous materials. The porosity of the final material is revealed by simple elution in water and the polyelectrolytes are partially recycled. Porous hybrid materials of various structures from cubic to lamellar have been obtained either by modulating the structure of the copolymers used or by adjusting the physicochemical conditions of the reaction medium. The control over the structure of the final material is expected to relate to the lyotropic behavior of the assemblies just as observed when micelles of amphiphilic molecules are used as templates. Lyotropic behavior has rarely been reported for PIC micelles. In an attempt to establish the relationship between the lyotropic behavior of micellar polyelectrolyte complexes and the structure of the materials obtained we characterize the lyotropic behavior of {PEO-b-PAA / oligoamine} systems. Micelles were studied by light, X-ray scattering in a broad range of concentration and dilution laws have been established. Mesophases formation was observed when the oligoamine is a well-defined molecule such as neomycine but not when it consists of oligochitosan. In the absence of silica, the lyotropic behavior of PIC micelles is less rich and also much less pronounced than the ones of amphiphilic copolymers. One possible explanation is the high compressibility of micelles the core of which contains water.

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