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Collection of Abstracts

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Ca’ Foscary University of Venice, Italy

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Surprises from polymer topology and hydrodynamics

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Linear polymers in shear flow display a dominant mode of dynamics known as tumbling, around the vorticity axis, whereby the two ends exchange their places, accompanied by a temporary compression of the chain in the gradient direction. We will demonstrate that topological polymers respond to shear in dramatically different ways, emerging from a coupling between topology and hydrodynamics. In particular, we will discuss ring polymers as well as rings connected either chemically (bonded rings, BR) or mechanically (catenated rings, CR). Rings display vorticity swelling and an inflated phase that suppresses tumbling and Brownian motion. BRs tumble around an axis parallel to the gradient direction, whereas CRs show slip tumbling while maintaining their overall orientation and shape. These unusual phenomena all result from proper consideration of hydrodynamic interactions and they disappear if the latter are (artificially) switched off.

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RIANA

The New Research Infrastructure for Nano-Material Characterization

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RIANA - Research Infrastructure Access in Nanoscience & Nanotechnology - is an infrastructure project funded by the European Commission providing scientists with access to cutting edge experimental and computational instrumentation. The RIANA consortium encompasses 56 beneficiaries organized in seven networks and offering access to 69 infrastructures in 22 European countries.

The available installations allow the characterization of both soft matter and solids and also include access to supercomputer computing time.

In addition to mere access to infrastructures, RIANA offers a service for inexperienced users to help them identify the most suitable experiments, conduct them and analyze and interpret collected data.

Figure 1: *The RIANA consortium provides access to 69 infrastructures in 22 European countries*

Applications for measurement time have to be submitted through the RIANA web-portal under `https://riana-project.eu/` and will be reviewed by a panel of international experts. After successful application, the usage of instruments will be free of charge for the users, and RIANA will make a financial contribution to the traveling expenses of two operators per experiment.

Direct observation and control of non-classical crystallization pathways in binary colloidal systems

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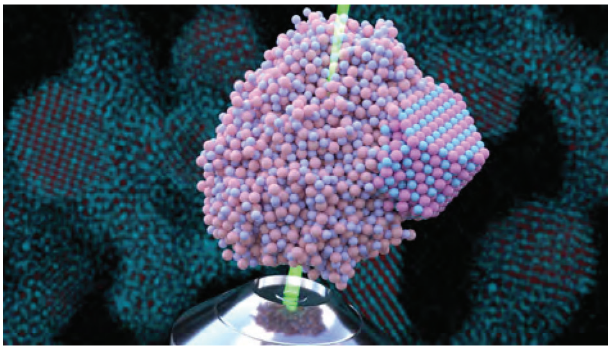
Crystallization stands as a prime example of self-assembly, where elementary building blocks converge to orchestrate order from chaos. Although classical theories frame this process as a particle-by-particle addition, recent evidence points to more complex, non-classical pathways. Using index-matched colloids bearing stable positive and negative charges, we directly observe ionic crystallization in real time through confocal microscopy, effectively granting "X-ray vision" at sub-micron length scales. We not only confirm structures by comparing their simulated scattering patterns to known atomic arrangements, but also track defect motion and twinning events as crystals form and melt.

Our findings reveal that ionic colloidal crystals form predominantly via a two-step process. First, metastable amorphous "blobs" condense from the dilute phase; these blobs then reorganize internally to seed small crystalline domains. Subsequently, these crystalline seeds grow into large faceted structures through three parallel mechanisms: addition of monomers from solution, absorption of surrounding blobs, and oriented attachment of other crystals. These processes occur both in bulk and on surfaces across a range of particle sizes and interaction strengths.

Leveraging a continuous dialysis strategy, we gain precise spatiotemporal control over the salt concentration—and thus over the colloidal interaction potential—allowing us to uncover diverse crystal polymorphs in a single experiment. Intriguingly, we observe a previously unreported open-lattice phase with hollow channels and complex heteroepitaxial growth of composite architectures. Taken together, these results illuminate the intricate pathways of non-classical crystallization and provide a robust experimental platform to image, analyze, and tune crystallization in real time.



Colloidal clusters from confined self-assembly: Structure – Thermodynamics – Formation kinetics

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The spontaneous organization of individual building blocks into ordered structures is extensively used in nature and found at all length scales, from crystallization processes, via composite materials, to living cells constituting complex tissue. Understanding the relationship between building blocks, environmental conditions, and resulting structure is of fundamental importance for controlling materials properties. Confining elements imposed upon the self-organizing particles can significantly alter the assembly process and may lead to entirely different structures. Of particular interest are spherical confinements, such as emulsion droplets, where curvature and interfaces impose restrictions on the ability of colloidal particles to crystallize.

Here, we explore the surprising diversity of crystal structures and symmetries that can form in this confining element. We create a phase diagram of observed crystal phases in dependence of the number of colloidal particles within the confinement and support our model by event-driven molecular dynamics simulations of hard-spheres in a spherical confinement. A closer look at the thermodynamics in such systems shows that certain configurations exist as minimum energy structures, a signature associated with magic number clusters which are well known in the atomic world, but have not been observed in the colloidal realm. Importantly, and differing from their atomic analogues, the occurrence of such magic number states is not driven by the mutual attraction of the individual building blocks. Instead, the thermodynamics in our colloidal system is entirely governed by entropy maximisation. In this presentation, I introduce synthetic requirements that are necessary for the self-assembly of magic colloidal clusters and present a detailed study on the structures, thermodynamics and formation kinetics of this confined self-assembly process.

Colloidal quasicrystals: How entropy can lead to forbidden symmetries

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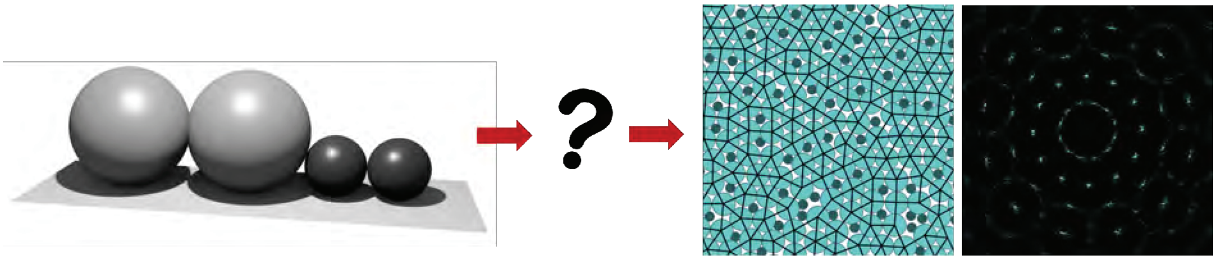
Quasicrystals are one of the most intriguing phases in classical matter. Defying the normal constraint of periodicity that applies to normal crystal structures, they can exhibit rotational symmetries that are forbidden for periodic crystals, e.g. 10-, 12-, 18-, 20-fold symmetries. These symmetries can be easily recognized in their diffraction patterns – and it is this characteristic that led to their original discovery by Dan Shechtman in 1984. While quasicrystals were first found in atomic systems, they have since been found in a growing number of soft-matter systems, including nanoparticles, polymers, and micelles. How simple can a system be and still exhibit such an atypical, complex phase? Why does such a phase form in the first place? In this talk I will address these questions, and many others regarding the appearance of quasicrystals in soft matter. In particular, I will show how even a simple system of hard spheres can be designed to form quasicrystals. I will discuss the huge importance of entropy in their stabilization, and end the talk by delving into the appearance of point defects in these systems – and introducing new simulation methods that will help us address them.



Chemical Design of Self-Regulation in Soft Materials

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Locomotion and the handling of objects requires finely tuned and tightly controlled motions. Such complex motions, while abundant in nature, remain fundamentally challenging to design for in synthetic materials in absence of electronic circuitry and traditional control concepts.[1] Too often, motions are simplistic, fixed, and not tunable, because traditional stimuli-responsive materials show single, material-specific deformations. New approaches for multimodal, multistep deformations in single materials are needed together with new control concepts that build on feedback.[2] In this presentation, I will highlight our recent work on self-regulated and chemically controlled microscale soft actuators. Building on liquid crystalline elastomers, we show how opto-chemo-mechanical feedback can lead to user-defined, on-the-fly tunable deformation trajectories resembling beating cilia and how the same feedback mechanism can give rise to patterned dynamics in arrays of such microactuators.[3] I will then highlight how molecular design specifically can be utilized to impart more than one motion in single material actuators and how such multi-step deformations can interesting motions at both the micro- and macroscale.[4] The novel approaches described herein open up new possibilities for the rational design of complex motion in soft robotics and the simplicity of the underlaying material design raises questions about how complex functions can arise from minimalist designs.

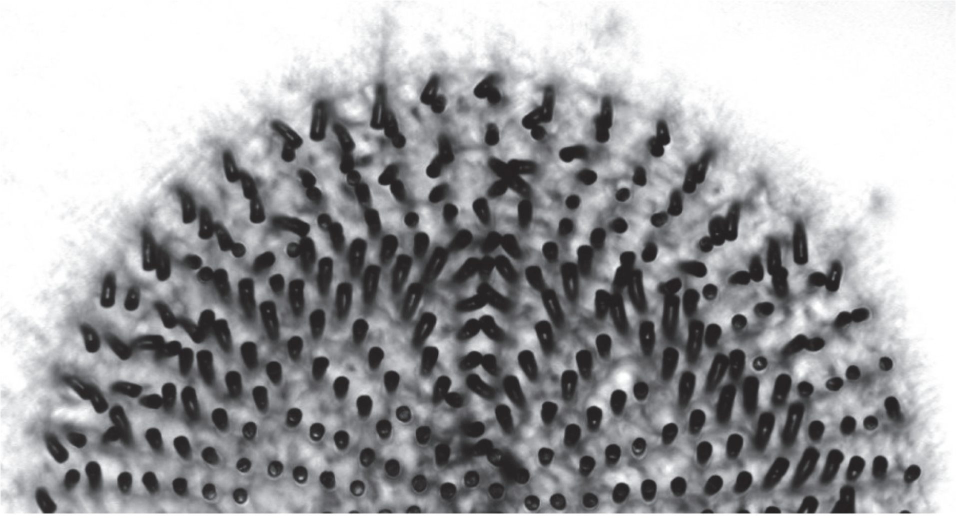

Figure 1: *Light-triggered deformation wave in an array of liquid crystalline elastomer microposts as a result of communication and feedback.[3]*

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The Ductile to Brittle Transition in Jammed Suspension Solids

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Suspensions of non-Brownian particles jam when the volume fraction is above some maximum, ϕ_m, due to inter-particle friction [1]. Such jammed suspensions are solids with finite yield stress, and occur, e.g., in various confectioneries worldwide, from Indian burfi to Scottish fudge and tablet. Such confectionaries are prepared by boiling and cooling a sugar-cream-butter mixture to some $T_{\max}$ with various degrees of mechanical agitation. We find that there is a transition from fluid to ductile solid and then to a brittle solid as $T_{\max}$ increases. In a compression test, the emergence of brittleness is accompanied by the appearance of abrupt fractures, see Figure, and a stress overshoot in the force-distance curve. Comparison with the behaviour of suspensions of non-Brownian calcite particles [2] and simulations shows that this transition is a generic one driven by volume fraction, with the system becoming solid at ϕ_m, and the solid transitioning from ductile to brittle as the particle packing becomes progressively more homogeneous *en route* to random close packing. We discuss these results in the context of growing interest in ductile to brittle transitions in a broad range of amorphous systems, including many soft materials [3].

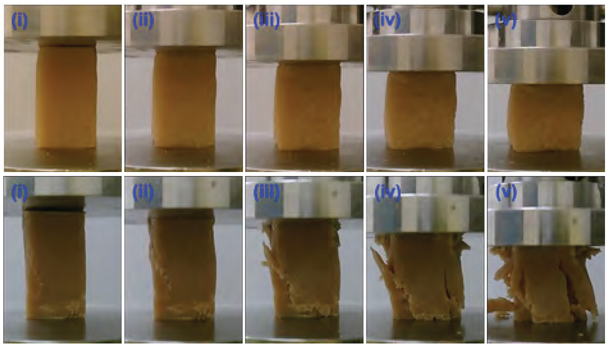

Figure 1: *The ductile (upper sequence) and brittle (lower sequence) failure of fudge – a high-volume-fraction suspension of sugar crystals – as it is compressed in a universal testing machine.*

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Glasses, Colloids and Self-Assembly I

Monday 19 May 15:20 – 16:00

Interdiffusion of polymer and water in waterborne polymer latex films

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Waterborne latex films, obtained from the dispersion of latex particles are of particular interest due to the non-content of volatile organic compounds (VOC), often mandatory under environmental legislation [1]. However, abrupt water penetration inside the films restricting their lifespan and deteriorating the shining of the coating. In order to prepare efficient and solvent-free coatings with the low glass-transition temperature (Tg < the drying temperature) but with higher mechanical strength, we have integrated hydrophilic layers (Acrylic acid/ Poly(acrylamide)) around the hydrophobic cores (mixture of Methyl methacrylate and Butyl acrylate) and also hard shell around the soft core in the latex film. Latex particles with different morphology (hairy layer variants and core-shell particles) have been synthesized using emulsion polymerization [2]. Polymer latex films have been prepared in the next step by evaporating water in a thermo-humidistatic chamber at temperature 25 oC. The structure formation of polymer latex films in the dry state (crystallinity) and in re-swelled state (change in crystallinity and whitening or blushing) have been studied to propose a recipe for the preparation of efficient latex coatings. Small-Angle Neutron Scattering (SANS) study shows the FCC-like structure formation by the latex film, which gets better ordering with the inclusion of the hydrophilic shell. The hydrophilic shell also promotes the formation of the homogeneously water-swollen film and slows down the development of water "pockets", preventing the deterioration of the latex film over time. On the other hand, the inclusion of hard shell protects the latex films from water whitening and provides additional mechanical strength. The interdiffusion between the latex particles has been analyzed by mixing H/D polymers. The transfer of polymer chains through interparticle boundaries that vanishes the crystalline structure and results in a formation continuous material.

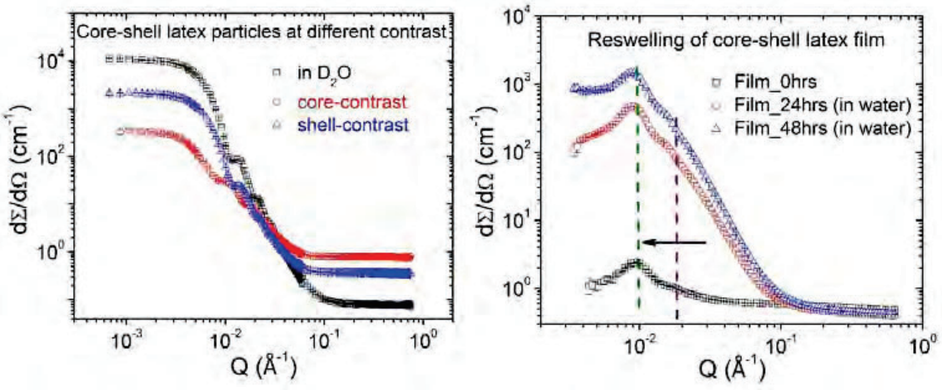

Figure 1: *Small-angle neutron scattering results of (Left) core-shell dispersion under different contrast (Right) structure and hydration of latex films made of core-shell dispersions.*

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First insight on the self-assembly behavior of tannins

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Tannins display very peculiar properties, such as antioxidant and antimicrobial activity, which make them promising compounds for a wide range of applications beyond oenological and tanning products. In particular, the generation of tannin nanoparticles would represent a significant advancement towards the valorization of this class of natural polyphenols. Tannins display a complex and heterogeneous structure which hinders the rationalization of their aggregation behaviour.[1]

Nevertheless, due to structural similarities with lignin, it can be hypothesized that the aggregation mechanism is based on the same driving forces. Specifically, the presence of different functional groups, mostly represented by hydroxyl groups and phenolic units, could give rise to both long-range interactions (e.g. *van der Waals*, electrostatic forces and hydrophobic interaction) and short-range interactions (e.g. H-bonds and π-π interactions). Moreover, the interplay between the mentioned interactions depends strongly on the solvating environment.[2]

In this framework, this study aims at unveiling the self-assembly behavior of tannins by investigating two model compounds, namely tannic acid and quercetin, as these molecules are often found in natural tannins. To this end, a wide range of solvents, sample concentrations, and other variables including pH, ionic strength, and temperature were modified exploiting previous knowledge on lignin aggregation.[3] Through the change of the ionic strength of the antisolvent water used in the synthesis of the nanoparticles, we were able to tailor the size of tannins aggregates, which was found to depend on the tannin structure and on the presence of impurities.

The size of the prepared tannin nanoparticles was analyzed by DLS and correlated to the structure of the starting materials, determined by ^{31}P NMR, ^{1}H/^{13}C HSQC, GC-MS, and HPLC,[4,5] to comprehend the main structural features triggering self-assembly and to derive structure-property correlations.[6]

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Polymers and Biopolymers I

Monday 19 May 15:20 – 16:00

Direct Observation of Short-Time Welding in UHMWPE Powders via X-ray Photon Correlation Spectroscopy and Magnetic Hyperthermia-Induced Sintering

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We at elucidating the rapid welding of ultra-high molecular weight polyethylene (UHMWPE) grains during their sintering by monitoring the dynamics of magnetic nanoparticles located at the interfaces by means of X-ray photon correlation spectroscopy (XPCS). The observation of abnormal fast NP's dynamics emphasizes the decoupling between the rapid material welding above its melting point (≈ 1 min) – crucial for engineering applications – and the much slower re-entanglement process emphasized by Yang et al. (≈ 20 h) [1], resulting in thermodynamic equilibrium. XPCS measurements are performed under both convective and inductive heating [2], and further supported by in-situ SEM observations, offering a comprehensive perspective on the behavior of weakly-entangled polymers.

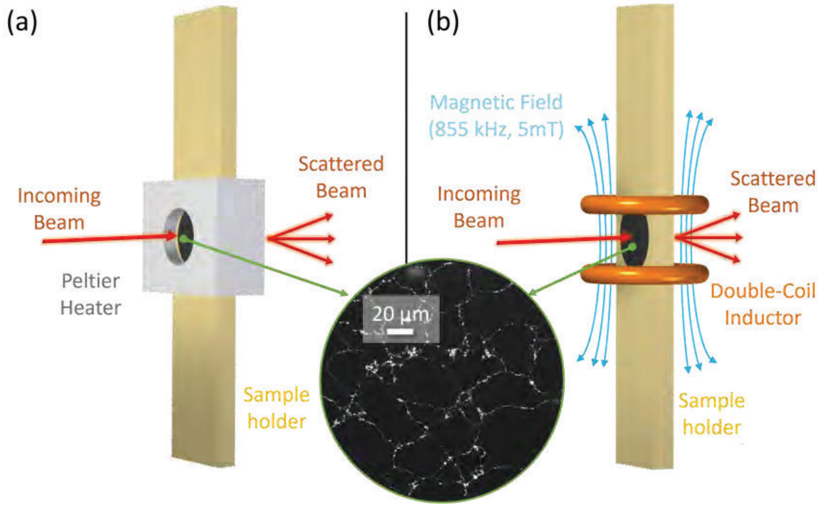

Figure 1: *Schematic view of the Time Resolved-USAXS and XPCS experimental setup including (a) a Peltier heating stage, and (b) a double-coil inductor (orange rings), serving to heat the sample (black disk) held by an epoxy strip. Inset: SEM micrograph of a sintered nanocomposite. NPs appear in white, localized at the polymer grain interfaces.*

References

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Static & Dynamic Behavior of Interacting Polymer Brushes in High Molecular Weight Matrices

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Filler dispersion in rubber compounds may be facilitated by grafting polymer chains on the surface of the filler particles, supposedly altering the interparticle interactions by giving rise to repulsive forces between approaching particles. If these forces are of sufficient magnitude, particle aggregation/agglomeration may be prevented [1]. By zooming onto the thin interface formed between nearby particles, we obtain physical system that may be represented by polymer brushes placed on opposing flat planes. Force/Energy profiles for such a setup have so far been studied theoretically and measured experimentally just for the case of brushes immersed in a low molecular weight (good) solvent. Available results confirm the existence of repulsive interactions that increase monotonically with brush overlap [2]. The proposed mechanism is that brush overlap increases the local concentration of grafted chains, leading to an increase of the solvent osmotic pressure. As a response, the solvent rushes within the pore, causing the observed repulsion. In this work we aim to examine the current picture and further expand it to include the case of brushes surrounded by a chemically identical polymeric solvent. To this end, we perform coarse-grained molecular dynamics simulations, exploring how the properties of the system evolve with separation for different sets of parameters and in both quiescent and dynamic conditions.

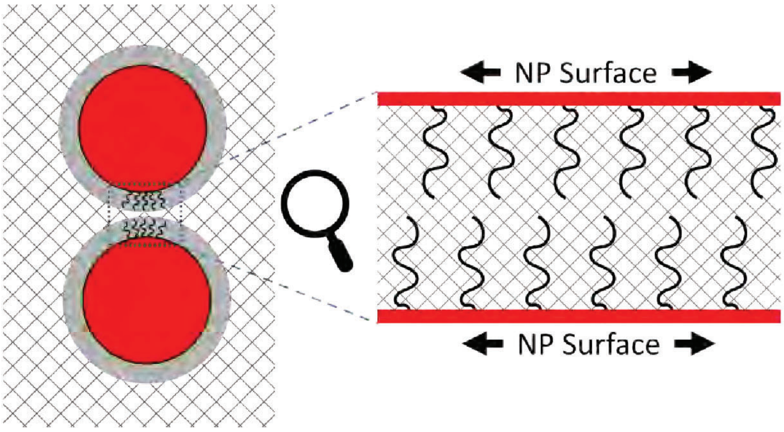

Figure 1: *Opposing polymer brushes forming on the surface of approaching particles*

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Glasses, Colloids and Self-Assembly II

Monday 19 May 16:30 – 18:10

The plastic flow dynamics of silicate glasses at room temperature is *almost* normal

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Silicate glasses (amorphous silica, window glass etc...) undergo plastic flow at room temperature (Fig.) but, in keeping with their brittleness, only at small lengthscales (microns). In the last two decades, we have developed tools to quantify this local plastic response [1]. One of them, pillar microcompression in situ in a SEM [2], turns out to be affected by a curious phenomenon : the yield threshold decreases under electron irradiation [3].

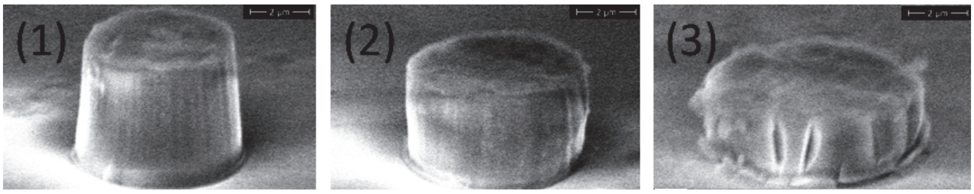

Figure 1: *In situ microcompression of amorphous silica in a SEM (pillar height 4 µm) at room temperature : 1) before compression ; 2) moderate plastic strain ; 3) large plastic strain. At the micron scale, even at large strains, the permanent deformation primarily consists of plastic flow, with very limited damage or fracture. From [2]*

We have carried out extensive experiments which demonstrate that it is the *viscoplastic flow dynamics* which scales with current density : by gradually increasing the irradiation current density, we can peer into the lower and lower strain rate regimes. The results establish a convincing parallel with the flow dynamics in the supercooled liquid state (at temperatures around the glass transition temperature T_g) : we find time-current density superposition (in place of time-temperature superposition), a non-newtonian, homogeneous flow regime with a logarithmic dependence of yield stress with strain rate, and a transition to a localized plastic flow at high strain rates. Interestingly, all these results have parallels in the plastic flow of *e.g.* metallic glasses in the supercooled liquid state, and likewise can be described by standard rate theory [4].

Does it mean that rate theory applies to the viscoplastic flow of silicate glasses at room temperature ? Probably not : very recently, we have measured the *temperature dependence* of the yield stress (as a function of strain rate *i.e.* under irradiation) and we have found that this temperature dependance is *not* consistent with simple rate theory. More precisely, within an Eyring description [5], our data call for a marked increase of the *effective* activation volume with temperature.

In summary, we have found the remarkable result that the dynamics of the viscoplastic flow of silicate glasses at temperatures much lower than T_g is very similar to the dynamics of the viscous flow near T_g. Only much slower... For the post part, it can be described by rata theory.

These considerations echo recent theoretical claims which evidence strong connections between the two regimes [6, 7]. However, in detail, we find that rate theory does not apply because the temperature dependence is not conventional. This observation certainly calls for more advanced descriptions of plastic flow in the glassy state, which are yet to be developed.

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At ultra-low temperatures, glass-forming liquids relax in a simple way

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Glass-forming liquids have only a modest tendency to crystallize and hence their dynamics can be studied even below the melting temperature. The relaxation dynamics of most of these liquids shows at a temperature T_c (close to the theoretical Mode-Coupling Temperature), somewhat above the glass-transition temperature T_g, a crossover, which indicates the conjunction of two different dynamical regimes. For temperatures slightly above T_c, experiments and computer simulations have extensively probed this dynamics on the particle level and identified several universal scaling laws that are often compatible with theoretical predictions.
In this presentation, I will show that those studies can be successfully extended to temperatures below T_c using large scale computer simulations [1]. The main finding is that the relaxation mechanism is qualitatively different from the one found at higher temperatures and that it is characterized by new scaling laws that allow to give a simple description of the relaxation dynamics at very low Ts. Specifically we reveal that the cage-escape process is related to rare but large particle displacements that give rise to a distinctive sub-diffusive power-law in the time correlation functions. This insights will help to advance our understanding on the relaxation dynamics of glass-forming systems at temperatures that are close to the experimental glass transition.

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Aggregation dynamics of colloidal particles in tin perovskite crystalline film formation

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Perovskite materials have emerged as highly promising candidates for several applications due to their distinctive electronic, optical, and structural properties. In the solar cells sector, they are very attractive because of their low costs and excellent performance [1]. Lead-based perovskites have been extensively researched for their superior electron mobility and carrier properties, even though lead causes environmental and health risks. Substituting lead with tin, a viable alternative from the same periodic table group, holds promise. Nevertheless, the elevated crystallization rates of tin-based perovskites make the control of the material's morphology challenging.

We present a novel approach to understand the crystallization of tin-based perovskite films for photovoltaic applications starting from precursor suspensions subjected to spin-coating [2]. By coupling the principles of colloidal theory and suspension fluid dynamics, our approach provides insight into the effects of both chemical and process-related variables on the crystallization mechanism of perovskite suspensions and on the microstructure of the resulting solid materials, thus allowing to identify which ones contribute to the formation of films with enhanced optical and photovoltaic properties [3].

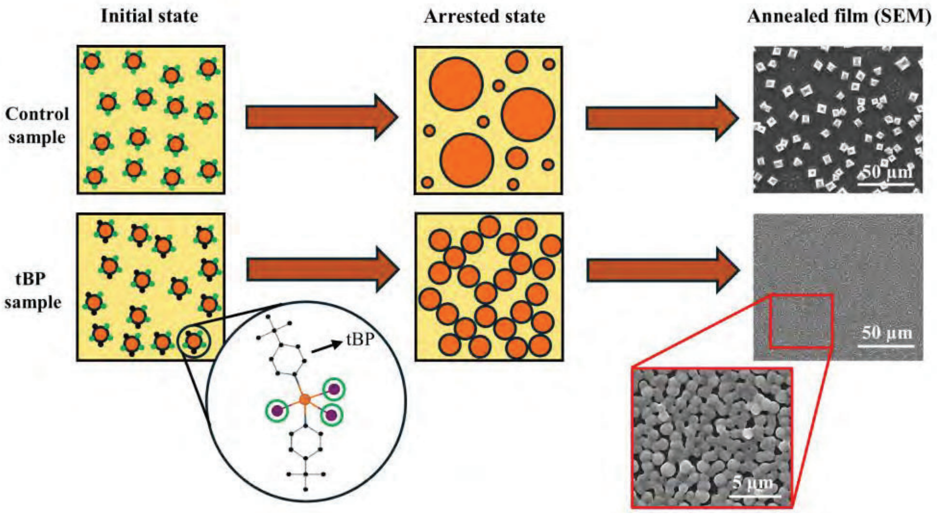

Figure 1: Explanation of the perovskite crystallization mechanism for the control sample (top) and the sample with tBP (bottom) during spin-coating.

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Realizing Quantitative Quasi-Particle Simulations of Skyrmion Dynamics in Arbitrary Potentials

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Magnetic skyrmions are two-dimensional whirls of magnetization which can be topologically stabilized in thin films and visualized with magneto-optical Kerr microscopy. Skyrmions can exhibit thermal diffusion in a spatially inhomogeneous effective energy landscape that stems from pinning effects caused by random material defects [1]. They are also easy to manipulate which makes them promising candidates for novel devices in the context of unconventional computing [1, 2] and for the study of phase transitions in two dimensions [3]. In the so-called Thiele framework, skyrmions are described as two-dimensional quasiparticles which evolve according to an overdamped Langevin equation similar to chiral colloidal systems. In previous work, we have obtained effective interactions between skyrmions and boundaries using Iterative Boltzmann Inversion on experimentally determined radial distribution functions [4]. Here, we exploit experimentally observed diffusion of magnetic skyrmions in pinning potentials to realize fully quantitative Thiele model simulations on experimentally relevant time and length scales [5]. By determining the underlying pinning landscape, we can calibrate experimental and simulation time scales and current-induced forces.

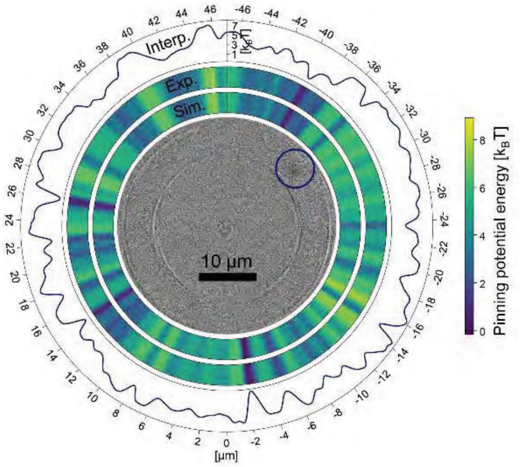

Figure 1: The central Kerr microscopy image displays a single skyrmion (a dark spot within a blue circle) constrained within a ring having an inner radius of 12.5 mm and a width of 5 mm. Surrounding this are two heatmaps representing the one-dimensional effective pinning energy landscapes for both the experimental and simulated systems. [5]

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Universal click-chemistry approach for the DNA functionalization of nanoparticles for the development of new optical nanoantennas

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Nanotechnology has revolutionized the fabrication of hybrid species with tailored functionalities. A milestone in this field is the DNA conjugation of nanoparticles, introduced almost 30 years ago by Mirkin's group via the deposition of thiol-modified DNA strands on Au NPs [1]. Since then, the reaction has been widely used, which has resulted in many modifications of the method. A particularly useful scheme is the freeze-thaw process, which results in both a shorter reaction time and a higher surface density of DNA [2]. In general, using thiolated-DNA provides a reliable way to functionalize only noble and coinage metals. Therefore, it is necessary to develop alternative techniques to address the DNA conjugation of NPs with surface compositions that are inert to thiols, including oxides, core-shell, polymeric NPs, and even NPs composed of active metals (Al, Ga) and metalloids (Si, Ge). Si NPs are an interesting example of such structures that cannot be functionalized using thiol chemistry. These high-index dielectric NPs have received growing attention in recent years, as theoretical and numerical studies have predicted novel properties including low heat losses and overlapping strong magnetic and electric modes in the visible range [3]. These features are key for applications in chiral sensing, unidirectional scattering, and the manipulation of magnetic dipole transitions, making Si NPs a convenient alternative to plasmonic NPs that exhibit relatively high losses and the absence of a magnetic response at optical frequencies [3]. However, in spite of the great potential of Si NPs, their stable conjugation with DNA strands has yet to be achieved.

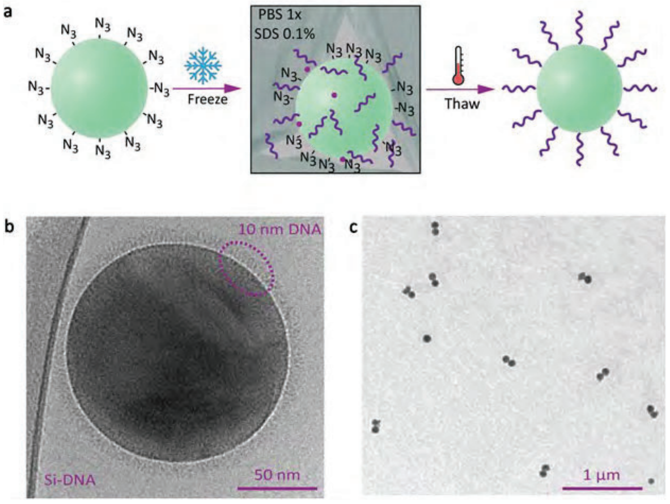

Figure 1: *(a) General protocol of the functionalization of various nanoparticles through freezing-assisted DBCO-DNA deposition. (b) Cryo-EM image of a DNA-functionalized Si NP with visible DNA shell of 10 nm (purple circle). (c) TEM image of Si dimer connected through DNA origami.*

Inspired by recent developments in the functionalization of Au NPs, we report here an approach to conjugate various nanomaterials with DNA by employing freezing-assisted SPAAC, with a special focus on Si and SiO_2 NPs (see Figure 1a) [4]. Impressively, this simple method of functionalization proved to tremendously improve the overall DNA deposition efficiency. When comparing with other approaches used for the DNA functionalization with SPAAC, we found that not only does freezing reduce the reaction time from about two days to only a few hours, but also that our method yields 2-10 times higher DNA density on the surface of the NPs, i.e., up to 0.2 molecules/nm^2 (see Figure 1b). The resultant DNA-Si NPs form highly stable colloids, which we incorporate into DNA origami structures via the formation of self-assembled NP dimers at pre-determined positions with an inter-particle gap of approximately 5-10 nm, as presented in Figure 1c. Our results show up to a 78% yield for the formation of NP dimers (i.e., versus unintended NPs monomers), thus representing the first step towards the synthesis of more complex geometries such as hybrid metal-dielectric antennas and the combination of high-index materials with other species in precise geometries. Finally, we demonstrate the versatility of this method by realizing the DNA functionalization of different NPs: oxide – TiO_2, Au@SiO_2, polymeric – PMMA and PS, as well as metallic Au NPs.

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Polymers and Biopolymers II

Monday 19 May 16:30 – 18:10

Confined chain dynamics in one component polymer nanocomposites (OCNC)

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Polymer nanocomposites (PNC) represent a broad class of modern materials with a wide spectrum of possible applications. Conventional nanocomposites are manufactured by blending nanoparticles (NPs) and polymers. The main complexity, especially at relatively high-volume fraction of fillers, relates to the required homogeneous and controlled distribution of the NPs in the polymer matrix. Recently, polymer hybrids such as the one component nanocomposites (OCNC) have drawn much attention both in basic as well as in application-oriented research due to perfect dispersion of the fillers.

In this work the polymer dynamics of grafted polymer chains in the OCNC, i.e. without an embedding matrix, in the weakly entanglement regime is investigated. The OCNC are built by self-assembly of block-copolymers of cross-linked deuterated 1,2-polybutadiene (1,2-dPB) in the core and poly (ethylene oxide) (PEO) grafts at outside. The resulting micellar core has a radius in the order of 14 nm with little dispersity. Using neutron spin echo (NSE) we address following points: (i) chain and segmental mobility of the grafted polymer chain; (ii) the role of the entanglements and confinement on the chain dynamics. (iii) the role of the heterogenous arrangement of micellar cores on the dynamic features.

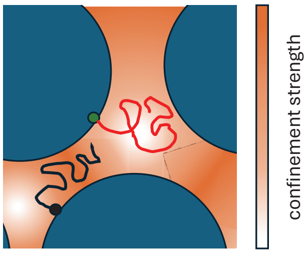

Figure 1: *Heterogeneities for the dynamics of a grafted chain: black chain is more confined than the red one.*

As already demonstrated for weakly entangled melts, we show that the role of the heterogenous environments created by the dense core melt and the entanglement behavior of grafted polymer chains could be captured using a Rouse mode analysis with suppressed amplitudes for modes with low numbers [1]. Even though the grafting density was low, our results show that the monomeric friction increases significantly close to the grafting point and decreases to constant friction in the direction of the chain ends. Furthermore, we demonstrate that dynamic heterogeneity needs to be assumed, in order to describe the experimental spectra (Figure 1). We relate this heterogeneity to the structural peculiarities of the dense core arrangement[2].

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The relaxation dynamics and role of dynamic facilitation in polymer glasses

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The glass transition is a long-standing unsolved problem in materials physics. For polymers, our understanding remains relatively poor because of the added complexity of chain connectivity and chain flexibility. Here, we use data from broadband dielectric spectroscopy, calorimetry and rheology, combined with simple computer modelling, to determine how the relaxation dynamics varies with key polymer characteristics, such as molecular chain-length and flexibility. Based on our results, we propose that in polymers, a set of generic molecular relaxations are linked through the mechanism of *dynamic facilitation* [1], by which a `local' relaxation facilitates adjacent relaxations, resulting in hierarchical dynamics; the important role of local conformational degrees of freedom paves the way for predictive polymer design based on monomer-scale metrics [1]. Importantly, we identify chain-length-dependent regimes where intra- and inter-molecular dynamics play different roles in defining the dynamics. Moreover, we demonstrate how dynamic heterogeneities (DH), a hallmark of glass-formation and of dynamic facilitation, depend on both chain-length and flexibility [2]: highly flexible polymers show a molecular weight (M) independent behaviour, whereas less flexible polymers show a complex regime behaviour, consistent with observations in both Tg(M) and conformational structure. Based on our DH results, we discuss and evaluate the detailed temperature-dependent evolution of relaxation dynamics in polymers. Finally, we illustrate how the fundamental relaxation behaviour of polymers link to important transport properties, with focus on ion transport relevant to the development of efficient polymer-based solid electrolytes in battery applications.

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Implementing diffusing wave spectroscopy to probe piezoelectric polymer dynamics under electric field

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Piezoelectric polymers are found in several applications as energy harvesters, acoustic transducers and tactile or inertial sensors due to their enhanced mechanical flexibility, lower costs and increased ease of production, as well as biocompatibility for uses such as implantable or wearable sensors. There is a wide range of piezoelectric polymer materials such as odd polyamides, poly(lactic acid), fluorinated polymers based on polyvinylidene fluoride (PVDF). Fluorinated polymers have the highest performance in terms of electromechanical coupling. These semi-crystalline materials draw their piezoelectricity from their polar crystal domains, notably from the beta phase. The piezoelectric character is thus obtained after a phase of polarization by electric field which serves to co-orient the dipoles of the crystals responsible for a residual polarization within the polymer. However, recent mechanical and structural analyses by X-ray scattering [1,2] have also shown a significant effect on the piezoelectricity of the amorphous phase mobility material, especially of the oriented one which is at the interface between the crystalline and amorphous isotropic phases. This contribution has been studied only by simulations [2] because it is not permanent and, therefore, can be observed only under electric field. A measurement system to couple the analysis of mobility with electric fields would remove the scientific barriers related to the optimization of electromechanical couplings.

We propose a new experimental approach by using diffusing wave spectroscopy (DWS) to study the polymer mobility under electric field. DWS is an optical technique that allows one to probe the mobility at the nanometre scale (also called microscopic dynamics) of opaque materials and, therefore, to reveal the presence of microscopic rearrangements in the material long before they can be detected by the change in macroscopic properties. We show the suitability of this technique by studying the effect of the electric field up to around 10 MV/m on the dynamics of PVDF-based copolymers. These results will help elucidating the role of the amorphous phase in the piezoelectricity of fluorinated polymers.

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Revisiting high frequency dynamics of wormlike micelles by DWS microrheology: determination of molecular weight and radius

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Wormlike micelles serve as excellent model systems for semiflexible polymers, with their structure-property relationships crucial for both academic and industrial applications. While scattering techniques have been used to investigate key structural parameters, microrheology offers a dynamic approach to probing their viscoelastic properties. By tracking Brownian motion, microrheology extends the accessible frequency range to $10^2 - 10^5$ rad/s, enabling the determination of persistence length from high-frequency rheological responses, as successfully demonstrated by previous microrheological studies. Revisiting and expanding upon these studies, this work extends microrheological analysis beyond persistence length measurements, demonstrating how the measured modulus can be employed to determine molecular weight and Kuhn segment radius in cetylpyridinium chloride/sodium salicylate wormlike micelles. Utilizing diffusing-wave spectroscopy microrheology, we identified the crossover between Zimm and bending modes, calculating relaxation times and moduli. We then estimated the molecular weight of Kuhn segments, obtaining results consistent with static light scattering and literature values. This work highlights the power of dynamic microrheology to extract structural parameters. These findings provide a refined methodology for molecular weight determination at dynamic mode boundaries. This method is applicable to semiflexible polymers. It shows that dynamic measurements can be used to extract structural information.

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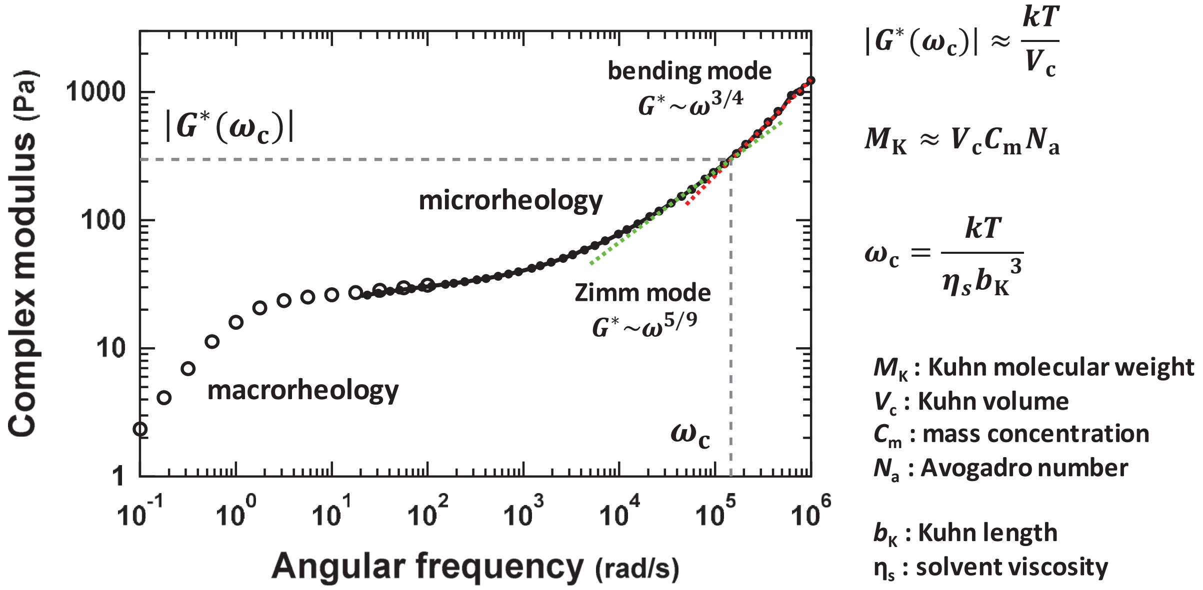

$$|G^*(\omega_c)| \approx \frac{kT}{V_c}$$

$$M_K \approx V_c C_m N_a$$

$$\omega_c = \frac{kT}{\eta_s b_K^3}$$

M_K : Kuhn molecular weight
V_c : Kuhn volume
C_m : mass concentration
N_a : Avogadro number

b_K : Kuhn length
η_s : solvent viscosity

Figure 1: *Amplitude of complex modulus for cetylpyridinium/salicylate wormlike micelles as a function of angular frequency. Determination of crossover point of bending mode and Zimm mode.*

Varying the core topology in all-glycidol hyperbranched polyglyerols

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Hyperbranched polyglycerols (hbPGs) have been extensively studied due to their promising potential for biomedical applications. They possess excellent biocompatibility and high functionality, which facilitates the attachment of a variety of bioactive molecules [1]. The core topology of hbPGs clearly plays an important role. For example, the use of low molecular weight initiators leads to the formation of compact globular structures with a three-armed star core and a random distribution of hydroxyl groups along the branches. In this study, we present a comprehensive synthetic route to the formation of a new architecture of this family of hyperbranched polymers, a cyclic graft hyperbranched polyglycerol [2]. To this end, we employed a hypergrafting strategy from a low molecular weight cyclic polyglycidol prepared by deprotection of a cyclic poly(tert-butyl glycidyl ether), synthesized by an electrophilic zwitterionic ring expansion polymerization mechanism [3]. The physical and biological properties of the doughnut-like structures obtained from a cyclic macroinitiator were compared with analogous structures obtained by hypergrafting a linear and a star-like initiator. This study highlights the influence of core topology on the properties of hyperbranched polyglycerols, advancing the field of water-soluble polymers and enabling tailor-made material design.

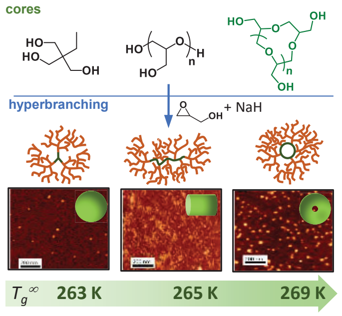

Figure 1: *Polyglycerol structures generated in this study combined with AFM images of 1MDa Mw and Tg∞ determined using the Kanig-Ueberreiter equation.*

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New Methods and Techniques

Tuesday 20 May 09:45 – 10:45

Uncovering mechanisms for ion transport in solid electrolytes

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A broad variety of soft materials are currently the subject of research for energy applications. For example, in new battery technologies, a very topical issue and a key societal challenge [1]. Many different electrolytes have been proposed for use in next-generation solid state batteries, with the potential to provide greener, more efficient energy storage. A key stumbling block in proposed electrolytes is often low conductivity. Therefore, when considering a new material, the first step is to understand ionic conductivity. Transport or diffusion of ions through a material can be a complex process, requiring knowledge on multiple length- and time-scales. A number of correlation effects have also been identified [2]. For example, the diffusion of individual ions depends upon the diffusion of neighbouring ions. Ions may also become trapped due to local structure and undergo movement but in a back-and-forth motion that does not not contribute to overall transport.

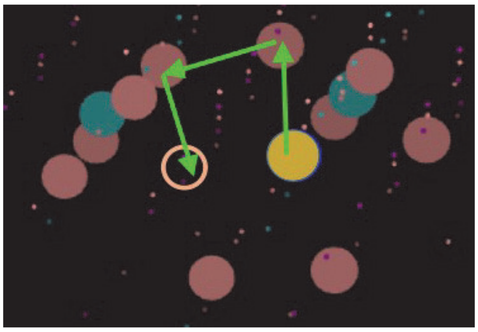

Figure 1: *Correlated motion of three Lithium ions*

Using methods developed for supercooled liquids [3], we identify ion ‘jumps’, particularly those that are productive for transport. Starting from any molecular dynamics trajectory, we can identify ‘inherent structures’ or local minima on the Potential Energy Landscape[4] and then analyse structural changes in the material. We study the interplay between structure and dynamics at different length and time scales, developing a fundamental understanding of experimental observations. Determining the relevant structural length scales is key to this approach. Here, we will demonstrate the methods for low-dimensional-networked Li-rich anti-perovskites, potential solid electrolytes [5]. Anti-perovskites have a remarkably versatile structure and their structures can be manipulated easily. Once we have a mechanistic understanding of ion transport mechanisms, structures can be tailored to improve conductivity. They are also an interesting class of materials because we can study the change in transport as the dimensionality of the network is reduced and the structure of the material changes from an ordered crystal to a disordered material.

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Microfluidic dead-end chamber devices for the study of water-in-water droplets and biological cells

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The formation of water-in-water droplets via liquid-liquid phase separation (LLPS) is crucial for drug delivery and biomanufacturing and has important implications for understanding the formation and dynamics of membraneless organelles in biological cells. However, studying LLPS in microfluidic systems is challenging due to the low surface tension of water-in-water droplets, which complicates their formation, stabilization, and long-term observation. Existing methods rely on surface treatments or encapsulating agents to stabilize the droplets, which can interfere with the phase separation process.

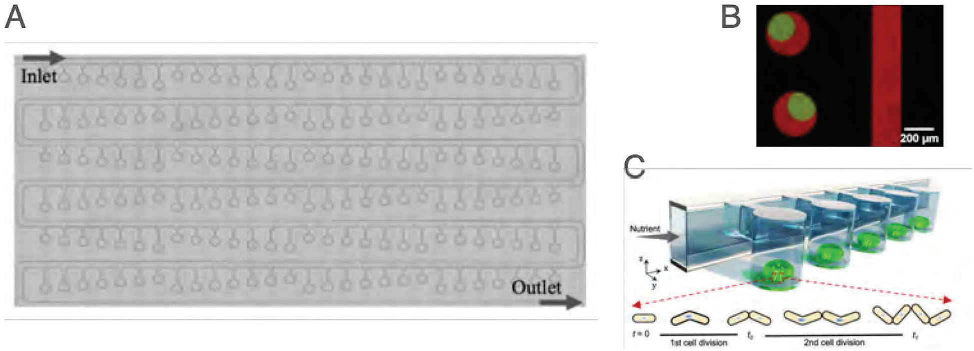

Figure 1: *Microfluidic dead-end chamber devices for the study of water-in-water droplets and biological cells. (A) Micosopy image of a typical device with main channel and dead-end chambers (B) Fluorescence microscopy imaging of water-in-water droplet formation in the chambers (PEG-dextran system). (C) Schematic illustration of dead-end chamber device used for cell studies, here focused on studying the cell cycle in different extracellular environments.*

In this study, we present a microfluidic platform in which water-in-water droplets can be generated and stabilized in dead-end chambers via controlled diffusion from a main flow channel (**Fig. 1A**). This approach eliminates the need for stabilizers and enables real-time fluorescence microscopy monitoring. We validate the platform by studying the phase separation of polyethylene glycol (PEG) and dextran solutions (**Fig.1B**) and employ it in the study of coacervate formation. We show that the platform enables us to reversibly form coacervates with multiple internal compartments in a two-component associative LLPS system, which has not been previously reported. We also show how the platform can be used in the study of biological cells. We demonstrate that cells can be efficiently delivered into the dead-end chambers by exploiting diffusiophoresis, driven by concentration gradients, without affecting their viability. Making use of this, we employ our platform for studying the influcence of physical parameters, such as the viscosity of the extracellular environment, on the cell cycle (**Fig.1C**).

Towards Absolute Interaction Measurements Using Optical Tweezers

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Accurate measurement of pair interaction potentials is crucial in colloidal and soft matter systems. Optical tweezers enable direct extraction of these potentials, but systematic errors—such as z-motion fluctuations, dynamic error from time-averaging, and static error due to tracking uncertainty—distort measurements. In this study, we develop a model to quantify these errors and validate it through controlled experiments that disentangle their individual contributions. Furthermore, we establish a correction framework that systematically removes distortions, allowing the recovery of true interaction potentials. As a demonstration, we apply this method to depletion interactions modelled by the Asakura-Oosawa potential, successfully reconstructing the true interaction strength. Our study provides a guideline for precise optical tweezer measurements and represents a step toward absolute interaction measurements in soft matter research.

Glasses, Colloids and Self-Assembly III

Tuesday 20 May 09:45 – 10:45

Development of Modular Sample Environment for Non-Equilibrium Soft Matter

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The primary scientific objective of this project is to develop a modular sample environment optimized for small-angle neutron scattering (SANS), a powerful technique for investigating the mesoscopic structural complexities of soft matter systems under non-equilibrium conditions. Additionally, the sample environment should allow for the integration of spectroscopic measurements, including fluorescence, UV-Vis, and infrared spectroscopy, enabling simultaneous monitoring of kinetics. This combined approach will provide complementary insights into structural evolution at molecular length scales [1]. The modular sample environment will offer precise temperature control and flexibility in adjusting the system's composition. This will facilitate continuous SANS and spectroscopic observations of non-equilibrium soft matter samples within a flow-through cell. A schematic of the flow-cell assembly is shown in Fig. 1(a). As a test characterization, we used an in-situ, non-stoichiometric mixture of two oppositely charged polyelectrolytes—polydiallyldimethylammonium chloride (PDADMAC) and sodium carboxymethyl cellulose (Na CMC)—to investigate the formation of interpolyelectrolyte complexes (IPECs) [2, 3]. This investigation was conducted as a function of the charge ratio, Z, which is defined as the ratio of molar charge per monomer of the polycation ([+]) to that of the polyanion ([−]). The time-resolved SANS profiles are shown in Fig. 1(b). Our analysis revealed that as the charge ratio (Z) increases, the complexes undergo compaction, transitioning from globular-shaped aggregates to lumpy-shaped aggregates.

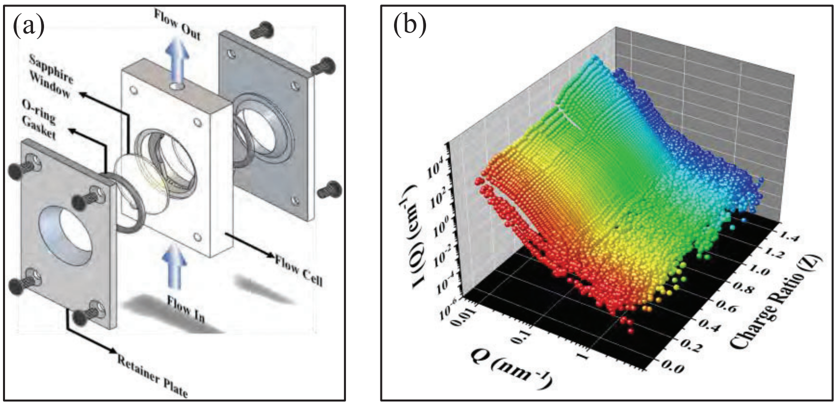

Figure 1: *(a) A schematic of the design of liquid flow-cell assembly, (b) Time-resolved SANS data of formed IPECs as a function of charge ratio.*

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Navigating Complexity: Designing Transient and Oscillating Systems with Nanoparticles

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Self assembly is nature's preferred means of forging both inanimate and animate matter.(1; 2) Inspired by nature, researchers have undertaken studies aiming to understand the design principles governing both equilibrium and non-equilibrium self-assembly processes. In this context, colloids—particularly gold nanoparticles (AuNPs) with their intense UV-Vis response—offer an ideal platform for studying the design principles of complex self-assemblies. In this work, we demonstrate the formation of transient and oscillating systems using AuNPs as building blocks.

To choreograph a transient self-assembly response, we capitalized on the pH-dependent electrostatic interactions between EDTA (linker) and positively charged AuNPs (bearing quaternary ammonium headgroups). EDTA effectively bound [+]AuNPs at pH 11 but failed to do so at pH values below 8 (see scheme left). Spontaneous absorption of CO_2 from the atmosphere induced an autonomous pH drift towards acidic values, facilitating transient switching between '*plasmonically off*' and '*plasmonically on*' states—a challenging feat owing to strong van der Waals interactions in AuNPs. Additionally, we demonstrate an efficient waste removal mechanism to mitigate damping effects across transient self-assembly cycles.(3)

Moving up the complexity ladder, we demonstrate an oscillating system comprising of thermoresponsive self-assembly of AuNPs.(4; 5) In this setup, DNA-coated AuNPs are glued together using a DNA-based linker (see right scheme). Upon photoirradiation, nanoparticles disperse due to photothermal heating, resulting in the melting of the H-bonding interactions that glue the AuNP aggregates. By selectively irradiating the bottom part of the NP dispersion, we induce convective flows, resulting in oscillations in the extinction intensity of AuNPs (see scheme right). This system operates akin to an electronic oscillator, transforming continuous NIR irradiation into an alternating optical signal with a period nearing 60 seconds. It is noteworthy that both these studies were designed so that the system could interact with the environment to show transient and oscillating behaviours.

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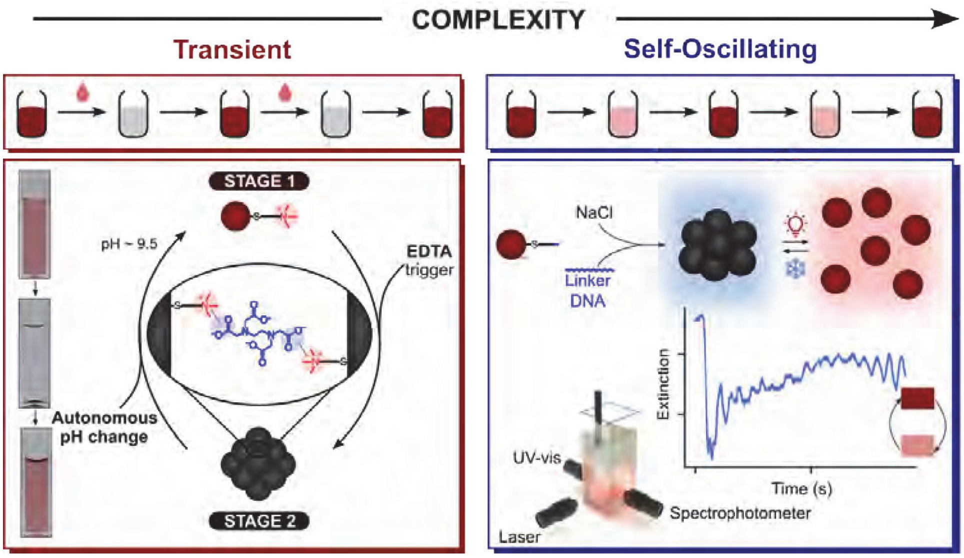

Figure 1: *Experimental designs illustrating the progressive increase in system complexity. The left panel shows a transient system where pH-induced changes drive the self-assembly and disassembly of AuNPs, while the right panel depicts an oscillating system enabled by photothermal heating and convective flows.*

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Using oscillations to stabilize particle strings and their fibers in flowing viscoelastic media

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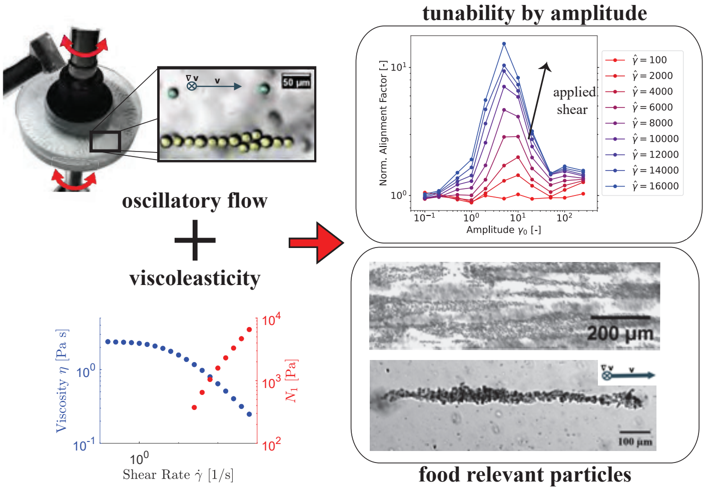
The formation of particle strings under shear in viscoelastic media is an elegant method to induce anisotropy in suspensions: Shear thinning and elastic stresses, quantified by the Weissenberg (Wi) number, arise in the medium under flow and drive the particles to arrange in pearl-necklace like string structures [1,2]. While alignment is desired for many applications, such as the formation fibers in biopolymer solutions, or of fibrous meat analogues, the required processing parameters for viscoelasticity-induced alignment precludes such implementations: traditionally is has been observed only at high shear rates or in strongly shear-thinning fluids. By applying oscillatory shear protocols we mitigate this and achieve more feasible processing conditions.



With counter-rotating rheometer with transparent geometries, we systematically investigate the shear-driven assembly of noncolloidal polystyrene (PS) spheres in a density-matched polyethylene oxide (PEO) solution. By varying the Weissenberg number (Wi), we map the stability and dynamics of string formation. Computationally, we extract a time-resolved structural evolution and identify key stability regimes and an instability mechanism. We found tuneable string morphology by controlling the number of particle-particle meeting events through controlling the amplitude of the oscillation. Beyond the model system, we extend this concept to food-relevant formulations and demonstrate anisotropic structuring in viscoelastic biopolymer suspensions (e.g., xanthan gum) with microalgae and protein particles. Our findings show that particle attraction enhances the stability of these aligned structures under flow, while retaining the tuneability found with the model systems.

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Polymers and Biopolymers III

Tuesday 20 May 11:15 – 12:55

Probing multiscale dynamics of pea protein gel during digestion

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Scattering techniques are non destructive and can be used to probe *in situ* structural dynamics of food upon digestion. Studying food resilience and digestion pathways comes handy when we focus on developing novel foodstuffs, in this work we focused on pea globulins-based hydrogels as platform for sustainable proteins. It is known that altering the pea protein profile, changing the legumin/vicilin ratio, will results in different mesostructure and rheological protein of the gel but in this work we focus on how this will affect the digestibility of the substrate. We probed hydrogels digestion *in situ* using neutron scattering techniques at the D33 beamline at ILL within a microfluidic device. The substrate is embedded within the chip while simulated gastric fluid is flawed through microfluidic pump system on top of it, and the scattering pattern is continuously acquired. Similar experimental setup has been adapted to synchrotron SAXS experiments at CoSAXS beamline at MAXIV and to confocal scanning laser microscope, in order to give us a multiscale picture of the process. Structural dynamics shows a slower structural collapse of the gel network when a higher concentration of vicilin is present in the protein isolates. While SAXS data support findings from the SANS profiles, slight differences emerges, possibly due to the different activity of pepsin in deuterated water. Developing new strategy to study *in situ* digestion of solid foodstuff is pivotal to deliver more nutritious and safe food. In this case we show how different way of sourcing globulins from pea seeds can affect the digestibility of the final product. Moreover, the present works aim to reflect on how x-rays and neutron result complementary in such study.

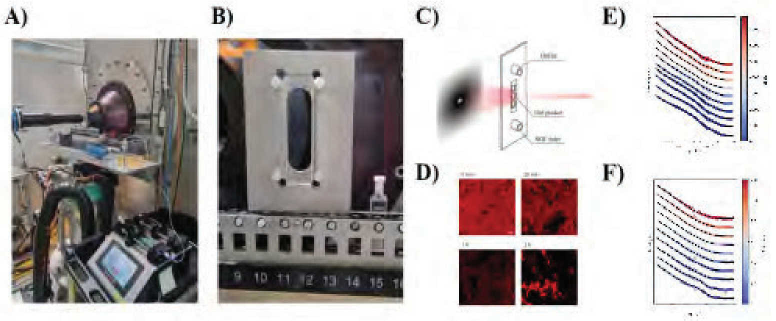

Figure 1: *A) Setup in use at D33 (ILL, Grenoble). B) Zoom on the sample holder. C) Sketch of the microfluidic device in use. D-E-F) Results of CSLM, SAXS and SANS experiments.*

Unlocking the Potential of Humins for Functional Materials: Chemical Modifications via Esterification and Diels-Alder Reactions

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Biorefineries are important for sustainability because they use renewable feedstocks to produce valuable chemicals and materials, allowing for a reduction in reliance on fossil fuels and minimizing environmental impact. One representative technology involves the acid-catalyzed conversion of polysaccharides into platform chemicals such as furfural, 5-hydroxymethylfurfural, levulinic acid, and formic acid. However, this process is inevitably compromised by the formation of a significant fraction of by-products, accounting for up to 50% of total carbon loss—humins. These black tar-like compounds originate from side reactions such as aldol condensation and transesterification, and, as a result, they possess dense polyaromatic structures with various functional groups. Their composition further depends on process conditions and raw materials, making commercialization difficult. Although their exact structure is not fully understood, it is known that humins contain furan rings, hydroxyl groups, aldehydes, and ketones.

Since humins are unavoidable by-products, this study explores ways to convert them into useful materials. In fact, finding ways to use these by-products not only reduces waste but also supports sustainable materials development and green chemistry principles. Pure humins are difficult to process because of their high viscosity, thermal sensitivity, and handling issues. To address these limitations, our strategy focused on selective chemical modifications of the humins, thus changing their properties.

Initially, our approach included the use of esterification reactions to modify abundant hydroxyl groups and suppress hydrogen bonding. Using short-chain monofunctional acyl chlorides, we were able to convert humins into fine powders, while the use of long-chain bifunctional acyl chlorides enabled the formation of low-viscosity liquids. These modifications proved efficient in enhancing solubility and thermal stability. We further extended this principle by attaching polymer arms onto the humins using the polymerization-onto method, giving rise to complex structures with distinct material characteristics.

The second approach focused on utilizing the furan rings in humins as dienes in the Diels-Alder reaction, a well-known [4+2] cycloaddition reaction in organic chemistry. Reacting the humins with a low-cost, commercially available dienophile-like bismaleimide, our experiments demonstrated the successful formation of dynamically covalently bonded networks, which could be further manipulated by shear. These new structures improved the mechanical properties of humins matrices, reducing their inherent brittleness and enhancing durability. Additionally, the reversible nature of the Diels-Alder adducts enabled a self-healing character of the material that can be initiated by heat. Our study is supported by a comprehensive set of analytical characterizations, including spectroscopic, calorimetric, rheological, mechanical, and microscopy tools.

Overall, we show that humins are valuable building blocks for making new materials with interesting properties.

Keywords: Biorefineries, Humins, Diels Alder Adduct.

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Seacrete: a soft composite for marine restoration

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Marine environments are typically challenging for good adhesive performance due to direct contact with adherent by water, biodegradation, and the promotion of swelling. There are few adhesives that work underwater; two-part epoxides and cyanoacrylates function, but both with deleterious effects on marine organisms. My laboratory has recently been investigating the use of soft, complex coacervates as marine adhesives for marine restoration. Inspired by the sandcastle worm, which utilizes a coacervate that “salt-switches” from a liquid to solid when exposed to marine water. The flat oyster (*Ostrea edulis*) once was an abundant key species in the North Sea that provided ecologically important reefs supporting biodiversity and productivity. These reefs disappeared due to destructive bottom trawling. Reintroduction of hard substrate is needed to kickstart oyster reef formation, and often concrete is used, however, its use is debated because of its high CO2 footprint and large amounts of fresh water and sand needed. As a solution to creating marine substrates, I will introduce our work on a complex coacervate composite, Seacrete, containing a liquid coacervate adhesive combined with solid aggregate. This composite salt-switches rapidly underwater creating long-lasting bonds and a solid substrate for oyster larvae settlement. I will discuss the design, function, and 3D printing of this composite. Field work shows a two-fold increase in oyster larvae settlement over other substrates likely due to surface roughness and hardness. This interdisciplinary work combines both soft matter design principles with marine ecology offering a new perspective on marine restoration and new opportunities for the Soft Matter community to contribute to a challenging global crisis.

Nanocomposites of Lignin and Poly(butylene succinate): the effect filler size and concentration on physicomechanical and functional properties

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Biodegradable polymeric materials have emerged as a promising alternative to conventional plastics due to their environmental benefits. However, their widespread adoption remains limited by challenges such as high production costs and suboptimal performance [1]. This study tackles these issues by integrating steam-explosion lignin, an inexpensive and widely available byproduct, into poly(butylene succinate) (PBS) to produce innovative biocomposites with enhanced properties.

Steam-explosion hardwood lignin (SEL) was thoroughly analyzed using advanced techniques like GPC, ^{31}P-NMR [2], and HSQC [3], confirming its structural resemblance to native lignin [4]. SEL exhibited impressive multifunctionality, including antioxidant activity (IC_{50} 28 ± 1 µg), broad-spectrum antimicrobial effects against gram-positive (*S. aureus*) and gram-negative (*E. coli*) bacteria, and effective UV protection comparable to industrial lignins such as kraft and organosolv lignins. To test its suitability in bioplastics, SEL was incorporated into PBS through melt blending, using varying filler sizes: microparticles (200–600 µm and <250 µm) and lignin nanoparticles (LNPs) produced via a sustainable hydrotropic process [5]. The effects of particle size and concentration (5, 10, and 15 wt%) on the composites' thermal, mechanical, and functional properties were evaluated and compared with composites reinforced with softwood kraft lignins and wood fibers.

The addition of SEL significantly improved PBS crystallinity and stiffness. Notably, LNPs acted as powerful nucleating agents, driving crystallization without compromising PBS ductility. Composites containing LNPs outperformed those with larger lignin particles in mechanical strength and overall performance. Furthermore, the functional benefits of lignin—antioxidant and antimicrobial activities—were retained in the composites, with LNP-based systems delivering the highest efficacy.

To investigate their environmental impact, the composites were subjected to accelerated aging tests (1,000 hours under simulated solar light at 50 °C and 50% RH). The results revealed that lignin, especially in its nanoparticle form, accelerated the degradation of PBS through mechanisms involving enhanced surface interaction and photocatalysis, which promoted hydrolysis and polymer chain scission.

These findings demonstrate that lignin, particularly as nanoparticles, can transform PBS into a material with superior mechanical performance, active functional properties, and tailored biodegradation behavior. This makes it a viable candidate for eco-friendly packaging and other sustainable applications, bridging the gap between performance and environmental responsibility in polymeric materials.

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Advancing Sustainable Protection: bio-based coatings from fractionated Kraft lignin

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The degradation of metal surfaces due to corrosion and fouling presents significant challenges in industrial and marine environments, leading to increased maintenance costs and material failures. Traditional protective coatings often rely on synthetic polymers and toxic additives, raising environmental concerns and facing increasing regulatory constraints. In this study, kraft lignin-an abundant byproduct of the pulp and paper industry- is investigated as a key component in bio based coatings designed to simultaneously inhibit corrosion and biofouling.

Kraft lignin's intrinsic antioxidant activity, metal chelation capability, and hydrophobic nature make it a promising candidate for protective barriers. Through fractionation techniques, its structure is refined by narrowing lignin's heterogeneity, allowing for tailored formulations that enhance coating stability and performance.[1] These biobased treatments effectively reduce surface wettability and prevent microbial attachment-critical factors in mitigating both corrosion and fouling. [2,3]

All lignin fractions were fully characterized via NMR spectroscopy (SSQC, HSQC0, 13C, 31P) and GPC analyses, providing a comprehensive overview of lignin structural features, including bonding patterns, measurements, and corrosion tests via electrochemical techniques demonstrated the effectiveness of these lignin-based coatings as sustainable alternatives to conventional protective systems. [4,5]

This research enhances high performance, eco-friendly metal protection while highlighting the significant potential of lignin valorization for industrial applications. By integrating circular economy principles and reducing reliance on fossil-based materials, these findings pave the way for a new generation of sustainable coatings with significant environmental and economic benefits.

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Glasses, Colloids and Self-Assembly IV

Tuesday 20 May 11:15 – 12:35

Investigating the Self-Assembly of Chitin Films Enhanced with Natural Polyphenols: Transforming Seafood Biomass Waste into Functional Materials

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Films made from fossil raw materials, used for food packaging and various other applications, pose a serious challenge to the environment. Due to their non-biodegradable nature, these materials substantially contribute to the growing problem of plastic waste. In fact, the packaging industry alone consumes the largest amounts of plastics derived from fossil fuels and is responsible for approximately half of the world's plastic waste.[1] On the other hand, the environmental impact of seafood waste is becoming increasingly significant over time. Between 6 and 8 million tonnes of crustacean shells are produced worldwide every year.[2] About half of shrimp goes to waste, resulting in overloaded landfills and causing environmental and health issues.[3] This situation urgently calls for sustainable recycling methods to reuse seafood by-products and mitigate the negative impacts of this biomass source. Polysaccharide-based biodegradable films offer an environmentally friendly alternative that not only reduces waste but also adds value to seafood waste biomass.[4]

The present study explores a sustainable method for producing 100% biobased, free-standing, transparent, and flexible films made from chitin nanocrystals (ChNCs) and surface-deacetylated chitin nanocrystals (CsNCs) derived from shrimp shells. The films were formed via slow solvent evaporation of nanocrystals (NCs) aqueous suspensions. To further enhance the functional properties of the films, tannic acid was used as crosslinker, strengthening the hydrogen bonding network and improving mechanical strength, water resistance, and bioactivity.[5] Additionally, glycerol was explored as a plasticizer to optimize flexibility and mechanical performance.[5], [6]

ChNCs were obtained by acid hydrolysis of shrimp chitin flakes and subsequently treated in an alkaline environment to produce surface-deacetylated chitin nanocrystals (CsNCs). The deacetylation reaction time was optimized to enhance the degree of deacetylation and consequently particle interactions. The data acquired from ^{1}H NMR in suspension, ^{13}C solid-state NMR, and elemental analysis revealed that after circa 12 hours, the degree of deacetylation reached a plateau (~80% of the surface groups). The effects of deacetylation on the size and morphology of the nanocrystals were examined using scanning electron microscopy (SEM). The SEM micrographs demonstrate the characteristic rod-like shape for both nanocrystals (**Figure 1A and B**) and their size analysis indicates that deacetylation does not significantly influence the size and aspect ratio of the NCs, with measurements of length, width, and aspect ratio being 367.9±227.0 nm, 40.9±17.6 nm, and 8.9±4.1 for ChNCs, and 369.2±245.9 nm, 46.4±21.0 nm, and 9.1±3.5 for CsNCs, respectively, obtained from fitting the data to lognormal distribution.

The presence of cholesteric liquid crystalline ordering was investigated in different formulations by preparing concentration series of NCs suspensions and by cross-sectional SEM of the freeze-fractured films. This allowed a quantitative evaluation of the liquid crystalline phase percentage relative to NCs concentration, and the size of the chiral nematic pitch both in suspensions and in films. Suspensions containing ChNCs without glycerol (ChNCs-WOG) showed phase separation starting at 3.5 wt% (**Figure 1C and D**), whereas suspensions containing ChNCs and glycerol (ChNCs-WG) showed phase separation starting at 4 wt% (**Figure 1D**), as expected given that adding glycerol to the formulation replaces some of the strong hydrogen bonds between NCs with the glycerol molecule. Polarized light optical microscopy (POM) of the round capillaries (internal diameter 1.00 mm) containing concentration series of ChNCs-WOG and ChNCs-WG showed the fingerprint pattern characteristic of chiral nematic liquid crystals, and confirmed the effect of glycerol. The pitch size of the helicoidal structures in the suspensions was determined to be twice the periodicity of the fingerprint pattern observed in POM.[7] This was compared to the pitch size of dried films determined from cross-sectional SEM micrographs of freeze-fractured films: for ChNCs-WOG, 42.04 ± 3.03 μm for a suspension at 7.0 wt% and 2.19 ± 0.33 μm for the dried film starting at 3.5 wt%; for ChNCs-WG (**Figure 1E and G**), 89.48 ± 4.17 μm for a suspension at 7.0 wt% and 4.96 ± 1.88 μm for the dried film staring at 3.5 wt% (**Figure 1F and H**).[8] The larger cholesteric pitch observed for ChNCs-WG confirms that glycerol molecules intercalate inside the chiral nematic structure, resulting in an increase in pitch size.[6], [8]

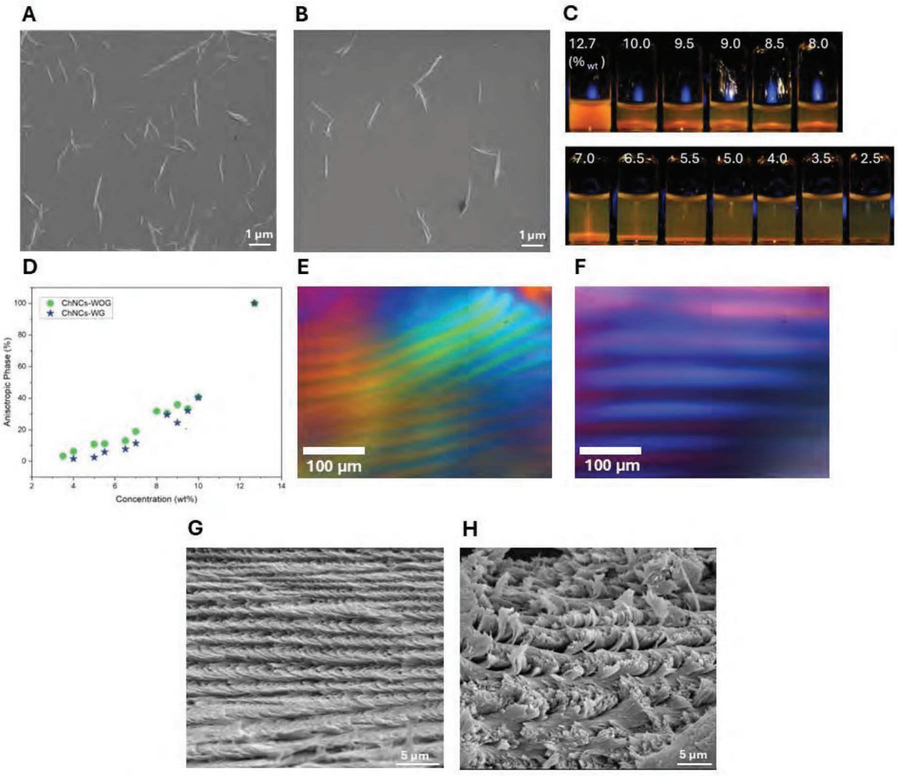

Figure 1: *Scanning electron microscopy (SEM) micrographs of Chitin Nanocrystals (ChNCs) (A), and surface deacetylated chitin nanocrystals (CsNCs) (B). Concentration series of ChNCs without glycerol (ChNCS-WOG) captured between two crossed polarizers (C). Phase diagram illustrating the increase of anisotropic phase fraction due to the increase of nanocrystal concentration for ChNCS-WOG (green circles) and ChNCS with glycerol (ChNCS-WG, blue stars). Fingerprint pattern characteristic of chiral nematic liquid crystals observed via polarized optical microscopy (POM) of the round capillaries containing 7 wt% ChNCs-WOG (E) and ChNCS-WG (F). Cross-sectional SEM micrographs of freeze-fractured ChNCs-WOG (G) and ChNCs-WG (H) films.*

In contrast, CsNCs suspensions and films did not exhibit evidence of cholesteric behaviour. This observation calls for further investigation to determine the conditions, such as pH or ionic strength, that might induce self-assembly into chiral nematic domains. Furthermore, the absence of cholesteric ordering in CsNCs films may contribute to superior mechanical and optical properties, suggesting a potential trade-off between structural ordering and functional performance, which yet remains to be studied.

In conclusion, this work highlights the significant potential of biodegradable films derived from shrimp shell chitin as a sustainable substitute for conventional fossil-based plastics, thereby addressing major environmental issues. Although this serves as an excellent foundation, further research is necessary, in particular regarding CsNCs.

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Morphological Transitions in Block Copolymer Surface Micelles via Solvent Immersion and the Effective Protein-salt Binding

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In this combined talk, first a comprehensive library of nanopatterns derived from a single block copolymer (BCP) exhibit wide range of structures from simple spheres to more intricate forms, including split micelles, flower-like clusters, toroids, disordered arrays, and other unique morphologies [1]. Using polystyrene-b-poly(2-vinylpyridine) (PS-b-P2VP) surface micelles deposited on SiOx surfaces, a distinctive transformation in morphology is triggered by direct immersion in various solvents. By varying the solvent type, BCP molecular weight, substrate interactions, and temperature, the thermodynamic and kinetic parameters are also affected by these driving morphological transitions. Furthermore, the work highlights the practical utility of BCP nanopatterns as templates for fabricating metal nanostructures via direct solvent immersion. This method provides a versatile and efficient strategy for producing diverse nanostructures, with potential applications in the fields of nanolithography, catalysis, electronics, membranes, plasmonics, and photonics.

Secondly, the protein crystallization (kinetics) and liquid−liquid phase separation (LLPS) are to be presented, showing the effective protein (lysozyme)-salt (NaSCN) binding in the phase diagram, with distinct crystal morphologies, by single- and multi-arm crystals, flower-like crystal structures, whiskers, and sea-urchin crystals [2]. Crystal morphologies exhibit significant variations in changes in protein and salt concentrations. Moreover, the adsorption of SCN^- ions to the surface of lysozyme is effectively enhanced by applying the weak AC electric field in protein crystallization processes.

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Flow and Phase behaviour of small and ultra-soft colloids

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The suspensions of small and ultra-low crosslinked (ULC) Poly(N-isopropylacrylamide) (pNIPAM) microgels studied here remain fluids well above overcrowding. We utilize shear rheology (steady, oscillatory mode) and small angle x-ray scattering (SAXS) to probe the evolution of relative viscosity, plateau modulus, apparent yield stress and nearest neighbour distance (NND) with generalized volume fraction (ζ). The PNIPAM particles have a radius of 100 nm (in the swollen state). The observed behaviour is not reproduced by microgels with same softness (0 mol%) and different sizes (R = 130 and 375 nm) and by microgels with the same size and different softness (synthesized with 5 mol% BIS). From the SAXS data we see a decrease in NND as ζ increases. Furthermore, the ULC microgels show a lack of dynamic arrest for generalized volume fractions well above 1; much higher than the liquid-glass transition observed in larger ULC particles and stiffer microgels.[1] Their linear rheological response shows similarities with the response of star-like micelles.[2] From large amplitude oscillatory shear, (LAOS) we observed a strain thinning behavior (decrease of G', G'' with shear strain) and this might indicate weak interpenetration of pNIPAM microgels.[3] Furthermore, the suspensions remain transparent for all studied concentrations making this system not only interesting to understand the role of softness on the flow properties of the suspension, but also for application as shear modifier for paints and coating.

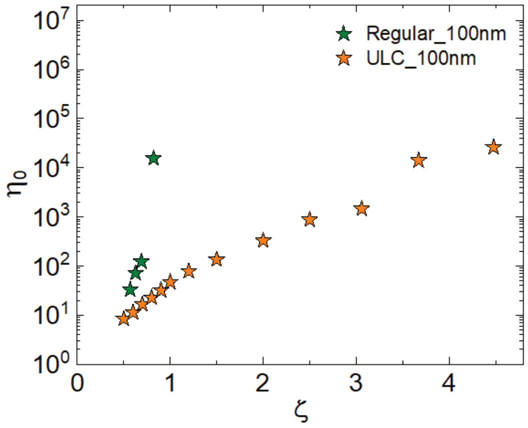

Figure 1: Relative viscosity, η_0, versus generalized volume fraction, ζ, for: ULC with radius 100 nm (orange stars) and 5 mol% crosslinked with radius 100 nm (green stars).

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Start-up response of wormlike micelles through rheology and rheo-microscopy: banding & scission

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We present the linear and non-linear rheology of a wormlike micellar solution containing a common surfactant molecule, cetylpyridinium chloride, and a penetrating salt, diclofenac sodium. The wormlike micelles (WLMs) mesoscopic lengths were evaluated through rheology. The micrometer long micelles were subjected to start-up flow experiments, both with a rotational rheometer and with a linear strain controlled shear cell coupled with a microscope. In non-linear regime, two Weissenberg numbers can be defined, one related to the terminal relaxation time, Wi_d, and one connected to the breaking and reforming time, Wi_b. When Wi_d is lower than one, the measured stress response is monotonically increasing up to a steady state value, and the resulting velocity profile is stable. When Wi_d is higher than one, the WLMs behave as polymer chains in fast flows, aligning and stretching in the flow direction. When $Wi_b>1$, WLMs show a very pronounced strain hardening behaviour, with a stress peak typical of an elastic chain response, appearing at the same strain units above a characteristic shear rate value. A sudden stress decrease appears after the peak, suggesting a breakage phenomenon. The strain at which the stress peak appears permits the evaluation of a WLMs scission energy. The reconstruction of the velocity profiles along the gap of the sample, thanks to a homemade rheo-optical device, and the reconstruction of the complete system flow curve, built with the help of a bio-printer used as a capillary rheometer, both suggest that a shear banding phenomenon appears when Wi_d is roughly equal to one. No connection between banding and scission has been experimentally found.

Surfaces Interfaces and Emulsions I

Tuesday 20 May 15:00 – 16:00

Creating Tailorable Substrates to Investigate High Curvature Effects on Model Biomembrane

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Biological membranes are structurally dynamic, undergoing continuous changes in curvature. Numerous cellular processes, including endocytosis, exocytosis, organelle formation, and extracellular vesicle formation, occur in regions of high curvature (below 300 nm) [1]. High curvature influences biomembrane properties, leading to phase separation, lipid sorting, packing, and domain formation, crucial mechanisms underlying physiological and physiopathological events at the cellular level [2]. However, we still lack a comprehensive understanding of some key mechanisms by which high curvature influences cell membrane behaviour. For example, little is known about the effects of curvature on the organization of ions adsorbed to a membrane or in its proximity, as well as the effects of curvature on the hydrogen bond network of water surrounding the cell. A full understanding of these mechanisms will provide deeper insight into the profound interplay between cell properties and the surrounding environment. Practically, investigating these effects remains experimentally challenging. One approach is to simplify the problem using supported lipid bilayers (SLBs). Model SLBs on highly curved surfaces (less than 100 nm curvature) can provide insights into these processes *in situ* and with high resolution owing to the control of membrane composition and increased spatial stability.

In this project, we devise a strategy for the systematic *in situ* investigation of membrane curvature. First, we develop a protocol to create hydrophilic substrates with precisely controlled curvatures (Fig.1). SLBs are then deposited onto these high curvature substrates. Finally, atomic force microscopy imaging and spectroscopy are employed to investigate the effects of high curvature on SLB properties. The results lay the foundation for depositing lipids onto the substrate, creating models of cell membranes with full control over their curvature and analysing their properties when exposed to a fluid environment with varying ionic strengths.

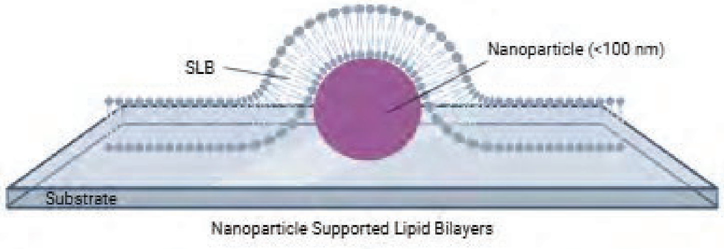

Figure 1: *Sketch of SLB with curvature on nanoparticle (Created with BioRender.com)*

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plic! ploc! : The impact of a water droplet in a surfactant solution to probe the Marangoni flow

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A drop impacting a liquid surface produces the famous “PLOC” sound, which is easy to identify, but becomes a “PLIC” if the liquid in the bath changes. One a surfactant is added to a water bath, the sound of the drop impact changes followed by a significant shift in the correlated hydrodynamic flow. We explore these features to reveal their correlation to the surfactant Marangoni flow at the air-water interface.

Functional Interfaces of Biomimetic Polydopamine and Transition Metal Oxides - Efficient Photocatalysis For Emerging Applications

J. Szewczyk[1,2], I. Iatsunskyi[1], T.Tjards[3], K. Załęski[1], P. P. Michałowski[4], A. Cabot[5], M.Bechelany[2,6], S. Veziroglu[3], E. Coy[1]

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Designing hybrid organic/inorganic materials by integrating biomimetic polymer polydopamine (PDA) with transition metal oxides such as TiO_2 is one innovative and promising approach in the advancing field of photocatalysis [1]. On a larger scale, this can be achieved through free-standing PDA films formed at the air/water interface (a/w-PDA). Here, we present various advanced nanoarchitectures utilizing a/w-PDA. First, by alternately transferring a/w-PDA and oxide nanolayers, we created an ultra-thin multilayer nanocomposite with well-defined interfaces and a gradient-like N-TiO_2 doping structure. Notably, with increasing layer numbers (1, 2, and 3) we observed progressively enhanced photoelectrochemical properties [2]. Next, we presented that the uniform, blanket-like deposition of a/w-PDA onto H:TiO_2 nanopillars significantly improved their already very competitive photoelectrochemical performance [3]. Both approaches combine the advantages of organic and inorganic materials within one nanocomposite (Fig. 1).

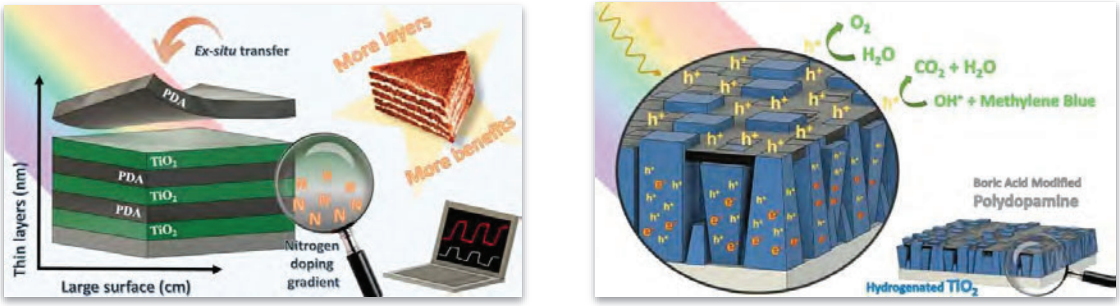

Figure 1: I*llustration of the multilayer [2] and nanocolumnar [3] hybrid nanoarchitectures facilitated with a/w-PDA films and* TiO_2.

This project was supported by the National Science Centre of Poland (NCN) with the grants 2021/41/N/ST5/00211 and 2019/35/B/ST5/00248. Authors acknowledge as well the financial support of project H2020-MSCA-RISE-2017 (project number: 778157).

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Young Scientists Session I

Tuesday 20 May 15:00 – 16:00

Extension and Retraction of impulsively induced viscoelastic jets

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The extensional behavior of saliva plays a key role in aerosol formation during speech, influencing the airborne transmission of viruses like SARS-CoV-2 [1]. To understand these dynamics, we investigate the extension and retraction dynamics of impulsively generated jets with a Drop-on-Demand system, as seen in figure 1a), using the viscoelastic model fluid of bridged micro-emulsions. While the retraction dynamics of Newtonian fluids are primarily driven by capillary effects, the retraction of viscoelastic filaments is accompanied by additional polymeric stresses that arise due to the flow deformation prior to the pinch-off [2]. This can be seen in figure 1b) as the terminal slope of the jet length. By quantifying these stresses one can extract the relaxation time of the model fluid, to establish a framework which allows to characterize the extensional properties of saliva and other viscoelastic fluids. This approach provides new insight into the mechanical response of viscoelastic fluids at high strain rates, with possible implications for disease transmission.

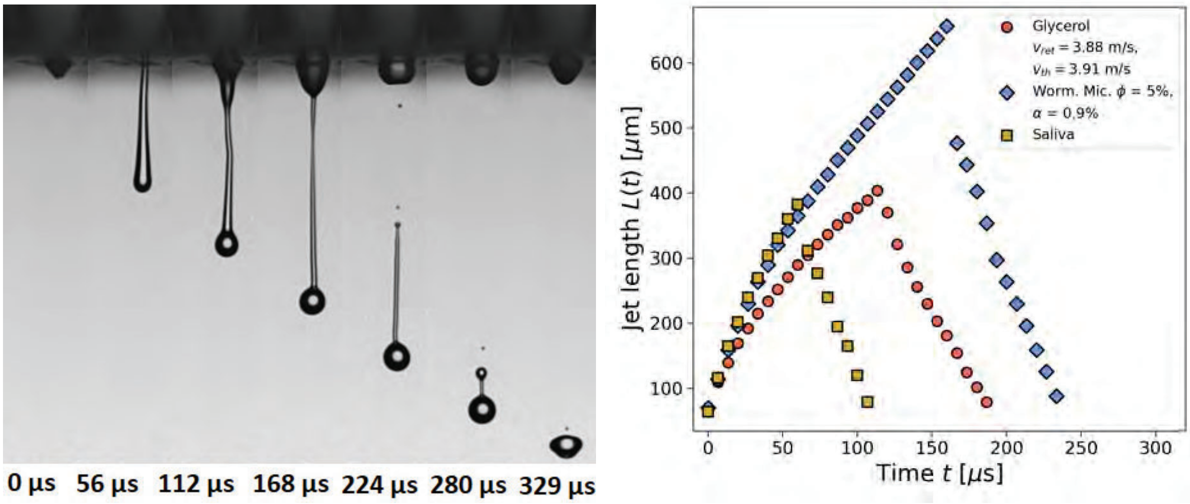

Figure 1: *a) The extension and retraction of a wormlike micellar jet b)Temporal evolution of the jet length $L(t)$ for several different fluids*

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Structure-yielding relationship of red blood cell networks

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Every day, we encounter materials that are neither perfect elastic solids nor Newtonian liquids. One class of such materials are the yield stress fluids, which behave like elastic solids until a stress threshold, commonly called the yield stress, is overcome, allowing the material to begin to flow [1]. A well-known yield stress fluid is blood. In the absence of flow, the red blood cells (RBCs) aggregate and eventually form percolated networks due to proteins like fibrinogen in the blood plasma. Due to their natural origin, RBCs are remarkably uniform, monodisperse discoidal particles, making them an ideal model system to study the network structure and resulting yielding behavior of anisotropic colloidal gels.

In this work, we investigated RBC aggregation, the resulting percolated network structure, and the yielding behavior of RBCs in aggregation agent-containing iso-osmolar solutions. RBC aggregation is induced by either dextran or fd-virus. The fd-virus is a pure depletant, while dextran also bridges neighboring RBCs, resulting in either a disordered house-of-cards [2] or ordered rouleaux network structure [3] (see figure 1). Further, the network density is controlled through the RBC volume fraction, while the interaction strength is mediated by the dextran or fd-virus concentration.

We first establish RBC phase diagrams in terms of RBC aggregate and network structures by adjusting the aggregation agent type and concentration and RBC volume fraction. Here, we focus on establishing the percolation threshold and characterizing the percolated networks in terms of structural descriptors, such as the number of percolation paths. Next, we investigate the yielding behavior in terms of yield stress and strain of various percolated RBC network structures. Our results highlight the intricate connection between network structure and the yielding behavior of colloidal gels.

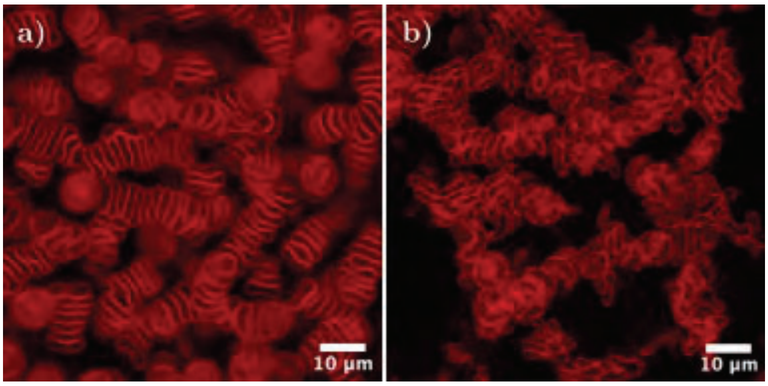

Figure 1: *Percolating network of a) rouleaux aggregates by dextran and b) house-of-card-like aggregates induced by fd-virus.*

References

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Combination of Coevolutionary Information and Supervised Learning Enables Generation of Cyclic Peptide Inhibitors with Enhanced Potency from a Small Data Set

Cristian Fregonese[1]; Ylenia Mazzocato[1], Nicola Frasson[1], Matthew Sample[2], Marta Simeoni[1,3], Alessandro Scarso[1], Petr Šulc[2,4], and Alessandro Angelini[1,3]
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Computational generation of cyclic peptide inhibitors using machine learning models requires large size training data sets often difficult to generate experimentally. Here we demonstrated that sequential combination of Random Forest Regression with the pseudolikelihood maximization Direct Coupling Analysis method and Monte Carlo simulation can effectively enhance the design pipeline of cyclic peptide inhibitors of a tumor-associated protease even for small experimental data sets. Further in vitro studies showed that such in silico-evolved cyclic peptides are more potent than the best peptide inhibitors previously developed to this target [1]. Crystal structure of the cyclic peptides in complex with the protease resembled those of protein complexes, with large interaction surfaces, constrained peptide backbones, and multiple inter- and intramolecular interactions, leading to good binding affinity and selectivity.

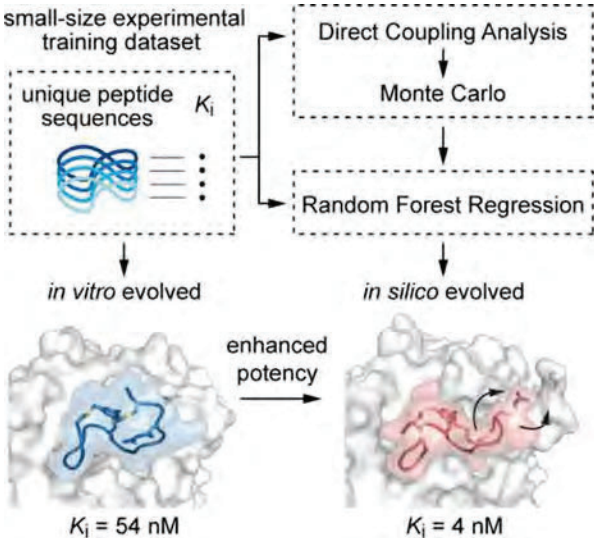

Figure 1: *Combination of statistical and computational approaches enables rapid and cost-effective generation of potent cyclic peptide inhibitors of a human cancer associated protease.*

References

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Gels and Networks

Tuesday 20 May 16:30 – 18:30

Permeation and selective retention of photopolymerized hydrogel membranes with added free polymer chains

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Hydrogels, which are networks of polymer chains in water, have been the subject of recent work in the context of filtration [1]. For example, thin coatings of hydrogels (<100 nm thick) deposited on classical filtration membranes allow to increase the hydrophilicity of the membranes and to decrease their fouling by hydrophobic proteins. An important remaining challenge is to control the selectivity and permeability of the hydrogels. We have developed a series of hydrogels of controlled permeability obtained by a simple and robust method. We photopolymerize poly(ethylene glycol) diacrylate, (PEGDA) under UV light in the presence of non-crosslinkable PEG free chains. We find that the PEG chains are trapped in the matrix and nevertheless enable to increase the permeability by orders of magnitude [2,3]. To account for this behavior we suggest that the PEG chains induce nanodefects in the cross-linking density that controls the permeability of water through the hydrogels. Furthermore taking advantage of the fact that the free polymer chains are trapped in the PEGDA hydrogels we vary the type of free polymer chains to functionalize our hydrogels and selectively retain solutes through hydrogel bonds or electrostatic interactions.

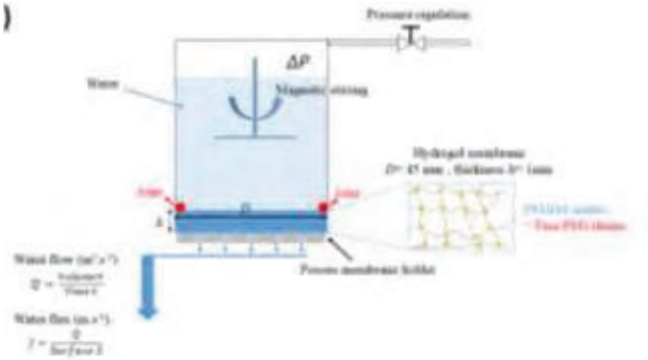

Figure 1. Permeation of water through PEGDA hydrogel membranes containing free PEG chains

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Thermo-Rheological Characterization of Konjac Glucomannan aqueous solutions

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Konjac Glucomannan (KGM) is a high molecular weight, non-ionic polysaccharide, which has attracted considerable interest in drug delivery, tissue engineering, biomedicine, and food processing. KGM is known to form thermally irreversible gels only upon alkaline driven deacetylation, and thermally reversible gels when compounded with other polysaccharides. When KGM alone is dissolved in neutral water, however, it is not known to form a gel and, despite its widespread use, a thorough rheological characterization is still lacking. In this paper, we explore the rheology of KGM aqueous solutions under rotational shear flow both in the linear and non-linear regime, within a wide range of concentrations and temperatures. The results show that the rheology of KGM is complex, due to a relatively sharp conformation transition of the polymer chain as a function of temperature. Based on previous observations reported in the literature, we show through rheology that the transition separates a random coil conformation at higher temperatures from a more rigid structure, where helical chain segments are present, at lower temperatures. The conformational changes reflect also in a qualitative change of the rheological response in the concentrated regime, where the solution becomes viscoplastic at lower temperatures and thermorheologically not simple, suggesting the formation of a weak gel.

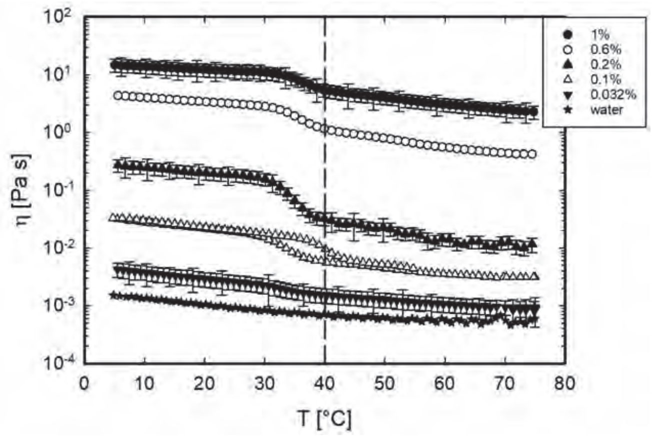

Figure 1: *KGM transition behavior in terms of viscosity*

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Multifunctional Supramolecular Gels with Strong Rheological Performances Formed by Self-assembly of Polyoxometalate-based Coordination Polymers

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Metallogels built in a bottom-up approach by metal coordination and supramolecular interactions have appealing potentialities for the elaboration of smart materials, such as self-healing an enhanced homogeneity compared to polymer gels. [1,2] The formation of supramolecular coordination polymers driven by complexation of cobalt (II) or zinc (II) ions with polyoxometalate-based hybrids displaying two terpyridine ligands in a linear arrangement will be presented. Thanks to the electrostatic interactions between the polyoxometalate cores and the metal nodes, the supramolecular polymer chains self-assemble into fibers that physically cross-link to form gels above a critical concentration. The supramolecular organization of the chains in the fibers and the resulting processes leading to gelation will be shown based on spectroscopy, microscopy, X-ray scattering, and rheometry, complemented by molecular dynamics simulations. These gels display high stiffness and ability to self-heal, as well as appealing physical properties, such as birefringence, luminescence and spin crossover, paving the way for their use as building blocks for multifunctional smart materials. [3]

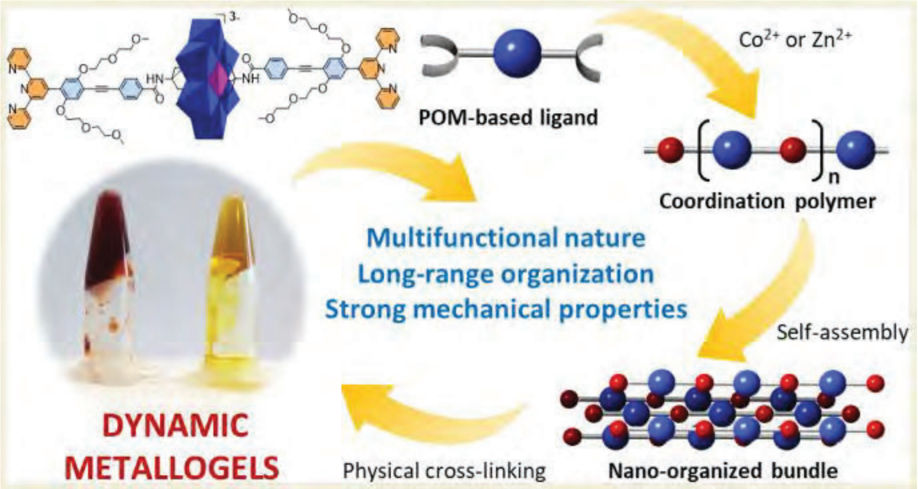

Figure 1: *Molecular structures of the organic and hybrid compounds, and schematic representation of the coordination-driven gelation.*

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Phase Diagram and Structural Transitions in Multiblock Amphiphilic Copolymer Hydrogels

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Multiblock alternating amphiphilic copolymers (AAPs) [1,2] exhibit unique self-assembly and gelation behavior, making them highly promising for applications in biomedicine, soft robotics, and functional materials. In this study, we investigate the structural and mechanical properties of P(CnEGm) AAPs, which consist of alternating hydrophobic segments derived from dicarboxylic acids and hydrophilic segments from poly(ethylene glycol) (PEG), forming hydrogels in water across a wide concentration range. The phase behavior and structural properties are analyzed using small-angle X-ray scattering (SAXS), differential scanning calorimetry (DSC), and rheology. Our findings reveal a concentration and temperature dependent phase transition from a disordered micellar gel at low concentrations to highly ordered BCC phase at intermediate concentrations, followed by micellar overcrowding and phase separation at high concentrations [3]. Rheological analysis confirms a nonmonotonic trend in elasticity, with the 50 wt% system exhibiting the strongest gel-like behavior, while both 30 wt% and 70 wt% systems display weaker mechanical responses due to insufficient interaction or excessive phase separation, respectively. These findings provide a comprehensive phase diagram that correlates structural evolution with thermal and mechanical properties, offering new insights into the design of composition-tunable hydrogels with tailored elasticity, viscoelasticity, and thermoresponsive behavior.

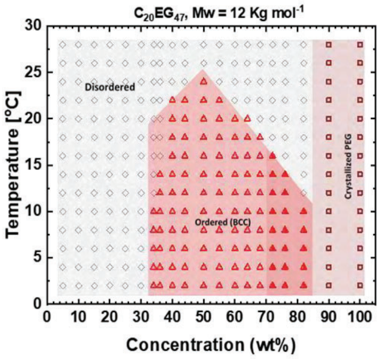

Figure 1: The phase diagram of 12 kg/mol P($C_{20}EG_{47}$) AAP shows morphological transitions from a disordered gel at low concentrations to a BCC ordered gel at higher concentrations, and finally to a partially ordered lamellar phase at the highest concentrations.

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Elastocapillarity in spinning polydimethylsiloxane (PDMS) droplets: Transition from liquid droplets to soft crosslinked beads

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Rotating droplets are among the most studied cases of liquids deformed by an external field as they are relevant in many situations such as large-scale flows taking place in oceans, atmosphere, and in the very body of planets and stars.[1] In this experimental study, we have characterized the interplay between interfacial energy and bulk elasticity in milimeter-size spinning polydimethylsiloxane (PDMS) liquid droplets and elastic beads, which were cured in situ. When subjected to the external centrifugal force, elastic bodies change their shape due to the interplay between the destabilizing centrifugal force and the restoring bulk elastic and surface tension forces [2-3]. Considering the beads as neo-Hookean solids up to large deformations, we have determined the shear modulus (G) and interfacial energy (γ) from their equilibrium shape, varying progressively the crosslinker concentration (C_i). We were able to determine the critical gelation concentration (C_{gel}) of crosslinker above which G' increases with increasing C_i, in agreement with shear rheology data. Quite intriguingly, we have observed that close to the liquid-to-gel transition, a very small shear modulus G' is accompained by a decrease of the interfacial energy γ, which eventually becomes zero at $C_i \approx C_{i,cr} > C_{gel}$. Our approach for the first time shows the adaptation of a spinning drop tensiometer (SDT) as a tool to investigate the interplay between bulk elasticity and surface tension effects in elastomer beads as a function of their molecular structure.

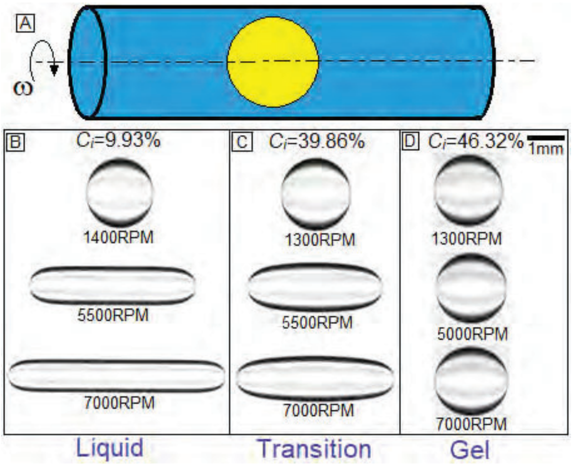

Figure 1: (A)Schematic of the experimental setup of a spinning bead/droplet inside a rotating capillary; (B-D)Snapshots of the evolution of the spinning PDMS droplets with varying crosslinker concentrations.

Keywords: Elastocapillarity, Soft bead, Viscoelasticity, Spinning drop tensiometer (SDT), Elastic modulus, Surface tension.

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Biobased, biodegradable, tough hydrogels from itaconic acid monomer

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Hydrogels are three-dimensional cross-linked polymer networks capable of retaining significant amounts of water in their structure while still maintaining the properties of solid materials. They can help reduce the need for traditional raw materials while retaining interesting mechanical and functional properties. Hydrogel materials are used for various applications including hygiene products, agriculture, biomedicine and even robotics[1]. However, most of these hydrogels are petroleum-based and not degradable. Existing biodegradable hydrogels are made from high molar mass biobased polymers (such as chitosan, alginate, cellulose, etc.) which makes it difficult to control and tune their architectures. In this context, we have developed fully biobased hydrogels using itaconic acid, a renewable monomer analogous to acrylic monomers, extracted by fermentation of lignocellulosic biomass with fungi[2]. By designing hydrogels from the monomer, we gain control over the structure and the functionality of the material, offering a renewable and eco-friendly alternative to traditional petrochemical-derived acrylates.

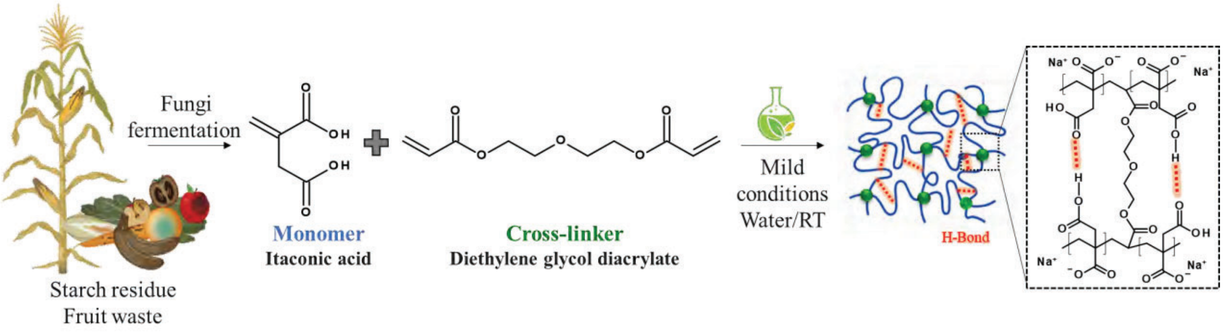

Figure 1: *Schematic representation of the synthesis of itaconic acid-based hydrogels.*

The specific interactions within the polymer matrix, due to the presence of a high density of hydrogen bonds combined with the polyelectrolyte aspect of the gel, are responsible for unique physico-chemical and mechanical properties. A detailed study was carried out to examine their swelling and mechanical behavior, in order to clarify how these interactions govern the overall performance of hydrogels. Complex non-Fickian diffusion was also observed during transient swelling, marked by a unique morphological evolution. Finite element method (FEM) simulations were conducted to model the coupling between solvent migration and polymer deformation, allowing better predictions of stress-induced diffusion, network swelling, and mechanical responses. Furthermore, preliminary tests according to OECD 301F standard highlight the promising biodegradation potential of these hydrogels.

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Young Scientists Session II

Tuesday 20 May 16:30 – 18:30

Environmental effects on the spatio-temporal evolution of plasma-wrinkled PDMS thin films

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We investigate the surface plasma oxidation of polydimethylsiloxane (PDMS) elastomers and the impact of environmental conditions on the spatio-temporal evolution of the resulting wrinkling patterns. We employ a combination of small-angle light scattering (SALS), XPS, and optical and atomic force microscopy to examine the evolution of the crack density, and wrinkling profile following plasma exposure. Previous reports have rationalized the spontaneous isotropic wrinkling of PDMS supported thin films following plasma exposure in terms of a thermally driven differential deformation of the elastomer normal to the direction of incident exposure.[1,2] Our findings challenge this widely accepted mechanism and provide evidence that, while temperature plays a role, other environmental factors largely govern this process and can account for the considerable variability of results reported in the literature using this method. These findings open new possibilities for exceptional control of PDMS plasma functionalization, in terms of tunable surface patterning, adhesion, and wettability.

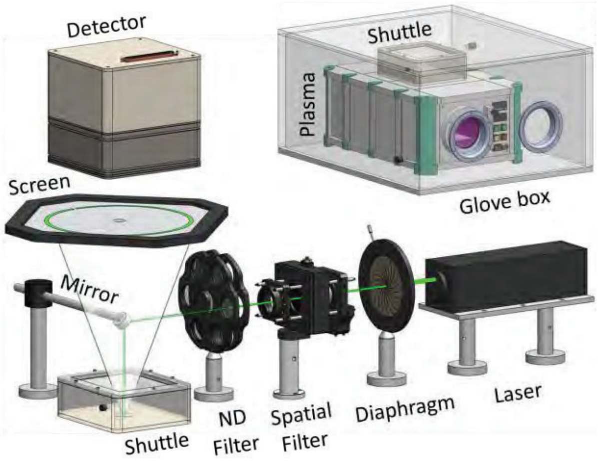

Figure 1: *Schematic of plasma exposure and small-angle light scattering (SALS) setup in controlled environment to study the environmental effects on spontaneous wrinkle formation in plasma oxidised PDMS.*

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Evaluation of disposable PDMS rheometer geometries for bulk rheology measurements on *P. putida* biofilms during growth

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Biofilms are surface-associated bacterial communities of cells enclosed in a matrix of self-produced extracellular polymeric substances. While undesired in many domains such as wound healing, food production, medical treatments and industrial fouling, biofilms are also growing in importance as a positive tool in biotechnological applications including bioremediation, biofertilization and energy production. Biofilms are viscoelastic in nature, allowing them to adapt to a changing environment, and significantly complicating their mechanical removal.

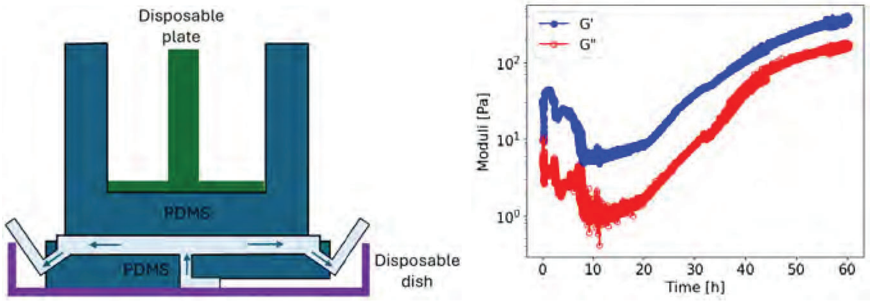

Figure 1: *Illustration showing the geometries used. (Left). Rheological time sweep of a P. putida biofilm growing under flow (Right). The test was conducted at a 150 µm gap and 30 °C, and supplied with 1.1 mL/h of LB medium (10 g/L tryptone, 5 g/L yeast extract, and 5 g/L NaCl).*

Understanding the mechanical properties of biofilms is thus a crucial aspect to many applications. However, their rheological characterization is difficult due to the inherent change of the biofilms heterogeneous structure when transferring and loading onto conventional measurement tools [1]. In this work we demonstrate how to adapt conventional rheometry to the study of biofilms. More precisely, the evaluation and use of disposable PDMS geometries, where the biofilm can be grown under relevant conditions (flow of nutrients, temperature) is shown. For the geometries, custom-made 3D-printed molds are used to prepare biocompatible and oxygen-permeable PDMS plates. Special care has to be taken to ensure alignment of the PDMS geometries, as the measurements occur at low gaps (∼ 100 µm). The setup developed here allows to grow biofilms *in situ*, directly on the PDMS geometries, eliminating the need to scrape and transfer the biofilm. Furthermore, the setup allows to feed the biofilm with nutrients via a channel structure integrated into the PDMS geometries, allowing to obtain long-term, time-resolved information of the rheological properties and the role of nutrient conditions.

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Hydrogel Design for Cornea-on-chip Drug Screening Model

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[2]*UGent, Polymer chemistry and Biomaterial Group, Gent, Belgium*

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Organs-on-chips are biofabricated devices that simulate a more realistic human organ environment compared to traditional cell cultures, offering the potential for more accurate insights from in-vitro testing before progressing to clinical trials. Our group aims to fabricate a full-thickness cornea-on-a chip, incorporating the three cellular layers within a microfluidic device that simulates all relevant physiological flows. This artificial cornea is created by photocrosslinking a 500µm thick hydrogel embedded with human keratocytes, followed by seeding epithelial and endothelial cells on each side. Candidate hydrogel biomaterials and human corneas are characterized to provide insights for both fabrication and accurately mimicking the human cornea. This opportunity to intensely focus on comparing the physicostructural properties of both hydrogels and human corneas using a plethora of techniques has provided understandings that benefit the field of tissue engineering.

We begin by addressing common misconceptions in tissue engineering regarding hydrogel structure.[1] Next, we demonstrate how more accurate insights can be gained using the theory of swollen networks, originally proposed by Flory in the 1940s. Building upon it, we developed a new theoretical framework for the understanding of hydrogels, suited to crosslinked gelatins, allowing prediction of hydrogel structure through swelling, elasticity and diffusion measurements and vice-versa. We compare this hydrogel structure to that of the human cornea, using *ex-vivo* samples. This comparison underscores limitations of hydrogels as extracellular matrix substitutes, as their homogeneous networks do not replicate some key properties of human tissue. We demonstrate that these shortcomings can only be overcome by using composite materials. Finally, our latest advancements in the engineering of the cornea-on-a-chip will be presented. These findings pave the way for more accurate models in tissue engineering, bringing us closer to replicating the complex structures and functions of human tissues.

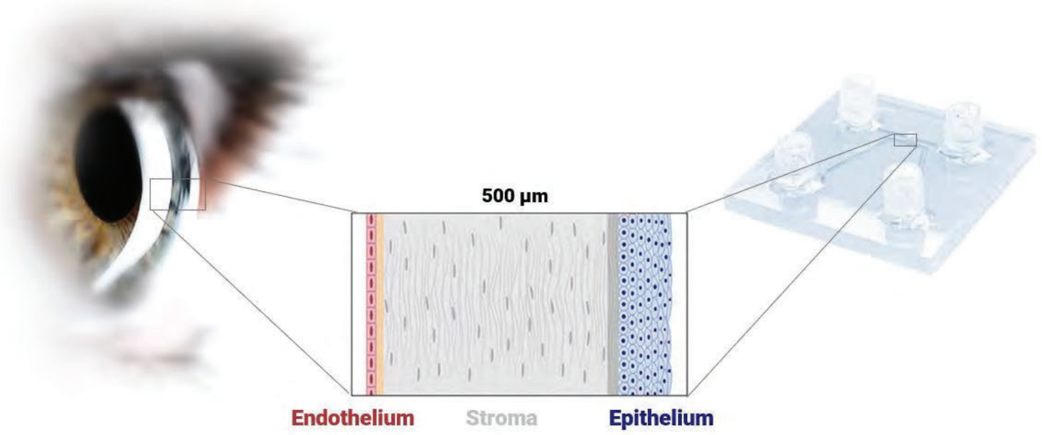

Figure 1: *Cornea-on-a-chip: a microphysiological model is fabricated by photolithographic bioprinting a cell-laden hydrogel inside a microfluidic chip.*

References:

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Direct Ink Writing of PANI Structures for Electromagnetic Shielding

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The increasing electromagnetic interference (EMI) from electronic devices due to rapid technological advancements poses a growing concern. Conductive polymers, known for their lightweight, environmental stability, durability, and tunability, offer a promising solution for EMI shielding. Polyaniline (PANI) is a particularly effective conductive polymer but is limited by its poor processability. The dispersion of the PANI emeraldine base (PANI-EB) in dodecylbenzenesulfonic acid (DBSA) overcomes this limitation by forming a viscous paste, where DBSA acts as a dispersant and dopant, enabling 3D printing[1].

This study investigates the rheology and 3D printing of polyaniline doped with DBSA considering the enhancement of electromagnetic shielding as a priority. The PANI is synthesized through chemical oxidative polymerization of aniline and then mixed with DBSA to form a 3D printable ink. The 3D printability is then confirmed using rheological shear rate ramps tests, creep tests, and oscillatory amplitude sweeps. Using direct ink writing (DIW), we fabricate samples to study their dielectric behavior (10^{-3} to 10^{7} Hz) and EM response(8 to 12 GHz). We systematically analyze how different 3D printed designs such as; plain filled and patterned geometries (honeycomb, grid) affect the EM shielding performance.

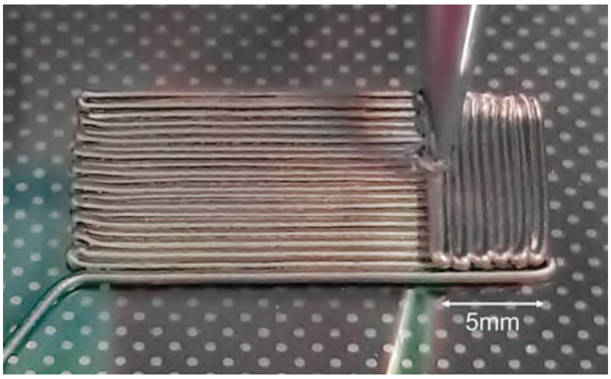

Figure 1: *DIW of PANI, fabricating a rectangular grid structure for EM shielding*

Through precise characterization, we aim to demonstrate that EMI shielding can be maintained with minimal material content and we explore the transition of the EM shielding mechanism from reflection to absorption by manipulating material properties and 3D printed geometries. This study demonstrates the potential of 3D printable PANI to provide customizable EM shielding solutions for advanced technological applications.

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Reductive catalytic fractionation-derived lignin nanoparticles and cellulose as multifunctional composite components for the removal of ciprofloxacin from wastewater

Giorgia Peroni[1], Federico Bugli[2], Sam Thomas[2], Alessio Baldelli[2], Matteo Gigli[1], Claudia Crestini[1], Fabrizio Cavani[2], Tommaso Tabanelli[2], Massimo Sgarzi1[1]
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Among the most concerning emerging pollutants in wastewaters, an eminent role is played by antibiotics,[1,2] whose adequate treatment cannot be currently provided by most wastewater treatment plants.[3] In the context of sustainable water remediation, novel multifunctional materials can be produced using wood from agroforestry waste, exploiting adsorptive and photocatalytic behaviour of cellulose and lignin.[2,4] These two components of the lignocellulosic biomass can be effectively separated with structure preservation using Reductive Catalytic Fractionation (RCF), which does not utilize harmful solvents.[5] In this work, the simple and inexpensive hydrotropic (HTNP) and solvent-antisolvent (SASNP) methods were used to convert RCF lignin into nanoparticles, employing nontoxic reagents. These nanoparticles were subsequently combined in a 1% w/w ratio with RCF cellulose to produce nanoparticle-loaded cellulose thin films.

These films were put in contact with 5 mg/L solutions of the antibiotic ciprofloxacin (CIP). Circa 25% of CIP was removed by adsorption in 150 minutes, followed by a 14% photocatalytic degradation upon a 150 minute UV irradiation ($0.245\ mW\cdot cm^{-2}$), resulting in a total 39% removal.

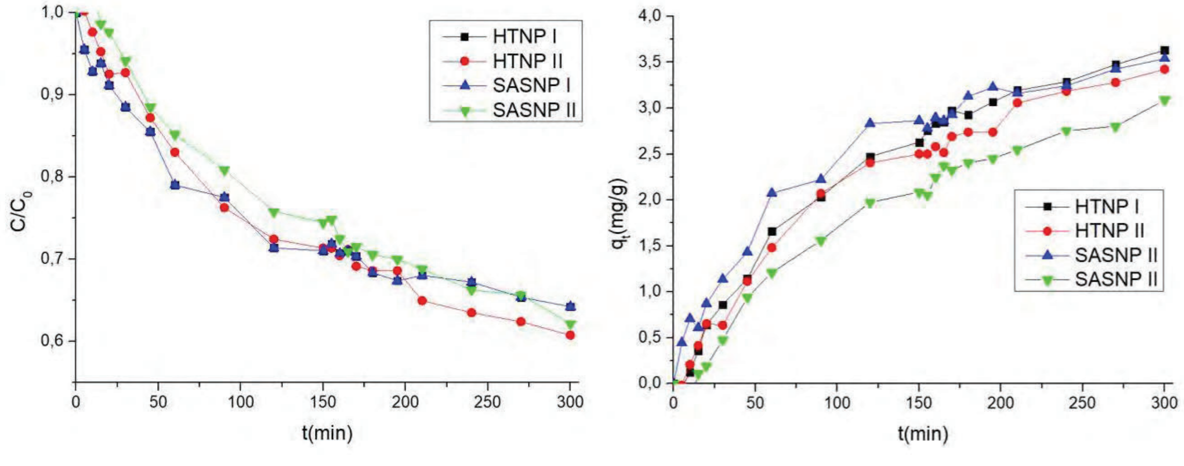

Figure 1: *Left: concentration variation over time for CIP; right: adsorption capacity over time for CIP. Black and red lines: experimental trends of the hydrotropic nanoparticle-loaded films; blue, green lines: experimental trends of the solvent-antisolvent nanoparticle-loaded films*

SASNP-loaded films yielded a significantly higher adsorption kinetic constant (7.6·10-2 min^{-1}) than both HTNP-loaded and pure cellulose films (1.5·10-2 min^{-1} and $1.1\cdot 10^{-2}\ min^{-1}$, respectively), paralleling the better adsorption performance of SASNPs with respect to HTNPs in aqueous dispersion.[4]

No photocatalytic activity was recorded for pure cellulose films, demonstrating that RCF cellulose acts only as an adsorbent. Conversely, the photocatalytic activity of HTNP-containing films

($5.5 \cdot 10^{-3}$ min^{-1}) was significantly higher than SASNP-containing films ($9.0 \cdot 10^{-4}$ min^{-1}), again confirming the behaviour registered in aqueous dispersion.[4]

In conclusion, multifunctional composite thin films for ciprofloxacin removal from wastewater were successfully manufactured from second generation lignocellulosic biomass combining innovative biomass fractionation and lignin valorisation methods.

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This study was carried out in the framework of the projects ENCAPSULATE (CUP N. H53D23003890001) (Mission 4 Component 2 Investment 1.1) funded by the European Union Next-GenerationEU – National Recovery and Resilience Plan (NRRP) – Fondo per il Programma Nazionale di Ricerca e Progetti di Rilevante Interesse Nazionale (PRIN).

Multifunctional antioxidant and UV-shielding coating based on highly engineered ultra-small CeO_2 NPs for complex surface protection

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Surface protection is a shared concern of different fields, since the exposed facets of different substrates are most commonly subjected to a variety of deteriorating environmental factors. The issue calls for novel coating strategies, able to counteract complex mechanisms and withstand interaction with different agents.

With this in mind, we designed a multifunctional biopolymer nanocomposite based on cerium oxide nanoparticles (CeO_2 NPs) and we tested it in a challenging case study. First, we engineered the NPs to achieve ultra-small sizes, resulting in a maximization of the surface-to- volume ratio and finally increasing the number of available active sites on the surface. Then, we deeply studied their catalytic activity and we linked it to the Ce(III)/Ce(IV) redox cycling established at their surface. We investigated the interaction of the NPs with different factors (*i.e.*, O_2 atmosphere, H_2O_2 solution, UV light, and high temperature) by means of Environmental XPS, proving the potential of the nanocatalyst for long-lasting protection.

To fully exploit the functionality of the NPs, we incorporated them in an optimized chitosan formulation taking care to guarantee the active sites on the surface of the NPs are not hindered by the matrix. We demonstrated that the synergistic action of CeO_2 NPs and chitosan gives rise to optimal UV-shielding and antioxidant action, by testing the coating in the context of pigments degradation. Both redox and UV-induced mechanisms, induced by simultaneous presence of UV light, chlorine-based salts and high relative humidity, are efficiently limited thanks to the barrier effect of the coating.

Artificial Intelligence and Machine Learning in Soft Matter I

Wednesday 21 May 09:45 – 10:45

Mechanisms of spontaneous translocation through lipid bilayers

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A critical challenge in nanotechnology lies in understanding and controlling how nanomaterials interact with lipid bilayers, the fundamental barrier of biological cells. Traditional understanding posits that passive translocation via thermal motion is hindered for objects larger than 5 nm, with smaller, hydrophobic nanoparticles typically accumulating within the bilayer core and larger nanoparticles (> 5 nm) relying on slow, energy-dependent endocytosis. However, emerging evidence reveals a more complex interplay between nanomaterial properties and bilayer interactions.

We present several instances illustrating that the physicochemical characteristics of nanoparticles significantly influence their passive translocation across lipid bilayers. For example, tuning nanoparticle hydrophobicity can induce reversible perturbations in bilayer structure, increasing permeability to water and small solutes. In stark contrast, lipid-coated hydrophobic nanoparticles have been observed to directly penetrate lipid membranes within milliseconds, suggesting a bypass of traditional size limitations. To further elucidate the complex relationship between molecular structure and membrane translocation, we utilize a computational approach [1] that combines exhaustive enumeration of polymer sequence space with neural network prediction. Specifically, we investigate the translocation time of a copolymer through a lipid membrane as a function of its sequence of hydrophilic and hydrophobic units. Massively parallel Rosenbluth sampling of all possible polymer sequences enables us to establish a comprehensive benchmark of translocation times and develop a meaningful dynamic interpretation based on mean first passage times using training neural network [2]. Further exploration and data assessment, suggest sensitivity of results versus parameter choice in training which needs to be consider in order for the networks to achieve robust outcomes and performance outside of validation-sets parameters.

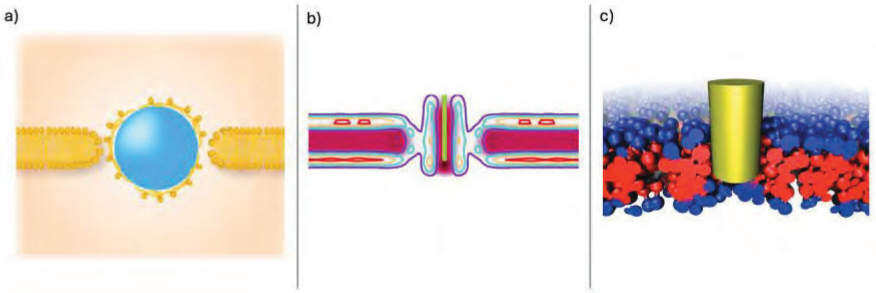

Figure: *The mechanisms of spontaneous translocation through a lipid bilayer by: a) hydrophobic nanoparticle* [3]*;graphene nanosheets* [4]*; carbon nanotubes* [5]

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Learning Classical Density Functionals for Colloidal Systems

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We discuss possible strategies to obtain classical density functionals for the description of inhomogeneous colloidal systems with methods of machine learning [1]. In particular, we will concentrate first on neural density functional theory introduced by Sammüller and coworkers [2] which has recently been shown to be capable to describe 2D and 3D inhomogeneous systems [3]. Secondly, we use the example of patchy particles to describe approaches for anisotropic particles, where the inhomgeneous density profile depends on position and orientation of the particles [4]. As an example, the machine learned functional is able to describe drying at a hard wall with full account of the particle orientations.

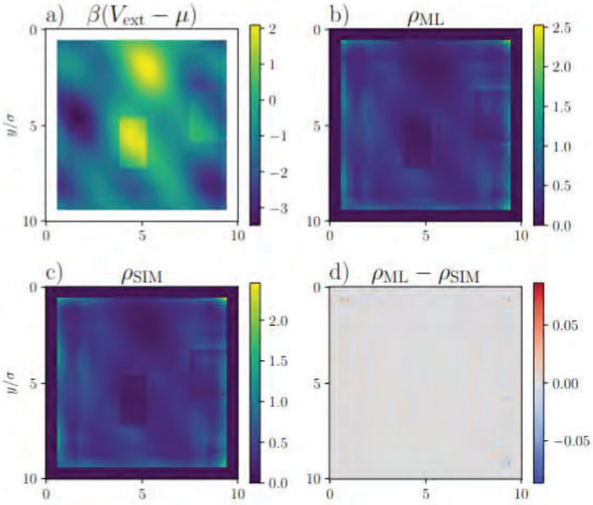

Figure 1: *Machine learning for 2D hard disks: (a) external potential with a hard surrounding box, density profile from (b) a selfconsistent minimization of an ML functional and (c) simulations with (d) showing the difference.*

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Deep-learning-based instability detection of formulated liquids

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Formulated liquids are multiphase systems constituted by aqueous dispersions containing complex chemical species. Being suspensions, such formulations are metastable systems, thus prone to the development of deterioration phenomena. Instabilities lead to changes in appearance and performance of the products and are not welcome. Rheological properties are altered as well.

Therefore, several approaches to assess the stability of formulated liquids have been proposed in literature, but none of them is reported to be reliable or easy to deploy[2]. In this work, we propose a cheap, easy-to-perform and non-intrusive model for the stability assessment of formulated liquids. Such method exploits FLUID[1], a publicly available dataset composed of snapshots of vials containing fabric softeners that develop several instabilities (e.g. phase separation, creaming, cracking, flocculation). Hence, we use FLUID to build a deep-learning-based detector for the instability of liquid formulations. The detection of instability on formulated liquid is of great relevance in both industrial and rheological field. In fact, monitoring 'macroscopically' their stability can help identifying the time-to-failure, which is helpful for the depiction of the properties of the material.

The proposed method can in principle be used to support the rheological characterization of liquid formulations and other systems which are subject to visual observation.

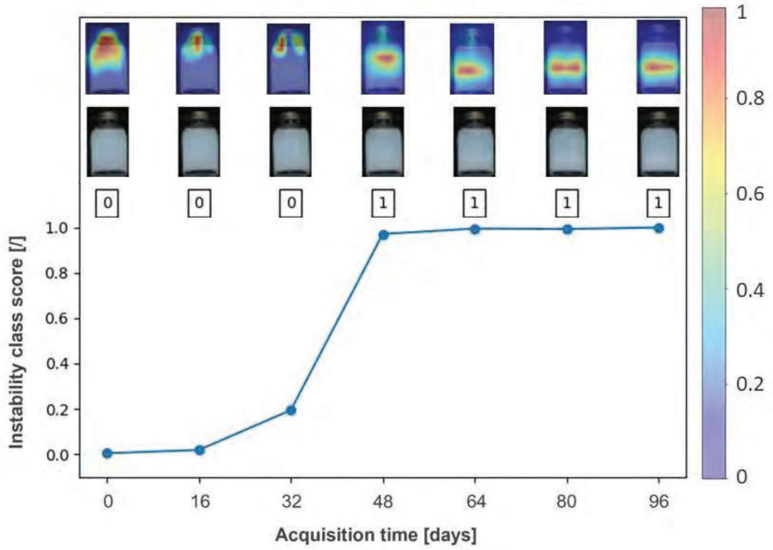

Figure 1: *Evolution of the instability score over time of a formulated liquid subject to phase separation with the corresponding interpretability analysis (heatmaps) and ground truth (0 for stable, 1 for unstable).*

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Biological Soft Matter I

Wednesday 21 May 09:45 – 10:45

Untangling plant cell walls biomechanics and the control of transport

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A key property of all living systems is intercellular communication which coordinates growth in response to external cues. Communication in plants is restricted by rigid walls. Plasmodesmata evolved as a route for the symplastic (cytoplasm-to-cytoplasm) transport of small and large molecules including signalling proteins and RNAs, metabolites and hormones. These are small pores that appear in cell wall domains enriched in the β-1,3 glucan polysaccharide callose. We studied the mechano-physical properties of callose underpinning its function in the regulation of the plasmodesmata [1]. Applying computer simulations and experimental approaches, the properties of callose in improving cellulose hygroscopicity and visco-elasticity were identified [2]. We also developed molecular probes and identified proteins that modify callose accumulation in cell walls [3]. These tools allowed us to uncover the structural properties of these cell walls microdomains that can be exploited in plant biotechnology and in biomaterial development. I will share how we build on this knowledge and discuss potential applications in understanding the soft matter of biological systems and tackling environmental sustainability challenges.

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Unveiling Myelination, Brain Microstructure and Connectivity: Combining 3D-scanning SAXS, WAXS, and 3D-PLI

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We used 3D-scanning SAXS[1] on a fixated Ara brain slice to reconstruct the 3D orientation and density of myelinated nerve fibers with a spatial resolution of 50 and 20 µm². Furthermore, we demonstrate that SAXS and WAXS can reveal a wealth of anatomical features without the need for staining by carefully analyzing up to a million scattering curves using a fast and reliable fitting routine. These features include the degree of myelination, the 3D orientation of nerve fibers, different cellular layers, and crystalline myelin.

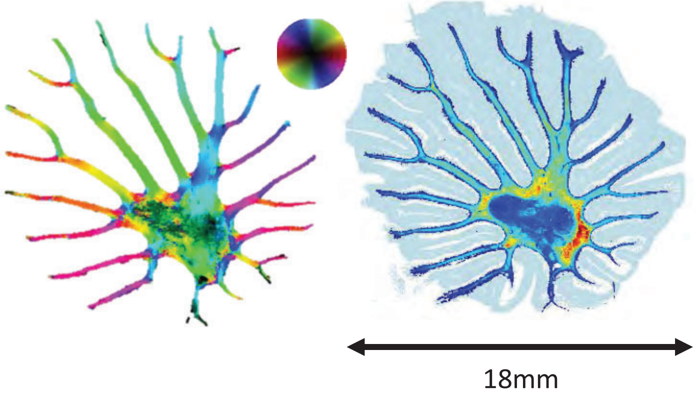

Figure 1: *left: in plane orientation of nerve fibres colorcoded as in the colorwheel, right: myelination degree of the white matter fiber tracts.*

We then quantitatively correlate these findings with data obtained from 3D-PLI[2]. While 3D-PLI offers the advantage of being significantly faster, achieving higher resolution, and being compatible with optical microscopy, it lacks the ability to unambiguously differentiate myelination from other retardation effects inherent to brain tissue. By integrating the two techniques, we aim to establish a direct correlation between the myelination detected via X-rays and the information contained in 3D-PLI data. This could allow for the determination of myelination levels from 3D-PLI data alone, significantly advancing our ability to map neural connections across different species, including humans.

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Interaction of the Parkinson's-Related Intrinsically Disordered Protein α-Synuclein with Biomembranes: A Neutron Reflectivity Examination

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Intrinsically disordered proteins (IDPs) lack a defined three-dimensional structure but can adopt specific conformations upon interaction with binding partners. α-Synuclein (α-Syn), a neuronal IDP, is closely associated with neurodegenerative disorders such as Parkinson's disease, where its aggregation into fibrils plays a key pathological role [1]. α-Syn's interaction with lipid membranes is a critical factor influencing its aggregation, toxicity, and contribution to disease progression [2]. However, the molecular mechanisms underlying these interactions remain poorly understood.

Neutron Reflectometry (NR) was employed to investigate the interaction of α-Syn with supported lipid bilayers of varying charge compositions. The bilayers, composed of partially deuterated DMPC and DMPG lipids, were analysed to assess structural changes upon protein binding. Circular Dichroism (CD) and Infrared (IR) spectroscopy were used as complementary techniques to examine secondary structure formation and membrane changes during protein-membrane interactions.

NR revealed that membrane charge plays a crucial role in α-Syn binding and its ability to disrupt membranes, showing detergent-like behaviour. The binding interactions were relatively weak, maintaining an equilibrium between free and membrane-bound proteins. Temperature-dependent effects were observed, with distinct behaviours at different temperatures. CD spectroscopy demonstrated a formation of α-helical structure upon binding to charged membranes, while IR spectroscopy confirmed α-Syn's detergent-like activity and its ability to destabilize membrane integrity.

This work addresses critical gaps in understanding α-Syn's interactions with membranes and its associated toxicity. It emphasises the importance of membrane charge, pre-existing damage, and temperature in modulating α-Syn behaviour. These findings enhance our understanding of α-Syn's role in neurodegenerative disorders and provide valuable insights for identifying potential therapeutic targets to mitigate its pathological effects.

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Artificial Intelligence and Machine Learning in Soft Matter II

Wednesday 21 May 11:15 – 12:15

Machine learning of glass transition temperature from chemical structure in conjugated polymers

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Conjugated polymers (CPs) have diverse applications in organic photovoltaics, field-effect transistors, secondary batteries, and light-emitting diodes. The mechanical properties of polymers undergo significant changes at the glass transition temperature (T_g). Below T_g, polymers are hard and brittle, while above T_g they become flexible and elastic. For applications in soft electronics, designing CPs with a T_g below room temperature is highly desirable to optimise on flexibility in their operational temperature range. This highlights the importance of predicting T_g from chemical structure, enabling cost-effective material design. CPs have a complex molecular structure, featuring a stiff backbone that is primarily composed of aromatic groups. Chain flexibility is controlled through the introduction side chains, which effectively plasticize the polymer. The side chain topology can vary through size, branching and the incorporation of ringed groups. Furthermore, the presence of multiple distinct side chains within the repeating unit adds another layer of structural complexity.

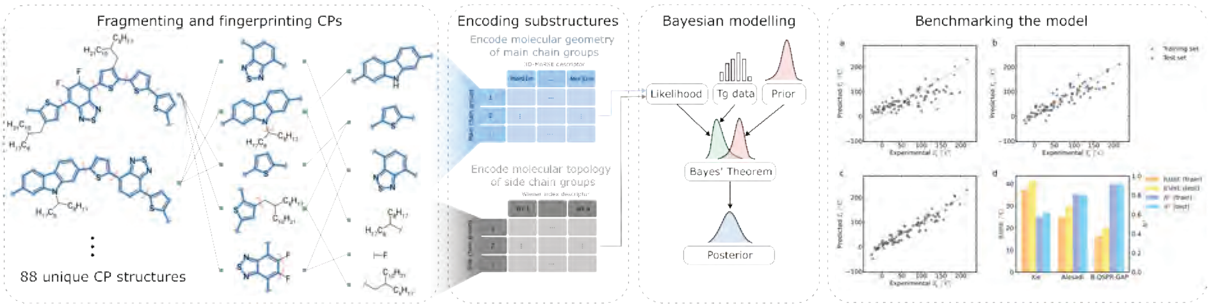

Figure 1: Illustration of this work, from the monomeric units to the final T_g prediction.

To address this challenge, we propose a probabilistic machine learning model powered by Bayesian inference to predict T_g directly from chemical structure. We benchmark our model against existing CP models in the literature, demonstrating a leap in predictive performance for new, unseen CPs; e.g., achieving an R-squared of 0.91, compared to the next-best CP model at 0.80. Our modelling framework demonstrates a simple yet general approach to complex polymer structures by breaking down the chain repeat unit into smaller, more manageable, substructures. Then, a combination of group contributions (GC) and quantitative structure-property relationships (QSPR) approaches is used to link the substructures to T_g. To validate the generality of our framework, we also demonstrate its application to the prediction of T_g from chemical structure in poly(aryl ether ketones) (PAEKs) [1].

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A Machine Learning Approach to Identify Carbon Dioxide Binding Proteins for Sustainability and Health

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Carbon dioxide (CO_2) has a fundamental role in biological processes throughout the biosphere. Although much is known about the impact of CO_2 on the overall physiology of an organism, much less is known about how its interaction with specific biomolecules may affect their function [1, 2]. Our work focuses on carbamylation, a non-enzymatic reversible protein post-translational modification (PTM) where CO_2 binds onto the neutral lysine ϵ-amino groups [3]. A novel mass spectrometry-based experimental technique, TEO trapping, has recently been used to demonstrate that, for reasons yet unclear, CO_2 does not bind to all lysines [2]. While informative, this experimental technique is laborious, which limits our ability to gain a clear and comprehensive view of the interactions between CO_2 and proteins. For this reason, we are conducting extensive molecular dynamics simulations and developing a computational method to predict which lysines, in any protein, may undergo carbamylation. We found that standard metrics (pKa, solvent accessible surface area, and amino acid depth) are insufficient to single-handedly explain why some lysines may be modified, but not others. Therefore, we are now exploring other scalar metrics and the usage of atomic environment vectors, commonly used in machine learning, for this classification task.

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Machine Learning many-body potentials for charged colloidal suspensions

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The Derjaguin-Landau-Verwey-Overbeek (DLVO) theory has been remarkably successful in describing the phase behavior of charge-stabilized colloids in solution. However, it fails to explain phenomena that suggest attractive interactions between like-charged colloids, particularly in low-salt solutions [1]. Simulations can give insight into these interactions. Unfortunately, simulations of charged colloidal suspensions with explicit ions, known as primitive model simulations, are computationally intensive. To overcome this challenge, we use machine learning (ML) techniques to develop an effective coarse-grained many-body potential for charged colloids, thereby eliminating the need to simulate explicitly the ions in solution, as illustrated in Fig. 1. We construct these effective many-body potentials by expressing them as a function of all colloid coordinates, using a set of two-body and three-body symmetry functions, and fitting them with a simple linear regression scheme.

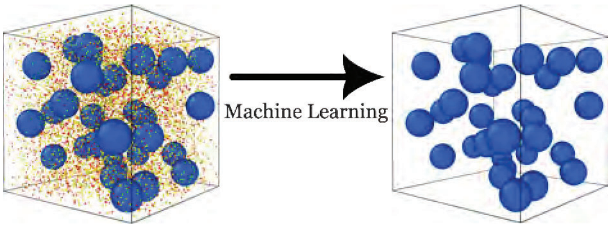

Figure 1: *Using a machine learning framework we generate a coarse-grained many-body potential, eliminating the need to explicitly consider ions in colloidal suspension when performing simulations.*

Using these ML potentials significantly accelerates the simulations while providing new insights into the intriguing ‘like-charge attractions’ observed between colloids in solution. In this study, we present the framework for developing these effective potentials and compare them to other potentials used to describe suspensions of charged colloids. Additionally, we investigate the gas-liquid and gas-solid coexistence behavior arising as a result of colloidal ‘like-charge attractions’ using simulations employing ML potentials. Our simulations reveal that phase coexistences found in low-charge electrolytes cannot be directly extrapolated to high-charge electrolytes, a conclusion that contrasts with predictions based on Poisson-Boltzmann theories [2].

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Biological Soft Matter II

Wednesday 21 May 11:15 – 12:15

Adhesion-driven vesicle translocation through membrane-covered pores: modeling the host-cell entry of apicomplexan parasites

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Translocation across barriers and through constrictions is a mechanism that is often used in vivo for transporting material between compartments. A specific example is apicomplexan parasites invading host cells through the tight junction that acts as a pore, the most prominent apicomplexan is Plasmodium falciparum that causes malaria. A similar pore translocation is involved in drug delivery using lipid vesicles to penetrate intact skin.

Here, we use triangulated membranes and energy minimization to study the translocation of vesicles through pores with fixed radii. The vesicles bind to a lipid bilayer spanning the pore, the adhesion-energy gain drives the translocation, and the vesicle deformation induces an energy barrier. In addition, the deformation-energy cost for deforming the pore-spanning membrane hinders the translocation. Increasing the bending rigidity of the pore-spanning membrane and decreasing the pore size both increase the barrier height and shift the maximum to smaller fractions of translocated vesicle membrane. We compare the translocation of initially spherical vesicles with fixed membrane area and freely adjustable volume to that of initially prolate vesicles with fixed membrane area and volume. In the latter case, translocation can be entirely suppressed. Our predictions may help rationalize the invasion of apicomplexan parasites into host cells and design measures to combat the diseases they transmit.

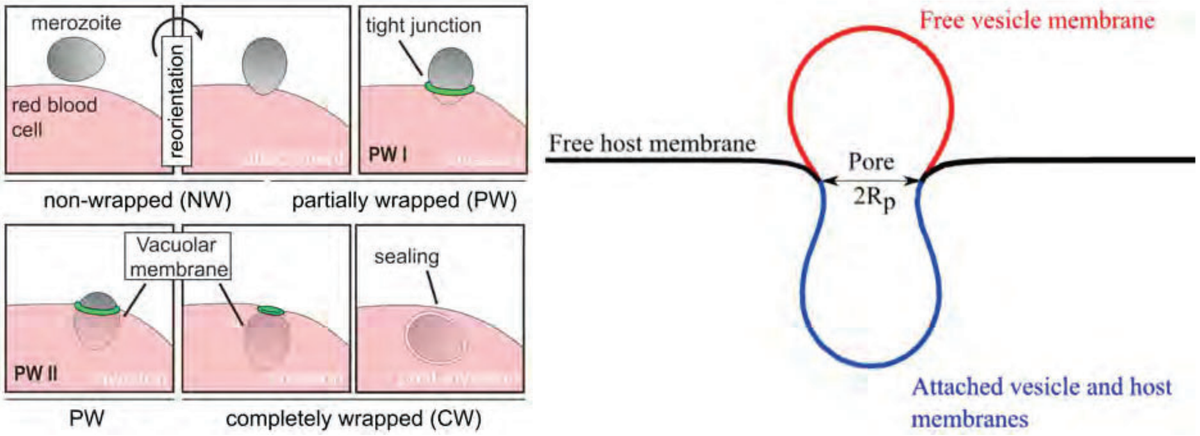

Figure 1: *Host-cell invasion of apicomplexan parasites. (left) The stages of Plasmodium falciparum merozoite invasion [1]. Schematic representation depicting different wrapping phases of the merozoite from reorientation through to invasion and postinvasion. (right) Vesicle-pore transition as a model for a deformable parasite squeezing through the tight junction [2].*

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Antimicrobial Peptide Interactions with Solid Supported Lipid Bilayer to Combat Antimicrobial Resistance

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Antimicrobial resistance in society has led to an increase in research in finding alternative methods and drugs to treat infections. Antimicrobial peptides (AMPs) have emerged as a promising alternative to traditional antibiotics due to their ability to disrupt bacterial cell membranes and avoid development of resistance. The mechanisms of many AMPs interaction with bacterial membranes have yet to be fully understood, however, various models have been proposed. In our experimental studies we explore a novel AMP (IK8) which has been shown to have antimicrobial properties against antibiotic resistant strains of bacteria [1]. However, the exact mode by action of which the peptide interacts with the bacterial membrane has yet to be established. Therefore, in this study, we use various techniques, including Quartz Crystal microbalance, Atomic Force Microscopy and Neutron reflectometry to study the interaction of the AMP with, supported lipid bilayers (SLBs). It was observed using these techniques that IK8 interacts more strongly with bacterial membrane mimics, with little or no interactions observed on the mammalian mimicking membrane. It was observed using AFM at a high concentration of IK8 on the bacterial membrane resulted in the removal of lipids from the surface creating lipid tubules on the surface. QCM-D results showed an increase in absorption on the bacterial SLB but with little mass change after the absorption on the bilayer. Therefore, we propose that IK8 disrupts the membrane of bacteria through the interaction of the cationic peptide inserting itself into the membrane and causing a reorganisation of the membrane creating secondary structures but not removing any lipids from the surface.

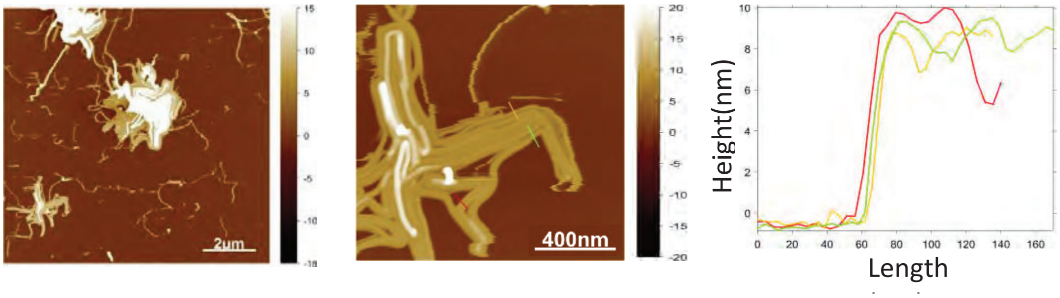

Figure 1: *AFM results showing the addition of 100uM IK8 to a supported bilayer POPC:POPG (3:1) creating secondary structures around 8-10nm in height.*

Additionally to understanding the mechanism of the AMP, in the study we aim to enhance the therapeutic potential of IK8 by incorporating it into nanobubbles encapsulated into liposomes for targeted delivery S.aureus biofilms [3]. The encapsulation of nanobubbles within drug-loaded liposomes provides the unique advantage of acoustic-triggered release using ultrasound, facilitating a more efficient delivery of the AMP to bacterial biofilms, and offering a novel approach for treating infections.

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Hybrid lipid-polymer materials for biotechnology applications

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Membrane- and vesicle-based soft materials have wide-ranging applications in biotechnologies, having already led to successes such as biomedical drug delivery formulations. However, there is still much scope for new innovations that solve challenges related to their stability and durability that are currently limiting their potential. We have been developing hybrid vesicle materials by blending natural phospholipids with synthetic block copolymers, which combine biofunctionality with enhanced mechanical robustness (Figure 1) [1,2]. These hybrid vesicles have been shown to enhance the functional lifetime of integral membrane proteins [3] as well as enabling broad tunability of drug release rates for long-acting parentheral therapies.

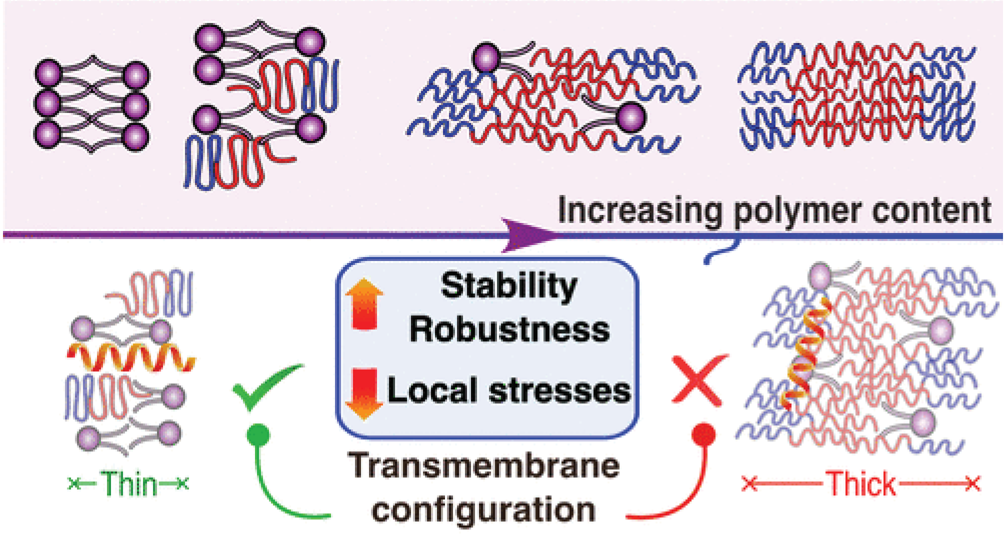

Figure 1: *Composition-dependent tunability of hybrid membrane properties formed from a blend of phospholipids and amphiphilic diblock copolymers [2].*

Here, I will present some of our recent work on two aspects of technology develop for hybrid vesicles. Firstly, we have screened a broad range of polymers to gain an improved understanding of their structure-property relationships in vesicles for membrane protein reconsitution. This presents a nuanced picture where the most important properties to be optimised such as reconstitution efficiency, protein activity or orientation within the membrane will dictate the optimal choice of polymer. Secondly, we have developed methods for rapid microfluidic formulation of drug-loaded hybrid vesicles. We find that solvent-solvent and solvent-amphiphile interactions play a critical role in successful formulation of composite particles with a narrow size range. Our new data provide both new insights but also new questions that are of interest to the soft matter community and will stimulate discussion.

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Polymers and Biopolymers IV

Wednesday 21 May 12:15 – 12:55

Study of the Piezoelectric Properties of PVDF/PLA Blends

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Piezoelectricity is a valuable property that enables a material to convert mechanical energy into electrical energy, and vice versa. As a result, piezoelectric materials are employed in a wide range of applications, such as sensors, actuators, battery-free systems, and even in cell regeneration. Generally, piezoelectric materials are ceramics, but these materials exhibit several drawbacks, including brittleness, high cost and, in some cases, toxicity. Polymeric materials, on the other hand, are of interest because they are flexible, cost effective and potentially biocompatible, despite having much lower piezoelectric properties.

Among polymers, PVDF (Polyvinylidene fluoride) is the most piezoelectric and can crystallize into several crystal phases, with the most common being α, β, and γ. The β-phase is the most desirable, as it has the highest piezoelectric coefficient, while the α-phase, although exhibiting no piezoelectric properties, is the easiest to obtain through traditional processing methods.

Several approaches have been developed to increase the β-phase content and piezoelectricity in PVDF. One of the most common methods is stretching an α-phase film. Recently, researchers have reported that incorporating PLA (Polylactic Acid) into a PVDF matrix can enhance the β-phase content, particularly after stretching the blend [1, 2]. This improvement is attributed to polar interactions occurring at the PLA-PVDF interfaces in the immiscible blend but, a detailed investigation on the PLA effect is still lacking.

Our research aims to rationalize the effect of incorporating PLA into PVDF on the β-phase content and the piezoelectric properties of PVDF. In particular, we studied the effect of PLA content and blend morphology as well as different processing conditions allowing us to obtain polymeric films or membranes. Several characterization techniques (i.e. differential scanning calorimetry, wide-angle x-ray scattering, infrared spectroscopy) have been employed to assess the amount of β-phase content in the samples whereas their piezoelectric properties were evaluated by measuring the piezoelectric coefficient d_{33}.

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Environment-Sensitive Thin Polymeric Membranes

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Using plasma-assisted polymerization technique, we demonstrate the fabrication of freestanding membranes made of poly-1,3-diaminopropane (pDAP), with thicknesses below 100 nm. The membranes are highly elastic as indicated by the Young modulus reaching 8 GPa, and exhibit significant durability at temperatures from 20 to 80 °C. In particular, we show that the 50 nm-thick membranes can be suspended over a millimeter-sized holes and even endure the temperature of 420 °C for 10 minutes.

The layers exhibit high sensitivity to environmental conditions related to humidity level. Therefore, they reversibly change their volume due to variations in air moisture, temperature, or when they are illuminated with light.

Our study shows that by covering silicon membranes with pDAP layers, we can develop cantilevers reacting to laser irradiation via photothermal heating. The bending of the cantilevers is primarily based on reversible water sorption mechanism. The relaxation times of the actuators are below 10 ms. After a million actuation cycles, the performance of the cantilevers remains unchanged – the deflection amplitude and relaxation times do not deteriorate.

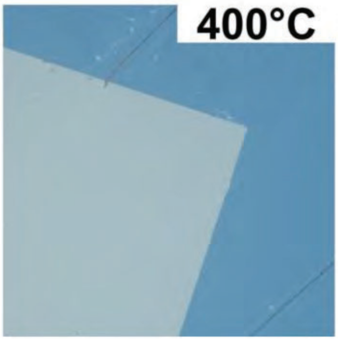

Figure : *Optical microscope image of the freestanding ≈100 nm thick pDAP membrane transferred onto a 2 mm hollow window in Si substrate at 400 ˚C.*

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Acknowledgements

A. K., M. P., and B. G. acknowledge the National Science Centre of Poland (NCN) for the OPUS grant UMO-2021/41/B/ST5/03038. P.P and P.M. acknowledge financial support from the National Science Centre of Poland (NCN) program Sonata Bis, grant number UMO-2020/38/E/ST5/00328.

Polymers and Biopolymers V

Wednesday 21 May 15:00 – 16:00

Breakdown of polymer self-similarity induced by polar activity in diluted and melt conditions

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Polar polymer activity is a fundamental mechanism behind a large number of cellular dynamical processes. The number and location of the active sites on the polymer backbone play a central role in their dynamics and conformational properties [1], as well as the presence of additional interparticle interactions [2]. Globular conformations for high motor densities change to stretched ones for the more realistic moderate or low density of motors. A small difference in the position of the first motor, or the motor distribution, can also dramatically modify the polymer typical conformations. Furthermore, polar activity induces a progressive local deformation of the polymer chains, making a clear distinction between head and tail [3]. For conformations that get more compact than equilibrium, the tail is more compact that the tail, while for stretched configurations the head is more extended. This effect persists in melt conditions, which coexists with a local induced alignment between chain segments [4].

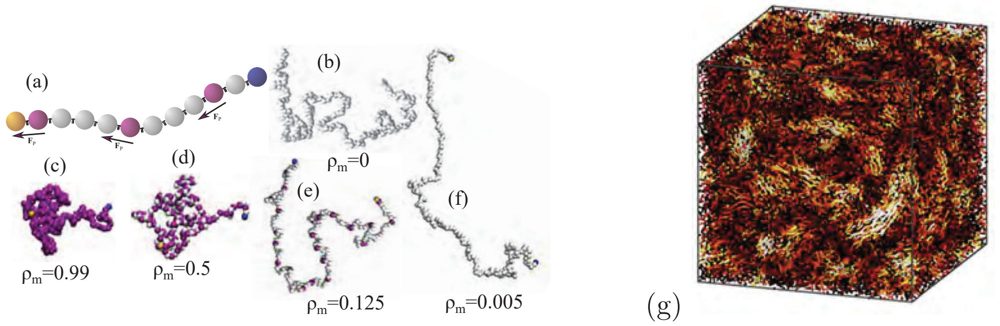

Figure 1: *(a) Polar polymer sketch with head bead in yellow, purple motor beads (arrows for the force direction), blue tail, and white linker beads. Snapshot for the passive polymer is depicted for reference in (b), while snapshots from (c) to (f) illustrate globule to stretch conformations with decreasing motor densities ρ_m. (g) Simulated melt snapshot with color coded bond alignments.*

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Symmetry of loop extrusion by dimeric SMC complexes is DNA-tension-dependent

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Structural maintenance of chromosome (SMC) complexes are involved in genome organization and regulation via DNA loop extrusion. During extrusion SMC proteins reel DNA from one or both sides and a loop forms and increases. At low DNA tension (< 0.1pN), Smc5/6 and Wadjet extrude DNA from both sides of the loop. At higher tension, however, they transition to a behavior akin to one-sided extruders, yet still capable of extruding from one or the other side thereby switching the direction of extrusion [1].

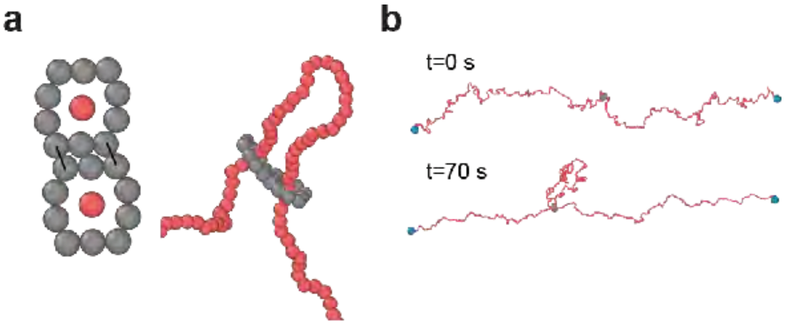

Figure 1: *The schematic in (a) illustrates the handcuff model used to simulate the loop extrusion experiment. The simulation begins with the handcuff already bound to the polymer, and (b) shows the system's state after the loop has been fully extruded.*

We combine single molecule experiments as well as simulations to investigate the symmetry of loop extrusion and show that it is driven by tension. In order to model this process in simulations, we propose a coarse-grained model for DNA loop extrusion using a Kratky-Porod chain as a basis for DNA [2] and a handcuff for SMC proteins. By matching stalling forces, we are able to simulate loop extrusion on experimental time and length scales. We find that the observed switching from two- to one-sided behavior does not require a change in motor activity, but can be explained as an interplay of extrusion, stalling and thermal fluctuations. Our findings challenge the previous view of loop extrusion symmetry as a fixed characteristic, and reveal its dynamic nature and regulation by both intrinsic protein properties and extrinsic factors.

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How loop extrusion affects the flow properties of dense DNA

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Loop extrusion is one of the main processes shaping chromosome organisation across the cell cycle, yet its role in regulating DNA entanglement and nucleoplasm viscoelasticity remains overlooked. We simulate entangled solutions of DNA under the action of generic Loop Extruding Factors (LEF). We find that extrusion (when exclusively intrachain) drives the formation of bottle-brush-like structures and significantly lowers the entanglement and effective viscosity of the system. In marked contrast with unextruded linear DNA, the viscosity of extruded systems scales linearly with DNA length, yielding up to 1000-fold effective fluidification. Surprisingly, considering bridging (or interchain loop extrusion) between LEFs yields a marked slowing down, eventually gellifying the solution. In this talk I will also report on in vitro microrheology experiments that we performed using dense solutions of Lambda-DNA and yeast condensin, which agree with our simulations that include bridging interactions. Our results may help to understand how LEFs contribute to actively modulate genome entanglement and viscoelasticity in vivo.

Surfaces, Interfaces and Emulsions II

Wednesday 21 May 15:00 – 16:00

Peptide-guided Self-Assembly: Fabrication of Tailored Spiral-like Nanostructures for Precise Inorganic Templating

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Fluid interfaces provide a powerful platform for the direct and spontaneous self-assembly of colloidal systems including amphiphilic peptides, where the driving force is the reduction in interfacial energy. [1, 2] Interestingly, self-assembling peptides (SAPs) are versatile building blocks in the creation of hierarchical nanostructures, offering precise control over obtained morphology and their dimensions. [3] While mainly driven by non-covalent interactions, the design of SAPs poses a challenge due to the complex molecular interactions involved. Nonetheless, SAPs have found significant applications in research fields such as nanomedicine, biosensing, and nanofabrication. Using fluid interfaces as a template, we introduce here an original methodology for the construction of SAPs with precise control over their structural organization. [4] Fluid interfaces also allow fine-tuning of the ensemble of peptides by an external force, such as the presence of an imposed interfacial flow. As a consequence, a wide-ranging set of interfacial, functional structures of different morphology can be achieved. Notably, we report the fabrication of SAP-based spiral structures, inspired by natural architectures and derived from the SARS-CoV-2 fusion core, showcasing the potential for biomedical applications. [5] The assembly process, critically dependent on peptide sequence design, is elucidated through a comprehensive suite of characterization techniques including atomic force microscopy, neutron reflectometry, interfacial rheology, and Infrared Spectroscopy. Furthermore, we demonstrate the utility of these spiral structures as templates for metallic replicas, paving the way for peptide-guided fabrication of tailored nanostructured surfaces with sub-10 nm dimensions over cm^2 areas. [4]

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Anomalous particle transfer between immiscible liquids in concentrated suspensions

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Mixing a concentrated suspension with an immiscible liquid is integral to using formulated products, *e.g.* chocolate mixing with saliva, and processing them, such as diluting oil-dispersion agrochemicals. Interfacial adsorption of particles is usually only considered at dilute concentrations. Here particles wetted by both liquids adsorb with a stress-scale set by surface tension over particle size, 10^3 Pa, that seemingly restricts transfer between liquids.

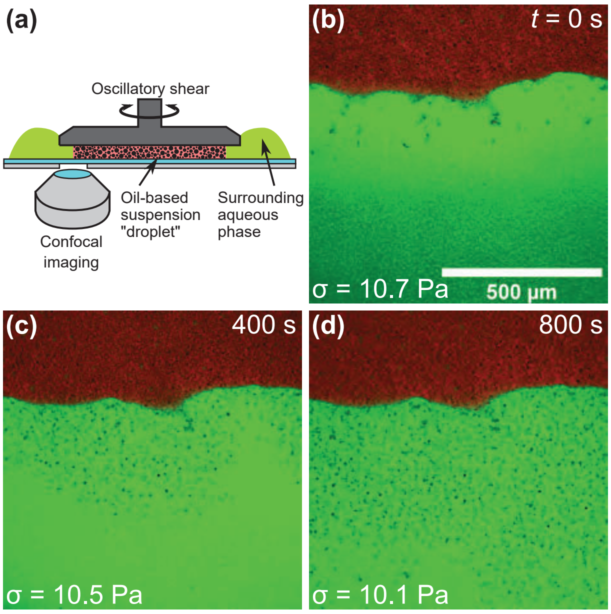

Figure 1: *Rheo-confocal imaging of the interface between oil-based suspension (red) and aqueous phase (green) under oscillatory shear, with particles (black). (a) Schematic diagram of experimental set-up (b)–(d) Movement of particles from a 40% solid volume fraction suspension over time from stroboscopic imaging.*

We show using rheo-confocal microscopy, Fig. 1, on a glass-in-oil suspension mixed with an aqueous phase that particles transfer at stress-scales three orders of magnitude below that predicted from the single particle picture. These microscopic measurements are supported by a quantitative model for macroscopic particle transport and the observed drop in viscosity. This necessitates a multi-particle picture for interfaces with concentrated suspensions that will control the structures formed in mixing from energy materials to eating chocolate.

Using Cruciferin Microgels for Microcapsule Formation

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Rapeseed is mainly used for oil extraction, with the remaining material often discarded or used as animal feed. However, it contains 30% to 45% high-quality protein, primarily com-posed of cruciferin and napin. While napin was found to be heat-stable, cruciferin is more sensitive. Remarkably, it forms stable suspensions of well-defined microgels (MG) when a suspension at 0.8 wt% is heated at 80°C for only 5 min. The diameter of the microgels varied between 0.1 and 0.4 µm depending on the pH. In the presence of 0.1 M NaCl such microgels could also be formed by a rapeseed protein isolate that contains both proteins in equal amounts. These cruciferin MG have potential applications in beverages and food products.

Additionally, the MG effectively stabilize water in water emulsions by adsorbing at the drop-let surface thereby preventing coalescence. Interestingly, the MG spontaneously slowly form permanent crosslinks within the layer leading a microcapsule (MC) that resists dilution. This crosslinking can be accelerated by heating the emulsion at 80°C for only 5 minutes. The size of the MC can be controlled by adjusting the volume fraction of the dispersed polymer phase or the concentration of the microgels (see fig). The MC can be washed to remove polymers and excess protein. They were found to be stable between pH 3 and pH 10 and after addition of up to least 0.1 M NaCl. They also resist mechanical stress such as vortex mixing, making them suitable for use as texturizers or carriers for encapsulation.

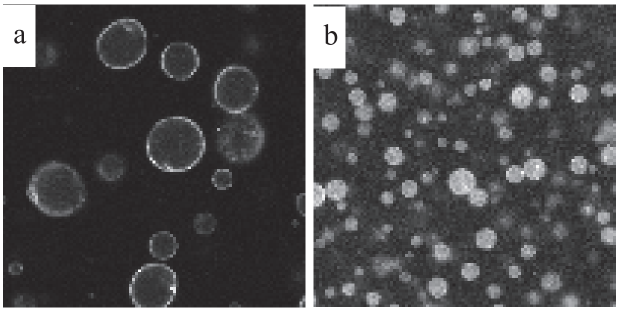

Fig.1 : CLSM images (40 × 40 µm) of a PEO (10 vol%) in dextran emulsion containing (a) 3 g/L, (b) 14g/L cruciferin MG. The protein microgels were fluorescently labelled.

Glasses, Colloids and Self-Assembly V

Wednesday 21 May 16:30 – 18:10

Vertical drying as a platform to study the role of polydispersity in 2D and 3D crystallization

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It is well-established that polydispersity is a deciding factor in the crystallization of colloids in both 2D and 3D. However, systematically varying polydispersity in a consistent, experimental system is challenging. Here, we systematically vary the polydispersity of Stöber silica dispersions, and we use vertical drying and bulk centrifugation to probe the effect of polydispersity on crystallization. The use of vertical drying is particularly intriguing, as it has been shown that it involves both 2D and 3D crystallization, and our quantitative bond-order parameter results seem to corroborate this. Our results may also have a bearing on growing colloidal crystals from particles synthesized through more sustainable methods, which often have higher polydispersities.

Quantitative 3D Real-Space analysis of Photonic Supraparticles

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Colloidal self-assembly into ordered structures is being extensively investigated through both experiments and simulations [1]. These supraparticles exhibit a variety of unique properties, stemming from the collective structure of the individual colloidal particles. One such example is the angle-independent photonic properties of icosahedral supraparticles, arising from their highly symmetric structure [1]. In the literature, icosahedral supraparticles are often characterized using scanning electron microscopy (SEM) [2], but this method has limitations. SEM can only examine the outer structure, which is the last layer to crystallize, making it unsuitable for determining the number and nature of crystalline layers present. Instead, we analyze these supraparticles using 3D stimulated emission depletion (STED) and confocal laser scanning microscopy; techniques that are already widely used in the field of colloid science. These methods enable real-space analysis, allowing differentiation between icosahedral clusters and onion-shell structures, where the interior is crystalline while the exterior remains disordered. Such structures are often misidentified using conventional SEM analysis. Besides distinguishing between these structures. STED and confocal microscopy also reveal features like an off-center symmetry point or local defects, as shown in Figures 1C and 1D respectively. This opens up the possibility of comparing experimental and simulated photonic properties for structures with defects, disordered domains, and onion-shell configurations for statistically relevant numbers of supraparticles.

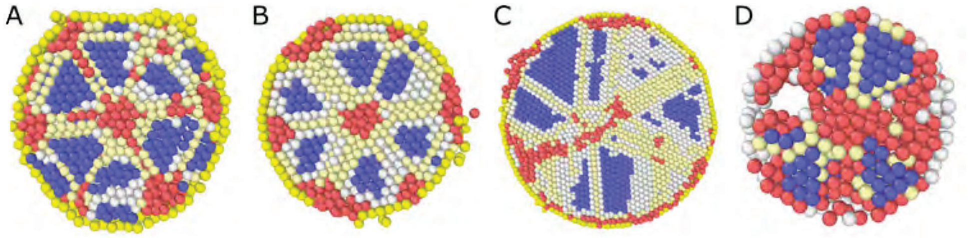

Figure 1: *Obtained particle coordinates, color coded by bond-order parameter machine learned clustering.* ***A.*** *Anti-Mackay icosahedral cluster.* ***B.*** *Onion shell icosahedral cluster.* ***C.*** *Off-center icosahedral cluster.* ***D.*** *Icosahedral cluster with a defect the size of several particles.*

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Dynamics and phase behavior of quasi-2D dispersions

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Dispersions of globular proteins or colloids with competing short-range attractive (SA) and long-range repulsive (LR) interactions have a rich phase behavior with different cluster phases. While three-dimensional SALR dispersions have been intensely studied also regarding their cluster dynamics [1], less is known about quasi-two-dimensional (Q2D) dispersions where the particles form a planar monolayer inside a bulk fluid.

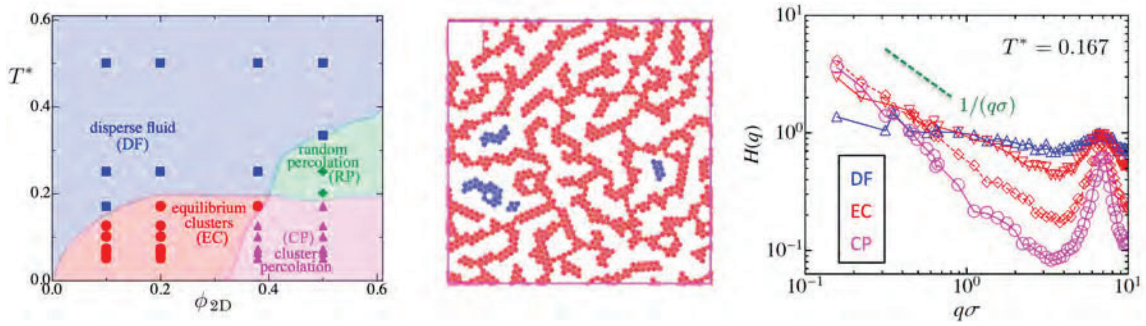

Figure 1: *Left: Phase diagram of Q2D - SALR dispersion. Middle: Simulation snapshot of a cluster-percolated Q2D system. Right: Hydrodynamic function $H(q)$ of indicated Q2D phases.*

Using mesoscale simulations without and with hydrodynamic interactions (HI) considered, we have determined the phase behavior and clustering properties of a generic Q2D model where the Brownian particles interact directly by short-range generalized Lenard-Jones and long-range repulsive screened Coulomb forces [2]. The Q2D phases resemble those in three dimensions but are different in their microstructure showing different levels of hexagonal ordering. With decreasing temperature, the cluster morphology crosses over from disk-like shapes in an equilibrium cluster (EC) phase to double-stranded anisotropic hexagonal clusters in a cluster-percolated (CP) gel phase. Indicators have been identified signaling the transition from a high-temperature dispersed fluid (DF) phase to the lower-temperature equilibrium cluster phase. Furthermore, we have uncovered the dynamics of Q2D – SALR systems including self-diffusion with its non-Gaussian statistics [3]. The interplay of Q2D motion and HI causes anomalously enhanced collective diffusion at small wavenumbers q related to out-of-plane fluid backflow. The spatio-temporal buildup of inter-proteins HI by vorticity diffusion and sound is revealed from time-resolved hydrodynamic and current (velocity) autocorrelation functions [3].

References

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Dielectric and Absorptive Doping of Photonic Icosahedral Supraparticles

Ruizhi Yang[1], Jesse Bückmann[1], Artyom Chirko[1], L.D. (Roy) Hoitink[1], Alptug Ulugöl[1], Laura Filion[1], Alfons van Blaaderen[1]
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Supraparticles are particles build up from smaller particles [1]. Introducing ”doping” particles into host supraparticles (SPs) without disrupting their structure is an important stop to broaden their applications[2-4]. Here, we synthesized (silica)core-(latex)shell-(silica)shell and (silica)core-(latex)shell-(polyelectrolyte)shell particles that can be doped into SPs without altering their self-assembly pathway because they were designed to have the same size and interactions with the host particles. The dopants can either be transformed into carbon black under high-temperature nitrogen treatment or be removed under oxygen, yielding air spheres. Incorporating these core- shell particles into silica spheres of comparable size via slow evaporation of aqueous dispersions enables the formation of icosahedral SPs. Doping with random latex particles introduces either vacancies or absorbers, enhancing structural color saturation by reducing incoherent scattering, or in the case of air spheres enabling tunable photonic band gaps and porosity. This versatile strategy establishes a general approach for functionalizing SPs, expanding their potential in photonics, biology, and catalysis.

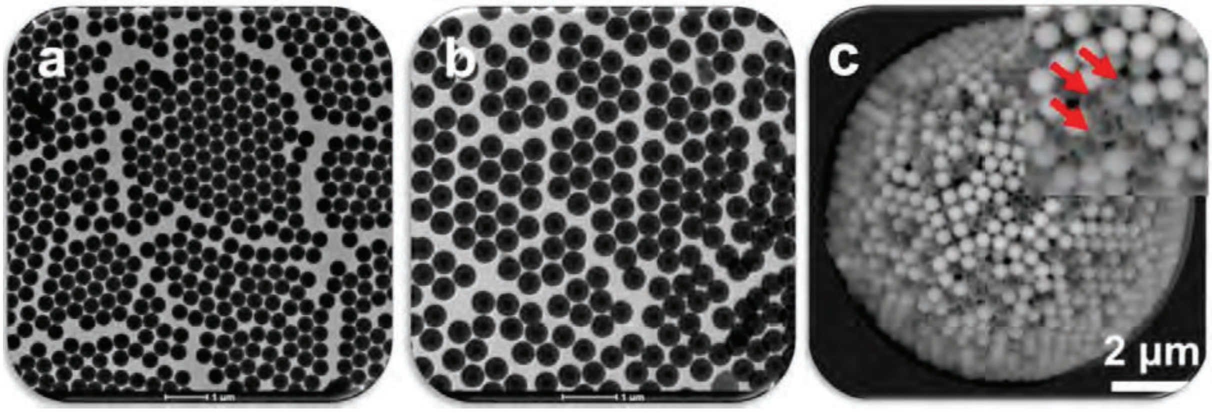

Figure 1: *Electron microscopy images showing (a) pure silica particles (TEM), (b) silica@latex core-shell particles (TEM), and (c) an icosahedral supraparticle (SEM), with inset highlighting dopant positions (red arrows).*

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A ferroelectric phase transition underlying the liquid-liquid phase transition in supercooled water

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A liquid-liquid phase transition (LLPT) between a High-Density Liquid (HDL) and a Low-Density Liquid (LDL) below water's freezing point, in the so-called supercooled regime, has emerged as a compelling hypothesis for explaining its equilibrium anomalies. MD simulations nowadays provided evidence for the LLPT, supported but not directly observed in experiments. On the other hand, a first proposal about existence of a ferroelectric phase transition in supercooled water can be dated back to 1977 by Stillinger. The two phase transitions, however, were never put in connection.

In this study [1], we show how the LLPT and ferroelectric phase transition in supercooled water can be designed as two facets of the same underlying phenomenon. Reanalyzing MD simulations of Ref. [2], we first identified a distinct correlation between density (ρ) and total polarization (P): HDL shows paraelectric and LDL ferroelectric characteristics. The subsequent analysis of pivotl indicators of liquid-liquid and ferroelectric phase transitions across the Widom line, the first order LLPT line and near the critical point (CP), showed that P and ρ are not interchangeable order parameters for the LLPT. MD results further confirmed ferroelectric order in LDL by observing collective modes resulting from spontaneous breaking of rotational symmetry. Finally, the development of a classical density functional theory in mean-field approximation, valid for polar liquids, allowed to underpin the potential role of ferroelectricity in promoting the LLPT, supporting P as the order parameter for the LLPT. The theory links annealed positional disorder to ferroelectricity. This work triggers experimental studies.

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Surfaces, Interfaces and Emulsions III

Wednesday 21 May 16:30 – 17:50

Stabilization of water-in-water emulsions by complex coacervate core micelles

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Water-in-water emulsions (WWE) form when aqueous solutions of incompatible polymers are mixed. These emulsions, that do not contain any organic phase, are of growing interest in soft matter due to their phase behavior and applications in encapsulation, separation and texturation. However, they possess a large interface and low interfacial tension, making the adsorption of molecular stabilizers, such as surfactants, difficult. Moreover, these all-aqueous systems require fully hydrophilic stabilizers displaying affinity for both phases (refered as bis-hydrophilicity). Therefore, using particles such as protein microgels or protein-polysaccharide complexes as stabilizers, namely Pickering effect, has proved effective in improving WWE stability. [1,2] However, the relationship between chemical composition of the particles and their ability to stabilize WWE is poorly understood. A recent work showed that poly(ethylene oxide) (PEO)/dextran WWE in the presence of dextran-derived microgels displaying bis-hydrophilicity had an excellent stability. [3]

In this context, complex coacervate core micelles (C3Ms), consisting of polyelectrolyte complexes stabilized by a neutral hydrophilic polymer segment attached to one or both polyelectrolytes, appear as promising candidates. Indeed, their size and surface chemistry (thus, bis-hydrophilicity) can be tuned by varying the copolymer composition. [4]

In this study, PEO/dextran emulsions in the presence of C3Ms with a PEO corona were developed. The role of the core in C3Ms partitioning will be highlighted. The ability of C3Ms to stabilize WWE varies with the length of the polymers and ionic strength.

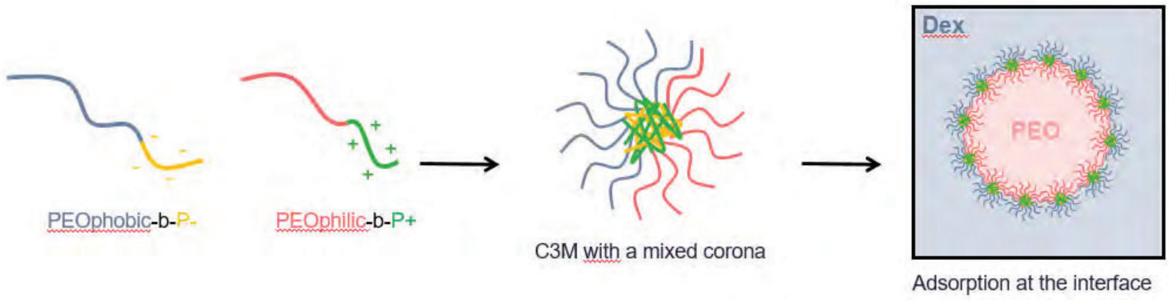

Figure 1: *Scheme of C3Ms stabilizing PEO/dextran emulsion.*

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Microgels wrapping at lipid-bilayer membranes

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The transport of soft deformable particles across lipid membranes by cellular engulfment plays a key role in biomedical applications, such as drug delivery [1]. The physico-chemical parameters of the particles that control engulfment are their size, shape, and deformability [2, 3]. Microgels are particularly versatile because their elasticity can be tuned in a wide range by changing the density of crosslinkers. Using a elastic network for the microgel and a continuum model for the membrane, we study microgel wrapping at lipid-bilayers, see Fig. 1.

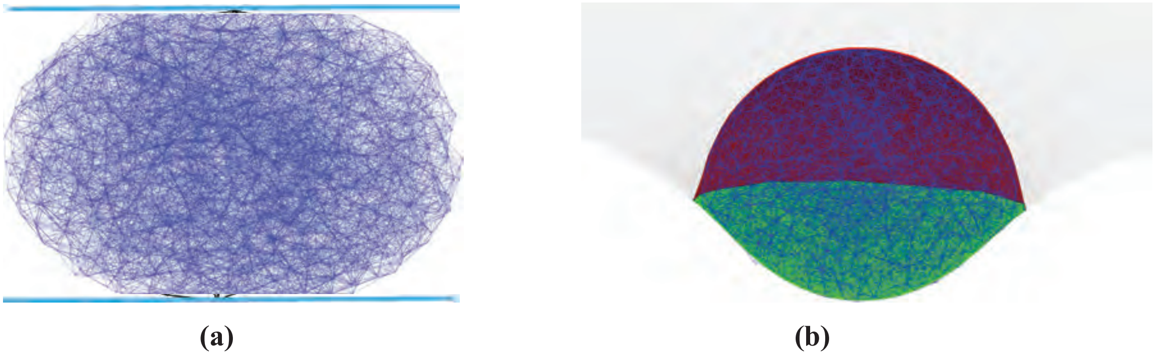

Figure 1: *An initially spherical microgel (a) in-between two parallel walls and (b) attached to a membrane.*

We use the Hertz theory to characterize the microgel's Young's modulus and Poisson's ratio. With the help of triangulated membranes and energy minimization, we determine the interplay of microgel and membrane deformation. Our calculations predict stable wrapping states for microgels with various Young's moduli at membranes with various tensions. There is a transition from oblate microgel states at low wrapping fractions to cup-like shapes at high wrapping fractions. A higher microgel deformability increases the stability of partial-wrapped states. Our results on this tunable and responsive system will allow the design of the microgels with optimal elastic properties for suitably tailoring novel biomaterials.

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Microgel stabilised aqueous foams

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Poly(N-isopropylacrylamide) microgel particles hold tremendous promise as thermo-sensitive surface-active agents for applications in catalysis, wetting behavior, emulsions, and particularly foam stability. Although emulsions stabilized by these microgels have been extensively studied, their role in foam stabilization—crucial for processes such as froth flotation—remains less explored. The inherent stiffness of microgels is assumed to significantly impact their arrangement and packing density at the air/water interface, which in turn modifies the resulting foam structure. In these studies, we aim at developing a quantitative understanding on how the crosslinker density of the microgels alters the stabilization of the macroscopic foams.

In this investigation, we systematically modulate the microgels' mechanical properties by adjusting crosslinker density and surface charge. We then quantify thickness of foam lamellas with small-angle neutron scattering (SANS) with fit model described in [1,2] combined with optical imaging, and nano-mechanical atomic force microscopy to correlate these properties with foam stability. Our results indicate that softer microgels enhance foam stability. Intriguingly, all formulations yield ultra-thin foam films approximately 32–38 nm thick, despite the swollen microgels being around 600 nm in size. This discrepancy raises a key question: do microgels flatten under confinement or assemble into discrete clusters within the Plateau borders? Answering this question is central to understanding particle-stabilized foams. To address this, simultaneous SAXS–SANS along with neutron reflectometry are employed to precisely locate the microgels within foam lamellas and understand how microgel deformation and spatial distribution govern foam stability. These investigations extend beyond fundamental science, having significant implications for industries such as froth flotation, emulsions, and sustainable material design.

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Doing the dishes with neutrons: micro-flow SANS of fat removal

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Understanding the mechanisms and kinetics of the removal of fat components such as triacyl glycerides (TAGs) from surfaces is of importance for the design of novel surfactant-based formulations for enhance performance optimized for lower temperatures and less water usage. Pairing of the physico-chemical properties of surfactant and fat components plays a role in dictating the macro-scale removal/cleaning mechanism[1-3] with simulations showing different nano-to-molecular scale pathways for these mechanisms.[4] There is therefore a need to experimentally examine these nanoscale pathways, the kinetics of structures formed, their transition towards thermodynamic equilibrium, and hence, the fat removal mechanism for the bottom-up design of formulations.

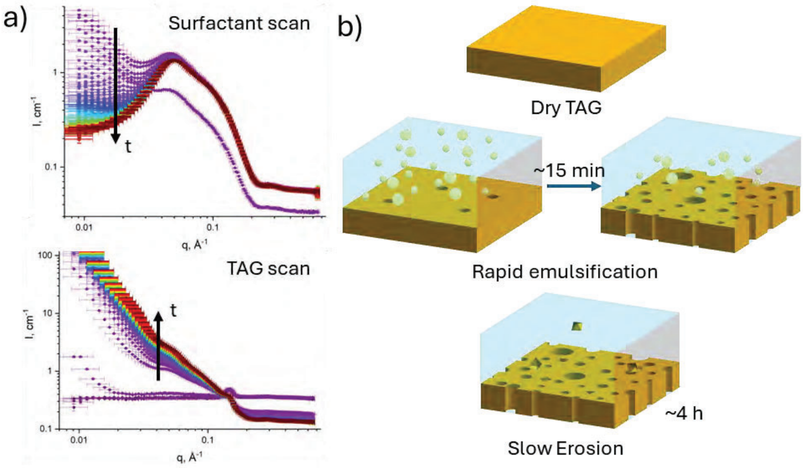

Figure 1: *a) Time evolution of SANS scattering profiles taken at the position of the 'wash/residues' (top) and of the deposited TAG (bottom) inside a micro-flow cell. b) Proposed TAG cleaning mechanism and timescales from SANS data. These are based on the kinetics of micelle shape/size and charge, emulsion size, interface fractal dimentions, and evolution of structure between the TAG and surfactant scan.*

Here, we present a dynamic SANS study with alternating measurement positions (Fig 1a) on the structures formed during the in-flow interaction of TAG (comprising glycerol units with 50% oleic, 30% palmitic and 20% stearic acid) deposited on a PMMA surface, and an aqueous 50 mM SDS-DDAO mixed surfactant solution ('TAG scan' in Fig 1a) and their evolution succeeding the cleaning ('Surfactant scan' in Fig 1a). Examining the sample transmission revealed a 2-step mechanism based on the rate of mass loss: an initial fast and short mechanism followed by a longer, and half as fast removal process. Examining the 'surfactant scan' profiles as comprising charged ellipsoids and larger spherical structures, reveals the presence of both ≈10 nm emulsions and loaded micelles, and the latter's evolution towards the structure and interactions of the neat micellar solution, pointing to an initial emulsification mechanism. Furthermore, combining a power law, gaussian-peak, and charged ellipsoid model for examining the 'TAG scan' profiles reveal a decrease in micelle size and interactions suggesting a depletion of surfactant concentration due to adsorption on the TAG surface. This is accompanied by changes in the TAG-solution interface (power law), the TAG crystalline peak (Gaussian-peak), and emergence of ≈8.5 nm holes in the structure, all pointing to an erosion mechanism. We combine these findings into an overall nanoscopic mechanism (Fig 1b) for the interaction of this model dish-washing surfactant solution with this model fat of the same TAG composition as beef fat.

Our work on this model non-equilibrium system gives us an understanding of nanoscale phenoma underpinning the interaction of surfactants and fats. This can be used to aid in the molecular design of formulations targeted at cruisial steps in this interaction such as optimisation towards more rapid mechanisms, e.g. emulsification and delamination,[1,5] increasing the loading of micelles, and their adsorption onto fat surfaces.

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Polymers and Biopolymers VI

Thursday 22 May 09:45 – 10:45

Polypeptide-Based Polymer Electrolytes for Lithium Batteries: A Structure-Property Investigation

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The increasing need for sustainable alternatives to petrochemical-based polymers has driven significant interest in bio-based polymers. These materials offer promising solutions to environmental concerns while enabling applications across various fields, from everyday consumer products to high-performance technologies. In this study, we investigate polypeptides with different architectures—homopolymers and block copolymers with polyethylene oxide—as polymer electrolytes for lithium battery applications. A comprehensive structure–property relationship analysis is conducted, focusing on ionic conductivity, thermal properties, and mechanical strength in polypeptide-lithium salt systems. Small-angle (SAXS) and wide-angle (WAXS) X-ray scattering techniques are employed to elucidate the material's structural organization, while ionic conductivity is assessed via electrochemical impedance spectroscopy. Thermal behavior is examined using differential scanning calorimetry (DSC), and rheological studies provide insights into mechanical performance. The findings of this work contribute to the development of bio-based polymer electrolytes with tailored properties for next-generation energy storage applications.

Acknowledgement: This work was supported by the Twinning project FORGREENSOFT (Number: 101078989 under HORIZON WIDERA-2021-ACCESS-03). SLB is thanked for permission to publish this work.

Knots in polymer molecules under Couette & Poiseuille flow

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Knots in polymers and biological macromolecules, such as DNA and proteins, are crucial to their structure and function. However, the effect of external forces on knots existing in polymers is still insufficiently understood. Here, we investigate the impact of shear and Poiseuille flow on knotted flexible polymers using Molecular Dynamics (MD) and Multi-Particle Collision Dynamics (MPCD) simulations [1].

We find that under simple shear (Couette flow), initially loose knots in polymer coils tighten beyond a critical shear rate. Further increase of the shear rate leads to tumbling motions of the chains in flow and fluctuating knot sizes. In contrast, knotted polymer globules subjected to shear unfold into pearl-necklace-like conformations, whereby knots spread across multiple sub-globules and undergo dynamic topological transitions.

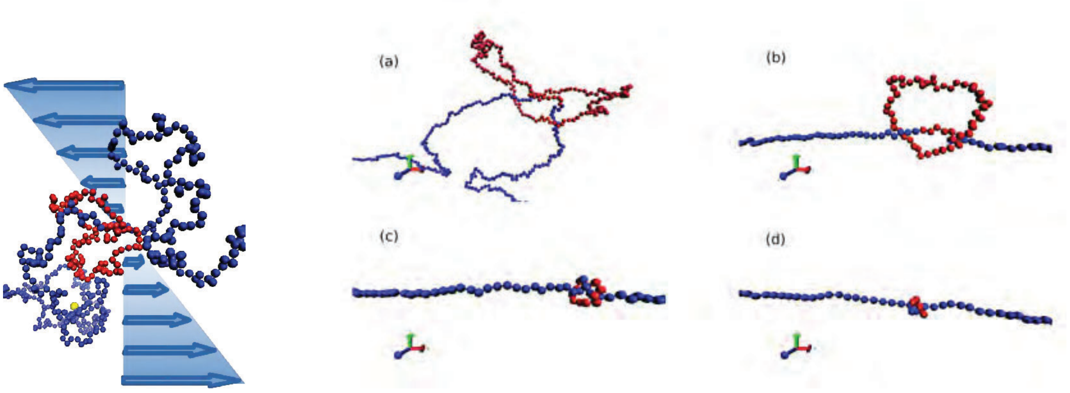

Figure 1: (left) A polymer chain experiencing Couette flow represented by the blue arrows, with a trefoil knot highlighted in red. (right) A typical time evolution of the knotted region of the polymer chain under Couette flow. The initial trefoil knot is tightened over time while the chain stretches.

In Poiseuille flow, knots also tighten under increasing flow strength, with fluctuations in knot size emerging at high shear rates. Unexpectedly, the channel width significantly influences the tightening process even at constant mean shear rate.

These findings reveal that both Couette and Poiseuille flow induce major structural and topological transformations in knotted polymers, offering insight into the behavior of knots under hydrodynamic forces in confined and driven environments.

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Pressure-induced phase transition in polymer systems

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The coil-to-globule transition of polymers with a lower critical solution temperature (LCST) is widely exploited in the design of responsive materials and surface functionalization strategies. While temperature is the primary control parameter for this transition, it can also be triggered by pressure variations within a specific temperature range. In contrast, the pressure and temperature response of polymers with an upper critical solution temperature (UCST) remains largely unexplored, despite the expectation that their phase behavior may exhibit stronger pressure dependence due to the large excess volumes associated with the transition.

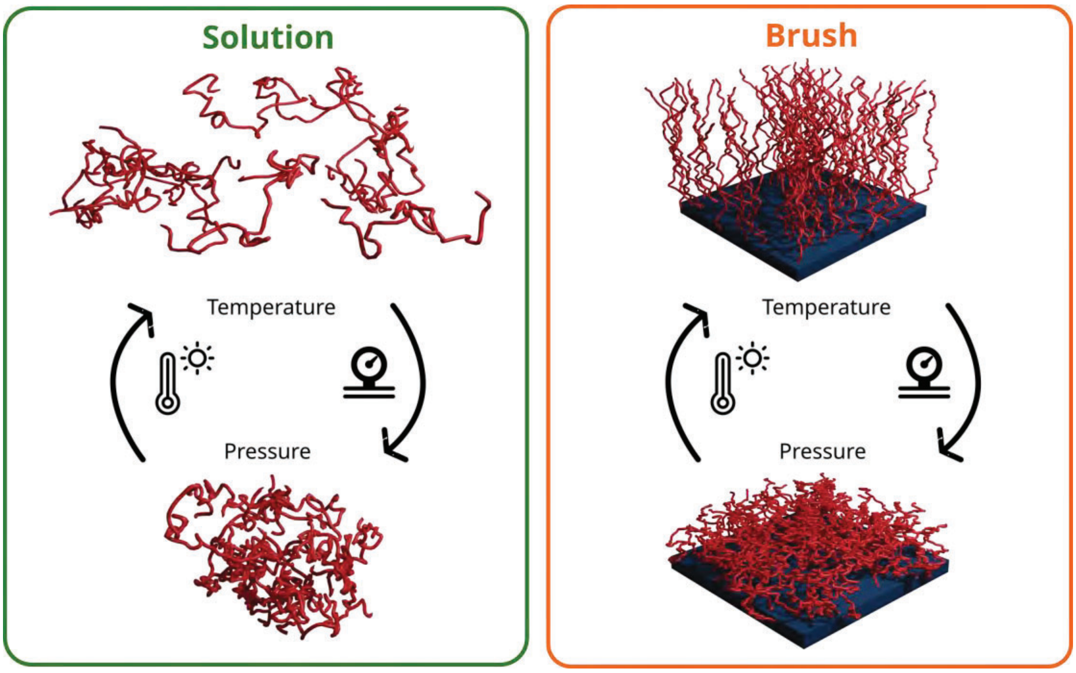

Figure 1: *Graphical representation illustrating the structural transition of polymer semidilute solution and polymer brushes covalently anchored to a solid surface as the solvent quality changes from good to poor due to variations in temperature or pressure of the system.*

In this contribution, we provide an overview of pressure effects on temperature-responsive LCST and UCST systems, spanning from semi-dilute solutions to polymer brushes. We discuss the experimental methods available to investigate these phenomena and highlight key open questions that need to be addressed to advance our understanding of pressure-induced phase transitions in polymer systems.

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Biological Soft Matter III

Thursday 22 May 09:45 – 10:45

A sTicky Situation: Phase Separation and Ageing of Disordered Protein from Tick Adhesive

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Hard ticks feed on their host for multiple days. To ensure firm attachment, they secrete a protein-rich saliva that eventually forms a solid bioadhesive, also known as the cement cone. The underlying mechanism of this liquid-to-solid transition is currently not understood. In this talk, I will focus on the phase transitions of a disordered glycine-rich protein (GRP) from a protein family that is abundant in the cement cone. I will show that GRP undergoes liquid-liquid phase separation via simple coacervation to form biomolecular condensates in salty environments. Cation-π and π-π interactions mediated by periodically placed arginine and aromatic amino acid residues are the primary driving forces that promote phase separation. Interestingly, GRP condensates exhibit ageing by undergoing liquid-to-gel transition and exhibit adhesive properties, measured through force spectroscopy. Finally, given the significant changes in pH taking place during the tick bite, I will show that pH significantly influences the GRP hydrophobicity via ionic residues, driving notable variations in the coacervation behavior (propensity, progression) and in shaping the material properties (viscosity, interfacial activity) of the formed condensates. These findings provide a starting point to gain insights into the adhesion of ticks, develop novel tick control strategies, and towards biomedical applications such as tissue sealants.

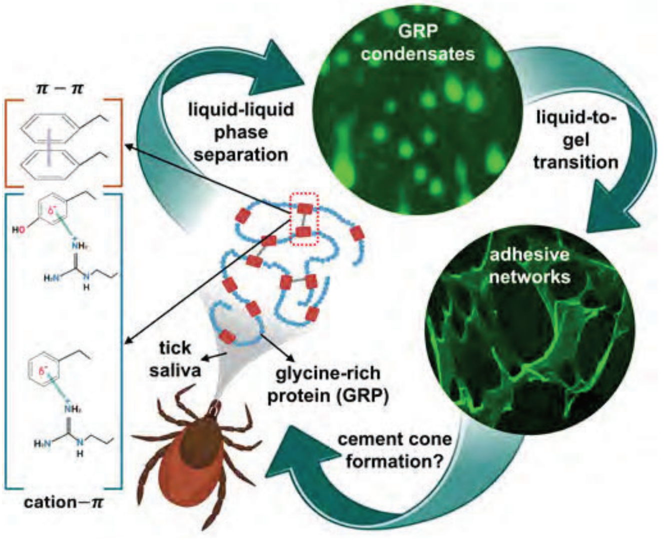

Figure 1: Schematic overview of GRP phase transitions and underlying driving forces, hinting at their role in tick cement cone formation.

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DOI: https://doi.org/10.1101/2025.01.09.632076

Advancing High-Speed AFM Analysis of Dynamic Systems

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Atomic Force Microscopy (AFM) is the only technique which can directly visualise individual molecules in biological membranes at high resolution and at video speed. Rapid imaging rates, resulting from the development of High-Speed AFM (HS-AFM) [1], provide the unique ability to record molecular and membrane dynamics. Alongside these improvements in instrumentation, novel analysis techniques must be developed to access and process information available from HS-AFM. This work firstly aims to enhance AFM resolution limits by optimising the technique of Localisation AFM (LAFM) [2], where small protrusions on biomolecules are localised by detecting thermal fluctuations in their height over multiple HS-AFM frames. The second objective is to provide a new automated Machine Learning (ML) method for quickly levelling movies of HS-AFM frames – a time-consuming step in data processing if each frame is levelled individually.

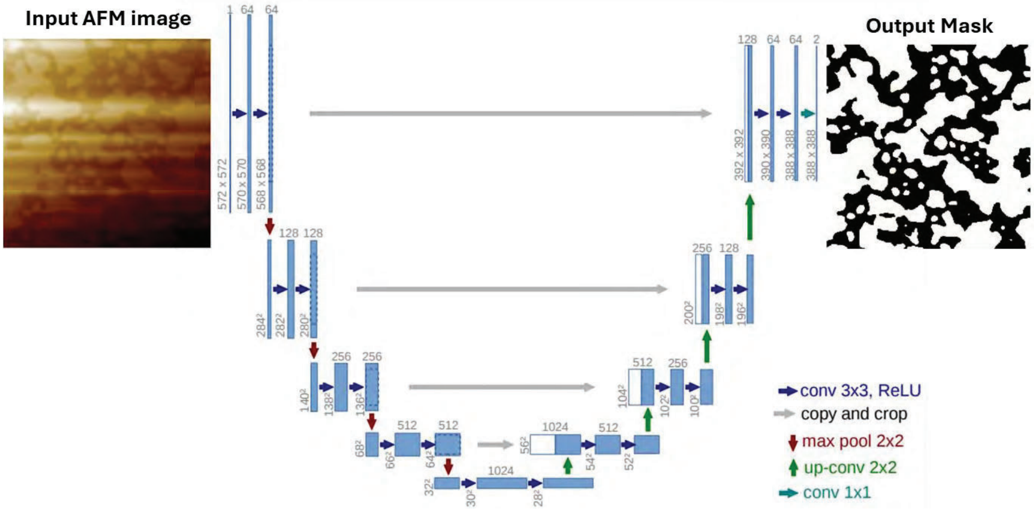

Figure 1: *Neural Network for AFM image levelling, using the U-net architecture developed for biomedical image segmentation [3]*

To this end, high throughput testing of optimal AFM image alignment parameters has been carried out, to improve the precision of protrusion detection in the LAFM workflow. Finally, a ML model has been trained to automatically detect features to mask before levelling in AFM images.

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Continuity of Short-Time Dynamics Crossing the Liquid–Liquid Phase Separation in Charge-Tuned Protein Solutions

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Liquid–liquid phase separation (LLPS) constitutes a crucial phenomenon in biological self-organization, not only intervening in the formation of membraneless organelles but also triggering pathological protein aggregation, which is a hallmark in neurodegenerative diseases. Employing incoherent quasi-elastic neutron spectroscopy (QENS), we examine the short-time self-diffusion of a model protein undergoing LLPS as a function of phase splitting and temperature to access information on the nanosecond hydrodynamic response to the cluster formation both within and outside the LLPS regime [1]. We investigate the samples as they dissociate into microdroplets of a dense protein phase dispersed in a dilute phase as well as the separated dense and dilute phases obtained from centrifugation. By interpreting the QENS results in terms of the local concentrations in the two phases determined by UV–vis spectroscopy, we hypothesize that the short-time transient protein cluster size distribution is conserved at the transition point while the local volume fractions separate (Figure 1). The presentation will provide insights into the colloid picture of protein solutions.

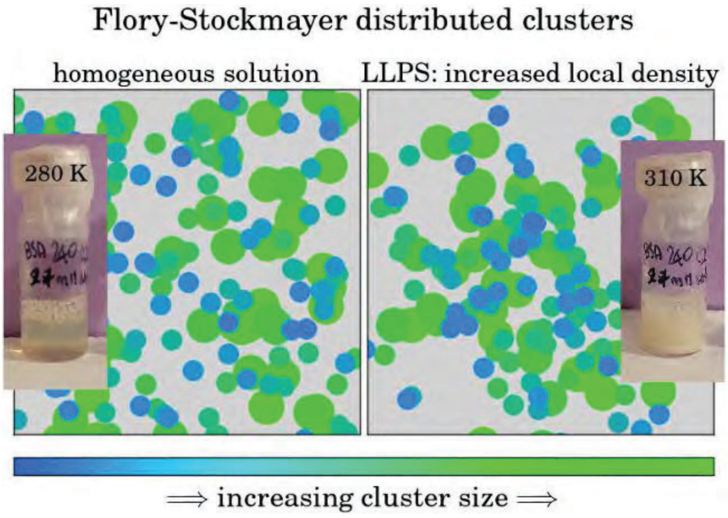

Figure 1: *The schematic illustrates the cluster distributions outside (left) and within the LLPS regime (right) with local dense and dilute regions described by the Flory–Stockmayer model [1]. The inset photographs display the associated protein solution in the visually transparent (left) and turbid (right) states, the transition being achieved by warming from* 280 *K to* 310 *K.*

References

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Polymers and Biopolymers VII

Thursday 22 May 11:15 – 12:35

Phase behavior and self-assembly properties of semiflexible polymers in solution

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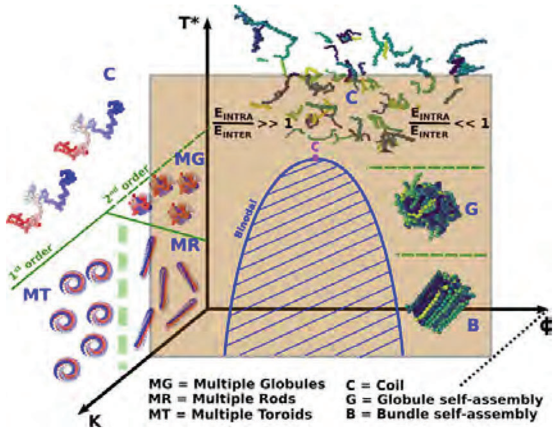
We explore the phase behaviour and self-assembly properties of semiflexible polymers in solution, focusing on temperature dependence and bending constraints [1]. We first examine the phase behaviour of a single semiflexible polymer, comparing two types of bending constraints. The first is the traditional elastic penalty used in the worm-like chain model, while the second is an entropic constraint arising from steric effects introduced by side spheres [2]. We will show that these constraints lead to markedly different phase behaviours at low temperatures. Further, we extend the analysis to multiple polymer chains in solution, investigating their self-assembly properties under each bending constraint. Although the detailed low-temperature behaviour differs between the two constraints, the general self-assembly mechanism appears to exhibit universal characteristics.



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Periodic cooking of eggs

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Egg cooks are challenged by the two phase structure of eggs. Indeed, albumen and yolk have two very different compositions and, therefore, they require two different temperatures for optimal cooking (85 °C for albumen and 65 °C for yolk) [1,2]. Separation or a compromise temperature to the detriment of food safety or taste preference are the options. This study shows that it is possible to cook albumen and yolk at two temperatures without separation by using periodic time-varying boundary conditions in the energy transport problem. Through mathematical modeling and subsequent simulation, we designed the novel cooking method, namely *periodic cooking*, and, through comparison with established cooking procedures – hard-boiling, soft-boiling and sous vide – with a plethora of characterization techniques, including Sensory Analysis, Texture Profile Analysis and FT-IR spectroscopy, we confirmed the different cooking extents and the different variations in protein denaturation with the novel approach. We also explored the impact on gel formation and egg texture from a rheological point of view with the aim of understating how different thermal histories can affect the sol-gel transition. In all cases, we found an optimum when egg yolk and albumen are cooked at their corresponding denaturation temperatures, as it happens in the newly designed periodic cooking.

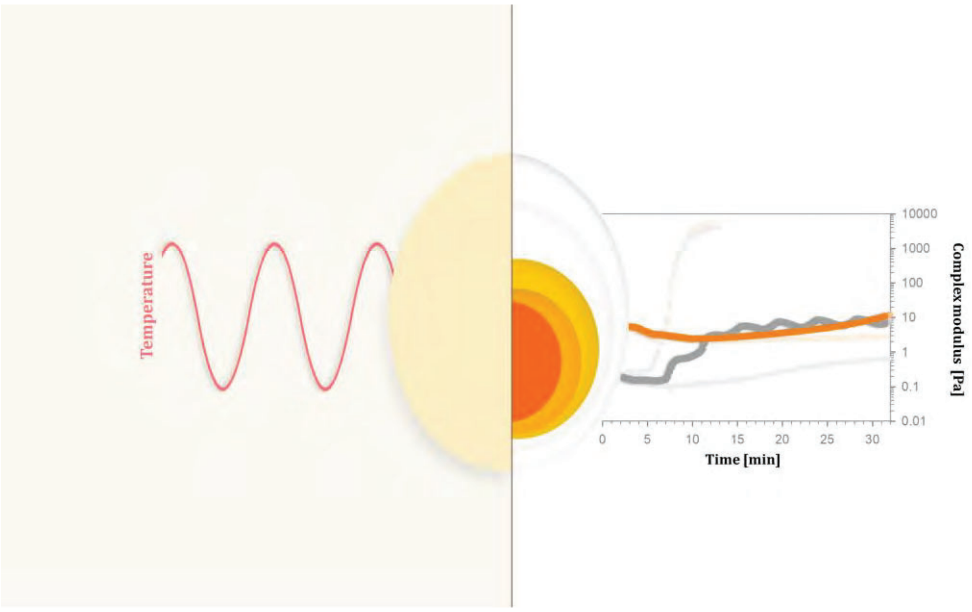

Figure 1: *Schematics of how the use of a periodic time-varying temperature in the cooking of an egg can affect texture development.*

Overall, the novel method optimizes egg texture and nutrients, but it also holds promise for innovative culinary applications and materials treatment: curing, crystallization and structuring of materials are only a few of the applications for which a thoroughly designed thermal treatment is at hand.

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Rheology of Block Copolymer Solutions in Organic Solvents

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Block copolymers (BCPs) are macromolecules with distinct block sequences, combining properties from different monomers[1]. Their self-assembly in solution is influenced by several variables, among them solvent selectivity and chains architecture, making them useful in various applications[2]. Phase diagrams help understand their stability, with key factors including the Flory-Huggins interaction parameter, block volume fraction, and molecular architecture[3]. In solution, BCPs undergo self-assembly into structured arrangements (microphase separation), due to the intrinsic incompatibility between their blocks. This process is highly temperature-dependent, typically shifting from a uniform isotropic state to microphase separation and, at lower temperatures, eventually leading to macrophase separation. This study examines two types of styrene-butadiene BCPs—one monodisperse and one polydisperse—interacting with two similar model systems composed of a mixture of solvents of different selectivity. Rheological analysis was used to track phase separations, complemented by optical techniques. The results indicate that polymer dispersity, concentration, and solvent properties significantly affect phase separation behavior.

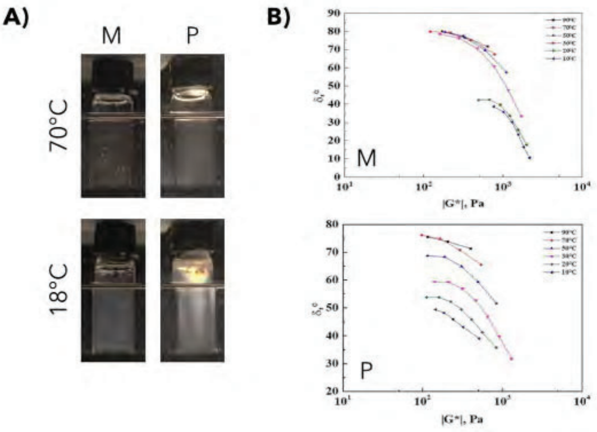

Figure 1: *Optical results (A) and Van-Gurp Palmen plots (B) of BCP solutions under investigation*

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From Monomer to Nanostructures by Polymerization-Induced Electrostatic Self-Assembly (PIESA): Revealing Structural and Morphological Features

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Reaction-assembly networks have recently received a great deal of attention to direct one-pot self-assembly of macromolecular building blocks. A more recently introduced pathway is the so-called polymerization-induced electrostatic self-assembly (PIESA) procedure in which a neutral-charged block copolymer is synthesized in the presence of an oppositely charged macromolecular template [1]. Following the polymerization, at a certain block length, the charged components undergo an electrostatically driven phase separation to a micellar core which is stabilized and restricted by the neutral block of the copolymers to the nanoscopic scales. In this work, we report a morphological study on the generation, evolution, and structural features of the complex coacervate nanostructures in a PIESA system. The polymerization kinetics and monomer conversion are followed by nuclear magnetic resonance. Furthermore, the morphology of the micelles is explored by small-angle X-ray scattering. Finally, the detailed structural features of the assemblies at different stages of the polymerization are investigated by least-square fitting of small-angle X-ray scattering data on the absolute scale, facilitated by NMR relaxometry and density measurements.

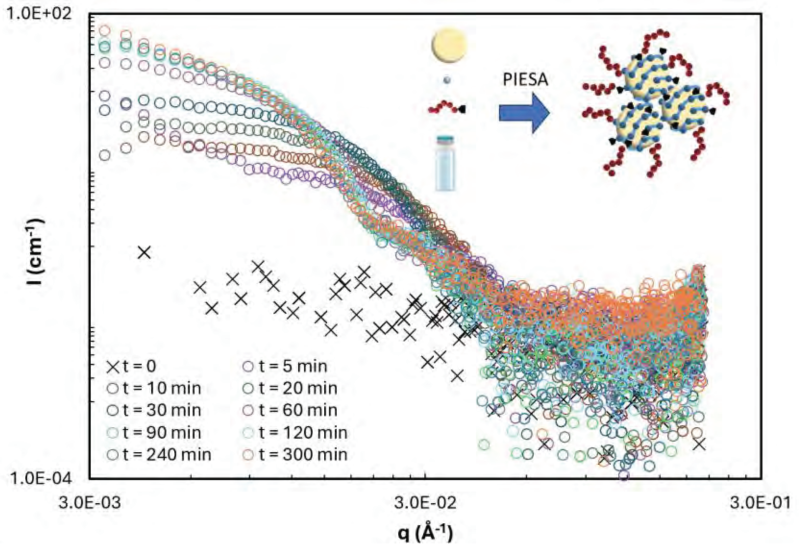

Figure 1: *Morphological evolution in a PIESA system by Time-resolved Small-Angle X-ray Scattering (SAXS)*

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Biological Soft Matter IV

Thursday 22 May 11:15 – 12:35

Rheological characterization of heterogeneous mucus samples obtained by disulfide bond reduction

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Disulfide bond reducing agents have been used as therapeutic drugs (mucolytics) in mucus hypersecretions. Breakage of disulfide bridges in mucus is known to cause a drop in viscoelastic properties and to affect its structure on both micro- and meso-scale. We recently demonstrated that a disulfide reducing agent, namely, the tris(2-carboxyethyl) phosphine hydrochloride (TCEP), induces macro-heterogeneities in native mucus [1], which we attributed to the tendency of the mucus to phase separate [2,3].

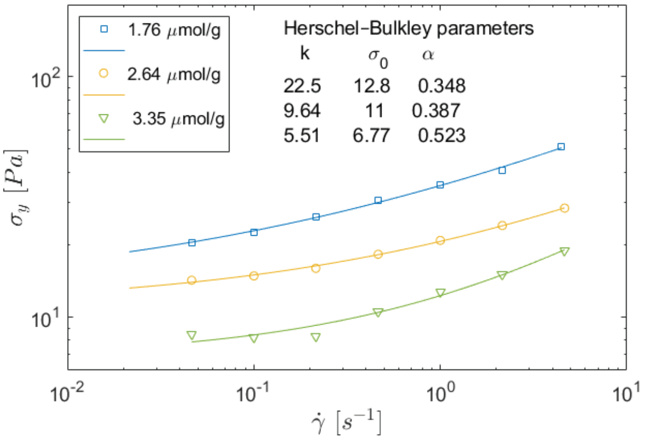

Figure 1: *Static yield-stress vs. shear rate for mucus samples treated with different TCEP concentrations (see legend). The lines are the best fitting with a Herschel-Bulkley model.*

In this work, we further explore the effect of TCEP on the mucus properties. We first assess the fractions of the different phases obtained upon treatment with different TCEP concentrations. We then analyze the nonlinear behavior of the different reduced mucus samples, classifying the rupture processes in relation to the hypothesized gel structure. This analysis was conducted on both the macro-separated system (reduced mucus as a whole) and the mucin-rich phase, i.e., on the residue of the reduced mucus obtained after centrifugation. The results confirm the heterogeneous disaggregation effect of TCEP on mucus and provide further insights into the molecular interactions underlying heterogeneous phases formed inside the mucus.

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The sedimentation role on bacteria asymmetrical distribution in confined environments

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The proliferation of bacterial biofilms is a recurring issue in sanitary, industrial and space exploration [1] fields due to fouling and contamination phenomena.
Biofilm is an example of bio-soft matter where sessile bacteria cells produce a complex matrix made of polysaccharides, eDNA, and proteins [2], colonizing interfaces and changing their interfacial properties [3]. Flow induced stresses shape this bio-soft matter enhancing nutrients transport rate, but also drag forces. Also, bulk stresses such as gravity have an interesting role separating microswimmers cells from senescent or no-motile bacteria.
We investigated Pseudomonas fluorescens SBW25 cell motility and biofilm morphology on the bottom and upper walls of a rectangular microfluidic at different wall shear rates. Differential analysis of the two walls revealed that gravity-driven sedimentation phenomena exert a substantial influence on local bacterial density at channel walls, impacting bacterial swimming behavior. These findings underscore the role of gravity vector on biofilm shaping suggesting the need of microgravity investigations.

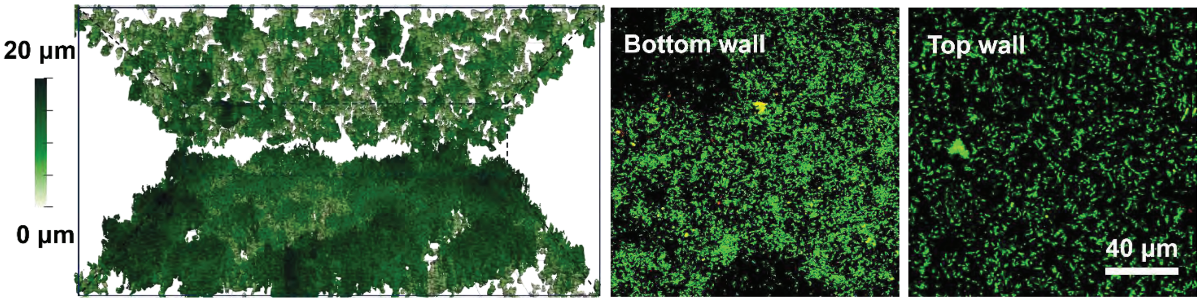

Figure 1: *CLSM images and 3D reconstruction of biofilm grown at the bottom and top part of a microfluidic channel in flow.*

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Charge effects in antibody solutions – insight from analogies to star polyelectrolytes

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Looking at globular proteins with the eyes of a colloid scientist has a long tradition, in fact a significant part of the early colloid literature was focused on protein solutions. However, it has also been recognized that proteins are much more complex than the typical hard sphere-like synthetic model colloids. This is particularly true for monoclonal antibodies [mAbs], where the anisotropic Y-shaped structure combined with a complex and heterogeneous charge distribution requires much more refined models.

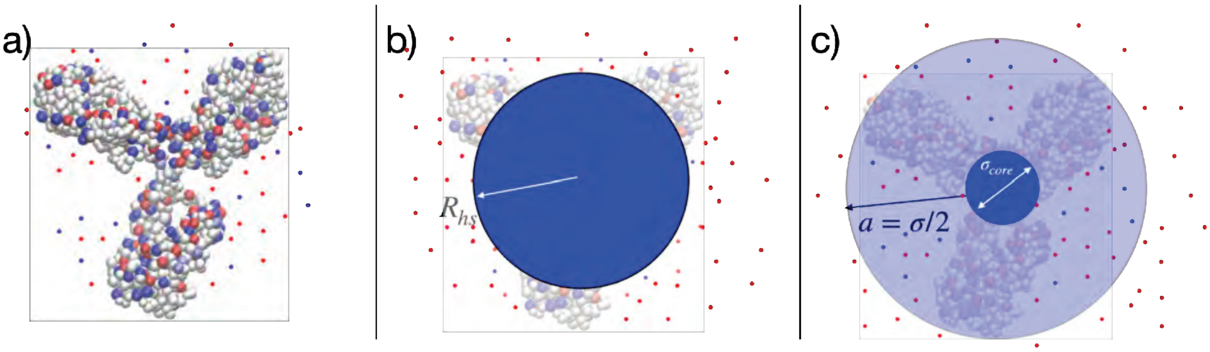

Figure 1: *Models used for describing protein-protein interactions in mAb solutions: a) weakly coarse grained model for MC simulations where each amino acid is treated as a bead (red: negative, blue: positive, white: neutral); b) classical colloid model with a hard colloid interacting with an additional screened Coulomb potential and a short range attraction; c) soft penetrable sphere model with a soft excluded volume interaction, screened Coulomb contributions based on star polyelectrolyte theory and an attractive ramp potential.*

Here, we report the results of a systematic investigation of the solution properties of a charged IgG1 mAb as a function of concentration and ionic strength using a combination of electrophoretic measurements, static and dynamic light scattering, small-angle x-ray scattering (SAXS), and tracer particle-based microrheology [1,2]. We analyse and interpret the experimental results using established colloid theory and coarse-grained computer simulations. Particular attention is given to the role of charges and their distribution on the antibody surface. Using a multi-scale coarse graining strategy that starts from the known molecular structure of the protein, we develop models based on analogies to soft particles such as star polyelectrolytes. We describe a model that is capable of reproducing the experimental results by using the overall charge and molecular dimensions obtained from an initial molecular simulation of the antibody structure and charge distribution only.

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Targeted Thrombolytics using Red Blood Cell derived Drug Delivery Systems

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Vascular thrombosis, encompassing ischemic stroke and myocardial infarction, remains a major cause of global mortality and long-term disability. These conditions arise from the formation of occlusive thrombi in blood vessels, necessitating rapid restoration of blood flow to prevent tissue damage. While intravenous thrombolytic therapy is an established treatment for thrombosis, its clinical efficacy is significantly limited by the rapid enzymatic degradation of thrombolytic agents and systemic off-target effects, leading to a severe haemorrhagic risk [1]. A promising approach to address these limitations is to employ drug delivery systems capable of enhancing thrombus targeting and improving drug stability.

To that end, we propose Red Blood Cell Vesicles (RBCVs) for the targeted delivery of a potent thrombolytic agent – streptokinase (SK). RBCVs are naturally derived nanotherapeutics composed of red blood cell membranes and functionalized with cyclic RGD peptides. The natural membrane confers RBCVs with innate biocompatibility and prolonged circulation [2], while the targeting peptide enables high-affinity binding to activated platelets in thrombi [3]. This design facilitates the precise delivery of SK to a thrombus site while minimising systemic and immunogenic toxicity [2].

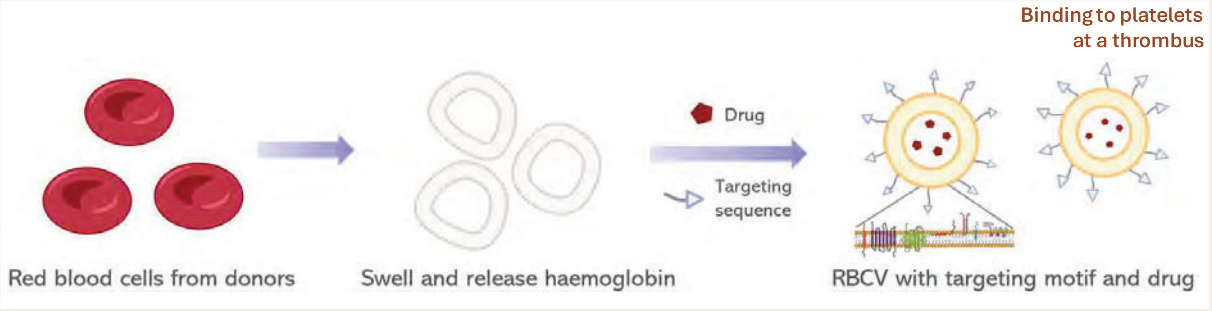

Figure 1: *Schematic of RBCV production from red blood cells.*

In *in vitro* clot models, SK-loaded RBCVs (SK-RBCVs) demonstrate a significantly enhanced thrombolytic potential compared to free SK, with up to 5 times greater clot dissolution rate. SK-RBCVs also effectively lysed platelet-rich clots, which are typically resistant to conventional thrombolytics, initiating clot breakdown approximately 15 minutes earlier, at a threefold higher rate, and to a greater extent than free SK. Furthermore, in a physiologically complex medium containing blood serum, SK-RBCVs exhibited a twofold increase in both thrombolysis rate and initiation compared with free SK, highlighting the protective role of RBCVs as drug carriers. These findings emphasize the potential of RBCVs to improve the safety and efficacy of thrombolytic interventions – ultimately leading to better patient outcomes.

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Posters

Optimization of the Refractive Index for Quantitative 3D Real Space Analysis using Confocal (STED) Microscopy

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To better understand the structure of colloidal assemblies and the nucleation pathways that lead to them, quantitative information is essential. One way to obtain this information is through confocal microscopy. However, this technique is diffraction-limited, meaning there is a minimum particle size below which data cannot be acquired. Smaller particles often exhibit interesting properties, requiring the use of super-resolution techniques such as stimulated emission depletion (STED). Our group has previously demonstrated that obtaining information about silica particles below the diffraction limit is challenging [1]. We have investigated the optimal fluorescent core size in core-shell silica particles, as well as the required index-matching solution. Surprisingly, the latter depended on the age of the particles. Recently, Fan et al. [2] identified the cause of this issue: large amounts of ammonia are incorporated into silica particles during synthesis, leading to a time-dependent refractive index when dispersed in water. To mitigate this issue, we optimized imaging conditions by precisely measuring and stabilizing the refractive index of silica. Under optimal conditions, where the fluorescent cores are sufficiently separated and the refractive index mismatch is below 0.005, quantitative real-space data can be obtained for particles with diameters of approximately 300 nm. This enables the analysis of structures with interesting photonic properties. As a proof of concept, this approach has also been extended to supraparticles, which exhibit red structural color with the aforementioned particle size.

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Self-Healing Under Stress: Zooming Into The Response Of Reversible Materials

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Adaptive materials, also known as smart materials, typically found in nature and living organisms, have the capability of responding to external or biological triggers, such as temperature, pH, magnetic field, enzymes, mechanical stress, etc., by altering their physical and chemical features. Conversely, most synthetic chemically cross-linked materials lack adaptive responsiveness due to the inability of their static covalent bonds to reestablish after rupture. Thus, designing synthetic materials with dynamic, non-covalent bonds with the goal of mimicking the self-healing ability of natural materials has garnered attention in the scientific community. Although investigations have revealed the molecular origin of self-healing, much less is known about the bonds' cooperative behavior on the meso/macroscale resulting in the self-healing of smart materials. Detailed descriptions of mesoscopic and macroscopic adaptive responses are required to fully understand how multiscale dynamics, applied stimuli, supramolecular bond chemistry, and self-healing kinetics work together (**Figure 1**). Herein, several adaptive materials will be synthesized based on published protocols and characterized, utilizing in-situ experimental tools, to realize the cooperative mechanisms in these materials. Furthermore, hierarchical dynamic features, such as bond rearrangements and network defects, and self-healing relaxation mechanisms after mechanical stimuli will be studied in detail. Employing multi-technique approaches based upon conjoint magnetic resonance imaging (MRI) and nuclear magnetic resonance (NMR) measurements and materials characterization tests like rheology and confocal microscopy, we will evaluate the fabricated materials and determine how bonds dynamically adapt at the mesoscale to restore the macroscopic physical integrity of a material.

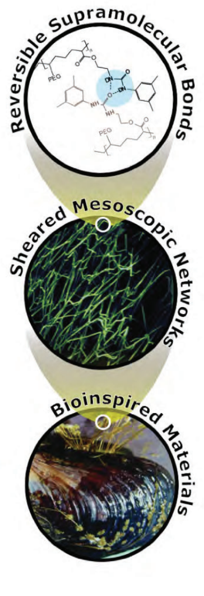

Figure 1: *Hierarchical materials' adaptive dynamics on multiple length scales: reversible supramolecular bonds on nanoscale (1 nm-10 µm), sheared network cross-links on mesoscale (10-100 µm), and bioinspired materials on macroscale (100 µm-1 mm).*

Bicontinuous morphologies formation - structural transition model for double diamond and gyroidal architecture

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Since the first discoveries made by Hermann Schwarz, triply periodic minimal surfaces (TPMS) have attracted significant attention from scientists and engineers. These unique architectures can spontaneously develop in so-called self-assembling systems. It has been shown that thermodynamically driven structural transitions can lead to the formation of various morphologies, including TPMS, such as double diamond (DD) and double gyroid (DG) structures (see Fig. 1). These bicontinuous, domain-like structures have been experimentally observed in synthetic copolymers and lipids [1, 2]. Notably, the remarkable gyroidal morphology, discovered by Alan Schoen, has also been found in biological systems, where it plays the role of a natural photonic crystal. Recently, with the rapid advancement of 3D printing technology, these fascinating geometries have gained particular interest among engineers working in the fields of metamaterials and novel structural designs. It has been documented that intertwined truss geometries based on TPMS exhibit unique mechanical and optical properties.

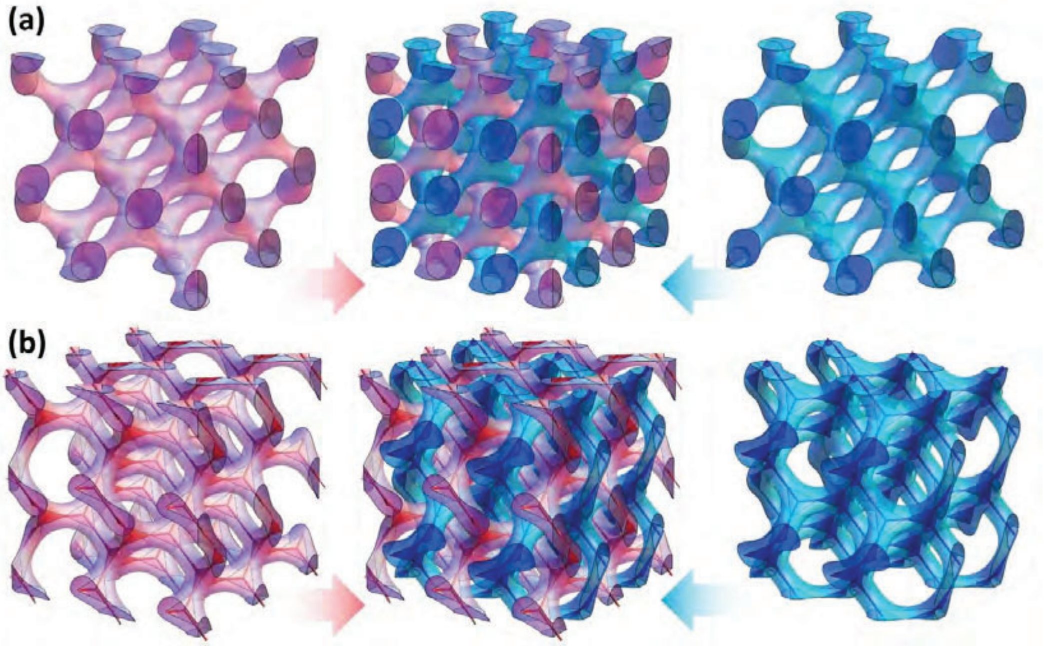

Figure 1: *a) DD and b) DG morphology*

Although TPMS are well-defined and well-documented, it is difficult to find a clear structural transition model in the literature that explains the formation of these exotic architectures. Here, I would like to introduce two geometrical transformation models that propose alternative pathways leading directly from lamellae to either DD or DG domain morphology.

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Structural characterization of asymmetric myelin membranes and their interaction with Myelin Basic Protein

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Myelin, an asymmetric multilamellar membrane enveloping axons, comprises alternating extracellular and cytoplasmic leaflets. Its composition includes phospholipids, sphingolipids, and cholesterol, with Myelin Basic Protein (MBP) playing a key role in stabilizing the structure by acting as an intermembrane adhesion protein between the cytoplasmic faces, forming the major-density-line [1].

Structural alterations in the myelin sheath, particularly demyelination, are indicative of various inflammatory neurological disorders, such as Multiple Sclerosis (MS) [2]. Experimental autoimmune encephalomyelitis (EAE) serves as a recognized animal model for MS, characterized by significant changes in the overall myelin lipid composition [3].

This study focuses on generating flat asymmetric myelin membranes, suitable for Neutron Reflectometry (NR) analysis. Employing the Langmuir-Blodgett and Langmuir-Schaeffer techniques, we successfully adsorbed asymmetric bilayers onto silica wafers.

Our findings reveal that the asymmetric myelin membranes demonstrate minimal differences in Scattering Length Density (SLD) when using non-deuterated lipids. However, deuterated lipids, such as d45-cholesterol, significantly enhance the detection of asymmetry. When comparing the pathological and native membranes, Myelin Basic Protein (MBP) adheres less to the EAE-modified membrane than to the native one and also induces less compaction between the membranes (Figure 1). Both reduced compaction between bilayers and membrane swelling may serve as potential destabilizing factors for the Myelin Major Dense Line in EAE-modified myelin.

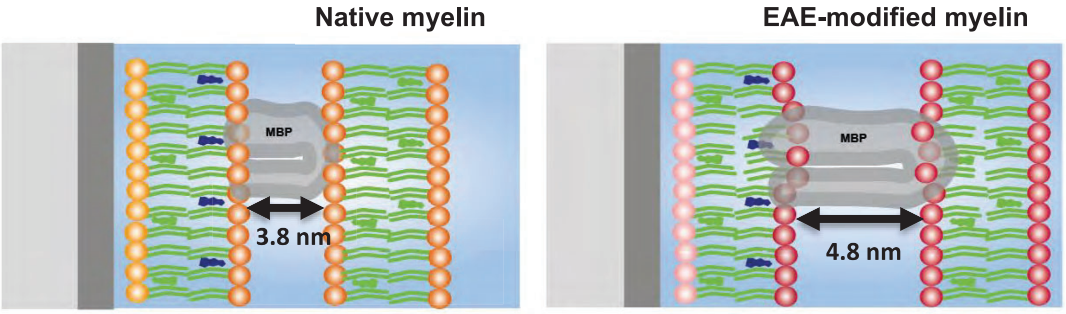

Figure 1: *MBP has less affinity for EAE-modified myelin, and the membranes adhere less tightly.*

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Structural and dynamical properties of PS-PNIPAM block copolymer micelles in concentrated samples

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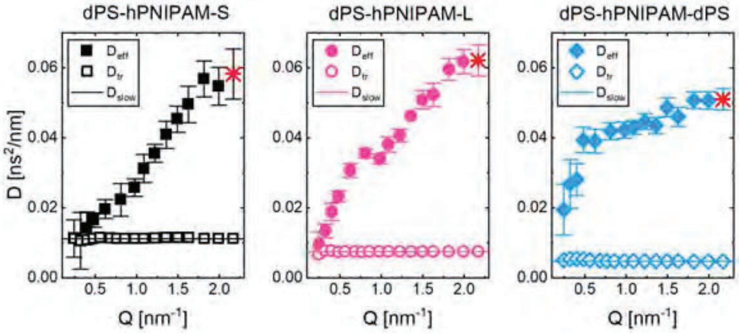

Fig.1: Full symbols: effective diffusion coefficient from NSE. Open symbols: translational diffusion coefficient, corrected of structure factor effects. Solid line: diffusion coefficient of the core as observed on NBS.

Concentrated solutions and gels of block copolymer micelles demonstrate both the cohesive strength of solids and the diffusive transport characteristics of liquids, and are subject of theoretical and technological interest [1]. Special attention is devoted to polymers containing a stimuli-responsive "switching" block that imparts the system the ability to undergo sharp, reversible changes in response to variation of external control parameters (e.g. temperature)[2]. In this work we investigate the behaviour of diblock and triblock copolymers containing thermoresponsive poly-N-isopropyl acrylamide (PNIPAM) and persistently hydrophobic polystyrene (PS) blocks, in concentrated water solution [3]. The polymers form spherical micelles in water, whose strucural parameteres have been determined by small-angle neutron scattering (SANS). At high concentration, the micellar samples show a completely different rheological behaviour depending on block size and, most importantly, architecture. In particular, a “soft” gel is formed in triblock-containing samples as the triblocks form junctions bridging different micellar cores. At the microscopic level, the PNIPAM chain mobility is influenced by the size and architecture of the polymer as well as topological constrains induced by the grafting on the PS core (Fig.1). Neutron spin echo (NSE) data shows how the single-chain dynamics is at best described by Zimm with one end fixed and additional internal friction between monomers. Moreover, the data clearly show an additional slow diffusion contribution that results from static correlation between on the micellar core grafted chains. Additional information is provided on the analysis of PNIPAM dynamics on shorter time an length scales by neutron backscattering (NBS). The evaluation confirm that backbone dynamics is mostly frozen on the nanosecond time scale due to internal friction, whereas side chains are mobile on the scale of hundreds of picoseconds.

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From Rice Biomass Waste to Bioactive Lignin Nanosystems

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Rice is a staple food worldwide, with a per capita consumption of about 65 kg/year. Its production, which is expected to increase significantly in the near future to feed the growing human population, generates enormous amounts of non-food biomass, primarily in the form of straw and husk. Currently, less than 20% of these residues are effectively utilized, while most are still burned on-site or incorporated into the soil.[1] However, both strategies have detrimental environmental effects and fail to harness the great potential of rice waste.[2] In this framework, the present study aimed at the development of bioactive nanoplatforms through the comprehensive valorization of the waste biomass from rice cultivation, with a specific focus on lignin and silica. These components were isolated, characterized, and transformed into high-value-added materials.

Specifically, once extracted by calcination, silica was characterized from a physicochemical standpoint and converted into porous silica microparticles, named Microscafs®, for the immobilization of transferases. On the other hand, lignin was isolated by dioxane extraction and thoroughly characterized using ^{31}P NMR, 2D HSQC NMR, and GPC. Subsequently, lignin was subjected to biocatalytic oxidation using a bacterial laccase at an alkaline pH of 10.6.[3] This process resulted in two fractions: i) low molecular weight lignin-derived molecules (12% yield) used as precursor for the synthesis of bioactives (LBMO) via the silica-immobilized transferases, and ii) a high molecular weight oxidized fraction (HMWL), showing a significant reduction in phenolic and aliphatic hydroxyl groups (approximately twofold and 4.6-fold, respectively), along with an increase in molecular weight with respect to the untreated lignin. HMWL was employed to produce LBMO-loaded lignin nanoparticles (diameter of ~ 200 nm) using a simple solvent-antisolvent precipitation process. Finally, the antioxidant and antibacterial properties of the prepared LBMO and of the lignin-based nanoplatforms were determined.

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P7

The LUCE project: can lignin be used to stimulate polysaccharides accumulation by microalgae?

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Recently, it has been demonstrated that the use of phenols and lignosulfonates derived from lignin in the cultivation of *Euglena gracilis* promotes the growth of microalgae and the accumulation of valuable metabolites.[1,2] Additionally, some photosynthetic microorganisms, such as cyanobacteria, are capable of accumulating cellulose. The LUCE project explores the use of technical lignins (TL), by-products of the paper industry and biorefineries, as growth stimulators for photosynthetic microalgae, with the aim producing of high value-added polysaccharides. Currently, TL are primarily incinerated for energy production due to their structural complexity and variability, which limits their commercial valorization. LUCE proposes an innovative and sustainable approach to transform TL into a biotechnological opportunity, promoting the accumulation of nanostructured cellulose and paramylon in *Synechococcus* PCC 11901 and *Euglena gracilis*.

Once grown, microalgal biomass will undergo selective extraction processes using environmentally friendly technologies (scCO_2, microwave, and ultrasound), with detailed characterization of the extracted polysaccharides through NMR, GPC, FT-IR, SEM/TEM and UV-Vis spectroscopy. The project includes the conversion of microalgal cellulose into nanocellulose (CNF/CNC) and the chemical modification of paramylon for the development of advanced bionanocomposites for smart packaging and functional materials. LUCE integrates multidisciplinary expertise in biotechnology, metabolic engineering, materials chemistry, and economic assessment, along with a techno-economic analysis to support industrial scalability. The expected results could revolutionize the valorization of lignin, mitigate environmental impact, and enable the development of novel functional materials for high-tech applications.

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Paramagnetic liposomes-based nanocarrier investigated by NMR and MRI methods

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Liposomes are widely used phospholipid-based biocompatible nanoparticles. The unique structures of liposomes allow them to be used as a nanocarrier for various components encapsulated in the aqueous core, between two phospholipid layers or attached to their surface [1]. These properties allowed us to create multifunctional hybrids containing gadolinium chelates-based MRI contrast agents (gadopentetic acid) and a hydrophobic photosensitizer (ZnPc) for photodynamic therapy. This system was prepared by the microfluidic method, which allows double the loading capacity of active components compared to the system made by traditional extrusion methods.

Using the NMR spin-lattice relaxation methods allows assessing the MRI contrast properties of the nanocarriers and asses the sensitivity of the contrast agent (relaxivity). Moreover, the T1 relaxation study in the wide range of magnetic field (H1 relaxation frequency) gives a proper tool for discovering the influence of the ZnPc component on the nanoparticle behavior, which corresponds to the increase in contrast properties. The results of the relaxivity parameters obtained for liposomal contrast agents were compared with the clinically used contrast agents.

To confirm the contrast properties at the images, the MRI studies were performed for the set of samples with dispersion of liposomes in different Gd concentrations, and finally MRI in vivo experiments at rodents.

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P9

Tailorable Gelatin Films from Fish Scales: A Sustainable Platform for UV-Blocking Wearable Patches

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The growing demand for sustainable materials has intensified research on valorizing fishery byproducts, a largely underutilized resource generating millions of tons of waste annually [1]. Among these, fish scales offer a rich source of biopolymers such as collagen and gelatin, which are highly desirable for biomedical applications due to their biocompatibility, biodegradability, and low immunogenicity [2]. This study focuses on the development of multifunctional gelatin-based films from mullet (Mugil cephalus) scales, designed for wearable healthcare patches with customizable properties, including UV-blocking, antimicrobial activity, and mechanical performance.

Through the addition of carefully selected additives, the properties of the gelatin films were finely modulated. Specifically, i) carbon dots (CDs), derived from seabass (Dicentrarchus labrax) scales,[3] imparted excellent UV-blocking capabilities, reducing transmittance to as low as 3% at 275 nm; ii) surface-deacetylated chitin nanocrystals (CsNCs) enhanced mechanical properties, achieving up to a 4.5-fold increase in tensile strength and maintaining high elasticity; iii) citric acid (CA) and chitosan (Cs) were used as cross-linkers, reducing water solubility to 3% while improving film rigidity; iv) polysorbate-20 (PS) acted as a dispersant for CDs, improving their distribution and contributing to enhanced UV protection and optical clarity; v) hydrophobic agents such as palmitic acid further tuned water resistance and permeability.

Film thickness was also varied (20–100 μm), allowing additional control over mechanical properties, UV shielding, and water vapor permeability. Thicker films exhibited superior UV-blocking performance and aligned with the moisture management needs of wound-healing applications.

Finally, biocompatibility tests on MRC-5 fibroblasts confirmed the non-cytotoxic nature of the films, while accelerated aging under simulated solar light demonstrated excellent durability with minimal property degradation. This study underscores the potential of fish waste-derived gelatin films as a sustainable solution for biomedical applications, addressing both environmental and healthcare challenges.

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Influence of Ionic Conditions on the Liquid-Gas Critical Point of the TIP4P/2005 Water Model

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We perform Molecular Dynamics simulations to investigate the liquid-gas critical point for the TIP4P/2005 water model. In the canonical (NVT) ensemble, density fluctuations are quantified and analyzed using a recently developed method [1] based on cumulant crossings. Complementary simulations in the isothermal-isobaric (NPT) ensemble yield density and energy distributions, which are mapped onto the universal 3D Ising master curve via histogram reweighting. The critical points determined by the two approaches exhibit very good agreement, highlighting the robustness of this methodology. This work establishes a reliable framework for accurately locating the critical point which is applied to test the influence of ionic conditions on the latter (see Fig. 1). We plan to extend our study to the second critical point of water, aiming to shift it to experimentally accessible regimes.

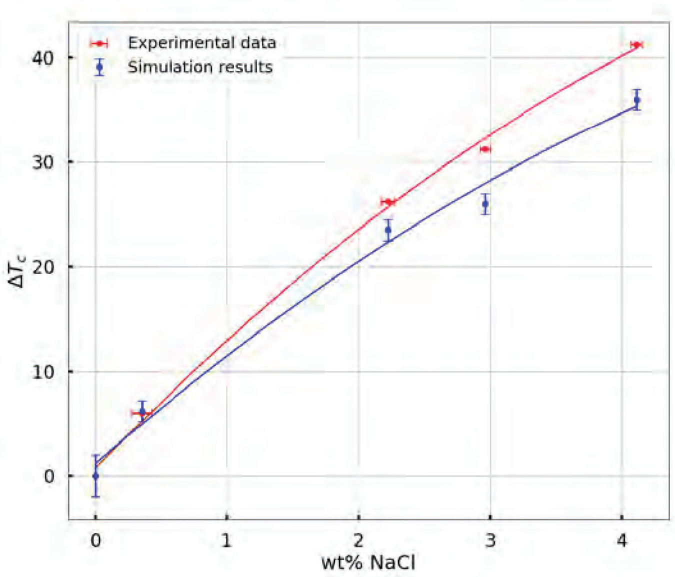

Figure 1: Shift in the critical temperature with increasing NaCl concentration. Experimental data taken from [2].

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Exploring Pathways of Supramolecular Self-Assembly at Liquid-Liquid Interfaces

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A self-assembly process usually occurs through thermodynamic pathways but the co-existence of different intermediates and diverse pathways of self-assembly have been observed. Liquid droplets are one such intermediate in many self-assembly processes, particularly in biological systems, which form through liquid-liquid phase separations (LLPS). Even though LLPS is a very well-known phenomena in polymer chemistry, only little is known about the possible role of LLPS in synthetic small molecule-based self-assembly.

In this project, we aim to study the possible mechanism of self-assembly at the surface of phase separated liquid droplets to explore the structure and dynamics at various stages of self-assembly and provide insights into how it can be controlled and manipulated for various applications. To that end, we develop a simple coarse-grained model for monomers in explicit solvent that self-assemble into fibers. The model is designed to represent BTA-like molecules in water. It is able to capture key characteristics of the experimental system such as the competition of fibril formation and liquid-liquid phase separation. We use it to examine the pathways of fibril formation from phase separated droplets.

Extension and Scission of polymers under high shear rates in microfluidic chips

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Microfluidic devices are ideally suited for the study of complex fluids undergoing large deformation rates in the absence of inertial complications [1]. Previous studies involving conventional narrow-width microfluidic channels faced challenges accurately measuring the flow-induced deformation of long-chain polymers, due to significant measurement uncertainties and limited precision [2]. In this research, we introduced a microfluidic chip fabricated specifically to provide accurate measurements of flow-induced birefringence in polymer solutions by polarized optical microscopy. As shown in Figure 1, the polymer solution (100K PEO, 1 wt % in water) was pumped into the microfluidic chip at a flow rate of 10 ml/h. In the central microchannel with a height of 20 µm and a width of 20 µm, the long-chain polymer molecules became oriented along a specific direction due to the flow-induced extension, forming flow-induced birefringence.

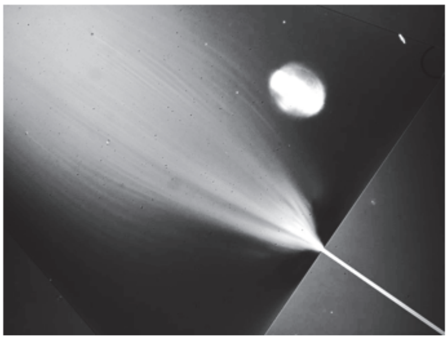

Figure 1: *Birefringence phenomenon observed under polarized optical microscopy (The direction of flow is from the top-left to the bottom-right).*

Another comparative viscosity chip, combined with high-speed micro-particle image velocimetry (µPIV), was used to precisely measure the viscosity changes under high shear conditions. Furthermore, Gel permeation chromatography (GPC) was employed to determine molecular weight distributions accurately. The sample, after the treatment in Figure 1, exhibited a clear decrease in molecular weight, with the number-average molecular weight (Mn) dropping to 45.14K. This indicates that the PEO sample underwent chain scission under high shear flow conditions. Moreover, the pronounced reduction in molecular weight demonstrates that the high shear flow is particularly effective at causing scission of longer polymer chains, confirming that high shear flow is more effective in breaking long-chain polymers. This comprehensive approach offers enhanced accuracy in analyzing the viscosity behavior of polymers under flow conditions. Such an approach provides valuable insights into polymer rheology, drug delivery systems, and industrial production processes.

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P13

Lipid nanoparticles loaded with Gd_2O_3 for enhanced Magnetic Resonance Imaging

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Nanoparticle-based contrast agents (CAs) for magnetic resonance imaging (MRI) have gained significant attention over the past few decades [1]. Among these, lipid-based ones serving as vehicles for the contrast-providing entity. These lipid-based carriers enhance the physicochemical stability, biocompatibility, and intracellular uptake of MRI CAs and can potentially improve relaxivity (r_1) [2]. The primary type of contrast agents used in MRI scans are using Gd-based formulations, and despite the health risks associated with Gd^{3+} ions, these cannot be entirely excluded from the use and further research [3]. Therefore, new solutions are being explored to enhance the biocompatibility of these Gd-based agents, including methods to reduce the leakage of free toxic ions and improve relaxivity. The promising direction is using lipid-based nanoparticles that may satisfy the mentioned issues.

Gd_2O_3 nanoparticles are considered in the study as CAs for T_1-enhanced MR imaging. The proper surface modification followed by embedding into a lipid-based carrier is proposed to ensure their biocompatibility and enhanced interaction with the surrounding water proton. Herein, lipid nanoparticles formed from glyceryl monooleate were used as carriers of Gd_2O_3 nanoparticles with hydrophobic (Gd_2O_3-oleate) and hydrophilic (Gd_2O_3-dimercaptosuccinic acid) properties, which were loaded in different weight ratios to lipid matrix. These nanosystems were studied regarding their physicochemical properties and positive T_1 contrast enhancement. However, the focus of the study was to investigate the Gd^{3+} ion loading efficiency, including calculations per nanoparticle, Gd^{3+} ion release in different conditions, and, finally, Gd^{3+} ion cell uptake, from prepared lipid-based nanoparticles as MRI CAs, which are critical characteristics regarding the development of CAs for MRI. These studies used the rapid and novel single particle and single cell (SP/SC)-ICP-MS method. This research tool greatly supports the development of nanoparticle-based formulations, including lipid ones loaded with metallic ions. Further, the *in vitro* cytotoxicity and functional assays were performed to confirm the biocompatibility of prepared lipid-nanoparticle-based carriers. Results indicate that after the precise refining of the composition and preparation method of the lipid-based nanoparticles loaded with Gd_2O_3, affecting the physicochemical and required relaxivity parameters, proposed nanosystems may be considered as new and biologically safe MRI T_1-enhancing CAs.

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Bacterial Motility and Membrane Deformation in Biohybrid Vesicles

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We want to understand the interaction of bacteria with lipid membranes in the context of intracellular bacteria invading host cells. Membrane deformations are critical for bacterial survival, immune evasion, and dissemination. To investigate this, we develop a biohybrid vesicle encapsulating *E. coli* within an artificial giant lipid vesicle. These vesicles serve as a minimal model to study bacterial-driven membrane deformations and self-propulsion [1]. The bacterial motion within the vesicle generates forces that deform the membrane, with deformation extent dependent on bacterial density and mode of motion [2]. We examine the vesicle's ability to undergo directed motion in chemical gradients, driven by the chemotactic motility of the bacteria inside.

To study this process, we designed a flow-free microfluidic system that establishes stable, diffusion-based pH gradients without introducing shear forces [3]. The vesicles, permeable to pH gradients, enable bacteria to exhibit chemotaxis within their lumen, leading to asymmetric bacterial distributions and vesicle propulsion [4]. We hypothesize two primary mechanisms for vesicle motion: (i) flagellar bundling forming helical propellers that couple with the membrane [1], or (ii) bacteria accumulating asymmetrically to generate a collective force for directed movement [2]. These behaviors are governed by bacterial swim pressure and vesicle membrane tension.

This study aims to enhance our understanding of bacterial interactions with lipid membranes and informs the design of smart drug delivery systems capable of sensing and responding to environmental cues.

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Expanding the reach of diffusing wave spectroscopy and tracer bead microrheology

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Diffusing Wave Spectroscopy (DWS) is an extension of standard Dynamic Light Scattering (DLS), applied to soft materials that are turbid or opaque. The propagation of light is modeled using light diffusion, characterized by a light diffusion coefficient that depends on the transport mean free path ℓ^* of the medium. DWS is highly sensitive to small particle displacements and local fluctuations in scattering properties, enabling the detection of subnanometer displacements. One of the most common applications of DWS is one-bead microrheology, where the motion of beads in a viscoelastic matrix is analyzed. Since its invention in 1987, DWS has seen significant advancements, including two-cell and multi-speckle techniques. However, challenges remain, such as merging single and multi-speckle data and improving accuracy for short correlation times. Here, we address these issues by improving the two-cell Echo DWS scheme. We propose a calibration-free method to blend and merge Echo and two-cell DWS data. Additionally, we demonstrate the use of regularized inversion algorithms to enhance data quality at very short times. Building on this, we introduce stable corrections for bead and fluid inertia, significantly improving the quality of microrheology data at high frequencies.

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Timelapse confocal microscopy of *P. putida* biofilms grown under flow in a microfluidic platform

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Biofilms are surface-associated bacterial communities of cells enclosed in a matrix of self-produced extracellular polymeric substances (EPS). While undesired in many domains such as wound healing, food production, medical treatments and industrial fouling, biofilms are also growing in importance as a positive tool in biotechnological applications including bioremediation, biofertilization and energy production. The conditions inside of a biofilm are completely different from the bulk fluid, as bacteria are present in a high concentration in the biofilm, and the secreted EPS make the whole biofilm very viscoelastic in nature.

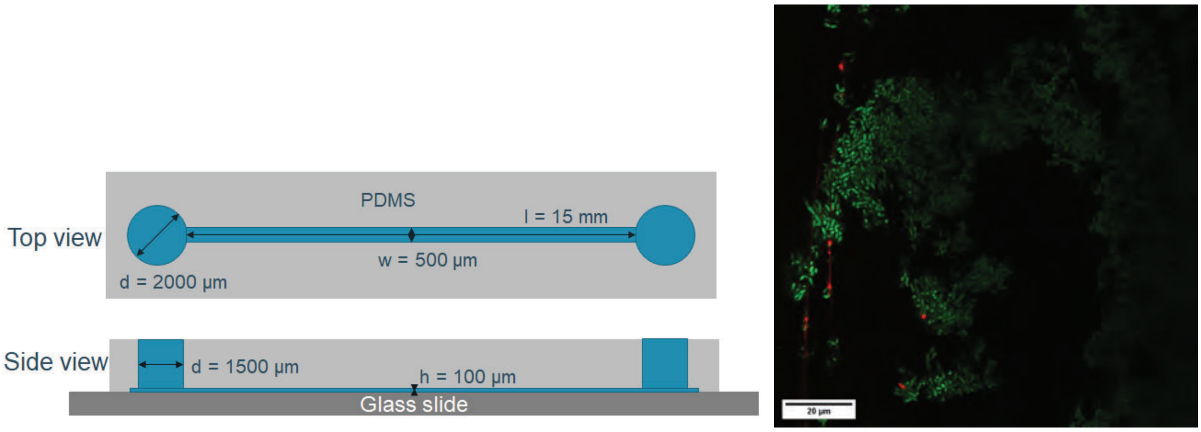

Figure 1: *Illustration showing the dimensions of the microfluidic channel used (Left). Confocal micrograph showing a P. putida biofilm 62 h after start of flow (Right). The live cells are visible in green, the dead cells in red. A 50 µm high channel was incubated at 30 °C, and supplied with 153 µL/h of LB medium (10 g/L tryptone, 5 g/L yeast extract, and 5 g/L NaCl).*

In this work we demonstrate how to use confocal microscopy to characterize the structure of biofilms under flow. More precisely, biofilms are grown in a microfluidic chip, allowing to grow the biofilm under relevant conditions (shear stress, replenishment of nutrients, temperature [1]. For the confocal microscopy, a high NA water immersion objective is used to image without any refractive index mismatch and avoid the associated data artefacts. Special care has to be taken to compensate for water evaporation, as the timelapse measurements are taken over long timeframes ($\sim$ 65 h). Furthermore, an image analysis pipeline is used to segment the images and obtain information on the count and orientation of the bacteria, allowing to obtain long-term, time-resolved information of the structure and growth of the biofilm.

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Characterization of the Local Mechanical Response of DNA Hydrogels via Mechanofluorescence

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Mechano-sensitive materials convert a mechanical signal into a physical or chemical signal. These materials are ubiquitous in nature, where they regulate cell spreading and are involved in senses such as hearing and touch [1,2]. Inspired by these materials, we developed DNA-based hydrogels that convert a mechanical signal in fluorescence response [3]. The hydrogels were assembled from sequence-controlled DNA strands produced by enzymatic synthesis. The mechanism of mechano-fluorescence relies on the functionalization of a DNA hairpin with a FRET pair (a fluorophore and a quencher that interact via Froster Resonance Energy Transfer, FRET), which becomes fluorescent only when the hairpin opens (Fig. A). We recently developed a new set-up to quantify the mechano-fluorescence response of the hydrogels in stress-controlled tests, using a shear cell with transparent windows coupled to a fluorescence imager. As shown in Figs. B-C, the fluorescence of the gel increases with the applied stress indicating that more and more probes open. We also observe a clear and strong correlation between the nonlinear response of the hydrogels—characterized by emerging strain stiffening and hysteresis—and the fluorescence signal. In conclusion, this work presents the first quantitative stress-fluorescence measurements on mechano-fluorescent DNA-based hydrogels and proposes an interpretation of their molecular origin.

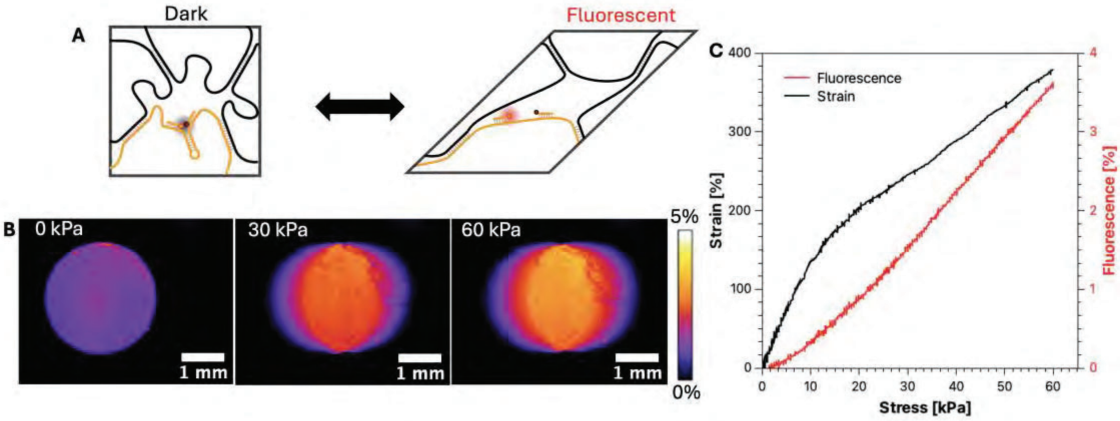

Figure 1: *(A) Schematic representation of mechanofluorescence mechanism. (B) Images collected during a mechanofluorescent test. (C) Quantitative strain (black) and fluorescence (red) measurements as function of the stress applied on a mechano-fluorescent DNA-hydrogel.*

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Introducing vitrimers in Electrolytes for Lithium-metal Batteries

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With properties in between thermosets and thermoplastics, vitrimers have many potential applications. Vitrimers are permanent polymer networks with dynamic covalent bonds that allow the network to change its topology while maintaining a constant number of chemical bonds. Therefore, these polymer networks have fascinating characteristics like shape memory, self-healing and recyclability [1]. One potential application of vitrimers is their use in lithium-metal batteries.

Lithium-ion batteries have become essential in our daily lives, powering a wide range of devices from smartphones to electric vehicles. The need for longer-lasting and more efficient storage solution has accelerated the search for batteries with higher energy density. While lithium-metal batteries (LMB) offer the potential to attain significantly higher energy densities, safety concerns related to the liquid electrolyte have hindered their widespread adoption. Solid-state electrolytes have emerged as a potential solution to enhance the safety and performance of LMBs. Solid state electrolytes are divided into three categories: inorganic ceramic electrolytes, solid polymer electrolytes and composite electrolytes. Among these, solid polymer electrolytes, like those involving polyethylene glycol, are known for their flexibility, low flammability, and easy processing [2]. However, several challenges are encountered in LMBs with solid-state electrolytes such as the loss of electrode-electrolyte contact due to volumetric changes in the electrode or cracks appearing in the solid-state electrolyte due to the formation of lithium dendrites [3]. In this context, incorporating vitrimers with self-healing and shape-memory properties into lithium-metal batteries presents a promising solution to these challenges (Figure 1). In this work, the solid polymer electrolyte with a vitrimer behavior is based on thioplast G131 crosslinked with poly(ethylene glycol) diglycidyl ether (PEGDE).

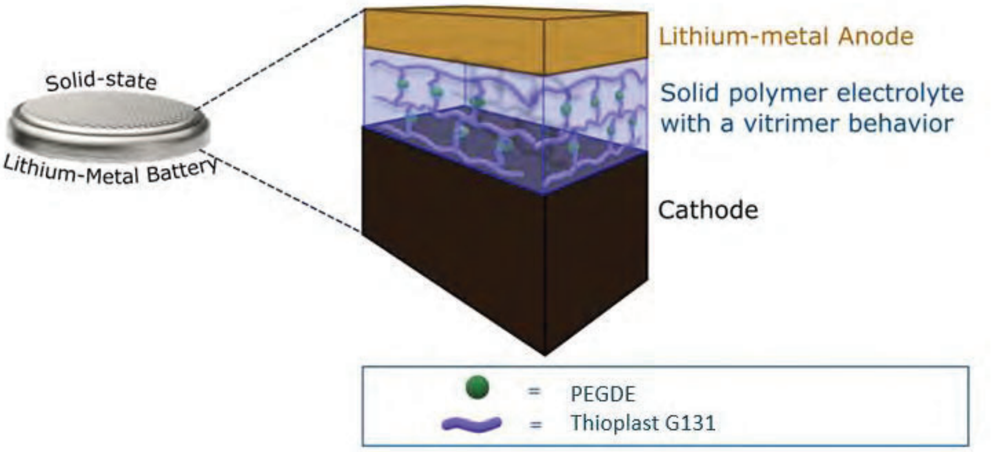

Figure 1: *Solid polymer electrolyte with a vitrimer behavior in LMB.*

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Diffusion Study in Hydrogels for Cornea-on-a-Chip Models

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To tackle the challenge of poor translation from preclinical studies to clinical trials, the scientific community has introduced organ-on-a-chip technology, innovative 3D microfluidic devices integrating living cells to mimic human organ functions. Building on this concept, our project focuses on developing a cornea-on-a-chip, designed to replicate the structural and functional complexity of the human cornea in a microfluidic device that simulates all physiological flows. By accurately reproducing its three layers (epithelium, stroma, and endothelium), this model aims to provide a physiologically relevant platform for ophthalmological research. The ability to better simulate corneal architecture and interactions could help reduce discrepancies between current preclinical tests and clinical outcomes, thereby improving ocular treatment development.

To recreate the structural and functional properties of the corneal stroma, the chip incorporates a gelatin-derived hydrogel. My task focuses on the study of diffusion mechanisms and potential interactions between solutes (anionic, neutral, cationic) and modified gelatin hydrogels. The diffusion of anionic, neutral, and cationic solutes through gelatin derivatives was analyzed under varying salinity conditions and at two different polymer concentrations. Additionally, the impact of solute size on diffusion within the hydrogel was investigated. These observations are then compared to the diffusion behaviour in porcine and human corneas to determine whether gelatin derivatives are suitable models for mimicking the stromal layer of the cornea. To achieve this, several methodologies have been developed, including the construction of diffusion chambers, epifluorescence microscopy and mathematical modelling.

Recent results have shown that steric and electrostatic interactions can significantly alter diffusion behaviour. Variation in solute size directly impacts the diffusion process, providing qualitative insights into the hydrogel structure. In addition, as gelatin derivatives are slightly negatively charged at pH >5, electrostatic repulsion is particularly noticeable in the absence of salts. In contrast, cationic solutes are more strongly attracted in salt-free solutions. Consequently, salts have a strong effect on diffusion and can also provide information about the structure of the hydrogel under study. These interactions, which modify the diffusion dynamics, highlight the need to refine the current mathematical models by taking this additional parameter into account. Incorporating these new data could improve the accuracy of simulations and, ultimately, increase the effectiveness of drug delivery systems, both for artificial corneas and for other tissue engineering applications.

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Effect of silica nanoparticles on the curing of HEA-based composite hydrogels

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The 2-hydroxyethyl acrylate (HEA) is characterized by a hydrogen bond that ensures its solubility in water and a high boiling temperature. Its polymeric form, the Poly(2-hydroxyethylacrylate) (pHEA), is extremely versatile and is widely used in both industries, as adhesive, coating or base for acrylic enamels, and in the cosmetic and medical fields, as carrier for drugs [1-2].

To improve the mechanical properties of HEA-based hydrogels, one of the strategies is to form a particle-loaded composite. In this work, pHEA-based composite hydrogels were prepared through the in-situ synthesis of silica particles from their alkoxide precursors in the presence of HEA. To evaluate the possible effect of the presence of silica particles on the curing of HEA, rheological measurements were performed following the polymerization kinetics: starting from the mixture before curing, until the obtainment of the final hydrogel. We run multifrequency time-sweep measurements to follow the mechanical spectra of the composite hydrogel during polymerization. We applied the Winter and Chambon [3] criteria to determine the gelation point of the mixture and demonstrate that the gelation point is a key parameter in the polymerization reaction. The obtained composites were characterized with Fourier transform infrared spectroscopy, electron microscopy, and calorimetric analysis.

The results indicate that the viscosity of mixture before curing increases strongly as a function of the alkoxide concentration, due to the interaction of the in-situ formed silica nanoparticles with the HEA monomer. Silica particles strengthen the mechanical properties of the hydrogels, as evidenced by the improved rheological properties. The obtained samples show an increase in swelling ratio compared to bare pHEA.

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Cellulose supported ionic liquids - synthesis, characterization and application in water treatment

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Cellulose-supported ionic liquids provide a complementary combination of ionic liquid's adaptability and biopolymer durability, making them a cutting-edge advancement in sustainable chemistry. In order to solve pressing environmental issues, these hybrid materials combine the special physicochemical characteristics of ionic liquids with the abundance and renewable nature of cellulose. Because of their adjustable architectures, thermal stability, and recyclability, the supported ionic liquids have drawn a lot of interest for their use in energy storage, pollutant removal, green chemistry, and catalysis. The use of natural-derived polymers on the other hand, thanks to their biodegradability, availability or mechanical properties, have gained significant interest in water technologies. They are effectively used as adsorbers for both inorganic and organic micropollutants, as well as for the removal of negatively charged contaminants, at a rate faster than in case of the conventional filters. By reducing problems like leaching, high viscosity, and costs, the immobilization of ionic liquids on cellulose should improve their usability while also offering a biodegradable and environmentally benign material. [1,2]

The preparation of cellulose nanostructures functionalized with ionic liquids (supported ionic liquids) is the main goal of the study that will be presented. Various cellulose nanostructures, prepared from both the microcrystalline cellulose but also prepared from the agricultural wastes were used in the first stage. Subsequently, the cellulose platforms were subjected to surface functionalization to produce cationized cellulose systems. Two methods were used to prepare cationized cellulose with varied alkyl chain lenghts (methyl, butyl and dodecyl), meaning the functionalization with the glycylidyl-trimethylammonium chloride, and with the use of silanization and thiol–ene chemistry. Both methods are simple, efficient, and versatile. We assessed the morphology, thermal stability, and structure of the prepared materials; moreover we evaluated the reactions efficiency, which is reflected by the type and quantity of surface available functional groups. The cationized cellulose were further subjected to ion exchange reaction to obtain cellulose-supported ionic liquids, which were characterized accordingly to their substrates. Further works were continued with the use of such materials in water treatment technologies, especially in the are of organic pollutants removal, namely organic dyes, antibiotics and micro- and nanoplastics.

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Isolated domains of ion pairs grow to form extended network in diphenyl phosphate-bis(2-ethylhexyl) amine mixtures

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Diphenyl phosphate (DPP)/bis(2-ethylhexyl) amine (BEEA) liquid mixtures are expected to show peculiar self-assembly behavior due to their, respectively, acidic and basic nature which can trigger acid-base reaction. The properties of DPP/BEEA mixtures of different compositions are explored by a combined theoretical (Ab initio calculations) and experimental (rheology, X-ray and light scattering) approach. A proton transfer from DPP to BEEA takes place with formation of cationic and anionic species in liquid phase (ion pair formation). Due to the strong and long-range electrostatic interactions, each of the two charged species is preferentially surrounded by the other one, in a picture resembling the structure of ionic materials. This gives rise to a striking viscosity increase takes place as DPP concentration increases. The composition dependencies of all measured parameters show clear deviations around DPP molar fraction of 0.2, indicating unequivocally the formation of isolated domains, formed by DPP-BEEA pairs which coalescence/percolate into a unique, extended, network at higher DPP concentrations.

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Probing local rheology during solvent sorption in polymers using Diffusing Wave Spectroscopy

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Solid polymeric foams are cellular materials widely used in everyday life and across various industries, including sport footwear, construction insulation, and automotive shock absorption. Among the foaming techniques, the use of supercritical (sc) CO2 as a clean and sustainable foaming agent presents clear advantages. However, producing polymer foams with scCO2 remains complex due to dual role of dissolved CO2—initially as a plasticizer and subsequently as a physical blowing agent—which significantly alters the rheological properties of the polymer matrix. In turn, these changes directly impact the rate of diffusion of CO2 in the matrix throughout the foaming process. Therefore, a better understanding of the rheology of polymer plasticized by CO2 under supercritical conditions is essential to optimize the foaming process and to control the final properties of polymer foams. As a preliminary step for subsequent studies on polymer-CO2 systems under high pressure, we use Diffusing Wave Spectroscopy (DWS) to characterize the rheological properties of a model system that is PDMS oils absorbing their good solvents. In a nutshell, DWS uses the generalized Stokes-Einstein equation to obtain the storage G'(ω) and loss G''(ω) moduli from the temporal evolution of the mean square displacement (MSD) of nanometric tracer particles (e.g. TiO2) dispersed in the sample. Being a contact-free tool that probes local properties with good spatial and temporal resolution, DWS allows us to measure G' and G'' of a polymer during the diffusion of solvent through the matrix, at various points along the z-axis, and from the initial polymer-solvent contact (t = 0) until saturation (t = ∞). Rheological measurements from DWS are validated against those obtained using a conventional mechanical rheometer. This study demonstrates that DWS can be a powerful contact-free tool for characterizing sorption kinetics and local rheological properties in polymer-solvent systems. It establishes a foundation for modeling the mechanisms involved in saturation and nucleation during physical foaming with $scCO_2$.

Tuning a soft repulsive glass with hard colloidal fillers

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Jammed suspensions of soft, deformable particles display solid-like properties at rest, whereas they flow like liquids above a yield stress. The properties of these soft glassy materials are typically adjusted by modifying either the particle volume fraction or their intrinsic characteristics, such as their individual elasticity. In this work, we explore a different approach by incorporating hard colloidal fillers (spherical silica of around 200 nm diameter) in a concentration range of 2 to 10 wt% into a jammed suspension of soft microgels (polyacrylic acid and polyacrylic acid-polyacrylamide microgels of between 10 and 20 µm diameter). We begin by examining the linear viscoelastic properties of these mixtures as the filler volume fraction increases, considering various microgel chemistries that influence interactions between the fillers and the microgels. We then analyze how the fillers affect the yielding transition and the non-linear flow behavior of the suspension exploring different flow protocols including shear startup and Large Amplitude Oscillatory Shear experiments.

Modeling Spectroscopic Properties of Biomolecules

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Spectroscopic techniques including infrared (IR) spectroscopy are widely used methods particularly useful in probing the structural organization of (bio)molecular systems, including proteins and nucleic acids. In fact, several IR bands are highly sensitive to the changes in the structural organization of biomolecular assemblies. One of the most used signals is that arising from C=O stretching vibrations as it proved to display relevant changes depending on e.g. the secondary structure in peptides and proteins or distinct structural motifs in nucleic acids. Nevertheless, the direct mapping between IR spectral bands and specific structural features is not straightforward, in particular for systems with a rugged conformational landscape. Here, we use a tandem quantum/classical computational approach based on the perturbed matrix method (PMM-MD) formalism [1] for the calculation of IR spectra. This approach has been already successfully applied to the calculation of IR spectra in polypeptides, both in the amide I region [2] and for specific side chains bands [3, 4]. Here, it is extended to compute IR bands in DNA nucleobases. More specifically, we compute the IR spectrum arising from C=O stretching modes of all carboxyl containing bases, i.e. thymine, guanine and cytosine, in both water and deuterated water. We use as a first benchmark the isolated bases in solution, both with and without the ribose and phosphate moieties. Then, we move to more extended systems and compute the spectra of single-stranded deca-homonucleotides. At last, we consider an aptamer whose sequence embodies the combination of all nucleic acid building blocks. The comparison of the calculated spectra with the corresponding experimental spectra shows an overall good agreement. Most importantly, our approach shows to be able to reproduce the experimentally observed variations in the spectral signals due to conformational diversity, including stacking and hydrogen bonding interactions among the bases.

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