Swiss Soft Days

30th Edition
October 20th 2022
Alumni Pavillion, ETH Zürich

How to get there:

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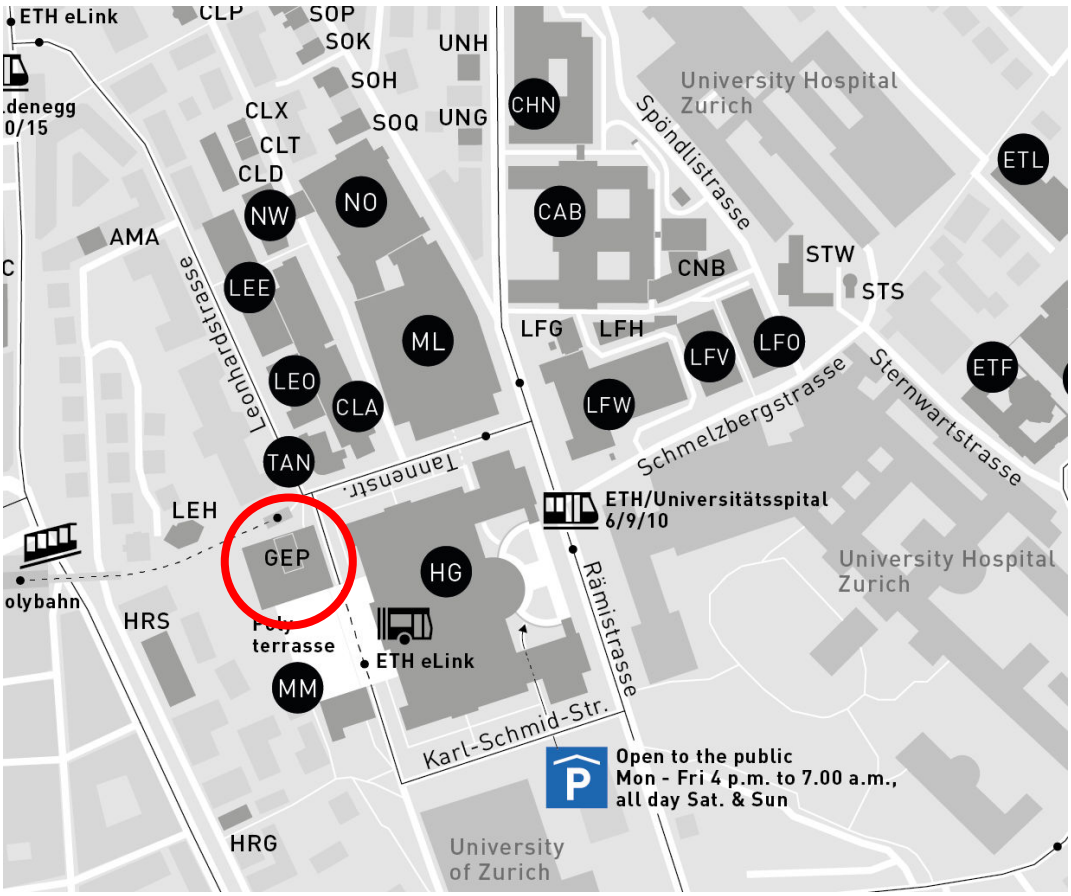


Program

09:30 – 10:00	**Coffee + Registration**
10:00 – 10:45	**Invited Talk**
	Simone Schürle «Engineering soft microrobots for drug delivery and local detection of pathological signals»
10:45 – 11:00	**Short break**
11:00 – 12:00	**Session 1**
	Viviane Lütz-Bueno «Hierarchical Porous Scaffolds for Culturing Complex Tissues: the Scattering of Foams»
	Dimitri Hürlimann «Bioactive Polymersomes Targeting Cancer-Associated Fibroblasts for Anticancer Therapy»
	Giovanni Savorana «Bacterial transport in polymer solutions»
	Viola Bauernfeind «Stable colors: how order and scale geometry make the colourful displays of Sternotomini longhorn beetles angle-independent»
12:00 – 13:30	**Lunch + Poster**
13:30 – 14:15	**Invited Talk**
	Cari Dutcher «Microfluidic Platforms for Studying Surfactant-laden Interfaces and Polymer Solutions»
14:15 – 14:30	**Short break**
14:30 – 15:15	**Session 2**
	Elisabeth Agoritsas «Mechanical response of dense amorphous materials: from passive particle systems to active matter»
	Lebo Molefe «Surface stress drives large material deformation to smooth compliant solids»
	Simon Scherrer «Measuring Rolling Friction of Microparticles using Lateral Force Microscopy»
15:15 – 15:45	**Coffee break**

15:45 – 16:45	**Session 3**
	Maryame Bina «Multicomponent solid-supported membranes based on amphiphilic block copolymers mixtures»
	Florence Müller «2 Functionalities -in-1: Embedded 3D printing of alginate into a SiO2-CaCl2 gel bath»
	Alexander Torzynski «Surface-Initiated Polymerization From Lipid Membranes»
	Xiang-Zhong Chen «Soft ceramics at the nanoscale: giant elastic strain and electrically sensitive shape-memory effect»
16:45 – 17:00	**Closing remarks**

List of posters:

[1] Unravelling the Mechanical Potential of Bioinformed Granular Hydrogels
Matteo Hirsch, Livia D'Onofrio, and Esther Amstad.

[2] Assembling Catalytic Nanocompartments into Artificial Signaling Cascades
Viviana Maffeis, Cora-Ann Schoenenberger, Catherine E. Housecroft, and Cornelia G. Palivan

[3] Hard-Soft Assembly Clusters for Bio-Applications
Voichita Mihali and Cornelia G. Palivan

[4] Zero-shear viscosity for yield stress fluids
G. Pagani, M. Hofmann, L. E. Govaert, T. A. Tervoort, and J. Vermant

[5] Thermo-responsive nanocellulose hydrogels as a universal drug release platform
Qiyao Sun, Garam Han, Gilberto Siqueira, Luca Müller, Pascal Bertsch, and Peter Fischer

[6] What is the impact of phase separation in elastic polymer networks?
C. Lorenz, C. Fernández-Rico, and E. R. Dufresne

[7] Selectively permeable viscoelastic composite microcapsules
Chuen-Ru Li, Pavel Kalinin, and Esther Amstad

[8] Polymeric Giant Unilamellar Vesicles for Bacterial Culture
Lukas Heuberger and Cornelia G. Palivan

[9] Multiplexed Analyte Detection in Droplet Interface Bilayers
Robert Strutt, Simon F. Berlanda, Petra S. Dittrich

[10] Inverting glucuronidation of hymecromone in situ by catalytic nanocompartments
Maria Korpidou, Viviana Maffeis, Ionel Adrian Dinu, Cora-Ann Schoenenberger, Wolfgang P. Meier, and Cornelia G. Palivan

[11] Tough Double Network Granular Hydrogels
Tianyu Yuan and Esther Amstad

[12] Bottom-up cell mimicry: ATP synthesis in giant polymersomes formed by microfluidics
Olivia Eggenberger

[13] Extensional Rheology of Mucin-Particle Networks
Caroline E. Giacomin, Dimitra Founta, Dimitri Vlassopoulos, and Peter Fischer

[14] Photoreversible Resins For Digitally Printed Protective Films
Morris Wolf and Mark W. Tibbitt

[15] Probing small molecule uptake with planar polymer membranes
Moritz Muthwill, Flavien Sciortino, Riccardo Wehr, Saziye Yorulmaz Avsar, Daria Sokolova, Konrad Tiefenbacher, and Cornelia Palivan

[16] Illuminating the structure of iron carbohydrates in complex biological environments
Leonard Krupnik, Neda I. Anaraki, Marianne Liebi, Jonathan Avaro, Joachim Kohlbrecher, Antonia Neels, and Peter Wick

[17] Controlling expansion dynamics in bacterial biofilms through modulation of extracellular polymeric matrix components
Sam Charlton, Dorothee Kurz, Steffen Geisel, Joaquin Jimenez-Martinez, and Eleonora Secchi

[18] Light Scattering of Micron-sized Self-assembled Colloidal Aggregates
Pavel Yazhgur, Geoffroy J. Aubry, Luis S. Froufe-Pérez, Nicolas Muller, and Frank Scheffold

[19] Host–guest-based Polymer–Nanoparticle Hydrogels for Modular Materials Design
Stéphane Bernhard, Marco Müller, Giovanni Bovone, Elia A. Guzzi, Wenqing Guo, and Mark W. Tibbitt

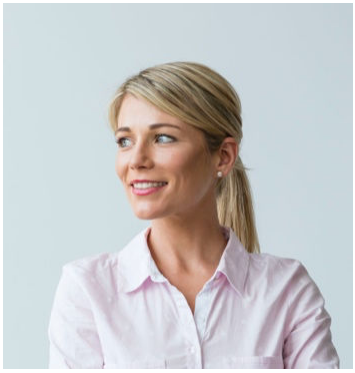
[20] Entropy links molecular and macroscopic behavior in dynamic covalent networks
Lucien Cousin, Bruno Marco-Dufort, Mark W. Tibbitt
[21] iSCORR a new technique using interference scattering microscope to enhance rheology techniques
Raphaël P.B. Jacquat, Quentin A.E. Peter, Georg Kreiner, Jan Heck, Tuomas P.J. Knowles, and Paolo Arosio

Invited Speakers:

Engineering soft microrobots for drug delivery and local detection of pathological signals

Prof. Simone Schuerle, ETH Zurich

Site specific diagnostics and effective, localized therapy delivery remain challenge tasks in today's medicine. To address this need, my laboratory develops micro- and nanoscale systems that respond to disease-specific biochemical cues or non-invasive external stimuli like magnetic fields such that they focus their action at the site of disease. In this talk, I describe a synthetically engineered soft microrobot that reports information about molecular activity at a disease site via acoustic fields. Moreover, I will show how swarms of soft living magnetic microrobots can help to locally enhance transport of nanodrug shuttles to tumor tissues, and how they can be further engineered to function as controllable therapeutic vectors.



Bio: Simone Schuerle is assistant professor at ETH Zurich, Switzerland, where she heads the Responsive Biomedical System Lab. With her team, she develops diagnostic and therapeutic systems at the nano-and microscale with the aim of tackling a range of challenging problems in medicine. Prior to taking this position, she researched at MIT on nanosensors for *in vivo* tumor profiling as well methods to wirelessly enhance drug transport (2014-2017). She is recipient of several awards, such as the Prix Zonta in 2019 for Women in Science, and fellowships from the SNSF, DAAD and Branco Weiss foundation, and was honored with the distinction of "Young Scientist" by the World Economic Forum (WEF) for her scientific contributions to society. In 2014 she co-founded the spin-off MagnebotiX that offers electromagnetic control systems for wireless micromanipulation. She earned her PhD degree with specialization in microrobotics in 2013 at ETHZ and a masters in industrial engineering with specialization on microsystems and nanotechnology at the Karlsruhe Institute of Technology in Germany.

Microfluidic Platforms for Studying Surfactant-laden Interfaces and Polymer Solutions

Cari Dutcher, University of Minnesota

In this talk, recent advancements in use of microscale flow fields will be highlighted for studies of aqueous multiphase, interface-rich systems. Microfluidic contractions, traps, and cross-slots are used to measure thermodynamic and material properties of surfactant-laden interfaces and polymer solutions. Dynamic interfacial tensions measurements were performed using a microfluidic tensiometer, demonstrating a dependence on if the surfactant approaches the interface from inside (dispersed) versus outside (continuous), implying phase dependent surfactant transport to curved interfaces at the microscale. Droplet coalescence and film drainage experiments are also performed in a microfluidic Stokes trap across a range of viscosity ratios and surfactant concentrations. Finally, microfluidic geometries will be used to characterize the extensional properties of low molecular weight, low viscosity polymer and polyelectrolyte solutions. Specifically, filament stretching using a cross-slot microfluidic channel will be used to resolve extensional properties of polyelectrolyte solutions at varying salt concentrations.

Bio: Cari S. Dutcher is an Associate Professor of Mechanical Engineering (ME) and Chemical Engineering and Materials Science (CEMS) at the University of Minnesota, Twin Cities. Her research interests are in complex fluids and multiphase fluids, including aerosols, emulsions, and foams. Cari currently serves on the Executive Board of the American Association of Aerosol Research (AAAR) and Editorial Board of Aerosol Science and Technology. Since starting her faculty position in 2013, Cari has received the 3M Non-Tenured Faculty Award, NSF CAREER Award, AAAR Kenneth T. Whitby Award, George Taylor Career Development Award, and SERDP WP Project of the Year. Cari received her B.S from Illinois Institute of Technology (2004) and her Ph.D. from the University of California, Berkeley (2009), both in Chemical Engineering.

Talks:

Hierarchical Porous Scaffolds for Culturing Complex Tissues: the Scattering of Foams

Chaimaa FIKRY; Carlo ANTONINI; Viviane LUTZ-BUENO

In materials science, structure spans over different length-scales, and determines the macroscopic properties and functionalities of hierarchical materials. Similarly, the structural hierarchy of soft biological tissues define their function and performance. During growth, cells can interact, and adapt to their 3D environment, but are incapable of self-assembling over the macroscopic length-scales of complex tissues. Consequently, cells during in vitro culture are guided towards their macroscale assembly by 3D scaffolds that mimic the extracellular matrix (ECM) with appropriate structural, mechanical and chemical signaling at the nanoscale. Porous structures, such as foams, that can trap cells in their porosity, are exploited for bone-mimetics, but are rarely considered for soft tissue engineering, where hydrogels are the norm. However, hydrogels' structure is mainly isotropic, and the trapped cells only grow into small aggregates without hierarchy. Therefore, we optimize the 3D porous scaffolds, as a foam structure to enable the growth of complex soft tissues by tuning their formulation, processing and structuring at the nano- and mesoscale. These foams will support 3D cell culture of complex tissues, such as skeletal muscle, focusing primarily on ideal tissue structure rather than on functionality, being fundamental in the fields of medical, pharmaceutical, biological and even food sciences, due to the current rise of artificial meat.

Structural characterization correlates the material composition and organization to its macroscopic properties. Here, we combine small X-ray and neutron scattering to investigate ultraporous and lightweight biopolymer-based foams using cellulose nanofibrils (CNFs), as building blocks. We collect spatially resolved maps at the macroscale (info on foam density and porosity), at the nanoscale (info on foam structural compactness, orientation of the foam walls, and packing state), and at the molecular scale (info cellulose on crystallite dimensions and protein conformation). Specifically, we compare the impact of freeze–thawing–drying (FTD) fabrication steps, such as static/stirred freezing and thawing in ethanol/water, on foam structural hierarchy spanning from the molecular to the millimeter scale. As such, we demonstrate the potential of small angle scattering for hierarchical characterization of biopolymer foams [1].

Furthermore, we investigate the development and structure of freezing fronts of CNF suspensions in real time with neutron imaging. When freeze-dried, CNF suspensions form high-porosity foams, which often have regions of different densities, resulting in structural inhomogeneities and defects. Understanding how the freezing front advances in the presence of particles, and how it influences the porosity and structure of the foam is necessary for prescriptive analysis of freeze-drying, as it dictates not only the structure of the foam, but also the freezing velocity, time, and overall energy consumption of the process. By using "fractional freezing", we differentiate the solidified freezing front from the remaining liquid solution, using the contrast of hydrogenated solutes in deuterium oxide. These findings serve as a basis for the development of predictive models for the structures produced by freeze-drying, which are then employed for cell scaffolding.

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Bioactive Polymersomes Targeting Cancer-Associated Fibroblasts for Anticancer Therapy

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Keywords: Targeting; Nanoreactor; Anticancer Treatment

Abstract:

Despite constant development of anticancer therapies cancer remains a leading cause of mortality. Modern chemotherapies, as a main tool in anticancer treatment, have numerous side effects due to their lack of specificity or systemic toxicity. Innovative solutions in the field of nanomedicine can contribute to the development of more precise and effective therapeutics. Recently, the fibroblastic element in tumor microenvironments emerged as a valuable target for treatment and diagnosis of different types of cancer. Herein, we decorated polymersomes with a bioactive ligand to target fibroblast activation protein α (FAP). By shielding their content from their surrounding, polymersomes favor lower immunogenicity and prolonged activity. Polymersomes were prepared by the self-assembly of the amphiphilic diblock copolymer poly(dimethylsiloxane)-*block*-poly(2-methyl-2- oxazoline)(PDMS-*b*-PMOXA) followed by decoration with a bioactive ligand. The cellular uptake of decorated polymersomes in contrast to unmodified polymersomes significantly increased in a FAP positive human adenocarcinoma cell line (A549). These polymersomes display the ability to escape endosomes proving their potential as an efficient delivery carrier (Figure 1). Additionally, the insertion of the honeybee venom peptide melittin into the membrane offers stable pores to allow diffusion for a functional nanoreactor. Such decorated nanoreactors might prove their potential as an efficient delivery carrier.

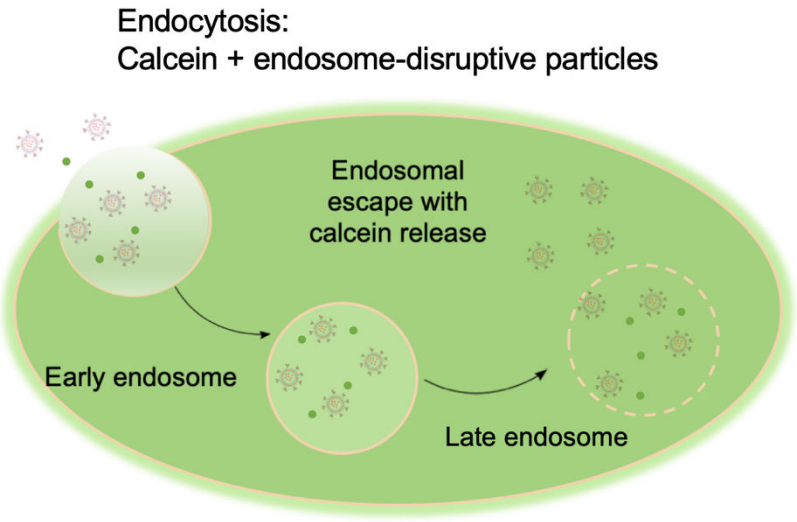

Figure 1: Endosomal escape of polymersomes decorated with a bioactive ligand.

Bacterial transport in polymer solutions

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Whether in environmental, medical, or biological settings, bacteria often live surrounded by fluid media, which are often in flow. In such habitats, trapping of bacteria in high-shear regions can arise from the interaction between fluid shear and cell motility. This interaction was shown to alter the spatial distribution of motile bacteria and the location and efficiency of surface colonization [1,2,3]. Although bacterial cells are often found in polymer fluids, bacterial transport and shear trapping have only been characterized in water [1]. Despite the presence of Newtonian and non-Newtonian polymer solutions in many bacterial habitats, the effect of their complex rheology on shear-induced trapping and bacterial transport in flow has remained unexplored.

Using microfluidic experiments and numerical simulations, we study how Newtonian and non-Newtonian polymer solutions affect the transport of motile, wild-type *Pseudomonas aeruginosa* in a Poiseuille flow [4]. Our results show that both bacterial swimming and polymer fluid rheology control the magnitude of shear-induced trapping, with potential consequences on surface colonization and biofilm formation in many relevant microbial habitats.

Acknowledgments

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Stable colors: how order and scale geometry make the colourful displays of *Sternotomini* longhorn beetles angle-independent

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Photonic nanostructures can vary in their degree of local order and their final optical appearance is often further tuned by pigments. Longhorn beetles display vivid colours and rely on varying degrees of (dis)order combined with pigments to create complex colour patterns via coloured scales that adorn the insects' bodies. Within the *Sternotomini* tribe, we found angle-independent colours ranging from blue to orange that originate from light interaction with elytral scales containing photonic crystals with difference in pigment content and scale shape. We employed light microscopy, FIB-SEM tomography and full-wave optical simulations (FDTD) to investigate the optical appearance and underlaying mechanisms in two *Sternotomini* subspecies. Green-blue/orange *Sternotomis amabilis* ssp. *sylvia* beetles (Fig. 1D) show angle-independent green-blue hues based on polycrystalline networks of recently discovered body-centred cubic (I-WP) photonic crystals (Fig. 1F) [1], combined with diffuser-like scale structures atop. The beetles' orange areas are due to scales that contain amorphous, quasi-ordered photonic networks (Fig. 1E) which produce angle-independent orange colours, where the diffuser-like scale structure plays a minor role. More importantly, we report that the non-iridescent green stripes and blue feet of *Sternotomis virescens* beetles (Fig. 1C) are produced by amorphous photonic crystals (Fig. 1A-B) with subunits with I-WP unit cells. Thus, we classify these networks in analogy to amorphous photonic diamond structures [2] as amorphous I-WP (AI-WP) photonic crystals. This work aims to illustrate the complex interplay of structural and pigmentary colour in longhorn beetles, and illuminate how angle-independence is achieved in different longhorn beetle subspecies.

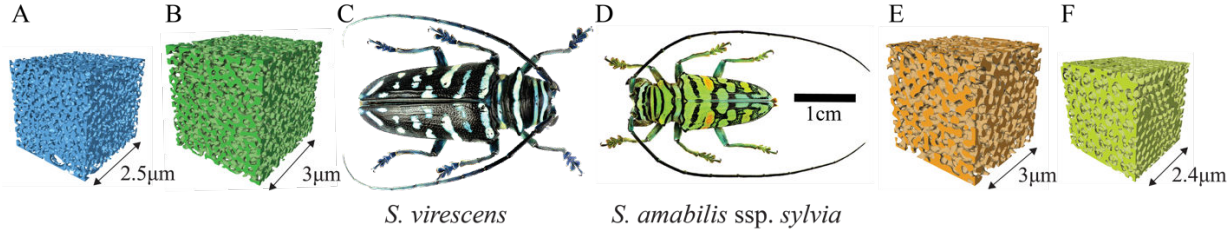

Figure 1. AI-WP photonic networks producing non-iridescent blue, **A**, and green, **B**, in *Sternotomis virescens* longhorn beetles, **C** [3]. **D** Photograph of a *Sternotomis amabilis* ssp. *sylvia* longhorn beetle [3], and corresponding amorphous, quasi-ordered and I-WP networks producing orange, **E**, and green-blue colors, **F**, respectively.

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Mechanical response of dense amorphous materials: from passive particle systems to active matter

Elisabeth Agoritsas

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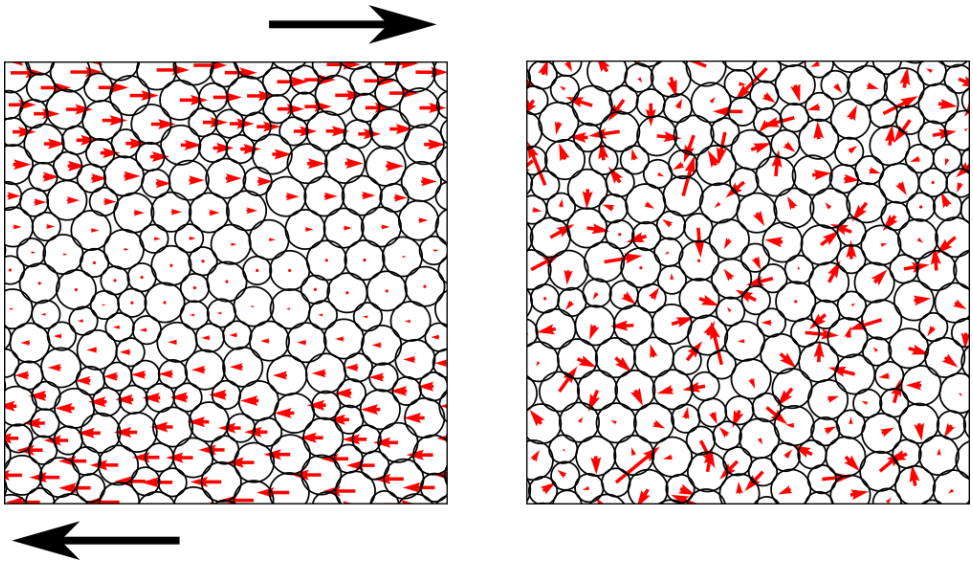
Amorphous materials are ubiquitous around us: emulsions as mayonnaise, foams, sandpiles or biological tissues are all structurally disordered, and this has key implications for their response to an external deformation. Nevertheless, theoretical descriptions of such 'driven' amorphous materials remain challenging, despite of decades of extensive analytical and computational studies. The difficulties pertain to the interplay of competing sources of stochasticity, and to the resulting out-of-equilibrium nature of these systems. A standard model for amorphous materials, which allows to focus on the key role of their structural (positional) disorder, is provided by dense many-body systems of pairwise interacting particles.

Furthermore, there has been recently many attempts to relate the important corpus of known results for such 'passive' amorphous materials, and their counterparts in active matter such as confluent biological tissues. One strong motivation is that the interplay between activity and structural disorder might in turn be related to biological functionalities.

Here I will discuss recent results on the exact mean-field dynamics of these many-body systems, that we have derived in the limit of infinite spatial dimension, for different driving protocols. We were in particular able to establish a direct equivalence between a global forcing (external shear) and a random local forcing (reminiscent of active matter), upon a simple rescaling of the control parameter (the accumulated strain). In this framework, global shear is thus simply a special case of a much broader family of local forcing, that can be explored by tuning its spatial correlations. Our predictions were moreover found to be in remarkably good agreement with two-dimensional numerical simulations. These results hint at a unifying framework for establishing rigorous analogies, at the mean- field level, between different families driven disordered systems, such as sheared granular materials and active matter.



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Surface stress drives large material deformation to smooth compliant solids

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With the increasing use of compliant materials in engineering applications, it becomes crucial to include effects of surface stress in our understanding of the interfacial mechanics of compliant solid structures. Several studies have investigated how surface stress deforms compliant solids' shapes and results in rounding and flattening of sharp features due to the relatively low elastic modulus [1, 2, 3]. Previous experiments have been largely limited to the two-dimensional (2D) and nearly planar case, studying rectangular ridges of height h_0 and initial width w_0, where h_0/w_0 is small ($h_0/w_0 << 1$). We introduce a microfabrication technique to produce three-dimensional (3D) compliant structures having non-negligible aspect ratio ($h_0/w_0 \sim 1$) and experimentally demonstrate that micropillar grids made of a compliant polymer in contact with air undergo large deformation at different spacings, where deformations are largest at low spacings and reach a constant value at large spacings. This spacing-dependent behavior suggests an elastocapillary interaction between the structures at small spacing, whereas the asymptotic value suggests a limiting distance over which long-range interactions between pillars apply.

The analytical solution provided by Hui et al. [4] can be used to theoretically calculate deformation when the height-to-width aspect ratio of micropillars is small. In high h_0/w_0 cases, this solution appears to underpredict deformation and no longer holds. Therefore, we numerically investigate the large deformation caused by surface stress using a 3D finite element modeling analysis (FEM) [5]. Our FEM results for high-altitude geometry are highly consistent with experimental measurements of pillar profiles and show large deformation and rotation within the material. Unlike the theoretical prediction, which underestimates the peak-to-valley distance of the deformed pillar shapes for large spacing, FEM can correctly predict this asymptotic value. Finite element analysis can also offer intriguing insights into the problem, such as predicting stress field maps and investigating the competition between internal and surface energy.

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Measuring Rolling Friction of Microparticles using Lateral Force Microscopy

Simon Scherrer [1], Shivaprakash N. Ramakrishna[1], and Lucio Isa [1]

[1] Laboratory for Soft Materials and Interfaces, ETH Zürich

The introduction of colloidal probes has expanded the capabilities of atomic force microscopes (AFMs) to allow measuring interactions between particles and surfaces directly. In particular, lateral force microscopy (LFM) provides access to friction coefficient measurements at the nanoscale by determining lateral forces between surfaces as a function of applied load. Typically, experiments are limited to AFM cantilevers with fixed colloids, and thus only enable measuring sliding friction. However, for unconstrained colloidal particles in a fluid, sliding as well as rolling motion is present, but the characterization of the latter remains elusive.

Using two-photon nanolithography, we have developed a colloidal probe that allows free rotation of an encapsulated particle and simultaneous imaging of the contact via fluorescence microscopy. The colloid releases from the cavity when the probe is not in contact with the substrate, making it reusable. Upon lateral spring constant calibration of the cantilever [1], a friction coefficient of the investigated colloid-substrate system can be determined. The three-dimensional rotation of the particle within the cantilever is confirmed by tracking fluorescent markers on the colloid, while simultaneously obtaining the lateral forces acting on it. Direct comparison to a fixed colloidal probe is possible by gluing the particle inside the cavity, therefore restricting any rotational movement, and enabling the observation of sliding and rolling friction.

We have first applied our method to a model system of rough, 12 μm silica particles and surfaces with matching asperities [2] and measured friction coefficients of rotating and fixed colloids in HEPES buffer, where the latter is significantly higher. Analysis of the rotational motion correlated with specific features in the friction force signals gives new insights into the behavior of contacting colloids that can be extended to a broad class of systems including the rheology of dense suspensions.

Contact events determine macroscopic rheological behavior such as thinning and thickening, which could be explained by a better understanding of the individual particle motion.

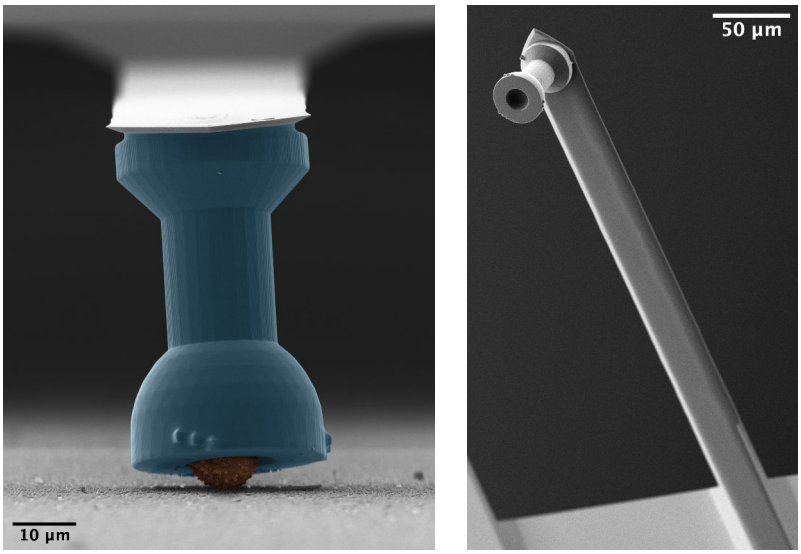

Figure 1. *False colored SEM images of the colloidal probe (blue) with rough silica particle (red).*

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Multicomponent solid-supported membranes based on amphiphilic block copolymers mixtures

Maryame Bina, Agata Krywko-Cendrowska, Wolfgang Meier, Cornelia Palivan

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Nanotextured surfaces are widely found in nature and represent an important class of materials as they display versatile properties such as superhydrophobicity, bacteria repellence, exceptional adhesion or structural colours. These textured surfaces are usually engineered through physical or chemical patterning e.g. *via* laser ablation or chemical vapor deposition respectively. These are however expensive techniques and require specialized equipment. Here, we present a biomimetic approach in which the formation of a patterned surface occurs by using the amphiphilic property of dissimilar block copolymers as the driving force for their self-assembly,[1] comparably to biological membranes formed by lipids. However, contrary to naturally occurring lipids, polymers offer good mechanical stability and chemical versatility thus allowing for the fine tuning of their properties. The obtained films in the form of mono- and bilayers are composed of PEO_{45}-*b*-$PEHOx_{20}$ and $PMOXA_{10}$-*b*-$PDMS_{25}$ as the two amphiphilic diblock copolymers mixed at various concentrations and displaying distinct properties. The solid-supported fully synthetic planar membranes, undergo phase separation into domains embedded within a continuous phase. The molar ratio of the copolymers in the mixture and the nature of the solid support were the key parameters to induce nanoscale phase separation of the planar membranes. Such soft nanotextured membranes taking inspiration from the domains in cell membranes open new avenues for simultaneous and controlled combination with active compounds such as proteins, drugs, catalysts to engineer multifunctional surfaces with nanoscale texture.

Keywords: multifunctional surfaces, textured surfaces, biosensor, amphiphilic diblock copolymer, self-assembly

Reference:

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2 functionalities -in-1 : Embedded 3D printing of alginate into a SiO_2-$CaCl_2$ gel bath

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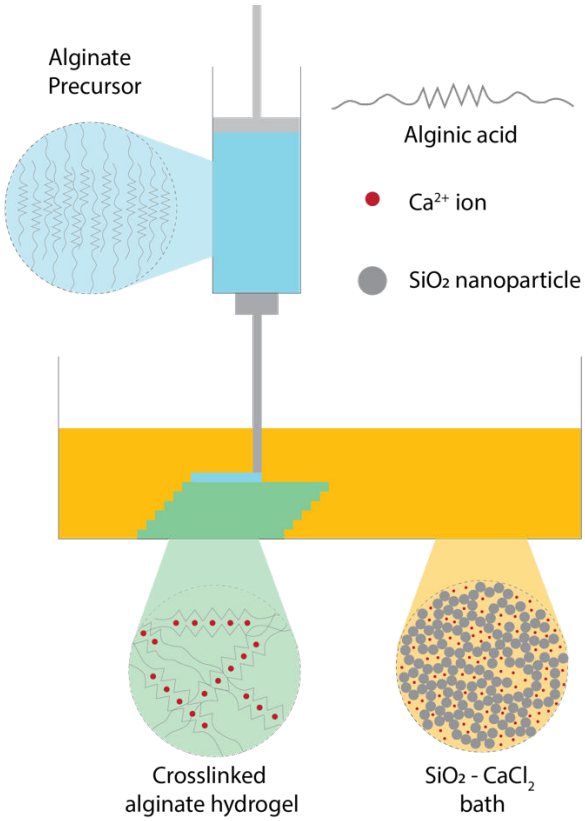
Extrusion 3D printing of soft materials is often limited in terms of shape, structure, size, and orientation due to printability constraints. One way to increase printability and shape fidelity is the use of a support bath, which will hold the extruded material in place until it reaches its final state of solidification. Here, we propose a 2-in-1 embedded printing approach, where the support bath gives stability to the printed structure and serves as a crosslinking agent at the same time. The approach is to print an alginate solution into an aqueous colloidal gel of Ludox SiO_2 particles and $CaCl_2$. The colloidal gel first serves as a support structure to the liquid alginate, while the $CaCl_2$ ions in the gel crosslink the alginate. Due to a very quick crosslinking of the outer layer of the alginate and outer mechanical support, it is possible to print more elaborate and even overhanging structures. This work first identifies the operating windows of this printing process in terms of material and system properties, using dimensional analysis. In terms of material properties, the primary parameter that can be tuned, are the mechanical properties of the support bath, by changing the SiO_2 and the $CaCl_2$ concentration – this also impacts the gelation time which impacts system properties such as the extrusion rate and printing velocity. In a second step, the optimized parameters are applied to fabricate various freeform architectures.



Surface-Initiated Polymerization From Lipid Membranes

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Abstract: Polymer brushes attached to lipid membranes are essential in biology and medicine, from the glycocalyx layer coating most cell membranes to PEG chains stabilizing liposomes for drug delivery. However, the synthetic methods commonly used to prepare decorated membranes typically rely on grafting pre-polymerized chains to a lipid bilayer. This strongly limits the surface density of polymer chains, keeping larger thicknesses and higher densities out of reach. Instead, we aim to grow polymer brushes of controlled density and thickness directly from the lipid membrane. To this end, we incorporate a novel controlled radical polymerization initiator into phospholipid bilayers, and initiate the polymerization of N-isopropylacrylamide (NIPAM) asymmetrically from their surface. We quantify the polymerization from supported lipid bilayers (SLBs) using quartz-crystal microbalance with dissipation monitoring (QCM-D), and from small unilamellar vesicles (SUVs) using dynamic light scattering (DLS). In both cases, the growth of a polymer layer is observed, whose thickness strongly correlates with the concentration of initiator in the membrane. SUV-initiated polymerizations run for longer and yield thicker brushes than their SLB counterparts, which we attribute to the difference in membrane curvature between these systems. Non-monotonic evolution of the brush thickness suggests degrafting at high surface densities. We expect that the exceptionally dense and thick polymer brushes obtained with this route will have strong effects on the membranes' stability and mechanical properties.

Soft ceramics at the nanoscale: giant elastic strain and electrically sensitive shape-memory effect

Xiang-Zhong Chen, Donghoon Kim, Minsoo Kim, Bradley Nelson, and Salvador Pané

Multi-Scale Robotics Lab, Department of Mechanical and Process Engineering, ETH Zurich

The rapid development in flexible electronics and soft robotics posed an urgent need for novel electronic and actuating materials. While flexible functional organic materials have been developed, many of their functional performances are still inferior to those of inorganic materials such as ceramics. However, inorganic materials are usually considered to be incompatible with flexible and soft devices due to their rigid nature. Yet, when a material shrinks its size down to the nanoscale, interesting mechanical properties emerge.

In this talk, I would like to present our recent findings on several peculiar mechanical behaviors in freestanding ceramic nanometer-thick microstructures that consist of epitaxial bilayers of magnetic cobalt ferrite and piezoelectric barium titanate. Contrary to their bulk counterparts, these films are flexible, superelastic, and even exhibit shape-memory features. Besides, their magneto-electric properties can also be continuously tuned thanks to their high endurance of the considerable strain.

Our results will potentially provide a way of developing novel ceramic-based high-performance flexible sensing units, memory elements, and actuating devices for flexible electronics and soft robots.

Posters:

[1]:

Unravelling the Mechanical Potential of Bioinformed Granular Hydrogels

Matteo Hirsch, Livia D'Onofrio, Esther Amstad.

3D printed hydrogels are increasingly often used as scaffolds for tissue engineering. However, despite tremendous progress in the formulation of hydrogel inks and their processing, it is still difficult to control their local composition. We recently demonstrated that the local composition of load-bearing hydrogels can be abruptly changed if inks are composed of microparticles with different compositions.1 However, these hydrogels are rather soft such that the range of mechanical properties that is attainable with them is limited. Inspired by nature, we increase the mechanical performance of these hydrogels by further reinforcing them with ions, through a metal coordination approach. By varying ion species and their relative concentration, we are able to tune the mechanical properties in a wide range, where the stiffest granular hydrogels display a Young's modulus up to 5 MPa. Additionally, the intrinsic spatial control of granular materials allows to easily control ions localization, thus fabricating materials with unique mechanical performance, such as core-shell structures displaying a clear yielding behavior and possessing fracture toughness as high as 8 MJ/m3. We believe that this versatility is key to bridging the gap between natural and soft materials.

[2]:

Assembling Catalytic Nanocompartments into Artificial Signaling Cascades

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The spatiotemporal separation of biochemical processes by compartmentalization plays a fundamental role in nature. Reproducing, but also manipulating hierarchically organized compartments with regard to their responsiveness and communication provides crucial information towards understanding biological systems. In the work presented, we highlight the preparation and characterization of catalytic nanocompartments (CNCs) that consist of polymeric nano-assemblies encapsulating active compounds (enzymes, proteins, catalysts, mimics) and explore their dual role in protecting the internal compounds from proteolytic attack and providing confined spaces for complex metabolic reactions. Above all, we used artificial nano-compartments to expand the cell's metabolic repertoire with a unique exogenous pathway. This extracellular "gain of function" is based on a robust decoration of cell membranes with complex "artificial organelles", which endow cells with non-native catalytic activity.1 The combinatorial und functional diversity of CNCs assembled into various supramolecular architectures can be further exploited either in suspension or immobilized on a surface, whereby surface immobilization offers the advantage of highly controlled spatial organization. As an example, we developed CNC-functionalized DNA microarrays where individual reaction compartments are kept in close proximity by a distinct geometrical arrangement to promote effective communication.2 Our data show that polymer-based nano-compartment assemblies offer an ideal scaffold for the development of the next generation responsive and communicative soft-matter analytical devices for applications in catalysis and medicine.

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[3]:

Hard-Soft Assembly Clusters for Bio-Applications

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The self-organization of nano-objects into complex architectures is a major strategy to produce novel systems with emerging properties and functionalities in fields such as chemistry, electronics and technology[1–3].
The self-organization of clusters between "hard" Janus nanoparticles (JNPs) and "soft" polymersomes represents a new approach for developing a multifunctional hybrid system for specific bio-applications. These polymer-based JNPs with anisotropic composition and orthogonally addressable functionality provide an asymmetric platform suited for directional interaction[4,5] with the soft polymersomes. The hybridization of complementary ssDNA strands attached to each component links them into clusters. The polymersomes are deformed upon adhesion to the "hard" JNPs surface but maintain their integrity, thanks to the inherent mechanical robustness of the block copolymer membrane. Importantly, the continued integrity of the vesicular architecture of polymersomes after assembly into JNP-polymersome clusters offers the possibility of encapsulating various kinds of functional cargo. Finally, the biocompatibility of the clusters and their interactions with cell surfaces, mediated by scavenger receptors, was investigated.

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[4]:

Zero-shear viscosity for yield stress fluids

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Simple yield-stress fluids exhibit a transition between a solid-like and a fluid-like behaviour at a precise stress [1]. However, the real existence of this critical transition is debated in the field literature since the origin of rheology. Several authors argue that these materials have a zero-shear viscosity several order of magnitude higher that the value at high shear rate [2]. Others dispute these findings and explain the appearance of a zero-shear viscosity with the presence of experimental time constrain [3].

In this work we apply to these materials a modified Maxwell model with a stress activated viscosity, previously developed to describe polymers behaviour [4]. In particular, a Carbopol dispersion is used to validate the model, carefully prepared according to Varges et al. [5] to avoid thixotropic effects.

Several rheological techniques were implemented in this study. Creep data were acquired both in the plastic regime and in the viscous regime, identifying how the material deform under different stresses. Orthogonal superposition (OSP) curves show the fluidization of the material with increasing stress, pointing out an actual fastening of the relaxation time. Two Mastercurve where generated from creep and OSP data, generalizing the observed behaviour to stresses where the steady state was not achievable. Start-up experiments were compared to the model prediction, with a good agreement in the steady state, Figure 1. In the end a flow-curve was composed from all the steady state measured or extrapolated, revealing a zero-shear viscosity and the absence of a critical yield-stress, Figure 2.

Figure 2 Start-up experiments (dots) and model predictions (lines) for different imposed shear rate. The model nicely predict the experimental steady state values.

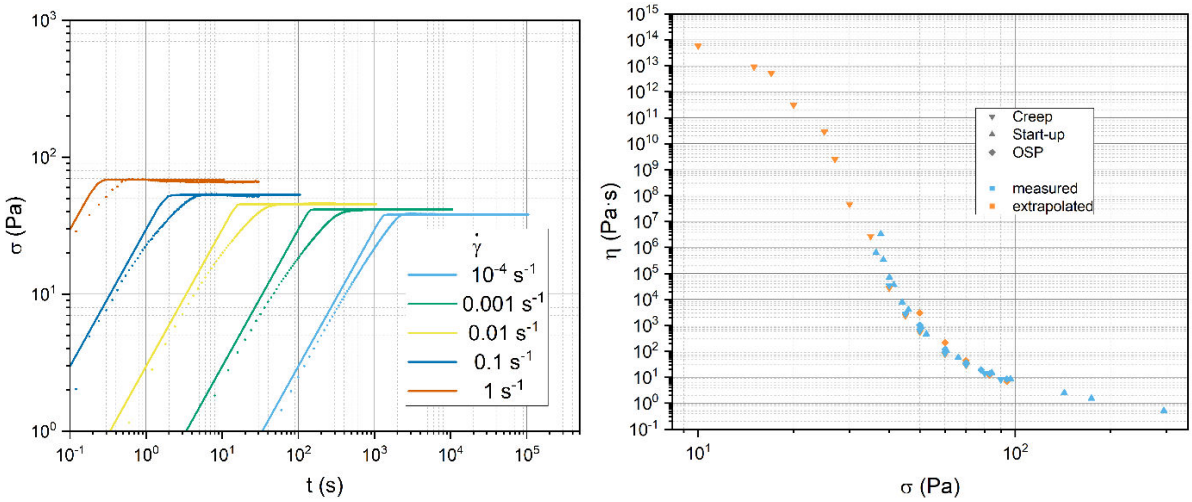

Figure 2 Flow-curve created by both experimental data (light blue) and extrapolated steady state from the Mastercurve (orange). No strong for yield-stress is visible.

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[5]:

Thermo-responsive nanocellulose hydrogels as a universal drug release platform

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Qiyao Sun[1], Garam Han[1], Luca Müller[2], Giovanni Bovone[3], Siyuan Tao[4], Pascal Bertsch[5], Mark Tibbitt[3], Qun Ren[4], Gilberto Siqueria[2] and Peter Fischer[1]*

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[2] Cellulose & Wood Materials Laboratory, Empa
[3] Macromolecular Engineering Laboratory, D-MAVT, ETH Zurich
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Hydrogels are a particularly applealing type of drug delivery system and have been used in many branches of medicines and tissues. The mechanical property is the key to its ability to maintain the structure and avoid fracture during use and after placing *in vivo*. To fulfill the requirements for various applications, a toolbox of biocompatible thermoreponsive hydrogel with tunable mechanical strength is established, from a liquid injectable hydrogel to a solid hydrogel. Thermo-responsive polymer PNIPAM (Poly(N-isopropylacrylamide)) was cross-linked at different degrees and reinforced by cellulose nanocrystals (CNCs) and cellulose nanofibrils (CNFs) to obtain different mechanical properties. Model therapeutic agents with different physiochemical properties were explored for *in-vitro* locally targeted sustained drug release in the liquid injectable hydrogel. The different release patterns are attributed to drug size, hydrophilicity and specific drug-cellulose interactions. Antimicrobial agents were included in solid form hydrogels for the purpose of a wound dressing patch. Interactions between the hydrogel and antimicalbial agents were thoroughly studied to obtain stable complex. Broad-spectrum anti-microbial activity is comfirmed aginst Gram-negative and Gram-positive bacteria as well as fungus. This work expands the application of cellulose in the biomedical field by enabling well-defined hybrid biomaterials with control over hydrogel mechanical property and drug release behavior.

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[6]:

What is the impact of phase separation in elastic polymer networks?

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With this project, we aim to create a new generation of bio-inspired materials based one lastically arrested phase separation. Arrested phase separation is a key mechanism also used by living systems to create well-controlled nanostructures. One of the most salient example is structural color as present in some bird and insect species. Next to fascinating optical properties, phase-separating composite systems can have astonishing mechanical properties: For example, liquid inclusions can stiffen polymer networks. These mechanical properties have been less studied compared to the optical properties. We probe for dissipated energy and elastic modulus depending on phase separation and liquid content of the system. With these experiments we can disentangle contributions from composition and structure to the mechanics of these systems.

[7]:

Selectively permeable viscoelastic composite microcapsules

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Microcapsules with a selectively permeable shell are attractive containers for sensing, microreactors, and synthetic cells. Despite the wide application range, it remains a challenge to develop mechanically stable microcompartments that are capable of repetitively and selectively uptake and release the cargo. Inspired by the mussel byssus' coordinated chemistry, we are developing surfactants that are functionalized with chelators such that they can be crosslinked with appropriate ions or nanoparticles (NPs). The resulting capsules possess a charge-selective permeability that enables the selective uptake of certain molecules and their up-concentration within the capsule core. The release of reagents can be repetitively triggered by cycling the pH. This platform opens up new possibilities to tune the permeability and functionality of viscoelastic capsules with crosslinking nanoparticles. Thus, these capsules have the potential used in wastewater treatment or catalysis.

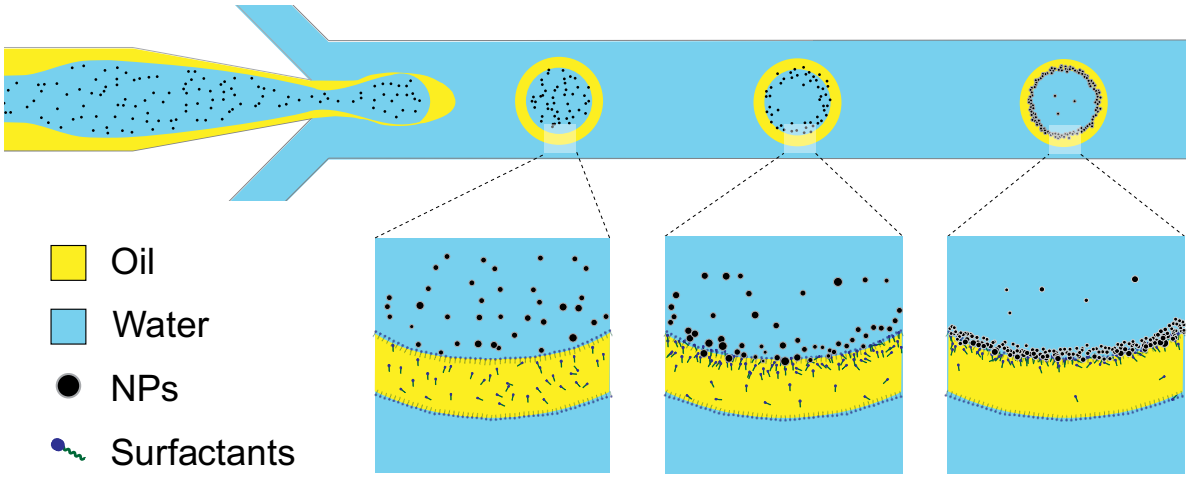

Figure 1. Water-oil-water double emulsions are converted into microcapsules through interfacial assembly of surfactants that are interlinked through metal oxide nanoparticles.

[8]:

Polymeric Giant Unilamellar Vesicles for Bacterial Culture

Lukas Heuberger and Cornelia G. Palivan

Department of Chemistry, University of Basel

Giant unilamellar vesicles (GUVs) are micrometer-sized vesicles that are applied as artificial compartments and can mimic cells.[1] Amphiphilic block copolymers can be synthesized with a wide range of compositions, block ratios, and functionalization and serve as excellent alternatives to lipids for vesicle formation. Double emulsion microfluidics can produce GUVs at high-throughput.[2] The modularity and design of this system facilitates the formation of monodisperse GUVs with controllable inner, polymer membrane, and membrane functionalization.[3] Employing high-throughput methodologies, GUVs can be applied for screening assays. By adjusting the inner composition, enzymatic cascades, smaller vesicles, organelles, and entire living organisms can be encapsulated within GUVs.

In the presented work, GUVs were produced using the diblock copolymer PDMS-*b*-PMOXA. Thanks to their size being comparable to cells, GUVs can be screened using flow cytometry. Using these techniques, tens of thousands of GUVs can be analyzed, making them a convenient alternative to w/o/w double emulsions. This allows for their applications as microreactors or cellular models. When permeabilizing their otherwise impermeable membranes, GUVs can be employed as incubation chambers for the growth of bacteria. This approach underlines the versatility of double emulsion templated polymer GUVs for studying compartmentalized biological systems, deepening the insights into fundamental biological processes.

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[9]:

Multiplexed Analyte Detection in Droplet Interface Bilayers

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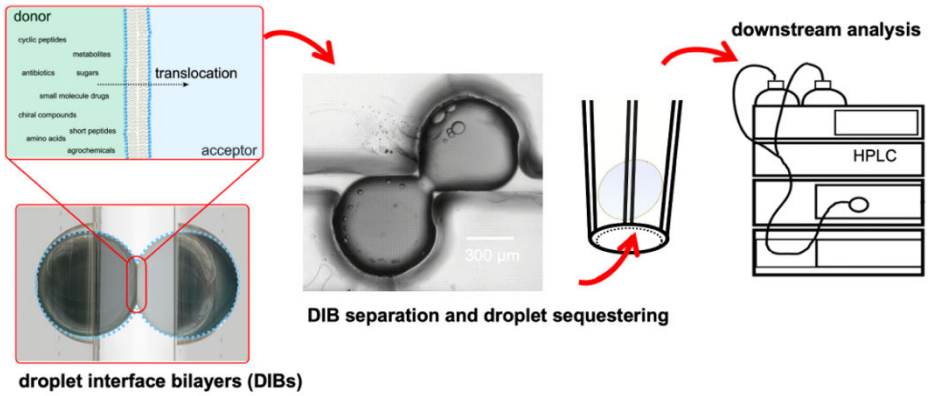
Abstract: Droplet interface bilayers (DIBs) are a powerful bottom-up synthetic biology technique for *in vitro* membrane formation.[1] These biomimetic membranes enable investigation of critical biological membrane features, such as diverse lipid types,[2] lipid asymmetry,[3] and membrane proteins.[4] In high nL volume DIBs, analytical techniques such as electrophysiology, UV and fluorescence spectroscopy can be performed.[5] At this scale, shear forces can be utilized to manipulate droplets for membrane formation and separation.

In this work, we have developed upon a platform for DIB separation and droplet sequestering and coupled this to a downstream high performance liquid chromatography (HPLC) measurement.[6] DIBs were formed with a cocktail of biologically relevant compounds broadly mimicking drug mixtures and molecular crowding, constituting a simple screen for assessing membrane permeable chemical space. The method can be modified for a simultaneous quantification of rate constants for the membrane permeable compounds within the mixture. With this strategy, we can enhance DIB models of biological transport phenomena by systematically adjusting detectable chemical complexity in volumes separated by membranes.

Graphical abstract:



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Inverting glucuronidation of hymecromone *in situ* by catalytic nanocompartments

Maria Korpidou [1], Viviana Maffeis [1,2], Ionel Adrian Dinu [1,2], Cora-Ann Schoenenberger [1,2], Wolfgang P. Meier [1, 2] †, Cornelia G. Palivan [1,2]

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Glucuronidation is a metabolic process that conjugates molecules such as drugs to the glucuronic acid, and ultimately lead to their excretion and elimination from the body (1). When this process takes place rapidly, the half-lives of molecules become shorter and thus the therapeutic effects of drugs wear off quickly. One strategy to overcome this limitation is administering higher or more frequent doses of the therapeutic compound, which could however lead to drug abuse or addiction (2). Polymer-based catalytic nanocompartments (CNCs) have been previously developed for biomedical applications and shown to shield encapsulated enzymes from their surroundings, prolong their activity and lower their immunogenicity (3,4). In this work, CNCs are employed for inverting the glucuronidation of hymecromone, a therapeutic compound, and the subsequent production of the active compound. (5) Our catalytic nanocompartments (GUS-CNCs) are made from $PMOXA_{10}$-*b*-$PDMS_{25}$ block copolymer and encapsulated ß-glucuronidase, the enzyme responsible for cleaving the glucuronide moiety. Moreover, the polymer membrane of GUS-CNCs is permeabilized with the pore-forming peptide, melittin. Once the glucuronide conjugate of hymecromone (4-MUG) is given, hymecromone is successfully produced by our system both in phosphate buffered solution and in cell culture medium. Furthermore, when incubated with HepG2 cells, CNCs demonstrated no toxic effects and after being taken up by cells, they produce hymecromone *in situ* over 24 hours. These promising results of reversion of a drug metabolite into its active form by catalytic nanocompartments offer them as a potential nanosystem for the production of pharmacologically active compounds locally and on demand.

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[11]:

Tough Double Network Granular Hydrogels

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ABSTRACT TEXT: Load-bearing soft materials that can be processed into 3D shapes gain increasing importance in the biomedical field. Granular hydrogels have the potential to be used as 3D-printable artificial tissues and their local composition can be tuned over micrometer length scales. However, the development of mechanically strong granular hydrogels that can be used for load-bearing applications remains challenging. To overcome this challenge, a secondary hydrogel network can be introduced to the granular hydrogel to form a percolating network, thereby resulting in double network granular hydrogels.[1] However, the interparticle interaction is weak and energy dissipation mechanisms are elusive, resulting in relatively brittle materials. In this work, I will demonstrate how the toughness of double network granular hydrogels can be increased by using two types of polyelectrolyte microgels, polyacrylic acid (PAA) and poly(3-Acrylamidopropyl)trimethylammonium chloride (PAPTAC). These two types of microgels electrostatically attract each other, thereby repetitively increasing the toughness of the material.

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[12]:

Bottom-up cell mimicry: ATP synthesis in giant polymersomes formed by microfluidics

Olivia Eggenberger, University of Basel

To investigate cellular mechanisms that require the cooperation of different membrane proteins, giant unilamellar vesicles (GUVs) made of amphiphilic block copolymers make an excellent cell mimic due to their similar size and increased stability over that of lipid vesicles. Microfluidic systems allow for the formation of a monodisperse population of GUVs at a rate of around 1 kHz with close to 100% encapsulation efficiency. For this study, we embed two different membrane proteins into the membrane of each GUV: NapA, a sodium-proton antiporter that functions to build and maintain a membrane potential, and ATP synthase, which generates ATP from ADP and free inorganic phosphate and is powered by the membrane potential. The formation of a contained cellular process system has far-reaching potential for the increased understanding of cell membranes and the cell as a whole.

[13]:

Extensional Rheology of Mucin-Particle Networks

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Mucin vesicles, sourced from the defense mucus of Atlantic Hagfish (*Myxine glutinosa*), are an uncontaminated and non-damaged source of isolated mucins [1]. Mucus is comprised of a mesh-like structure of mucins and limits conveyance of particles by pore size and viscoelasticity of the network [2] [3]. Mucus acts as a barrier to the epithelium throughout the human body and, depending on surroundings, mucus preferentially absorbs particles of different sizes or surface charges [4]. The ability of microbes, drugs, nutrients, pollutants, or other particles to reach the epithelium is dependent on their ability to be conveyed by this mucus barrier.

Evaluation of mucin-particle interactions that affect physical properties of mucus is performed with extensional, shear, and micro rheology on particle-laden mucin networks. Particles are classified with DLS and zetapotential measurements. Further, water retention measurements are performed to elucidate which nanoparticles can macroscopically stabilize a mucus network. Particles of varied charge, size, shape, and material are investigated.

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Photoreversible Resins For Digitally Printed Protective Films

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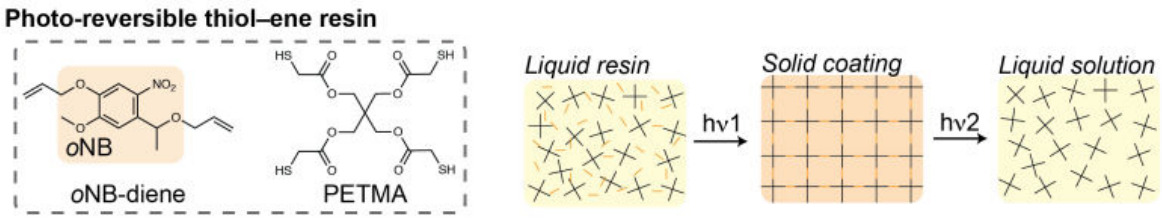
To date, the traditional Swiss watch industry has relied on handcrafted excellence. While precision manufacturing remains the focus of the industry, we now have advanced materials and processes that can replace some of the manufacturing steps that are still done manually. One such step is the manual application of resins (lacquers) onto dials, cases, and bracelet links that serve as temporary masks against mechanical processing, galvanic deposition (coloring), or scratching of the finished parts. Our project, DiPrintProtect in collaboration with EMPA and EPFL, aims to replace the manual application step of protective coatings within the watch manufacturing cycle with digital printing process by developing a printable ink based on a reversible photo-polymer that can be cured (hardened) with light and removed later via irradition with a second wavelength of light without leaving traces. Our approach is based on a thiol-ene resin with a photodegradable cross-linker. While a suitable photoinitiator initates the cross-linking through thiol-ene click chemistry at 405nm UV light, the photodegradable cross-linker degrades the network by cleaving under irradiation with UV light at 365nm. An *ortho*-nitrobenzyl (*o*NB)-diene was synthesized as the cross-linker, as the *o*NB chemistry has shown appropriate cleavage characteristics in the literature. The current challenge lies in advancing the cross-linker molecule to combine efficient cleavage capability with the desired properties of the final resin including printability, compatibility with nanoparticles, and solvent resistance.



Through thiol-ene chemistry, a photoreversible network is formed that de-cross-links by breaking the *o*NB-diene cross-linker

[15]:

Probing small molecule uptake with planar polymer membranes

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Planar polymer membranes are efficient models to study biochemical processes when combined with biomolecules, such as membrane proteins or enzymes[1]. Compared to protein transporter systems, self-assembling artificial capsules with a defined cavity have the major advantage of being host to specific guest molecules[2]. In this joint project, we aim for generation of hybrid membranes with selective uptake function by insertion of different capsule types into artificial, planar membranes. The major difficulty here is to provide strictly apolar conditions for capsule assembly as well as to assess their functionality inside the membrane. For this purpose, we delevoped a fluorescence-based assay as well as a surface-sensitive acoustics-based assay, which allow for the analysis of capsule assembly in polymer environment and for small molecule uptake apability from aqueous flow, resepctively.

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Illuminating the structure of iron carbohydrates in complex biological environments

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Intravenous iron carbohydrate nanoparticles are widely used nanomedicines to treat iron deficiency anaemia, which is associated with illnesses including chronic kidney disease and inflammatory bowel disease (1,2). A variety of clinical and biological studies on these products (ferric carboxymaltose and iron sucrose) are available (3); however, their undergoing structural changes and interactions with proteins during the early stages of entering the human bloodstream are not fully understood. Using a combination of small-angle x-ray and neutron scattering (SAXS/SANS), we investigated how size, shape and agglomeration of iron carbohydrates was influenced by interaction with proteins in biological environments. Hereby, SAXS was used to study the iron core and was complemented by SANS to investigate the much weaker scattering signature of the carbohydrate shell. Our SAXS experiments indicated formation of ellipsoidal agglomerates from single iron cores of iron sucrose, which increased their inter-agglomerate distance with increasing dilution in biological buffer. Higher hierarchical-order agglomerates were also observed upon increasing dilution. SANS measurements revealed a loosely bound and diffuse carbohydrate shell. Experiments on interaction of iron sucrose with two differently shaped plasma proteins (human serum albumin and fibrinogen) showed adsorption of the proteins onto the NP surface, which increased with higher incubation time. With this approach, we shine light on the correlation between physicochemical parameters of iron carbohydrates and their behaviour in biological environments for better prediction of clinical outcomes.

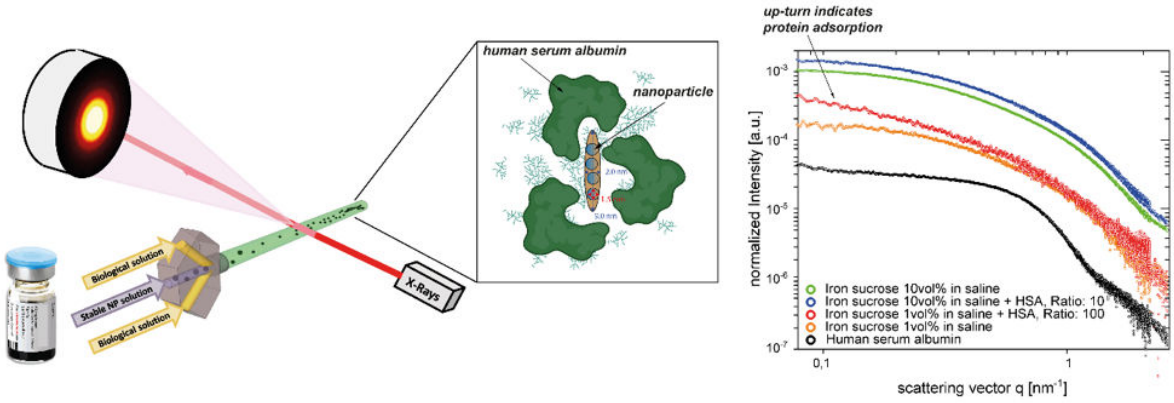

Figure 3 - Modelling the interaction between human serum albumin and iron sucrose through SAXS.

[17]:

Controlling expansion dynamics in bacterial biofilms through modulation of extracellular polymeric matrix components

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Biofilms are biological viscoelastic gels composed of bacterial cells embedded in a self-secreted polymeric extra-cellular matrix (ECM). In environmental settings, such as in the rhizosphere and phyllosphere, biofilm colonization occurs at the solid-air interface. The biofilms' ability to colonize and expand over these surfaces depends on the formation of osmotic gradients and ECM viscoelastic properties. The work we present herein, studies the influence of biofilm ECM components on its viscoelasticity and expansion, using the model organism *Bacillus subtilis* and deletion mutants of its three major ECM components, TasA, EPS, and BslA. Using a multi-scale approach, we quantified macro-scale viscoelasticity and expansion dynamics. Furthermore, we utilized a microsphere assay to visualize the micro-scale expansion patterns. We find that the biofilm expansion dynamics is best correlated the viscoelastic phase angle ϕ. Moreover, we quantify the sensitivity of the biofilm to changes in substrate water potential as a function of ECM composition. Finally, we find that the deletion of ECM components significantly increases the coherence of micro-scale colony expansion patterns. These results demonstrate the influence of ECM viscoelasticity and substrate water potential on the expansion of biofilm colonies on wet surfaces at the air-solid interface, commonly found in natural environments.

[18]:

Light Scattering of Micron-sized Self-assembled Colloidal Aggregates

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Strongly correlated heterogeneous disordered dielectrics can exhibit structural colouring, which is, for example, widely used by nature. However, for practical applications, it is crucial to compartmentalize such materials into small building blocks (with sizes ⩽ 5μm). This can be achieved by using spherical aggregates of nanoparticles, known as photonic balls (PB). The photonic balls not only serve as colourful supraparticles, but, due to their final size, present an ideal playground to study the fundamental aspects of light scattering by correlated disordered media.

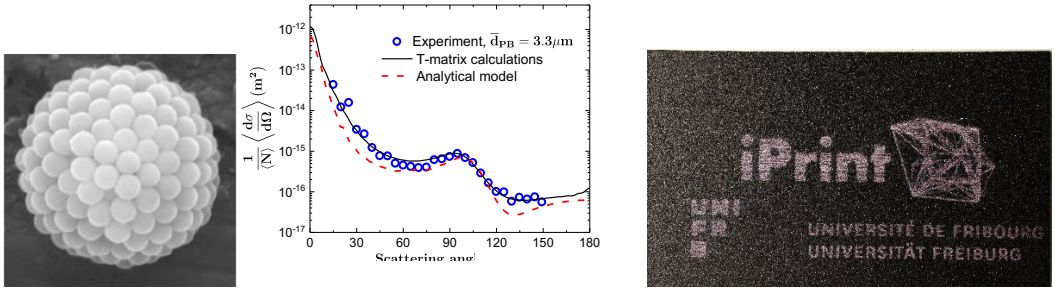

Figure 1: **Left**: SEM image of a single photonic ball (Diameter ~ 1 μm) **Centre**: Differential scattering cross-section vs scattering angle for experimental data, electromagnetic simulations and the analytical model **Right**: Inkjet-printed business cards of the author's affiliation

In our research, we experimentally study light scattering of PBs and developed a theoretical framework to explain a structural color formation by them [1]. We use photonic balls as a model system to elucidate fundamental aspects of phase delay and momentum transfer of light in optically soft heterogeneous dielectric materials [2]. Finally, we use the developed knowledge to demonstrate the PBs potential for graphical printing applications [3].

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Host–guest-based Polymer–Nanoparticle Hydrogels for Modular Materials Design

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Injectable hydrogels are increasingly used in the biomedical field for their shear-thinning and self-healing properties. Polymer–nanoparticle (PNP) hydrogels are a class of injectable nanocomposites based on physical interactions between polymers and nanoparticles (NPs). (1) Such hydrogels have been used as drug delivery platforms or bioinks for additive manufacturing. (2-3) Current design of PNP hydrogels is mainly based on hydrophobic polymer–nanoparticle interactions limiting the range of mechanical properties and functionalities that the material can achieve. Improved design of PNP hydrogels would be enabled by engineered supramolecular interactions of the constituents, enabling tailored rheological properties and functionalities for biomedical applications.
In this work, reversible host–guest interactions between beta-cyclodextrin (CD) and adamantane were used for the design of PNP hydrogels. For this purpose, hyaluronic acid (HA) was functionalized with CD and guest carrying NPs were synthetised through the nanoprecipitation of adamantane functionalized PEG-b-PLA block-co-polymer. PNP hydrogels with varying concentration of polymer and NPs were prepared and showed a limited window of concentration for gelation—insufficient or excess NPs hindered the formation of a solid material. We hypothesised that the network formation related to the NPs capacity to bridge efficiently disparate polymer chains and by extension the ratio of host to guest present. Additionally preliminary data showed the modular potential of the material through the combined use of guest label NPs with non-labelled NPs. This work demonstrates how tailored supramolecular interactions can be used to engineer shear-thinning and self-healing nanocomposites and provides fundamental insight into the design rules of this emerging class of materials.

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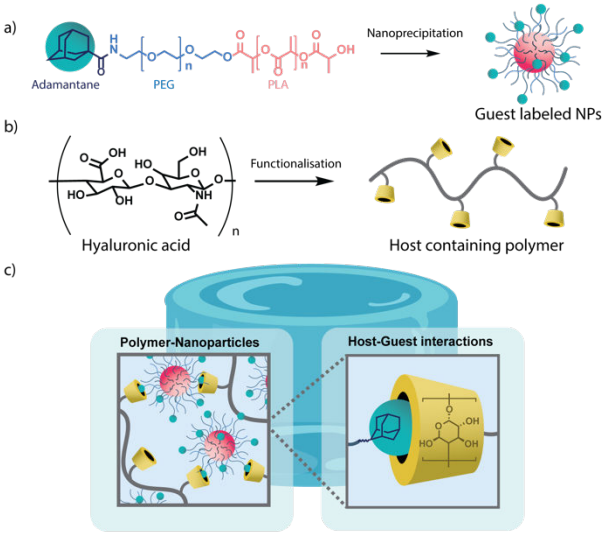

Figure 1: a) Guest containing nanoparticles are obtained through nanoprecipitation of adamantane functionalized PEG-b-PLA block-co-polymer b) Hyaluronic functionalization with cyclodextrin c) PNP hydrogel formation takes place though host guest complex formation.

[20]:

Entropy links molecular and macroscopic behavior in dynamic covalent networks

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Dynamic covalent networks have emerged as a promising class of materials offering advanced applications, such as self-healing hydrogels or for the thermal stabilization of biologics [1-2]. In dynamic covalent networks, the chemistry of the cross-links controls the emergent mechanical properties of the network. For example, the binding kinetics control the network dynamics while the bond stability influences the elastic modulus [3]. However, current models fail to accurately predict how the mechanical properties emerge from the dynamic behavior at the cross-links. Therefore, we developed a framework to better understand how the junction properties in dynamic covalent hydrogels influence their mechanical behavior, as quantified by shear rheometry. For this, dynamic covalent boronic ester-based hydrogels were selected as model networks (Fig 1A). The fraction of bound functional groups, *p*, defined the elastic modulus of the network, independent of chemistry (Fig 1B). A dynamic phantom network model (Fig 1B, black line), which accounts for the equilibrium binding constant of the reversible bond, has been developed to describe rubber elasticity in dynamic covalent networks. However, our observations deviated from this model—the gel point was delayed and the modulus scaled differently with *p*. We accounted for these differences by maximizing the entropy associated with the number of cross-links formed by each network component. In contrast to permanent covalent networks, entropy can be maximized in dynamic covalent networks as the bonds can rearrange after formation. We combined entropy maximization with a model of rubber elasticity derived from the phantom network model to relate the fraction of formed functional groups, *p*, or formed cross-links and the elastic modulus (Fig 1B, red line). In total, our results provide important insight into the molecular behavior of dynamic covalent networks by highlighting that entropy of the network components is a key phenomenon governing gelation in this class of polymer networks.

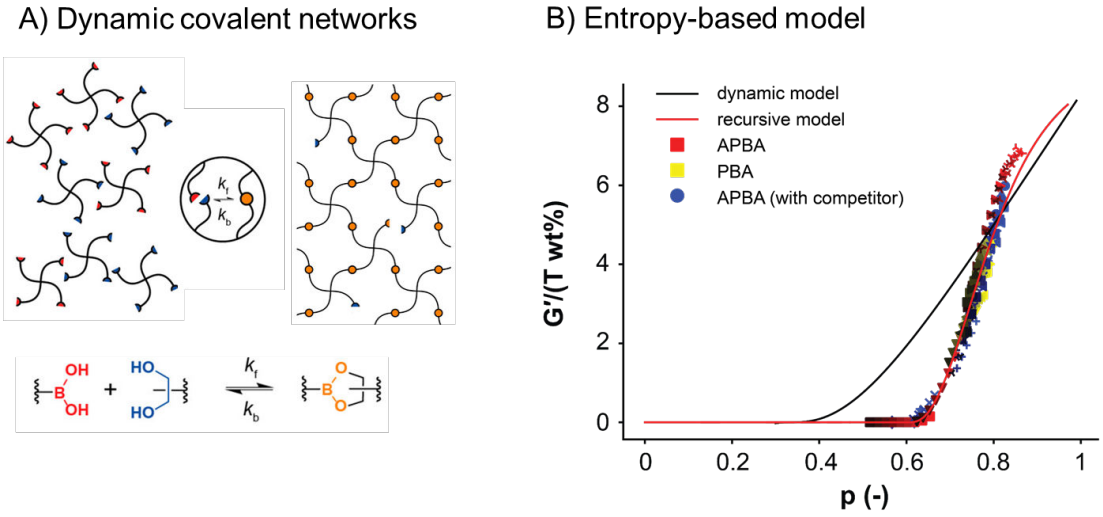

Figure 1 A) 4-arm polymer stars end-functionalized with boronic acids and diols assemble to form dynamic covalent networks. B) Comparison of the dynamic phantom network model (black line) and the newly developed model that accounts for entropy maximization (red line; recursive model) with

experimental data for normalized modulus as a function of the calculated fraction of formed bonds (data points).

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iSCORR a new technique using interference scattering microscope to enhance rheology techniques

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Interference scattering microscopy (iSCAT) has been firstly developed by Sandoghdar [1, 2], and extensively used by Kukura [3]. It is based on the principle that the signal of scattering term decreases with the radius on power 6, and the interference signal decreases only on power 3 of the radius. This allows to probe much smaller particles. This system therefore enables to see small particles close to the surface without using fluorescence. It is mainly used to determine the mass of proteins which adsorb on the surface [4]. We have developed a theory to perform interference correlation microscopy (**iSCORR**) on the iSCAT microscope. With this technique we were able to measure size of proteins in solution in a μm^2 field of view. This scale allows to zoom at aggregated or phase separated regions and capture nanoscopic behaviour. I present the work on measuring the size of proteins in solution, but I will discuss on the potential of this technique to measure complexes modulus, and the integration of this microscope within microfluidics as well as the use for cell probing.

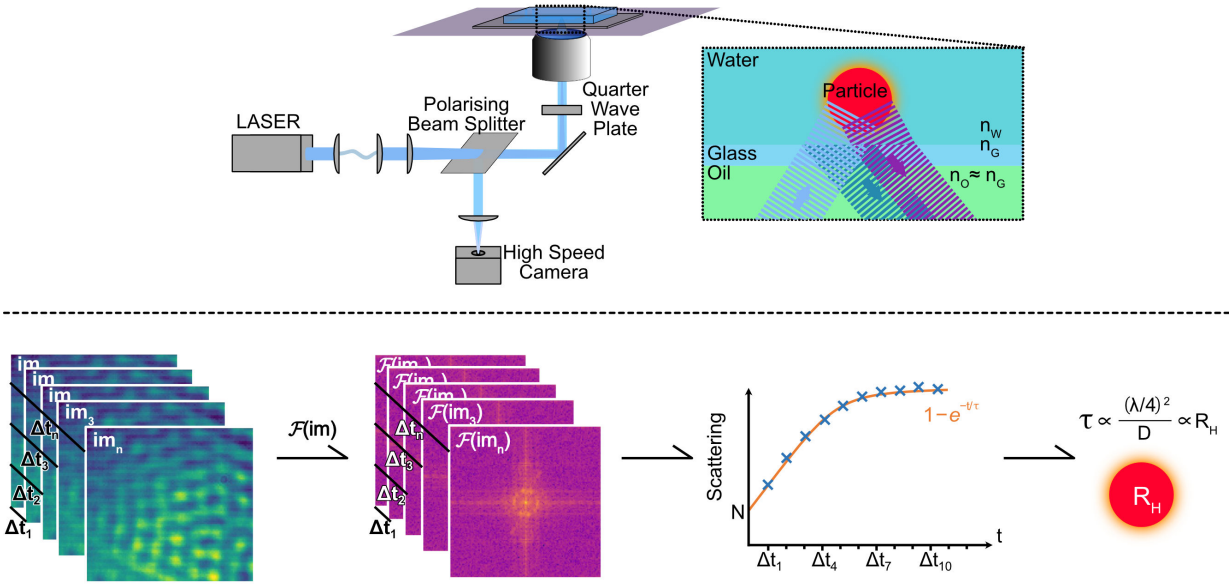

***Figure**. On top the configuration of iSCAT microscope, with a zoom on the reflected light at the interface glass/water and the scattering particle. At the bottom, the process of the iSCORR, the images are transformed into images in Fourier plane. The images correlation is analysed to extract the decay time τ. This decay time gives information of diffusion coefficient and therefore of the particle size.*

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