## dsm-firmenich



# 35<sup>th</sup> Swiss Soft Days

Monday 17.03.2025 | 9h15-17h | Satigny

## **Program**

9:15	Registration and coffee*	
9:55	Welcome	
10:05	Youwei Ma	Closed-Loop Recycling of Vinylogous Urethane Polymers and Their Composites
10:20	Dorina Opris	High-permittivity elastomers: Paving the way for next- generation applications
10:35	Gadi Slor	(De)polymerization from mixtures: towards natural recycling
10:50	Oya Tagit	Perfluorocarbon-loaded polymeric nanoparticles with tailored nanomechanical and surface properties for cell-specific uptake
11:05	Break-out sessions : Opportunities and choices in career development	
11:45	Boyang Zhou	Set number of plastic rearrangements at the onset to yielding
12:00	Yangxia Feng	Characterizing hydrogel behavior under compression with gel- freezing osmometry
12:15	Julien Bauland	Two-step aging dynamics in enzymatic milk gels
12:30	Lunch and poster session	
13:30	Madina Almukambetova	Double Symmetry Breaking in Filamentous Colloidal Tactoids
13:45	Savannah Gowen	Training and re-training disordered liquid crystal elastomer networks
14:00	Maryame Bina	Solid-supported textured polymer membranes as dual antifouling - antimicrobial functional surfaces



14:15	Anasua Mukhopadhyay	Fingerprinting Tau Oligomers with a 20 nm Diameter Nanopore from Pneumolysin
14:30	Yuechuan Lin	A New Method to Measure Pore Radius Distribution of Powders
14:45	Break and poster session	
15:15	Sara Catalini	Amyloid Aggregation in Mixed Whey Protein Systems: An Experimental Approach Across Multiple Length Scales
15:30	Simone Bertucci	Fabrication of Water-Based Photonic Paints Using Self- Assembled Block Copolymer Microparticles
15:45	Thomas Kainz	Colloidal Self-Assembly as Templating for 3D Second-Harmonic Photonic Crystals
16:00	Georges Formon	Expanding supramolecular polymers: from synthesis to responsiveness
16:15	Matteo Rutsch	Structure and dynamics of phytantriol-glycerol mesophases: Insights into the reverse micelle to lamellar phase transition
16:30	Closing remarks	

<sup>\*</sup> Train RL5 from Geneva main station arrives at Zimeysa train station at 8:57 and 9:27.

It is about 5min walk to the dsm-firmenich site at Rue de la Bergère 7.

The guards at the entry gate will guide you towards the reception to receive a visitors pass and we'll guide you towards the meeting room.

## **Poster overview**

P1	Francesca Bono	Enzyme-induced mineralization of calcium carbonate in 3D printable granular hydrogels
P2	Mie T. Pedersen	Jellyfish Soft Matter: Transforming Living Hydrogels into Culinary
		and Biomaterial Innovations
P3	Greta Cocchi	Enzymatic mineralization of 3D-printed granular hydrogels
P4	Matteo Darra	Mycelium-Cellulosenanofibrils Hybrid Materials
P5	Voichita Mihali	Hybrid Clusters Obtained from the Self-Assembly of Janus
		Nanoparticles and Polymersomes for Bio-Applications
P6	Eva Baur	Microstructured elastomers and their macroscopic properties



P7	Yiyao Hu	Exploring the colloidal properties of Bacillus subtilis spores at liquid interface
P8	María Li López Bautista	Non-equilibrium stabilization of proteins by chaperones
P9	Manuel Kraus	Solid-Supported Polymer Membranes: Influence of Deposition Methods on Morphology and Properties
P10	Shasha Zheng	An Aldehyde-Stabilization Strategy for Building Bio-based Consumer Products Around Intact Lignocellulosic Structures
P11	Mirela Malekovic	Solid-supported triblock copolymer mixtures as a platform for targeted molecule attachment and biosensing
P12	Bruno Silva	Quantifying Lipid Bilayer Coating on Polymer-Nucleic Acid Nanoparticles Using Fluorescence Cross-Correlation Spectroscopy
P13	Manuel	Reaction Principles for in-Situ Functionalization of Ion
	Stehrenberger	Chromatography Columns
P14	Taieesa Peshkovsky	Fabry-Pérot-Interferometry for Microscale Concentration Gradient Imaging
P15	Leonard Krupnik	Elucidating Interactions between Iron-Carbohydrate Complexes and Human Blood Serum using Small-Angle Scattering
P16	Ding Ren	Naturally Occurring Protein-rich Lipoprotein Nanoparticles as Tools for Melanoma Biomarkers Discovery and Detection
P17	Praveen Kumar	Recreating the color palette of nature: Bioinspired structural colors
P18	Lorenzo Lucherini	Patterning Electronically Conductive Features within Soft Hydrogel Substrates
P19	Anamarija Nikoletić	Polymer-inorganic nanohybrids for NIR-light triggering
P20	François Rivat	Lithium salt-containing elastomers towards soft robotics applications
P21	Giovanni Savorana	Bacterial extracellular DNA forms stress-hardening biofilm streamers
P22	Inyoung Yonah Lee	Exopolymers from the fermentation of Schizophyllum commune
P23	Lucien Cousin	Entropy links molecular and macroscopic behavior in dynamic covalent networks
P24	Laura Rüegger	Restructuring dynamics of pastes at the yielding transition
P25	Carol Bouvard	Hydrodynamics of plant based emulsion breakup, recoalescence & aggregation in homogenizers
P26	Antonia Georgopoulou	Bioprinting of Stable Bionic Interfaces Using Piezoresistive Hydrogel Organoelectronics
P27	Vincent Glauser	Computer-generated disordered networks for photonic bandgap

# ABSTRACTS TALKS



## Closed-Loop Recycling of Vinylogous Urethane Polymers and Their Composites

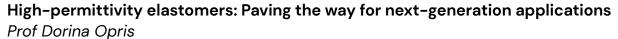
Youwei Ma,a\*

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The development of mechanically robust and closed-loop recyclable polymers and composites represents an essential undertaking in the context of advancing a circular materials economy. In this contribution, we show that vinylogous urethane (VU) polymer networks synthesized from bisacetoacetates with a skeleton of polyethylene glycol (aPEG) or polytetrahydrofuran (aPTHF), and tris(2-aminoethyl)amine (TREN), can be degraded by water or HCl, with the excellent recovery of bisacetoacetates (i.e., aPEG or aPTHF) and TREN components.<sup>23</sup> Specifically, the hydrophilic VU networks with an aPEG skeleton undergo depolymerization in water, while the hydrophobic VU networks derived from aPTHF are depolymerized in a HCI/chloroform solvent mixture. The rate of depolymerization is controlled by temperature, the amount of water or HCl concentration, molecular weight of building blocks (i.e., aPEG and aPTHF), and composition of the starting materials. These last two parameters also allow one to tailor the mechanical properties of the final materials, which exhibit plastic-like or elastomer-like tensile behavior for aPEGor aPTHF-derived networks, respectively. Moreover, incorporating fillers such as multiwalled carbon nanotubes (MCNs) and carbon fibers enhances the functionality of the resulting VU polymer composites without compromising their recyclability. This is corroborated by a case study, in which we demonstrate that the aPTHF-based VU composites containing 5 wt% MCNs can serve as a strain sensor, with the successful recovery of aPTHF and MCNs presented after its end of life.3 Overall, we anticipate that the VU chemistry explored here offers a versatile platform for synthesizing sustainable polymers and/or composites with closed-loop recyclability across diverse application domains.



- Ma, Y.; Zheng, C.; Slor, G.; Özkan, M.; <u>Gubelmann</u>, O.J.; Stellacci, F. *Angew. Chem. Int. Ed.* **2024**, e202410624. DOI: 10.1002/anie.202410624.
- 2. Ma, Y.; Jiang, X.; Shi, Z.; Berrocal, J. A.; Weder, C. Angew. Chem. Int. Ed. 2023, 62, e202306188.
- 3. Ma, Y.; Stellacci, F.; Macromolecules 2025, DOI: 10.1021/acs.macromol.4c03256.



Polymers with high and finely tunable dielectric permittivity are of great scientific interest due to their wide application potential, ranging from transducers and capacitors to Li-ion batteries. However, how much can the permittivity be increased, and how does this increase affect other properties? To answer these questions, we have modified the polysiloxanes with different types and amounts of polar groups and investigated how these chemical modifications affect different properties. We also explore the impact of filler addition on the properties of our polar silicones, focusing on enhancing the processability and functionality of thin films. The most promising polar silicone elastomers have also been investigated as dielectrics in actuators, sensors, energy harvesters, capacitive light-emitting devices, and electrolytes in solid-state Lilon batteries and thus demonstrated their functionality.

# (DE)POLYMERIZATION FROM MIXTURES: TOWARDS NATURAL RECYCLING

Gadi Slor

With approximately 400 million tons of plastic produced annually, the end-of-life (EoL) problem is one of the most pressing environmental issues today. Shockingly, only about 9% of this plastic is recycled, largely due to the challenge of managing mixed plastic waste streams that are difficult, if not impossible, to sort and process efficiently. In contrast, natural polymers like proteins, nucleic acids, and polysaccharides are produced on a billion-ton scale and are recycled in a perfectly sustainable way. These biopolymers undergo efficient and controlled synthesis and degradation within a highly complex matrix, in the presence of various monomers, organic molecules, salts, and more. If we could apply this synthesis and degradation scheme—paralleling a living cell to a reaction flask—we could significantly improve our recycling capabilities. Inspired by nature's remarkable ability to recycle its own polymers, in this talk I will present a closed-loop chemical recycling system capable of handling mixed plastic waste. The system is based on chemical recycling to monomer of polymers mixtures followed by selective polymerization of the yielded monomers mixture. This approach addresses one of the most critical challenges in plastic recycling: the efficient recycling of multi-material objects and films, and allows the recycling of complex mixed plastic waste streams.

## PERFLUOROCARBON-LOADED POLYMERIC NANOPARTICLES WITH TAILORED NANOMECHANICAL AND SURFACE PROPERTIES FOR CELL-SPECIFIC UPTAKE

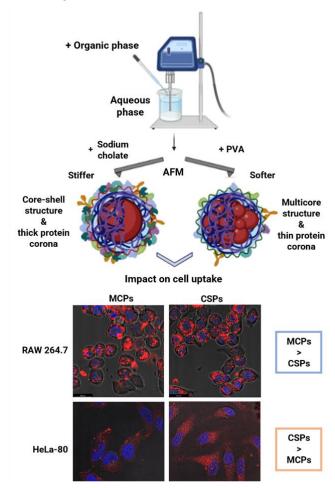
NAIARA LARREINA VICENTE<sup>1</sup>, MANGALA SRINIVAS<sup>1</sup>, OYA TAGIT <sup>2\*</sup>

<sup>1</sup>Cell Biology and Immunology (CBI), Wageningen University, De Elst 1, 6708WD Wageningen, Netherlands <sup>2</sup>Group of Biointerfaces, Institute for Chemistry and Bioanalytics, FHNW University of Applied Sciences and Arts Northwestern Switzerland, Hofackerstrasse 30, 4132, Muttenz, Switzerland – Email: oya.tagit@fhnw.ch

#### **Abstract**

Poly(lactide-co-glycolide) nanoparticles (PLGA NPs) loaded with Perfluoro-15-crown-5-ether (PFCE) have been developed for *in vivo* <sup>19</sup>F magnetic resonance imaging applications. A slight modification of the formulation in terms of

surfactant type led to different distribution profiles of PFCE phase through the polymer matrix with direct implications on the nanoparticle internal structure. While the non-ionic surfactant polyvinyl alcohol (PVA) facilitated the formation of multi-core particles with PFCE phase distributed as multiple small domains through the nanoparticle (multi-core particles, MCPs), the anionic surfactant sodium cholate (NaCh) packed the PFCE phase as a single domain within the core of the nanoparticles (core-shell particles, CSPs). This difference in the nanoparticle ultrastructure further impacted the hydration profile of the MCPs and CSPs, giving rise to significantly different nanomechanical properties (52 MPa and 120 MPa, respectively) as demonstrated by atomic force microscopy.<sup>2</sup> The impact of the surfactant on the NP surface chemistry was evidenced by their protein corona, which was significantly greater in CSPs. In vitro studies showed a higher uptake of MCPs by RAW macrophages but a preference for CSPs by HeLa cells. (Fig. 1). Overall, our study establishes a direct link between the type of surfactant used in nanoparticle formulation and the resulting internal structure, stiffness, and protein corona, all of which significantly influence particlecell interactions. Our findings underscore the potential of tailoring particle stiffness and architecture to achieve cellspecific delivery in therapeutic and therapostic applications, where uptake efficiency by the biological targets is paramount.



**Figure 1.** Surfactant-driven tailoring of the internal structure, stiffness, and protein corona of PFCE-loaded PLGA nanoparticles for cell-specific uptake.

<sup>&</sup>lt;sup>1</sup> Staal, et al. In vivo clearance of 19F MRI imaging nanocarriers is strongly influenced by nanoparticle ultrastructure. Biomaterials 2020, 261, 120307.

<sup>&</sup>lt;sup>2</sup> Vicente, et al. Perfluorocarbon-loaded poly(lactide-co-glycolide) nanoparticles from core to crust: multifaceted impact of surfactant on particle ultrastructure, stiffness, and cell uptake. ACS Applied Polymer Materials 2025, https://doi.org/10.1021/acsapm.4c03360
Published March 3, 2025



## Set number of plastic rearrangements at the onset to yielding

Boyang Zhou and Veronique Trappe University of Fribourg, Department of Physics, 1700 Fribourg, Switzerland

Soft jammed materials subjected to a constant strain rate exhibit a stress relaxation process upon shear cessation that eventually leads to a finite residual stress. Investigations revealed that when shear is stopped within the steady-state regime, where the stress has reached a constant value, the residual stresses systematically decrease with increasing shear rate (1-4).

We here explore the response to shear cessation when the application of a shear rate is stopped within the transient regime to steady state. Our experiments reveal that the residual stress depends on strain and is independent of the shear rate when the experiment is stopped at 'moderate' strains. This strain dependence of the residual stress contrasts with the strain-rate dependence of the stress observed during the application of the shear rate.

This suggests that strain encodes a fixed number of plastic rearrangements, which occur regardless of how fast this strain is reached. When the system is deformed too quickly, plastic rearrangements do not have time to occur within the experimental time scale, such that the system is elastically overloaded. Consistent with this interpretation, we find that the stresses obtained during the application of the strain rate matches the residual stress once the shear rate is sufficiently slow. Residual stresses thus effectively reflect the stresses observed under quasi-static loading conditions.

- 1.) L. Mohan, R.T. Bonnecaze, M. Cloitre, Phys. Rev. Lett. 111, 268301 (2013).
- 2.) L. Mohan, M. Cloitre, R.T. Bonnecaze, J. Rheol. **59**, 63-84 (2015).
- 3.) M. Ballauff, et al., Phys. Rev. Lett. **110**, 215701 (2013).
- 4.) H. A. Vinutha, et al., PNAS Nexus 3, pgae441 (2024).



# Characterizing hydrogel behavior under compression with gel-freezing osmometry

Yanxia Feng<sup>1\*</sup>, Dominic Gerber<sup>1</sup>, Stefanie Heyden<sup>2</sup>, Martin Kröger<sup>1</sup>, Eric R. Dufresne<sup>3,4</sup>, Lucio Isa<sup>1</sup>, and Robert W. Style<sup>1</sup>

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 Department of Civil, Environmental and Geomatic Engineering, ETH Zürich, 8093 Zürich, Switzerland
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 Laboratory of Atomic and Solid State Physics, Cornell University, Ithaca, USA

#### Abstract

Hydrogels are particularly versatile materials that are widely found in both Nature and industry. One key reason for this versatility is their high-water content, which lets them dramatically change their volume and many of their mechanical properties — often by orders of magnitude — as they swell and dry out. Currently, we lack techniques that can precisely characterize how these properties change with water content. To overcome this challenge, here we develop Gel-Freezing Osmometry (GelFrO): an extension of freezing-point osmometry. We show how GelFrO can measure a hydrogel's mechanical response to compression and shrinkage in response to an applied osmotic pressure, while only using small, 100µL samples. Because the technique allows measurement of properties over an unusually wide range of water contents, it allows us to accurately test theoretical predictions. We find simple, power-law behavior for both mechanical and osmotic responses, while these are not well-captured by classical Flory-Huggins theory. We interpret this power-law behavior as a hallmark of a microscopic fractal structure of the gel's polymer network and propose a simple way to connect the gel's fractal dimension to its mechanical and osmotic properties. This connection is supported by observations of hydrogel microstructures using small-angle x-ray scattering. Finally, our results motivate simplifications to common models for hydrogel mechanics, and we propose an updated hydrogel constitutive model.

## References

Yanxia Feng, Dominic Gerber, Stefanie Heyden, Martin Kröger, Eric R. Dufresne, Lucio Isa, and Robert W.Style, arXiv:2407.13718.

## Two-step aging dynamics in enzymatic milk gels

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**Abstract**: Colloidal gels undergo a phenomenon known as physical aging, i.e., a continuous change of their physical properties with time. To date, most of the research effort on aging in gels has been focused on suspensions of hard colloidal particles. In this work, we tackle the case of soft colloidal "micelles" comprised of milk proteins, in which gelation is induced by the addition of an enzyme. Using time-resolved mechanical spectroscopy, we monitor the viscoelastic properties of a suspension of colloidal micelles through the sol-gel transition and subsequent aging. We show that the microscopic scenario underpinning the macroscopic aging dynamics comprises two sequential steps. First, the gel microstructure undergoes rapid coarsening, as observed by optical microscopy, followed by arrest. Second, aging occurs solely through a contact-driven mechanism, as evidenced by the square-root dependence of the yield stress with the elastic modulus measured at different ages of the gel. These results provide a comprehensive understanding of aging in enzymatic milk gels, crucial for a broad range of dairy products, and for soft colloids in general.

## **Double Symmetry Breaking in Filamentous Colloidal Tactoids**

Madina Almukambetova, <sup>1</sup> Hamed Almohammadi, <sup>1,2</sup> Florine Schleiffer, <sup>1</sup> and Raffaele Mezzenga<sup>1,3</sup>

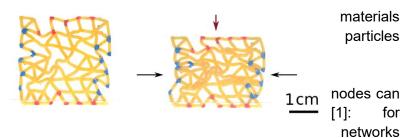
<sup>1</sup> Department of Health Sciences and Technology, ETH Zürich, Zürich, Switzerland <sup>2</sup> John A. Paulson School of Engineering and Applied Sciences, Harvard University, Cambridge, MA, USA <sup>3</sup> Department of Materials, ETH Zürich, Zürich, Switzerland

Understanding dynamics of liquid crystalline tactoids under external forces is of great importance due to their potential applications in optics, medical devices and displays. However, only recently have tactoids started to be studied systematically under external forces, in particular, by extensional flow [1]. Here, we subject tactoids to a shear flow field and study their deformation dynamics upon varying conditions of shear and time scales. Using amyloids and nanocellulose to form tactoids from model filamentous colloids with opposite sequence of chirality amplification (left-handed mesoscopic—right-handed cholesteric for amyloids [2]; right-handed mesoscopic—left-handed cholesteric for nanocellulose [3]), we show a complex deformation mechanism in their shape and internal structure under shear flow. When tactoids deform perpendicularly to their long axis, a double symmetry breaking occurs in both their contour shape, with the emergence of a kink, and their orientation of nematic field. We further show that the mesoscopic chirality of the building blocks directs the position of the kink, with the macroscopic tactoid asymmetry being mirrored when inverting the mesoscopic chirality of the constitutive filamentous colloids, e.g. from the left-handed amyloids to the right-handed nanocellulose.

- [1] H. Almohammadi, M. Bagnani, and R. Mezzenga, Flow-induced order-order transitions in amyloid fibril liquid crystalline tactoids, Nat. Commun. 11, (2020).
- [2] G. Nyström, M. Arcari, and R. Mezzenga, Confinement-induced liquid crystalline transitions in amyloid fibril cholesteric tactoids, Nat. Nanotechnol. 13, (2018).
- [3] G. Nyström, M. Arcari, J. Adamcik, I. Usov, and R. Mezzenga, Nanocellulose Fragmentation Mechanisms and Inversion of Chirality from the Single Particle to the Cholesteric Phase, ACS Nano 12, 5141 (2018).

## Training and re-training disordered liquid crystal elastomer networks Savannah Gowen

Disordered elastic networks are model for disordered systems from jammed to polymers. It has been shown that metamaterials formed from macroscopic disordered network arrays of bonds and be trained to exhibit novel elastic functions example, unlike most natural materials these



can be trained to display a negative Poisson's ratio in which they respond to compression along one axis by contracting along all the others[2]. Another novel elastic property is inspired by protein allostery: the binding of a molecule at one site in a protein triggers the ability of a distant site to bind to another molecule. Likewise, this action-at-a-distance can be implemented mechanically: Applying strain locally to a set of source nodes in the material triggers a strain response at a distant set of target nodes [3-5]. This work investigates both the ability to train for function and then to erase that function on-demand in macroscopic metamaterials made from liquid crystal elastomers (LCEs) [6]. We first show how macroscopic liquid crystal elastomer networks can be tuned via directed aging to induce an auxetic response. We then show that the arrays can be reset and re-trained for another local mechanical function, allostery, thus demonstrating pluripotent functionality.

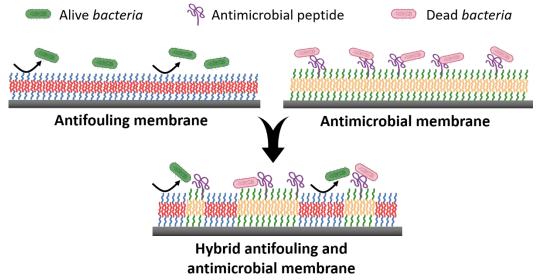
- [1] HM Jaeger, A Murugan, SR Nagel, Training physical matter to matter. Soft Matter 20, 6695–6701 (2024).
- [2] N Pashine, D Hexner, AJ Liu, SR Nagel, Directed aging, memory, and nature's greed. *Sci. advances* **5**, eaax4215 (2019).
- [3] D Hexner, AJ Liu, SR Nagel, Periodic training of creeping solids. Proc. Natl. Acad. Sci. 117, 31690–31695 (2020).
- [4] SD Gowen, Training allostery-inspired mechanical response in disordered elastic networks. arXiv preprint arXiv:2501.04838 (2025).
- [5] LE Altman, M Stern, AJ Liu, DJ Durian, Experimental demonstration of coupled learning in elastic networks. Phys. Rev. Appl. 22, 024053 (2024).
- [6] SD Gowen, E Ghimire, CA Lindberg, IS Appen, SJ Rowan, SR Nagel, Training and re-training liquid crystal elastomer metamaterials for pluripotent functionality. arXiv preprint arXiv:2502.17238 (2025).

# Solid-supported textured polymer membranes as dual antifouling - antimicrobial functional surfaces

Maryame Bina, John P. Coats, Michal Skowicki, Mirela Malekovic, Voichita Mihali, Cornelia G. Palivan

Department of Chemistry, University of Basel

Nanotextured surfaces are widely found in nature and exhibit a variety of properties, such as superhydrophobicity, bacteria repellence, exceptional adhesion or structural colours. Traditional methods for manufacturing these surfaces, including physical or chemical patterning, are costly and require specialised equipment. Here, we introduce a biomimetic approach utilising the amphiphilic property of two dissimilar diblock copolymers as the driving force for their self-assembly, similar to how amphiphilic lipids form biological membranes. However, contrary to naturally occurring lipids, polymers offer enhanced mechanical stability and chemical versatility, thus allowing for the fine tuning of their properties. The membranes are composed of PEO-b-PEHOx and PMOXA-b-PDMS as the two amphiphilic diblock copolymers mixed at various concentrations. PEO exhibits antifouling properties, while PMOXA features a functional azide end group suitable for bioconjugation. These fully synthetic, solid-supported planar membranes, undergo phase separation, forming domains embedded within a continuous phase. The dissimilar properties of each block and the molar ratio of the copolymers in the mixture were the key parameters to induce nanoscale phase separation of the planar membranes.1 Subsequently, by conjugating an antimicrobial peptide, KYE28, onto the phase separated polymer membrane, we create a multifunctional coating that significantly reduces bacterial attachment and growth. The combination of the antifouling PEO and the antimicrobial KYE28 produces a synergistic effect, resulting in a substantial decrease in bacterial proliferation.<sup>2</sup> Although this study presents only one example of application of phase separating polymer membranes, such hybrid polymer surfaces offer endless possibilities for the conjugation of biomolecules (enzymes, proteins, DNA strands, or antibodies) or nano assemblies, including nanoparticles or micelles.



- [1] Bina, Maryame, Agata Krywko-Cendrowska, Davy Daubian, Wolfgang Meier, and Cornelia G. Palivan. "Multicomponent Copolymer Planar Membranes with Nanoscale Domain Separation." Nano Letters 22, no. 13 (2022): 5077-5085.
- [2] Bina, Maryame, John P. Coats, Michal Skowicki, Mirela Malekovic, Voichita Mihali, and Cornelia G. Palivan. "Hybrid Planar Copolymer Membranes with Dual Functionality against Bacteria Growth." Langmuir 40, no. 44 (2024): 23178-23188.

## Fingerprinting Tau Oligomers with a 20 nm Diameter Nanopore from Pneumolysin

Anasua Mukhopadhyay, <sup>1,2\*</sup> Wachara Chanakul, <sup>1</sup> Yu-Noel Larpin, <sup>1</sup> Saurabh Awasthi, <sup>1,3</sup> Anna D. Protopopova, <sup>1</sup> René Köffel, <sup>4</sup> Alessandro Ianiro, <sup>1,2,5</sup> Michael Mayer <sup>1,2\*</sup>

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- <sup>2</sup> Swiss National Center for Competence in Research (NCCR) Bio-inspired Materials, University of Fribourg, Fribourg, Switzerland
- <sup>3</sup> Present Address: Department of Biotechnology, National Institute of Pharmaceutical Education and Research Raebareli (NIPER-R), Lucknow, UP, India
- <sup>4</sup> Institute for Chemistry and Biotechnology, Zurich University of Applied Sciences (ZHAW), Wädenswil, Switzerland.
- <sup>5</sup> Present Address: Department of Chemistry, Katholieke Universiteit Leuven, Leuven, Belgium
- \* Email IDs: michael.mayer@unifr.ch; anasua.mukhopadhyay@unifr.ch

Biological nanopores are emerging as powerful sensing tools for single-molecule analysis of nucleic acids, peptides, and proteins. However, the limited size of existing biological nanopores presents a long-standing challenge for transporting large-size, full-length proteins in their natively folded state. Here, we introduce a stable, low-noise, cylindrical transmembrane pore formed by self-assembly of Pneumolysin (PLY) toxin. In situ, assembly of the PLY nanopore occurred in a single step directly on a lipid bilayer upon application of a potential difference of 100 mV. Electrical resistance measurements revealed a diameter of approximately  $20 \pm 2$  (N = 50) nm of membrane-inserted PLY nanopores. This exceptionally large nanopore enabled accurate single-molecule resistive-pulse sensing for the estimation of the volume and shape of folded proteins, ranging in size from FAB ( $\approx 50$  kDa) to tetramers of concanavalin A (4 x 28 kDa = 112 kDa). We used PLY pores to detect differences in the volume of single proteins within a mixture and estimated the size distribution of tau protein (monomer  $\approx 45$  kDa) and its oligomers from dimers to hexamers in solution. By combining volume analysis with single-particle shape approximation, we uncovered details of Tau oligomerization at nanomolar concentrations. The novel PLY nanopore shows strong promise for advancing the quantification and characterization of heterogeneous amyloid oligomers as biomarkers.

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- 2. Mukhopadhyay, A.; Chanakul, W.; Larpin, Y.-N.; Awasthi, S.; Protopopova, A. D.; Köffel, R.; Ianiro, A.; Mayer, M. Pneumolysin Nanopores with 20 Nm Inner Diameter to Characterize the Size and Shape of Tau Oligomers. bioRxiv. February 8, **2025**, p 2025.02.07.637128.



Company: Nestle research, Route du Jorat 57, CH-1000 Lausanne 26

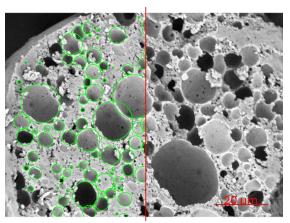
Author: Yuechuan Lin, Adam Burbidge, Josep Busom Descarrega, Ilaria Gaiani, Sidhanth Tyagi Abstract:

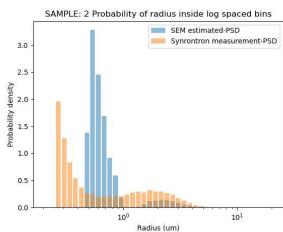
Pore radius distribution is one of the main metrics to characterize the internal porous structure of food [1,2]. State of the art techniques involve 3D tomography and 3D segmentation by thresholding [3,4]. However, resolution of x-ray tomography is limiting the minimal detectable pores. Voxel size of State-of-the-Art synchrotron X-ray tomography is on the order of 0.1 micron, meanwhile the smallest pores radius in the samples we want to study are around 0.5 micron. The resolution is not enough to resolve the smallest pores based on our experience.

To close the gap on resolution, <u>we developed a new technique based on automated Scanning Electron Microscope (SEM)</u>, <u>object detection model</u>, <u>and mathematical transformation to measure the pore size distribution of porous powder</u>.

We first prepare the sample by cutting the powders to expose the internal structure. We then developed a two-stage scanning routine combining SEM and custom-trained object detection model to detect exposed pores for each sample. We name the radius of exposed pores as chord radius to distinguish from the spherical pore radius. Finally, we developed a mathematical transformation to convert the chord radius distribution into pore radius distribution.

To validate our method, we measured the pore radius distribution of 5 food powder sample with our method and synchrotron x-ray tomography. We reached the conclusion that our method has better resolving power for smaller pores (~0.5um) at the same time agree with the synchrotron measurement for larger pores (~2um). Our method is also more cost effective than the synchrotron method.





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# Amyloid Aggregation in Mixed Whey Protein Systems: An Experimental Approach Across Multiple Length Scales

## Sara Catalini

The fundamental principles underlying the complexity of protein assembly, particularly in mixed protein systems and crowded environments [1], remain poorly understood. Despite significant progress in the field, the molecular mechanisms driving protein aggregation and gelation in these environments continue to present challenges. This study provides crucial molecular, structural, and viscoelastic [2] insights into the aggregation and gelation processes of pure and mixed aqueous solutions of  $\beta$ -lactoglobulin and albumin whey proteins [3]. Using a multi-technique approach that spans length scales from the molecular to the macroscopic [4], we present a more comprehensive understanding of protein aggregation in complex systems. Our results demonstrate that under low pH and heat denaturation,  $\beta$ -lactoglobulin tends to form highly ordered amyloid-like aggregates [3, 5], while bovine serum albumin forms distinct non-amyloid aggregates [3]. In the presence of crowding agents, all protein solutions tested undergo a transition to composite gel networks with molecular origins that vary depending on protein composition [3]. Notably, the study emphasizes the ability to control the amyloid aggregate content, which has a significant impact on the structural, rheological, and viscoelastic properties of these composite gels. Such control over gel structure and viscosity is critical for applications in the food industry, as these properties are key to determining the softness, texture, and overall quality of food products, offering valuable insights for the design of texture-modified foods and other protein-based systems.

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# Fabrication of Water-Based Photonic Paints Using Self-Assembled Block Copolymer Microparticles

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Since ancient times, colors and paints have played a fundamental role in human culture and technology. In modern days, paints primarily use synthetic pigments or dyes embedded in a polymeric binder, allowing for greater vibrancy and color control. These pigments, both organic and inorganic, owe their coloration to selective absorption of certain light wavelengths. Although versatile, commonly used pigments present dull tints and tend to fade over time. As an alternative to conventional techniques, recent research has concentrated on replicating the structural coloration found in nature. Structural color emerges from the interaction of light with a nanostructured material. In photonic crystals, light interacts with a structure consisting of a periodic nanoscale alternation of two materials with distinct refractive indices. This produces a spectral region of high reflectivity, known as Photonic Band Gap (PBG). Within this framework, we fabricated water-dispersed photonic pigments by leveraging the three-dimensional confined self-assembly of block copolymers (BCPs) within emulsion droplets. The resulting microspheres, composed of poly(2-vinylpyridine)-b-poly(methyl methacrylate) (P2VP-PMMA), exhibit structural coloration arising from their internal lamellar organization. To enhance their optical response, we introduced 2,4,6-triiodophenol, a high-refractive-index molecule that selectively binds with one of the polymer blocks. By precisely controlling the amount of this molecular additive, we achieved tunable coloration across the visible spectrum with enhanced color brilliance due to the increased dielectric contrast. Subsequently, we formulate a photonic paint by mixing a water-soluble polymer binder with the microparticles to enhance the system stability, while simultaneously improving the overall color vividness. This system was thoroughly characterized, with a focus on performance criteria governed by the balance between the components within the formulation.

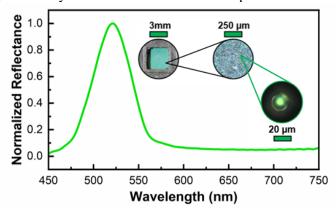


Table of Content. From macro to micro: digital photographs and microscope images of our paints at increasing magnification and relative reflectance spectrum

## Colloidal Self-Assembly as Templating for 3D Second-Harmonic Photonic Crystals

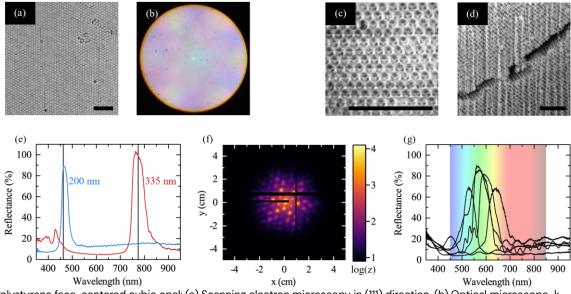
Thomas Kainz<sup>1</sup>, Helena Weigand<sup>2</sup>, Rachel Grange<sup>2</sup>, Ullrich Steiner<sup>1</sup> and Viola Vogler-Neuling<sup>1</sup>
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Three-dimensional nonlinear (second-harmonic) photonic crystals can simultaneously generate different non-linear processes, such as second-harmonic generation (SHG), optical parametric amplification, and other sum- and difference-frequency processes [1, 2]. However, creating large crystals in all three dimensions, mainly in the z direction, presents a considerable challenge, primarily due to the chemical inertness of metal oxides [3-5].

This study shows the first demonstration of colloidal-crystal-templating for a second-order optical material. We selectively synthesized monodisperse nanospheres with tunable unit sizes to self-assemble polystyrene opals with different band gaps (Fig.1 a, b, e). These serve as templates for infiltration with barium titanate sol-gel. After a calcination step, we obtained an inverse fcc network of tetragonal barium titanate (Fig. 1 c). We fabricated samples with unprecedented sizes for single domains: > 3000 unit cells in x, y directions and > 100 in z (Fig. 1 d, f). The achieved optical reflectivity values are above 80% throughout the fabrication. We can engineer the final photonic band gap over the entire optical range, matching it both to material and setup requirements (Fig. 1 g).

We successfully replicated the photonic network into a second-order material. For the first time, we demonstrated a linear photonic band gap from a fully scalable three-dimensional photonic crystal made of a  $\chi^{(2)}$  optical material. Our approach overcame previous limitations, especially stacking in the z-direction, while maintaining full tunability of the initial unit cell [3]. Reverse engineering of the final band gap wavelength to both avoid the materials absorption regime and match the available laser setups enables the experimental investigation of SHG generated within a band gap, such as inhibited spontaneous emission [6].



**Fig. 1** Polystyrene face-centered cubic opal: (a) Scanning electron microscopy in (111) direction, (b) Optical microscope k-space image, (e) Reflectance spectra with calculated bandgap positions (vertical lines) for different initial PS bead sizes; Barium titanate inverse fcc crystal: (c) SEM in (111) direction, (d) Focused ion beam-SEM cross-section, (f) Ultra-small angle X-ray spectroscopy 2D diffraction pattern; (g) Reflectance spectra for different inclinations with optical range color coding; All scale bars are 2 μm.

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Supramolecular polymers (SMPs) have garnered significant interest due to their inherent stimuli-responsiveness. Which imparts advanced properties such as self-healing, reprocessability, and shape memory.<sup>2</sup> However, synthesizing these materials typically requires de novo synthesis, limiting their mechanical properties. Additionally, endowing them with responsiveness to new stimuli (e.g., light) often necessitates a complete redesign of the supramolecular motifs, involving laborious synthetic efforts.<sup>3</sup> Recent developments in our lab aim to streamline the synthesis of responsive materials by either modifying commercial polymers or adopting an additive-based systems approach.

Creating supramolecular materials from commercially available polymers is a promising strategy to implement responsiveness into widely used materials, thereby increasing applicability and expanding their range of properties (Figure 1a). In our first approach, we glycol-modified polyethylene terephthalate (PETG) and end-capped with methylbenzimidazolyl)pyridine (Mebip)<sup>4</sup> tridentate ligand, in one or two steps. Metallosupramolecular polymerization yielded glassy polymers that displayed comparable stiffness to the parent polymer and excellent optical and thermal healability. Similarly, we created supramolecular networks by grafting azide-modified 6-(1'-methylbenzimidazolyl)pyridine (MBP)<sup>5</sup> bidentate ligand onto polybutadiene in a single step. The addition of metal ions (Zn<sup>2+</sup>, Ni<sup>2+</sup>, and Mg<sup>2+</sup>) yielded metallosupramolecular rubbers whose properties could be tuned by varying the metal center and the metal-to-ligand ratio.

Alternatively, by adopting a systems approach, we developed isothermally light-responsive supramolecular gels and networks (Figure 1b). These networks, which are supramolecularly crosslinked either by the self-dimerizing hydrogen-bonding ureidopyrimidinone (UPy) or metal-ligand complexes with Mebip, are typically irresponsive low-power light. By blending a photoacid generator into the system, light irradiation induces the release of hydrochloric acid, which protonates the supramolecular moieties, disrupting the supramolecular crosslinks. These isothermal transformations enable solid-to-liquid transitions by decrosslinking these networks. This approach was applied to achieve isothermal de-bonding on demand in a rapid and efficient manner. With these novel approaches to creating supramolecular polymers and systems, we aim to simplify and expand the applicability of this class of materials by enhancing their modularity and broadening the range of their properties.

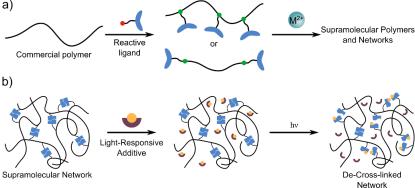
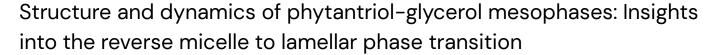


Figure 1: Schemetic representation of a) synthesis of supramolecular polymers from commercial polymers, b) systems approach to achieve light-responsive networks.

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## Matteo Rutsch, Department of Chemistry, University of Basel

Lipidic mesophases (LMPs) are lyotropic liquid crystals formed by the self-assembly of amphiphilic compounds in solvents [1]. LMPs typically use water as a solvent, offering a rich variety of phase symmetries with distinct physicochemical properties, making them valuable for both fundamental and applied research [2]. However, studies on LMPs in non-aqueous solvents are scarce [3,4] and the fundamental understanding of how the dynamics relate to the composition and structure remains limited [5,6]. In this study, we replaced water with glycerol as the solvent to form LMPs with phytantriol, a commonly used, uncharged amphiphile. Combined small-angle x-ray scattering (SAXS) and differential scanning calorimetry (DSC) reveal a weakly exothermic reverse micelle ( $L_2$ ) to lamellar ( $L_{\alpha}$ ) phase transition, which occurs at higher temperature as the glycerol content is increased. Using Broadband dielectric spectroscopy (BDS), we observed how the dynamics of phytantriol are governed by the composition and symmetry of the LMP: Increasing glycerol content decreases the relaxation time of the Debyeand α-relaxation, therefore exerting a plasticizing effect. The change in long-range order of phytantriol during the  $L_2 - L_{\alpha}$  phase transition reveals a decrease in the conductivity relaxation time. The introduction of a net orientation of phytantriol further reveals a new relaxation process—the dipole-matrix interaction—exclusive to the  $L_{\alpha}$  phase. These results highlight the value of combining BDS with structural and thermal analyses for a deeper understanding of the dynamics in soft matter systems. In addition, our developed system provides a straightforward platform to host non-aqueous reactions under soft nanoconfinement and opens new avenues for the study of biomolecular solvation.

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# ABSTRACTS POSTERS

# Enzyme-induced mineralization of calcium carbonate in 3D printable granular hydrogels

Francesca Bono<sup>1,2</sup>, Anna Puiggalí-Jou<sup>3</sup>, Lorenzo Lucherini<sup>1</sup>, Greta Cocchi<sup>1</sup>, Marcy Zenobi-Wong<sup>3</sup>, and Esther Amstad<sup>1,2\*</sup>

Many biological materials are structured as organic-inorganic composites. A paradigmatic example of this apparent dichotomy is bone, a biomineralized tissue with a polymeric matrix. Inspired by the natural design strategy, recent work has been done to encapsulate living components such as bacteria into a granular polymeric matrix to then trigger biomineralization [1]. Taking advantage of the intrinsic rheological properties of jammed microgels, organic/inorganic composites can be 3D printed. However, applications are limited if living organisms are involved.

In this work, we introduce a 3D printable inks that can be mineralized to yield load-bearing CaCO<sub>3</sub>-based composites after the 3D printing process has been completed. This is achieved by formulating enzyme-loaded hydrogels that are exclusively made of naturally sourced soft polymers as microgels. The enzyme-loaded microgels are jammed to obtain the required rheological properties. These microgel-based inks are 3D printed at room temperature through direct ink writing before they are converted into load-bearing mineralized scaffolds. We demonstrate how the local composition, mineral content, and porosity can be adjusted with the formulation of the granular ink. The obtained scaffolds are biocompatible with low cytotoxicity when tested with osteoblast cells showing great potential for tissue engineering applications such as bone reparation.

Keywords: microgels, 3D printing, enzymatically induced mineralization

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## Mycelium-Cellulosenanofibrils Hybrid Materials Matteo Darra, EPFL

## Abstract:

Cellulose nanofibers (CNFs) are a family of non-toxic, bio-based materials presenting superior properties such as large specific surface area, high strength, and low weight. Due to the numerous hydroxyl groups present on their surfaces, CNFs are suitable for a wide range of chemical modifications. These are commonly exploited to increase hydrophobicity, increase miscibility with polymers, generate crosslinks, and more generally to change CNF surface properties depending on the desired application. Amongst others, their implementation as strengthening agents and as films with oxygen barrier properties makes them appealing for the development of sustainable packaging materials. Unfortunately, due to the strong interactions of CNFs with water, certain properties can be compromised under humid conditions. Approaches such as chemical modification and various additives have been explored in recent years to modulate the water-holding capacity of CNFs. We propose an alternative approach to traditional chemical modifications based on a natural modification of CNFs with fungal mycelium to attenuate water interactions. CNFs can be incorporated into the growing mycelium network, creating a CNF-mycelium bio-nanocomposite. Here we present how the surface chemistry of the CNFs in the CNF-mycelium hybrids influences key features such as drainage rate and potential as a strength additive. It is expected that the CNF surface chemistry will influence the CNFmycelium interaction and consequently change the properties obtained.

## Non-equilibrium stabilization of proteins by chaperones

## María Li López Bautista, EPFL

Within living organisms, proteins are essential components. They are responsible for carrying out almost every function in the cell. Proteins must fold into a specific three-dimensional shape to perform their diverse roles effectively. Indeed, improper folding is the root cause of many diseases.

Not surprisingly, there is a specific group of proteins whose function is to assist and safeguard the folding process of other proteins. These are known as molecular chaperones. Here, we focus on the 70 kiloDalton heat shock protein, Hsp70, a molecular chaperone that has been under the spotlight of scientists for decades due to its ubiquitous presence across all living systems and its assistance in a wide range of cellular processes. It is well accepted that the mechanism of action of Hsp70 chaperones consists of a biochemical energy-consuming cycle, which allows them to drive the system out-of-equilibrium and escape the inherent limitations of equilibrium thermodynamics to perform their functions efficiently.

Considering both the molecular details of chaperones and their client protein, along with a correct inclusion of the energy consumed in each step of the cycle and all relevant conformational transitions, we present a kinetic rate model for the description of the functional cycle of Hsp70 chaperones in protein folding that aims to elucidate the fundamental principles that govern their complex behavior.

# Exploring the colloidal properties of *Bacillus subtilis* spores at liquid interface *YiyaoHu, ETH Zürich*

Spores, resilient dormant cells with size and shape akin to colloidal particles hold promise as novel building blocks. Their ability to endure harsh environments while retaining functionality distinguishes them from bacteria, offering exceptional tolerance for various manufacturing processes. Compared to the traditional colloidal particles, spores can become active cells in favorable conditions, introducing dynamism to the system.

While bacteria spores have found applications in biotechnology, their potential in constructing living materials has been explored only through the embedding them as additives. However, their utilization as nature particles at interfaces remains underexplored.

This study aims to fill this gap by investigating the feasibility of harnessing *Bacillus subtilis* spores as building blocks at liquid interfaces for the fabrication of dynamically evolving materials.

To comprehend the colloidal behavior of spores, zeta potential tests were conducted to assess their surface charge under various environmental conditions. Microscopy imaging was then employed to observe their self-assembly behavior at water-oil interfaces. For comparative purposes, ellipsoidal polystyrene (PS) particles with similar aspect ratios were fabricated and analyzed with the same techniques.

Our findings reveal similarities in the zeta potential curves across different pH and salt concentrations between spores and PS particles, as well as in their self-assembly patterns at the interface, as we characterized by computing radial distribution functions, orientation correlation functions, and time-dependent cluster size. Finally, we successfully demonstrated a dynamic Pickering emulsion stabilized by spores and managed to introduce bacteria growth in a Pickering emulsion.

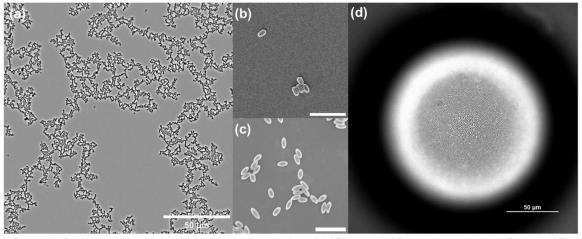
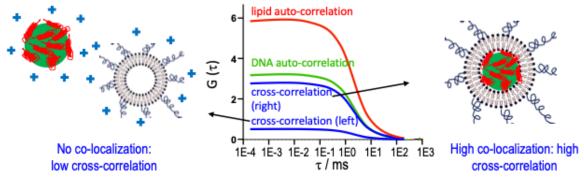


Figure. (a) Bacillus subtilis spores at water-hexadecane interface. (b) SEM image of Spores, the scale bar is 5  $\mu$ m, (c) SEM image of ellipsoid PS particle, the scale bar is 5  $\mu$ m. (d) Hexadecane-in-MilliQ droplets stabilized by PS533 spores.

## Quantifying Lipid Bilayer Coating on Polymer-Nucleic Acid Nanoparticles Using Fluorescence Cross-Correlation Spectroscopy

Juan Paris,<sup>1</sup> Ricardo Gaspar,<sup>1</sup> Filipe Coelho,<sup>1</sup> Pieter De Beule,<sup>1</sup> <u>Bruno F.B. Silva</u>,<sup>1,2,3,4</sup> <sup>1</sup> *International Iberian Nanotechnology Laboratory: Braga, Portugal.* 

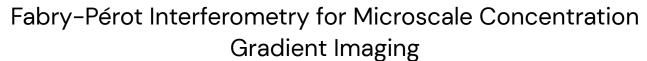
Hybrid core-shell lipid-polycation-nucleic acid nanoparticles (LPNPs) offer promising opportunities for nonviral gene delivery. However, their multi-component nature makes characterization challenging. In this study, we introduce a dual-labeled fluorescence cross-correlation spectroscopy (FCCS) method [1,2] to characterize the association between polycation-DNA cores and lipid bilayer shells [3]. By labeling lipids and polycations with distinct fluorophores, FCCS quantifies colocalization between the two components though the analysis of their spatial-temporal fluorescence correlations (Fig 1). Our results reveal that association between the cores and liposomes requires them to be oppositely charged. Additionally, we identify the liposome:core number ratio ( $P_N$ ) as a critical parameter, with  $P_N \ge 1$  being required to achieve colloidal stability. This new characterization approach provides deeper insights into LPNP assembly and stability, facilitating the design of more effective gene delivery systems.



**Figure 1.** Illustration of the use of FCCS to monitor the formation of hybrid core-shell lipid-polycation-nucleic acid nanoparticles (LPNPs). Liposomes and polycation are labelled with two spectrally-resolved dyes and their motions are analyzed simultaneously, which allows determining the individual auto- and cross-correlation functions. From the amplitude of the cross-correlation a quantitative measure of the co-localization between liposomes and cores can be obtained. Non-associated liposomes and cores have very low cross-correlation (left). LPNPs show high cross-correlation amplitudes (right).

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35th Swiss Soft Days Taieesa Peshkovsky March 17th, 2025

#### **Abstract**

Chemical concentration gradients at the microscale are omnipresent, shaping fundamental processes in a wide range of systems from cell signaling in nature, to transport phenomena in applied sciences. Despite their ubiquity, the majority of approaches to measure chemical gradients on a microscale are indirect, often relying on labels such as fluorophores or colorful dyes to visualize the compounds of interest. This technique suffers considerable limitations in that it is inherently invasive and thus impacts the observed system, it has limited temporal resolution due to photobleaching, and when used to resolve living organisms it can prove to be toxic. For systems that lack optical contrast, a label-free alternative is to use interferometric methods that relate refractive index (RI) to a compound's identity or concentration in situ. While interferometric techniques are very powerful and precise, they typically yield only pointwise data, and thus require a long time to resolve a small area. For processes that evolve on short timescales in more than one dimension, a full-field RI profile map over a two-dimensional area offers greater insight into the system at hand. Building upon a technique developed by Vogus et al [1], herein we describe a novel tool that expands on the principles of Fabry-Pérot interferometry to two dimensions, making for straight-forward and label-free concentration gradient mapping over a microfluidic chip. By tracking Fringes of Equal Chromatic Order (FECO) we achieve a RI resolution on the order of 1e-5 refractive index units (RIU), sensitive to 1 mM changes in concentration. The tool mounts onto a microscope, and the spatiotemporal resolution can be adjusted to the task at hand by exchanging the objective. For long range interactions a 4x objective magnification might be most suitable where the field of view is 4.6 mm by 4.6 mm and each pixel has a dimension of 8 μm. However, for short range interactions a 20x objective with a 0.92 mm by 0.92 mm field of view and a pixel dimension of 1.6 μm would work better. The temporal resolution is also adjustable: while a measurement at 4x takes 8 seconds, at 10x it takes 17.6 seconds, and at 20x it takes 23.8 seconds. Here we characterize the resolution and RI precision of the tool, as well as demonstrate its novel potential by mapping dynamically evolving concentration gradients between colaminar flow streams of aqueous NaCl solutions and water in a microfluidic chip.

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Butterflies have mastered producing vibrant colors using simple biological building blocks like lipids, proteins, and other metabolomes. Although several butterfly species have long mastered this colloidal self-assembly, our understanding of the responsible molecules and their self-assembling technique is still incomplete [1, 2]. Certain butterfly species produce different photonic structures [3], and my research is focused on understanding them by rearing them in a controlled environment that closely mimics their natural habitat.

The dull pupae transform into a vibrant butterfly after undergoing a metamorphosis cycle, and what happens inside the cocoon is still an enigma. The pupal stage is crucial because it represents a dramatic transformation in color, offering a unique opportunity to study. We are investigating how the interplay between chitin, proteins, and other biomolecules influences the formation of specific photonic structures during metamorphosis.

Firstly, we tried to comprehend what led to this transformation using tandem mass spectrometry and bioinformatic tools to narrow down the responsible building blocks. We will then study the interaction between these blocks and how they facilitate self-assembly to produce specific structures.

The ultimate goal of this research is to understand the self-assembly principles in biological systems better and apply this understanding to new, eco-friendly color production in cosmetics and food formulation. This can provide sustainable alternatives to toxic pigments, e.g., titanium dioxide, that are increasingly restricted.

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## Bioprinting of Stable Bionic Interfaces Using Piezoresistive Hydrogel Organoelectronics

Antonia Georgopoulou, Jakob Schreiner, Miriam Filippi, Robert Katzschmann, Esther Amstad

Bionic tissues combine artificial electronics, like sensors with live tissue. To monitor the contraction of tissue engineered muscle, soft, bioadhesive sensors are combined with a cell-laden construct. Due to the viscoelasticity of soft polymers soft sensors are susceptible to signal drift that limit their accuracy. We propose the use of double network granular hydrogels (DNGH) to combine softness with reliable sensor properties. In addition, DNGHs are compatible with 3D bioprinting, a method that facilitate the formation of stable interfaces, ensuring seamless deformation transfer from muscle to the sensor. Moreover, the composite displayed good bioadhesive properties, allowing for cells to adhere to the surface. Cell viability 87% and differentiation ability, as well as the integrity of the sensor-tissue interface, were preserved through all the culture duration. Due to its effective interfacing with soft tissue and facile fabrication method, this conductive hydrogel formulation can serve to realize various bionic tissues for biomedical applications, such as implantable electronics and organ-on-a-chip devices.



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A significant approach in various fields such as chemistry, electronics, and technology are the self-organization of nano-objects into complex architectures. This strategy aims to generate novel systems with unique properties and functionalities <sup>[1–3]</sup>. An important step in creating interconnected artificial organelles is the DNA hybridization between synthetic assemblies, including polymersomes, nanoparticles, and micelles. These assemblies facilitate cascade reactions among different encapsulated catalytic compounds and can imitate cell signaling and interactions<sup>[4,5]</sup>.

In this study, a new approach is proposed for developing a multifunctional hybrid system for specific bio-applications by investigating the self-organization of clusters between "hard" Janus nanoparticles (JNPs) and "soft" polymersomes. These polymer-based JNPs provide an asymmetric platform suited for directional interaction <sup>[7]</sup> with soft polymersomes. The clusters are modularly assembled through programmed DNA hybridization. Furthermore, the vesicular architecture of the polymersomes after assembly into JNP-polymersome clusters allows the encapsulation of various kinds of catalytic compounds. The asymmetry of the JNPs has unique advantages by allowing a precise arrangement of the polymersomes and enabling, in a modular manner, various reaction configurations, including single, parallel and cascade enzymatic reactions. Additionally, these clusters, which integrate imaging and therapeutic nanocompartments, support nanotheranostic applications by enabling precise in vitro detection and simultaneously producing reactive oxygen species (ROS) to induce apoptosis.

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## Exopolymers from the fermentation of Schizophyllum commune

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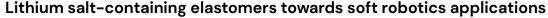
Sustainable Materials Laboratory (SML), Institute of Materials, School of Engineering, EPFL, Station 12, 1015 Lausanne, Switzerland The emergence of the bioeconomy has increased the contribution of bio-based products due to their renewable nature [1]. Among these, extracellular secretions from microbial species are of interest due to their material properties, such as oxygen barrier [2].

Schizophyllum commune (S. commune), an edible fungal species, produces extracellular substances, including the polysaccharide schizophyllan (SPG) and the mannoprotein hydrophobin. SPG has already been commercialized as a cosmetic ingredient (e.g., in a moisturizing cream from Paula's choice) [3], is actively studied for biomedicine [4], but is less explored in the field of materials science [5]. A major bottleneck in utilizing microbial-derived exopolymers is the high costs associated with conventional isolation processes (e.g., pullulan isolation by ethanol precipitation can cost up to \$1608.71 per kg of pullulan) [6, 7], as well as the use of tedious and toxic purification strategies (e.g., Sevag deproteinization) [8, 9]. Furthermore, the harsh purification methods used to isolate polysaccharides from the exopolymer mixture can deteriorate their properties (e.g., alkali extraction of fungal polysaccharides can lead to structural changes) [8]. Specifically, SPG extraction under alkaline conditions ([NaOH] > 0.25 M) can lead to the destruction of its triple helix structure [10]. In this study, submerged fermentation of S. commune is used to produce a minimally purified exopolymer stream enriched in SPG, avoiding traditional solvent-based precipitation approaches. The composition and supramolecular structure of our isolated stream are analyzed, and the effect of fermentation time is elucidated. Notably, as S. commune matures, not only does the yield of extracellular polymers change, but also their material properties, with implications for the functional applications of SPG-derived materials.



Figure 2. TEM image of minimally purified extracellular polymers enriched in SPG

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François Rivat, Esther Amstad

Soft Materials Laboratory, Institute of Materials, EPFL, Lausanne Elastomers are widely used in soft robotics due to their superior elastic modulus, toughness, and air stability compared to hydrogels. However, unlike hydrogels, elastomers lack responsiveness to external stimuli such as temperature, pH, and electric fields. To impart responsibility to elastomers, they can be functionalized with ionic liquids to form ionogels, which have improved air stability while retaining a high ionic conductivity of up to 10<sup>-2</sup> S cm<sup>-1</sup>. However, ionogels suffer from a low elastic modulus and toughness [1]. Here, I introduce a stiff and strong ionogel composed of xxxx, which is functionalized with lithium bis(trifluoromethanesulfonyl)imide (LiTFSI). The resulting materials display ionic conductivities of 10<sup>-10</sup> to 10<sup>-7</sup> S cm<sup>-1</sup> at room temperature. I envisage them to present a first step towards ionically conductive elastomers for soft robotic applications.

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## Restructuring dynamics of pastes at the yielding transition

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We investigate the yielding characteristics of an over-jammed dispersion of Carbopol microgels in propylene glycol by performing oscillatory strain experiments at fixed frequency and increasing strain amplitudes. The yielding transition is characterized by a smooth evolution of the storage and loss moduli typical for yield stress materials. By contrast, a parallel microscopic investigation of the restructuring dynamics using tracer particles reveals that the mean square displacement increases sharply at the yielding transition. A finer analysis of the distribution of the displacements reveals that not all particles move during a cycle. Our work reveals that the sharp increase of the mean dynamics at the transition is due to a combination of an increasing fraction of moving particles and an increasing displacement of the moving particles with the strain amplitude.



## Microstructured elastomers and their macroscopic properties

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The fast progress in soft robotics demands for elastic soft materials whose mechanical properties can be controlled and locally varied to design structures that can deform in a pre-defined fashion while bearing significant loads. Elastomers are hydrophobic polymeric networks and hold the potential to fulfil these requirements. Thereby, they could enable the design of soft robots that possesses high dexterity, can interact safely with humans, and adapt themselves to different environments. Unfortunately, the processing of most elastomeric liquid precursors is limited to casting, preventing localized changes in their composition and hence, mechanical properties. [3]

In my project, I introduce a new strategy to 3D print elastomers with complex structures. This is achieved by formulating elastomers as microparticles and loading them with additional elastomer precursors. [4] I will demonstrate how altering the composition of the 3D printing elastomer can unlock an extensive spectrum of mechanical properties: stiffness, toughness and fatigue. Through localized changes in the composition and properties, these materials deform in a pre-defined fashion, opening up new opportunities for soft robotics applications.

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## Bacterial extracellular DNA forms stress-hardening biofilm streamers

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In habitats with fluid flow, bacteria can secrete extracellular polymeric substances that self-assemble into biofilm streamers, slender polymer threads tethered to a surface and extending into the bulk fluid. Streamers can rapidly clog and disrupt flow in water filters, porous media or medical devices [1]. Thanks to their viscoelastic mechanical properties, they can elastically respond to quick mechanical stimuli, preserving their structure, while also adapting and dissipating persistent mechanical stresses through viscous deformation [2]. Despite their role in accelerating bacterial colonization, their properties remain poorly understood due to the lack of platforms and frameworks for systematic studies.

By integrating microfluidics, optical microscopy, numerical simulations and rheological models, we developed a microfluidic platform that enables the reproducible formation of biofilm streamers under controlled physico-chemical conditions, along with *in situ* characterization of their biochemical composition, morphology and rheology [3]. Using this platform, we discovered that extracellular DNA is the essential building block of biofilm streamers, self-assembling into millimeter-long threads under flow [4]. We demonstrated that this DNA mainly comes from explosive cell lysis, which is enhanced by antibiotic-induced genotoxic stress [4,5]. Moreover, we discovered that DNA confers biofilms streamers with a marked stress-hardening behavior, acting as an instantaneous, purely physical mechanism to increase their stiffness upon increasing mechanical stress [6]. These findings highlight the crucial structural and mechanical role of DNA in streamers, suggesting potential strategies for their removal and highlighting potential drawbacks of antibiotic treatment for infections [4,5].

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Solid-supported polymer membranes (SSPMs) formed by the self-assembly of amphiphilic block copolymers offer enhanced stability and chemical versatility compared to lipid-based membranes. They can be used to modify the surface properties of underlying substrates or serve as stabilizing platforms for (bio)macromolecules, making them highly relevant for applications in materials and life sciences. Despite their broad potential, a systematic comparison of how different deposition methods influence their internal organization and physicochemical properties remains lacking.

In this study, we compare SSPMs prepared using two widely employed methods—Langmuir monolayer transfer and solvent-assisted polymer deposition—to assess their impact on membrane morphology, thickness, wettability, roughness, and elasticity. We investigate amphiphilic di- and triblock copolymers based on poly(dimethylsiloxane) (PDMS) and poly(2-methyl-2-oxazoline) (PMOXA) to understand how polymer architecture affects membrane formation and properties. Our findings reveal that the choice of deposition method significantly influences SSPM organization, with triblock-based membranes exhibiting more pronounced structural and property variations than diblock ones.

These insights demonstrate that the selection of an appropriate deposition technique is crucial for tailoring membrane characteristics to specific applications. By identifying key advantages and limitations of each method, this study provides a framework for the rational design of SSPMs, supporting their further development for functional surface modifications, (bio)sensing, and other advanced applications.

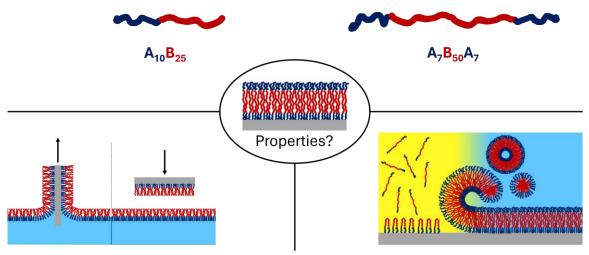


Figure 3: Schematic representation of the investigated polymer architectures and solidsupported membrane deposition methods.

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## **Enzymatic mineralization of 3D-printed granular hydrogels**

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Many biological materials are structured as organic–inorganic composites, achieving an exceptional balance of strength and toughness. A prime example is nacre, a highly resilient material composed of polygonal aragonite tablets arranged in a hierarchical 3D structure, interwoven with a biopolymeric matrix. This architecture efficiently dissipates energy, granting nacre its remarkable combination of stiffness and toughness [1]. Recent research in our lab has focused on reinforcing soft granular gels with minerals [2]. However, the load-bearing capabilities of the resulting structures remain limited due to the challenge of simultaneously achieving strength and toughness, which are typically mutually exclusive.

In this work, we introduce a hybrid composite that combines, to a certain extent, stiffness and toughness. This is achieved by formulating enzyme-loaded microgels exclusively from naturally sourced polymers. The enzyme-loaded microgels are jammed to enable their direct ink writing or casting. The resulting material is mineralized to form stiff, load-bearing scaffolds. To toughen the material, we backfill the interstitial spaces with elastomers. I will demonstrate how the composition and microstructure influence the mechanical properties of these granular materials that can be processed through direct ink writing or casting. Such bioinspired hybrid composites hold significant potential for their tunable mechanical properties, and hierarchical design, offering a promising pathway toward the development of advanced materials that bridge the gap between strength and toughness.

Keywords: microgels, enzymatically induced mineralization, elastomer, hybrid composite material

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# An Aldehyde-Stabilization Strategy for Building Bio-based Consumer Products Around Intact Lignocellulosic Structures

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The predicted shortage of fossil fuels and increased environmental concerns encourage the development of sustainable approaches for the exploitation and utilization of biomass for our daily use. Herein, we report an aldehyde-stabilization strategy for the fractionation of lignocellulose, where stabilized lignins, diacetyl-xyloses (DAXs), and cellulose are isolated as major products and find interesting applications in the modern society (Figure 1). Depending on the choices of aldehydes, the extracted lignins exhibit different properties, making them suitable for use as surfactants, adhesives, and coating materials. During the fractionation, hemicellulose is simultaneously converted into diacetyl-xyloses (DAXs), a new class of platform chemicals that are upgraded to valuable consumer products, including solvents, thermoplastic and thermoset polymers, surfactants, and food additives. Moreover, aldehyde-stabilization strategy provides a new approach for high-yielding production of sugars by tapping the sugar cores in the form of DAXs and Diacetyl-glucoses (DAGs).

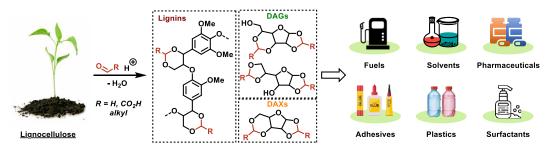


Figure 1. Aldehyde-Stabilization Strategy to produce daily consumer products from lignocellulose.

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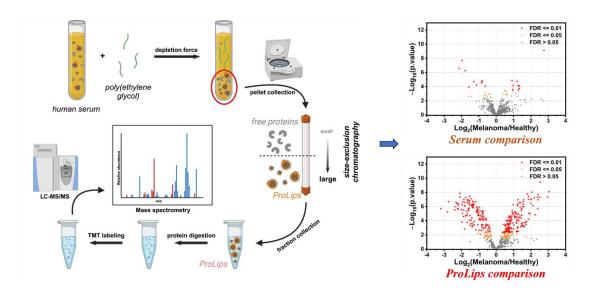


## Naturally Occurring Protein-rich Lipoprotein Nanoparticles as Tools for Melanoma Biomarkers Discovery and Detection

Ding Ren<sup>1</sup>, Francesco Stellacci<sup>1</sup>

## **Abstract**

Cancer detection from the analysis of peripheral blood samples is one of the holy grails in oncology. Protein-based detection methods are potentially rich in information on patient health but face two major obstacles: first, the identification and validation of suitable biomarkers, and second, the analytical sensitivity required to detect trace concentrations of cancer-associated proteins within the complex protein matrix of blood samples<sup>1, 2</sup>. Here we show that both hurdles can be overcome by a proteomic analysis of protein-rich lipoprotein nanoparticles (ProLips). We isolated ProLips from blood with a simple and effective method based on precipitation induced by polyethylene glycol<sup>3</sup>, redispersion, and fractionation with size-exclusion chromatography. Proteomic analysis showed that ProLips comprise a diverse variety of proteins (> 600) with markedly lower amounts of blood-abundant proteins when compared to serum proteomic profiles. Over 300 proteins were up- or down-regulated in melanoma patients (FDR < 0.05) compared to healthy patients. Through an analysis based on multi-group parallel runs across 50 samples, we identified six proteins that together are effective in serving as melanoma biomarkers. 100% accuracy in melanoma detection was achieved for nine new patients. The approach is validated by a rat model, showing that ProLips proteomics successfully distinguish melanoma at various stages.



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## Patterning Electronically Conductive Features within Soft Hydrogel Substrates

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Hydrogels, highly hydrophilic polymeric networks, have gathered significant attention in the biomedical field due to their exceptional water-retention capability, biocompatibility, and anti-biofouling properties. The mechanical properties of hydrogels can be tuned with their composition, and type and degree of crosslinking. Moreover, hydrogels exhibit excellent ionic conductivity due to their ability to swell to many times their dried volume in aqueous solutions. The synergy of these two features makes hydrogels ideal for mimicking natural soft tissues. However, their lack of electronic conductivity limits their applicability in bioelectronics. Moreover, many applications in bioelectronics would strongly benefit not only from the electrical conductivity in the hydrogel, but from the ability to pattern it, a feature that has not been demonstrated at the sub-µm length scale. In this presentation, I will present a method to fulfill these requirements. Exploiting two-photon direct laser writing (DLW), we trigger the in-situ synthesis of noble metal nanoparticles (NPs) through photoreduction. Thereby, we generate electrically conductive 50-250 µm wide tracks within natural and synthetic hydrogels possessing Young's moduli as low as 20 kPa, matching the mechanical properties of soft natural tissues. We envision this method could be used for the fabrication of hydrogel-based devices and functional substrates for tissue engineering. Lastly, the introduction of spatially-controllable conductivity within hydrogels would open new possibilities for interfacing soft materials with electronic equipment.

## Polymer-inorganic nanohybrids for NIR-light triggering

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Stimuli-responsive polymeric self-assemblies are promising drug delivery platforms offering on demand release with dose and spatiotemporal control over delivery. Thermoresponsive polymers with a lower critical solution temperature (LCST) are particularly promising as building block for such drug delivery systems. However, temperature can be used as an endogenous stimulus only in exceptional cases, creating a need for an external heating source that would trigger the thermoresponsive polymer. <sup>2</sup>

We aim to use near-infrared (NIR) light as an exogenous stimulus to trigger polymer nanocompartments (Fig. 1). NIR light is ideal for biomedical applications due to its biocompatibility, deep tissue penetration, and low phototoxicity.<sup>3</sup> To achieve this, gold nanorods (AuNRs) are conjugated with thermoresponsive block copolymers. Upon exposure to light at specific wavelengths, the AuNRs generate heat, triggering the thermoresponsive polymer compartments.

Herein, we present the synthesis of a novel thermoresponsive amphiphilic block copolymers based on polydimethylsiloxane (PDMS) and poly(di- and poly(tri(ethylene glycol) methyl ether acrylate) (PDEGA and PTEGA). Obtained block copolymers self-assemble into uniform nanocarriers in aqueous solutions and exhibit an LCST, with critical temperature spanning a range between 5 °C and 52 °C depending on block lengths. When heated above LCST, the hydrophilic PDEGA and PTEGA blocks undergo hydrophobic transition, leading to a collapse of assembled structure and cargo release. Through appropriate end-group functionalization, polymer nanocarriers are conjugated to AuNRs to construct polymer-inorganic hybrids. These nanohybrids offer a promising platform for NIR-light triggered drug release.

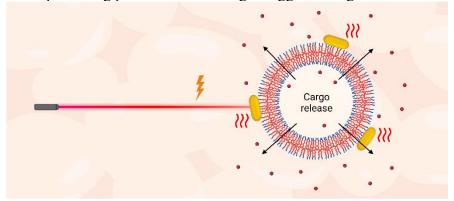
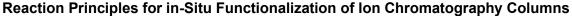


Figure 1. Triggered release from polymeric compartment conjugated with AuNRs.

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Stationary phases for ion chromatography undergo multiple reaction steps when functionalized with a grafting-from approach [1]. Reaction schemes with more than 10 steps are not uncommon [1-4] and usually carried out in labour-intensive batch processes. These processes also use big amounts of solvents and lose stationary phase during the work up. A new approach to functionalize ion chromatography materials was published by Pohl & Saini. Sulfonated styrene-divinylbenzene particles were electrostatically coated in a flow-through process in which the raw particle was packed into a 4 mm inner diameter PEEK column, followed by several reaction steps. The material was then unpacked and repacked into a column with smaller inner diameter to compensate for the material lost [4]. Our work strives to eliminate this repacking step to functionalize in-situ the final chromatography column and to generalize this approach to include covalently bonded functionalization.

Besides saving labour costs, reducing batch-to-batch heterogeneity is one of the main reasons for the new flow-functionalization approach. Steps carried out by a high precision pump and a controlled heating chamber should be more reproducible than manual synthesis in batch reactors. Additionally, recent work by Huckabee et al. [5] showed improved chromatographic performance for stationary phases functionalized after packing as a result of more homogeneous functional layers.

Raw columns of sufficient quality are necessary to avoid heterogeneous functionalisation as the result of channelling and dead volumes. Therefore, the packing process of raw particles was optimized with the systematic packing approach of Wahab et al. [6]. The resulting columns exhibited reduced plate heights of  $h_{min}(Acetone) = 3$  under non-retained iSEC conditions.

The functionalization heterogeneity along the column axis was studied with two different chemistries, namely with divinyl sulfone undergoing an oxa-Michael addition [7], as well as a thermal glycosylation of the particle with an activated and protected glucosamine [8]. Particles grafted with glucosamine appeared axially homogeneous after 36 h reaction time, whereas columns grafted with divinyl sulfone haven't been grafted homogeneously yet.

By using high quality raw columns and a non-polymerizing functionalization regime, columns can be functionalized automatically in a flow-through process with axial column heterogeneity comparable to columns produced in batch syntheses.

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## Computer-generated disordered networks for photonic bandgap

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A photonic bandgap (PBG) is a range of frequencies in which no propagating states of light exist inside a dielectric medium. PBGs can be engineered by dielectric arrangements that are structured on the length scale of the light's wavelength [1]. Many biological species, from plants to insects, use this mechanism to produce structural color through periodically ordered or amorphous networks [2]. These networks can be characterized by their coordination number – the number of edges joined at each vertex. Photonic band gaps in ordered networks are well understood but show an intrinsically anisotropic optical response. Specifically, the diamond network with coordination number 4 exhibits a large bandgap [3].

Since nature also employs other network geometries to create structural color, such as the chiral gyroid with coordination number 3 and its disordered variations [4], we analyze the effects of coordination number and disorder on photonic band gaps. To this end, we extend an established Monte Carlo algorithm for disordered network generation by altering the conventional bond length and bending energy and enabling arbitrary coordination numbers. We then investigate the statistical properties of the generated amorphous networks to obtain proper simulation parameters [5][6].

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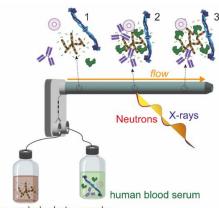
## Elucidating Interactions between Iron-Carbohydrate Complexes and Human Blood Serum using Small-Angle Scattering

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Intravenous (IV) iron-carbohydrate complexes (ICCs) are cornerstone nanomedicines for the treatment of iron deficiency anaemia, often associated with various medical conditions including chronic kidney disease or heart failure<sup>1</sup>. Though extensive physicochemical characterisation data and clinical studies are available for these nanoparticles (NPs), a clear evidence-based correlation between physicochemical properties and clinical results remains elusive. Studies on similar NPs suggest that early interactions with blood upon IV administration are crucial to understanding how physicochemical differences in ICCs influence bio-response and ultimately clinical efficacy<sup>2</sup>.

This work therefore investigated the nano-structure and surface morphology of two clinically used ICCs, iron sucrose (IS) and ferric carboxymaltose (FCM), and dynamics of their interaction with human blood serum using small-angle scattering. With this approach, the nano-structure of IS and FCM was defined in detail, revealing striking differences in cluster and surface morphology<sup>3</sup>. The differences in surface morphology were hypothesised to guide the dynamic



Iron-carbohydrate complex

ICCs with human blood serum, as IS formed agglomerates with serum components, while FCM was stabilised by adsorption of specific serum components. These important differences in nano-bio interactions may influence bioresponse of ICCs such as macrophage uptake and clearance kinetics in serum and sets a prerequisite towards complete correlation between physicochemical properties of ICCs and their clinical outcome.

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# Solid-supported triblock copolymer mixtures as a platform for targeted molecule attachment and biosensing

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Solid-supported planar membranes made of amphiphilic triblock polymers can replicate the structure and function of natural biological membranes, making them useful in biosensing and medical applications. When a membrane is composed of two materials, e.g. triblock copolymers with different hydrophobic and hydrophilic blocks, we observe lateral phase separation. The phase separation depends on the ratio of two triblocks. For example, triblock copolymers PMOXA<sub>12</sub>-b-PDMS<sub>40</sub>-b-PMOXA<sub>12</sub> (SK100) and PEO<sub>45</sub>-b-PEHOX<sub>30</sub>-b-PEtOz<sub>14</sub> (DTP10) show lateral phase separation. Furthermore, the hydrophilic top ends of each material can be functionalized with different end groups, allowing us targeted attachment of chemical moieties, thus enabling biosensing of suck polymer mixtures.

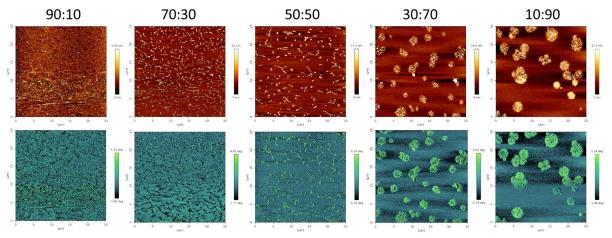


Figure 1: AFM images of different mixtures of triblock amphiphilic polymers, PMOXA $_{12}$ -b-PDMS $_{40}$ -b-PMOXA $_{12}$  (SK100) and PEO $_{45}$ -b-PEHOX $_{30}$ -b-PEtOz $_{14}$  (DTP10). With the increasing content of DTP10 copolymer (from left to right), lateral phase separation is observed in the shape of islands or patches. These domains are composed of SK100 copolymer, where the hydrophobic part of the copolymer is flexible PDMS. In contrast, the hydrophobic part of the continuous phase of copolymer DTP10 is more rigid PEHOX, which also has a side chain. Dimensions of the images:  $25x25 \, \mu m$ .

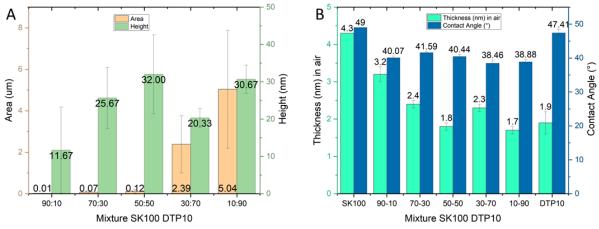
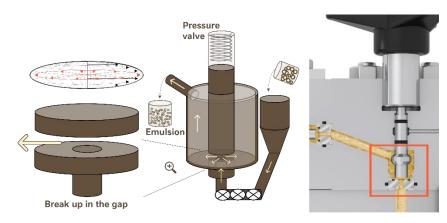


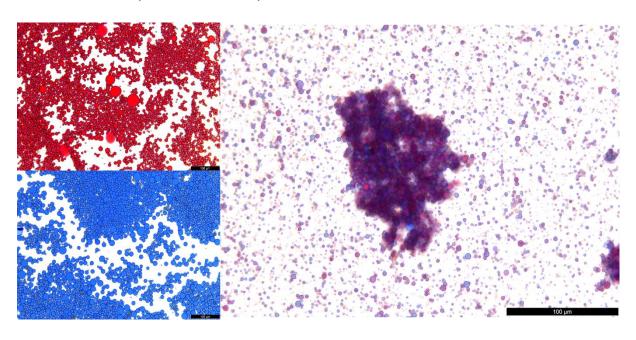
Figure 2: A) Size and height of patches of different mixtures determined from 25x25 μm AFM images. B) Thickness and contact angle of patches of different mixtures and pure SK100 and DTP10 polymers.

# Hydrodynamics of plant based emulsion breakup, recoalescence & aggregation in homogenizers



## Carol Bouvard, Nestlé

Homogenizers were originally designed for cow milk and require adaptation for plant-based milk alternatives to prevent droplet aggregation and coalescence during emulsification. A deeper understanding of the homogenization process and emulsion droplet interactions is crucial for improving stability and texture. Our study highlights the critical role of protein size and hydrophobic sites in aggregation behavior, while thin-film balance measurements reveal that larger protein aggregates contribute to droplet interactions. These aggregates not only prevent droplet coalescence but may also bridge the two interfaces by interacting with adsorbed proteins. By exploring plant-based protein adsorption mechanisms, we can enhance the formulation and performance of plant-based milk alternatives.





## Jellyfish Soft Matter: Transforming Living Hydrogels into Culinary and Biomaterial Innovations

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#### **ABSTRACT**

Jellyfish, one of Earth's oldest multi-organed organisms, are living hydrogels composed of approximately 95–98% water and with only about 1% biological matter. These organisms have evolved into a globally abundant species that can impose significant ecological and economic challenges in coastal areas. In traditional Asian cuisines, jellyfish are enjoyed as a delicacy after preservation in alum salt – a process that transforms their naturally swollen, gel-like structure into a distinctive crunchy-elastic texture. However, the conventional salted product's high aluminum content and unique texture may limit its appeal in other culinary contexts [1, 2].

Inspired by soft matter physics – particularly the collapse behavior of polyelectrolyte gels in poor solvents – we have developed a method to convert jellyfish into a crispy product without. Under controlled conditions (e.g., ethanol concentrations of approximately 60–70%, corresponding to a solvent polarity threshold of around 12.2–12.9 MPa^1/2), the jellyfish mesoglea undergoes a controlled collapse, leading to the formation of a finely structured network that yields a crisp texture upon drying [2, 3]. Moreover, by leveraging the naturally high water-binding capacity and structural features of jellyfish, we demonstrate that a simple extraction can function as a multifunctional structuring agent. This fraction exhibits Pickering-like stabilization properties and, under appropriate physicochemical conditions, can template oleogel formation and promote foaming [4, 5].

While our methods are promising for gastronomic applications – both in tailoring jellyfish textures and in designing novel food matrices with jellyfish as a structuring agent, challenges remain, including scalability. Collectively, these studies highlight that jellyfish embody rich soft matter characteristics that not only can challenge our understanding of biological gels but also offer innovative avenues for food and biomaterial applications.

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## Lucien Cousin, ETH Zürich

Dynamic covalent networks have emerged as a promising new class of materials combining the structural integrity of thermosets with properties usually specific to thermoplastics, with applications ranging from reprocessable materials to the thermal stabilization of biologics [1-2]. The emergent mechanical properties are controlled to a large extent by the thermodynamic and kinetic properties of the bond [3]. However, a quantitative understanding of some of the classic properties of dynamic polymer networks, such as elasticity and the gelation threshold, is still lacking. In this work, we investigate the rheology of a model network formed by 4-arm PEG stars cross-linked by dynamic covalent bonds. We show that the elasticity of such networks is determined by the proportion of formed bonds, p. We observe striking discrepancies between our experimental results and previously developed models concerning the elasticity and the gelation threshold. We show that the ability of the network to rearrange after formation leads to a different internal structure. This structure results from the maximization of the entropy of the network itself, independently of the conformational entropy of the single polymer chains. This, combined with previously published models, allows us to explain the delayed gelation and to quantitatively predict the elasticity of dynamic networks. We obtain similar results with networks formed by 8- arm PEG stars, showing the universality of our approach. The changes in mechanical properties due to network entropy maximization are irreversible, implying that reprocessable materials based on dynamic bonds can be permanently altered by reprocessing, even without damage to the bonds. We show this experimentally by triggering bond exchange in otherwise permanent covalent networks: depending on p we observe irreversible liquefaction or stiffening of the network. Overall, our results underscore the critical role of network entropy, distinct from chain entropy, in defining the properties of dynamic polymer networks.

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