SWISS SOFT DAYS

19th edition

Friday, 2nd of September 2016

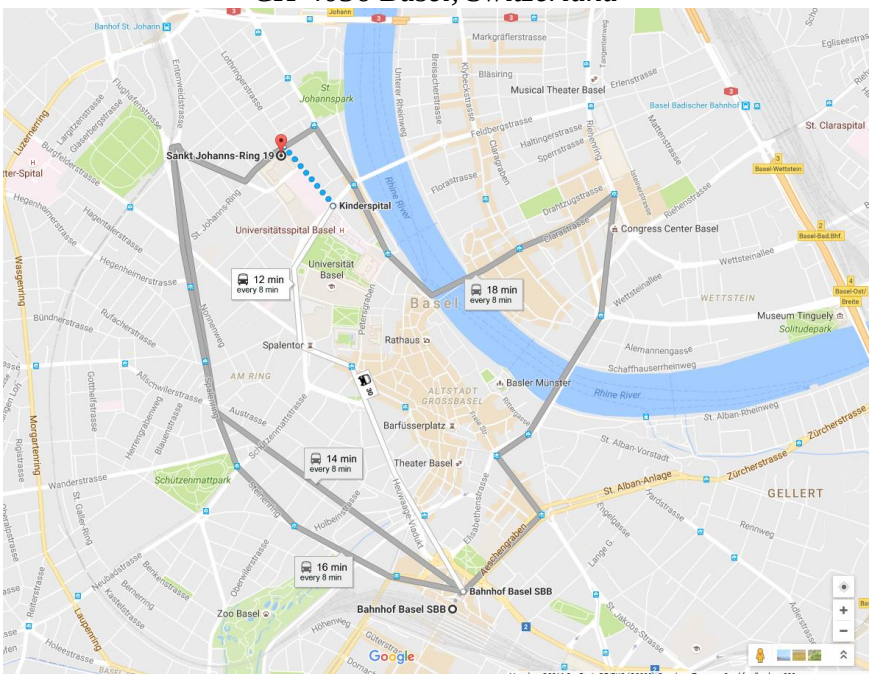
Basel

Welcome to Swiss Soft Days meeting in Basel!

Access Plan:

The conference venue takes place in University of Basel:

Department of Organic Chemistry, St.Johanns- Ring 19
CH-4056 Basel, Switzerland



<u>How to get there from Basel central train station:</u>

- You will need a ticket for Basel city-zone 1

1) **Bus 30** towards Basel Badischer Bahnhof, bus stop Kinderspital, then northwest to Spitalstrasse, turn left onto St-Johanns Ring. Approximately 12min.
2) **Bus 50** towards EuroAirport, bus stop Kannenfeldplatz, then southeast to Kannenfeldstrasse, continue to Metzerstrasse, turn left onto St. Johans ring and the destination is on the right. Approx. 15min.
3) **Tram 11** towards St-Louis Grenze, stop St-Johanns Tor, then southwest to St-Johanns Ring and the destination in on the left. Approx.18min.
4) **Tram 1** towards Dreirosenbrücke, stop Kannenfeldplatz, then southeast to Kannenfeldstrasse, continue to Metzerstrasse, turn left onto St. Johans ring and the destination is on the right. Approx. 18min.

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Program 19th Swiss Soft Days – 02.09.2016

09:00-10:00: **Welcome Coffee/Registration**

10:00-10:10: **Welcoming Remarks**

10:10-10:55: **Soft Sensitive Matter**: Structure, Dynamics, and Function of Supramolecular Polymer Gels (Sebastian Seiffert, Uni Mainz)

Session 1: Soft Matter Physics
11:00-11:20: Nanoparticle Transport and Nanostructure Assembly with Brownian Motors (Christian Schwemmer, IBM Zürich)
11:20-11:40: Dynamical and Structural Signatures of the Glass Transition in Emulsions (Chi Zhang, Uni Fribourg)
11:40-12:00: Particle Aggregation in the Presence of Mulitvalent Co-Ions and the Inverse Schulze-Hardy Rule (Gregor Trefalt, Uni Geneva)
12:00-13:30: **Lunch + Poster Session**

Session 3: Biomimetic Materials

13:30-14:15: Self Assembling Biohybrid Materials (Corinne Vebert-Nardin, Université de Pau)
14:15-14:35: Covalent Modification of Synthetic Hydrogels with Bioactive Proteins via Sortase-Mediated Ligation (Kasper Renggli, ETH Zürich)
14:35-14:55: Novel Small Molecule Heparin Mimics as Lead Therapeutics for the Treatment of Inflammatory Lung Diseases (Ioana Craciun, Uni Basel)
14:55-15:15: Active Surfaces Engineered by Immobilizing Protein-Polymer Nanoreactors for Selectively Detecting Sugar Alcohols (Xiaoyan Zhang, Uni Basel)
15:15-16:00: **Afternoon Coffee + Poster Session**

Session 2: Synthetic Materials in Nanotechnology

16:00-16:20: Using Biocatalytic Polymerizations to Detect and Quantify Blood-related Diseases (Nico Bruns, AMI Fribourg)
16:20-16:40: Curvature-induced Wrinkling Patterns and Defect Formation in Elastic Bilayer Systems (Norbert Stoop, MIT)
16:40-17:00: Surfactant –free Janus Nanoparticles Synthesis and Their Application in Pickering Emulsions (Dalin Wu, ZHAW)

17:00-17:05 **Closing remarks**

Invited Lecture

Soft Sensitive Matter: Structure, Dynamics, and Function of Supramolecular Polymer Gels

Sebastian Seiffert

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Supramolecular polymer gels consist of polymer chains connected by non-covalent interactions [1]. These materials are promising for a plethora of applications, ranging from adaptive and self-healing scaffolds [2] to designed extracellular matrixes [3]. To truly exploit the utility of these gels, it is necessary to understand the interplay between their structure, dynamics, and properties [1]. We address this challenge on the basis of macromolecular toolboxes derived from the same precursor polymer but functionalized with different crosslinkable motifs, thereby exhibiting greatly varying strength of association without perceptible alteration of other parameters [4, 5]. We find that a prime factor of impact on the mechanics and responsive performance of the resulting supramolecular gels is their nanometer-scale polymer-network architecture. To specifically account for this circumstance, we prepare our sample platforms such to either exhibit irregular, heterogeneous distributions of their supramolecular crosslinking nodes [4] or model-type, homogeneous and regular supramolecular chain interconnection [5]. These different gels are then further studied to correlate their macroscopic properties to their supramolecular binding–unbinding equilibria [4–6], nano- and mesoscopic polymer-network topologies [4, 5], and chain+junction dynamics [7, 8] on multiple scales of length and time [9].

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[7] S. Hackelbusch, T. Rossow, P. Van Assenbergh, S. Seiffert, *Macromolecules* **2013**, *46*, 6273.
[8] T. Rossow, A. Habicht, S. Seiffert, *Macromolecules* **2014**, *47*, 6473.
[9] S. Seiffert, *Macromol. Rapid Commun.* **2016**, *37*, 257.

Talks

Nanoparticle Transport and Nanostructure Assembly with Brownian Motors

C. Schwemmer[1], S. Fringes[1,2], C. Rawlings[1], A. W. Knoll[1]

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Molecular motors play a crucial role in the directed transport of molecules inside cells [1,2]. There has been a remarkable research effort on both the underlying principles and the technical implementation of artificial molecular or so-called Brownian motors over the past few years [3]. The main requirements for a functioning Brownian motor are a spatially asymmetric potential and an unbiased external driving force which brings the system out of thermodynamic equilibrium.

We have developed an artificial Brownian motor for the transport of 60 nm spherical gold particles inside a nanofluidic slit. First a 3D ratchet topography is written in one of the confining surfaces of the slit using thermal scanning probe lithography [4]. The topography then translates into an asymmetric effective potential by means of the interaction between the negatively charged surface of the gold nanoparticles and the negatively charged surfaces in the nanofluidic slit. The system can be driven out of thermal equilibrium by applying an oscillating external electric field which, as a result, leads to particle transport. The trajectories of the particles inside the slit are measured by means of interferometric scattering detection. This technique provides a spatial resolution of < 10 nm and a temporal resolution of < 2 ms [5]. In recent experiments, we observed drift velocities of 100 microns per second for the gold nano particles. In these experiments, it has also been observed that the performance of directed particle transport strongly depends on the particle size and the gap distance of the slit.

To demonstrate the utility of our motor design, we positioned 60 nm gold nanoparticles with a pitch of 200 nm to form the IBM logo by delivering the particles to an assembly site, see Fig. 1. In detail, a well-defined number of nanoparticles were transported by a ratchet into a compartment which confined the particles and contained 10 nm deep holes to trap the particles at the intended positions. After some diffusion time, the particles were trapped in the holes and could be immobilized on the surface with a positioning accuracy (1σ) of less than 10 nm.

In the next step, we plan to develop a fast and highly selective particle sorting device by exploiting the observed dependence of ratchet behavior on particle geometry. This device would be able to sort a wide range of charged nano particles e.g., carbon nanotubes or DNA.

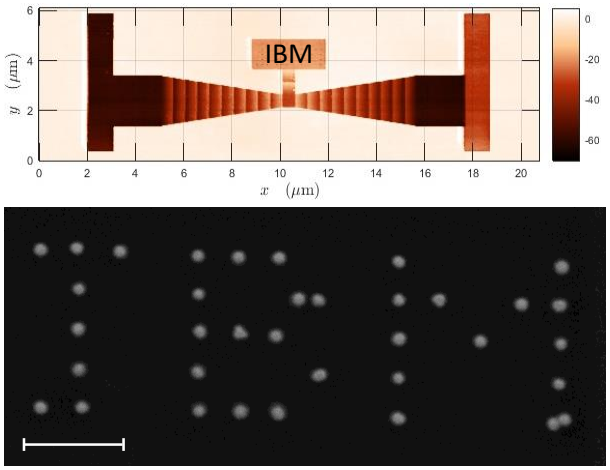

Fig. 1: (upper part) AFM picture of the experimental structure used to assemble the IBM logo. (lower part) SEM picture of the assembled nano particles. The scale bar is 500 nm.

REFERENCES: [1] Kodera et al., Nature **2010**, 468, 72, [2] Vale et al., Science **2000**, 288, 88, [3] Hänggi et al., Rev. Mod. Phys, **2009**, 81, 387, [4] Pires et al., Science **2010**, 328, 732, [5] Fringes et al., J. Appl. Phys. **2016**, 119, 024303

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Dynamical and structural signatures of the glass transition in emulsions

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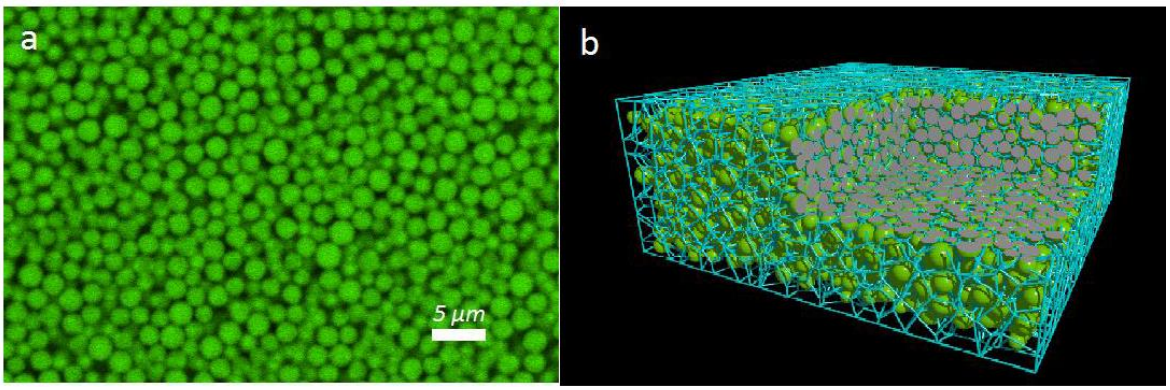

We investigate structural and dynamical properties of moderately polydisperse emulsions across anextended range of droplet volume fractions φ, encompassing fluid and glassy states up to jamming. Combining experiments and simulations, we show that when φ approaches the glass transition volume fraction φg, dynamical heterogeneities and amorphous order arise within the emulsion. In particular, we find an increasing number of clusters of particles having five-fold symmetry (*i.e.* the so-called locally favoured structures, LFS) as φ approaches φg, saturating to a roughly constant value in the glassy regime.

However, contrary to previous studies, we do notobserve a corresponding growth of medium-range crystalline order; instead, the emergence of LFS is decoupled from the appearance of more ordered regions in our system. We also find that the static correlation lengths associated with the LFS and with the fastest particles can be successfully related to the relaxation time of the system. By contrast, this does not hold for the length associated with the orientational order. Our study reveals the existence of a link between dynamics and structure close to the glass transition even in the absence of crystalline precursors or crystallization. Furthermore, the quantitative agreement between our confocal microscopy experiments and Brownian dynamics simulations indicates that emulsions are and will continue to be important model systems for the investigation of the glass transition and beyond.

Fig. 1: (a) A two-dimension confocal image of the emulsion droplets. (b) A three-dimension reconstructed image. Green spheres represent the oil droplets; Cyan rods show the Voronoi radicaltessellation.

This research was supported by the Swiss National Science Foundation through project number 149867 (ZC, FS). NG and EZ acknowledge support from MIUR ("Futuro in Ricerca" ANISOFT/RBFR125H0M) and from the European Research Council (ERC Consolidator Grant 681597, MIMIC). TGM acknowledges support from UCLA.

Particle Aggregation in the Presence of Mulitvalent Co-Ions and the Inverse Schulze-Hardy Rule

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More than a century ago Schulze and Hardy discovered that multivalent counterions strongly affect the aggregation of charged particles in suspensions.[1,2] The theoretical explanation of this behaviour was put forward by Derjaguin, Landau, Vervey, and Overbeek (DLVO) in their seminal works.[3,4] The DLVO theory assumes that the sum of double layer and van der Waals forces governs the stability of colloidal suspensions and determines the critical coagulation concentration (CCC). This assumption enabled the derivation of the *Schulze-Hardy* rule as $\mathrm{CCC} \propto 1/z^6$, where z is the valence of the counter-ion and particles are highly charged. The mirror situation where multivalent ions act as co-ions received much less attention. In this presentation, we propose an analogous *inverse Schulze-Hardy* rule $\mathrm{CCC} \propto 1/z$, where z represents the valence of multivalent co-ion. This rule is based on experimental observations where the aggregation of latex particles in the presence of multivalent co-ions is measured by light scattering techniques, see Fig. 1.[5] Highly charge sulfate latex particles follow the 1/z dependence, while the dependence for the less charged amidine latex particles is weaker and closer to $1/z^2$ (Fig. 2). The latter case can be understood with the Debye-Hückel theory, which is valid for particles with lower charge. We further show that it is possible to theoretically derive the *inverse Schulze-Hardy* rule by understanding the soft and long-range interactions present in the multivalent co-ion systems.[6]

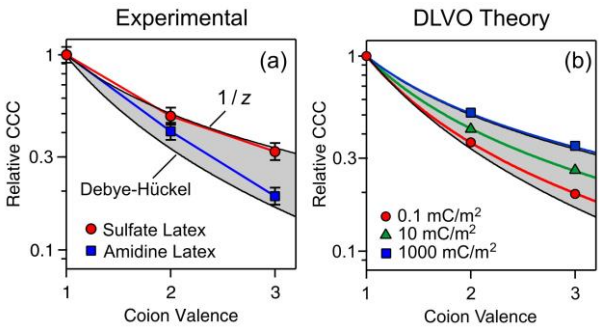

Fig. 1: Dependence of the CCC on the co-ion valence for sulfate and amidine latex particles (left). The DLVO calculations of the CCC for different surface charge densities of the particles (right).

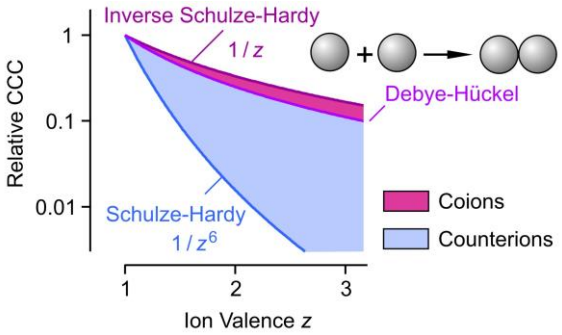

Fig. 2: Dependence of the CCC on the ion valence. For highly charged surfaces the Schulze-Hardy rule is reached for the multivalent counter-ion case, while the inverse Schulze-Hardy rule is reached for the multivalent co-ion case. If the surfaces are weakly charged the Debye-Hückel dependence is valid in both cases.

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SELF-ASSEMBLING BIOHBRID MATERIALS

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Abstract: The synergetic combination of synthetic and natural materials is an exciting topic of research since, on one hand, the theories and methods developed in polymer science might enable the establishment of the mechanisms of structure formation and modes of interaction of biological polymers and macromolecular self-assemblies thereof. On the other hand, the biological soft matter is a source of inspiration to develop materials of academic interests and of high potential for various applications in particular in biology and medicine.

DNA-copolymer self-assembly and the formation of interpolyelectrolyte complexes between nucleic acids and proteins are two examples which illustrate this duality. Self-assembling DNA-copolymers, which are composed of a nucleic acid sequence coupled to a hydrophobic polymer, do self-assemble in dilute aqueous solution and undergo crystallization in thin films like their fully synthetic counterpart. Of higher interest though is that these hybrids do assemble into functional structures, i. e. the nucleic acids engaged in self-assembly retain their capacity of hybridization or of specific recognition. These hybrids are especially designed to develop vectors for either sustained, targeted drug delivery or for gene therapy. Fine tuning of the DNA-copolymer composition to enhance intra-molecular interactions between nucleic acid grafts and the synthetic polymer backbone of a graft/comb DNA-copolymer of suitable composition enabled mimicking the nucleation polymerization into fibrillar structures according to which amyloid proteins self-assemble. These proteins have been identified in several neurodegenerative diseases. Prevent or disrupt their pathogenic macromolecular assembly is the current approach to identify a potential future cure. However, although the analogy between nucleic acids or polypeptides and charged polymers is obvious, few reports in the literature address the formation of interpolyelectrolyte complexes between nucleotide and peptide sequences to prevent or disrupt the macromolecular assembly of amyloids. Interpolyelectrolyte complex formation is an established mechanism in polymer science, which we are exploring to counterbalance the pathogenic association of amyloid proteins.

COVALENT MODIFICATION OF SYNTHETIC HYDROGELS WITH BIOACTIVE PROTEINS VIA SORTASE-MEDIATED LIGATION

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INTRODUCTION: Synthetic extracellular matrices are widely used in regenerative medicine and as tools to represent the physiological features of tissue architecture in vitro.1 The progress in biomaterials to include tailored functional peptide sequences and growth factors in synthetic scaffold and hydrogels increased biocompatibility and supported long-term cell encapsulation.2 Indeed, the incorporation of biochemical and mechanical cues provides a useful toolbox for mimicking properties of the extracellular environment and permit functionalization of the matrix with active sites for cell adhesion,3 matrix degradation4 and matrix stiffness modulation.5 Moreover, growthfactors are essential signaling molecules that affect cell migration, proliferation and differentiation in a strictly regulated spatiotemporal manner.2 Growth factors have a critical role in tissue regeneration and are frequently used in tissue engineering for (i) release and diffusion into adjacent tissues, (ii) incorporation to a matrix via covalent conjugation or (iii) demanded release of the growth factor upon external stimulus.6 However, synthetic hydrogels are challenging to modify with large peptides or proteins.

RESULTS: Sortase-mediated ligation was used to conjugate human epidermal growth factor (EGF) engineered with a GGG ligation motif (GGG-EGF) to poly(ethylene glycol) (PEG) hydrogels containing the sortase LPRTG substrate (see Figure 1).7

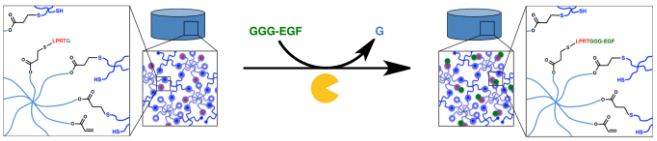

Fig. 1: Sortase-mediated ligation of GGG-EGF topre-formed PEG hydrogels containing LPRTG peptide. Reprinted with permission from 7.
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The reversibility of the sortase reaction was then exploited to cleave tethered EGF from the hydrogels for quantification. Analyses of the reaction supernatant and the post-ligation hydrogels showed that the amount of tethered EGF increases with increasing LPRTG in the hydrogel or GGG-EGF in the supernatant. Sortase-tethered EGF was biologically active, as demonstrated by stimulation of DNA synthesis in primary human hepatocytes and endometrial epithelial cells.

CONCLUSIONS: Here, a facile, mild enzymatic post-grafting approach is presented. The simplicity, specificity, and reversibility of sort as emediated ligation and cleavage reactions make it an attractive approach for modification of hydrogels.

REFERENCES: 1 L. G. Griffith, M. A. Swartz; **2006**; *Nat. Rev. Mol. Cell Biol.;* 7:211–224. 2 D. E. Discher, D. J. Mooney, P. W. Zandstra; **2009**; *Science;* 324:1673–1677. 3 M. B. Rahmany, M.Van Dyke; **2013**; *Acta Biomater.;* 9:5431–5437. 4 M. P. Lutolf, et al.; **2003**; *Proc. Natl. Acad. Sci.;* 100:5413–5418. 5 F. Brandl, F. Sommer, A. Goepferich; **2007**; *Biomaterials;* 28:134–146. 6 K. Lee, E. A. Silva, D. J. Mooney; **2011**; *J. R. Soc. Interface;* 8:153–170. 7 E. Cambria, K. Renggli, C. C. Ahrens, C. D. Cook, C. Kroll, A. T. Krueger, B. Imperiali, L. G. Griffith; **2014**; *Biomacromolecules*; 16:2316–2326

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Novel Small Molecule Heparin Mimics as Lead Therapeutics for the Treatment of Inflammatory Lung Diseases

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INTRODUCTION: Neutrophil serine proteases (NSPs) play an important role in the innate immune system. However, when the balance between NSPs and their endogenous protease inhibitors (PIs) is disrupted, they also play a critical role in the pathogenesis of inflammatory lung disease (Fig 1.).[1] Excessive release of NSPs such as human neutrophil elastase (HNE), proteinase 3 (Pr3) and cathepsin G (CatG), leads to destruction of the lung matrix and continued propagation of acute inflammation.[2-3] Under normal conditions, endogenous PIs counteract these effects by inactivating NSPs. In inflammatory lung diseases, including chronic obstructive pulmonary disease, cystic fibrosis, emphysema, and acute lung injury, there are insufficient levels of endogenous PIs to mitigate damage. Therapeutic strategies are needed to modulate excessive NSP proteolytic activity in conditions of inflammatory lung disease, in order to restore the NSP-endogenous PI balance and decrease the inflammatory response.

METHODS: A chromogenic peptidolytic assay was employed to screen a recently synthesized panel of *N*-arylacyl *O*-sulfonated aminoglycosides for their ability to inhibit each of the NSPs. The inhibitory profile of each *N*-arylacyl *O*-sulfonated aminoglycoside with respect to HNE, CatG and Pr3 was characterized. The *N*-arylacyl *O*-sulfonated aminoglycosides were also evaluated for their ability to inhibit the proteolytic activity of the three NSPs in a cell based assay that evaluates the ability of test compounds to inhibit NSP-mediated detachment of A549 lung epithelial cells from the surface of a 96-well plate. A mouse model for acute pulmonary toxicity and inflammation was used for evaluating the potential of one lead compound to decrease LPS-induced inflammation.

RESULTS: *N*-carbobenzyloxy *O*-sulfonated kanamycin was identified as a novel inhibitor of HNE, CatG and Pr3, and *N*-carbobenzyloxy *O*-sulfonated neomycin was shown to be a potent dual inhibitor of HNE and CatG. NSP-mediated proteolytic detachment of A549 lung epithelial cells was significantly inhibited by both test compounds in the case of each protease. Finally, the ability of *N*-carbobenzyloxy *O*-sulfonated

kanamycin to inhibit the three NSPs and restore the protease-protease inhibitor imbalance was explored and confirmed using *in vivo* mouse model for acute lung toxicity and inflammation.

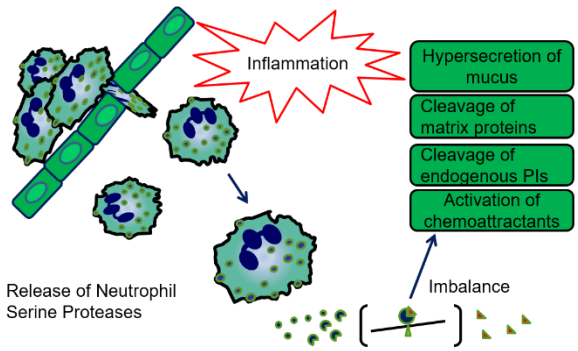

Fig. 1: Role of neutrophil serine proteases in inflammation

DISCUSSION & CONCLUSIONS: Several of the compounds screened had moderate activity towards all three NSPs, thus allowing us to construct initial structure–activity relationship models. This proof-of-concept study demonstrates that *N*-arylacyl *O*-sulfonated aminoglycosides, and likely other similar *N*-arylacyl *O*-sulfonated saccharide-based scaffolds, can be exploited as a promising new type of lead structure. The results obtained in the *in vivo* studies are encouraging considering that *N*-carbobenzyloxy *O*-sulfonated kanamycin was identified using *in vitro* screens as a modest inhibitor of the three NSPs and was not further optimized prior to the *in vivo* studies, yet it was able to decrease LPS-induced acute pulmonary toxicity and inflammation. This successfully demonstrates that *N*-arylacyl *O*-sulfonated aminoglycosides, as novel unique molecules, merit further exploration as potential lead therapeutics for the treatment of inflammatory lung diseases.

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ACKNOWLEDGEMENTS: Robert Kerns and Kerns Lab, Peter Thorne, Meng Wu and University of Iowa HTS Facility.

Active Surfaces Engineered by Immobilizing Protein-polymer Nanoreactors for Selectively Detecting Sugar Alcohols

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INTRODUCTION: Sugar alcohols (*e.g.* D-sorbitol and D-mannitol) are essential metabolic intermediates involved in biological processes. The detection and concentration of sugar alcohols in aqueous solutions is necessary for a variety of applications, in biochemical, medical or even industrial domains. An elegant way to detect specific molecules with high sensitivity is by designing nanoreactors, which encapsulate active molecules that sense their presence inside polymer supramolecular assemblies (polymersomes, micelles, dendrimers, and capsules) with sizes in the nanometer range.

METHODS: First, such selective nanoreactors were engineered in solution by simultaneous encapsulation of specific enzymes (ribitol dehydrogenase, RDH) in copolymer polymersomes (poly(2-methyloxazoline)-*block*-poly(dimethylsiloxane)-*block*-poly(2-methyloxazoline, PMOXA-*b*-PDMS-*b*-PMOXA), and insertion of membrane proteins (*E. Coli.* glycerol facilitator, GlpF) for selective conduct of sugar alcohols. Ribitol was selected as a model sugar alcohol. Second, to obtain "active surfaces" for detecting sugar alcohols, the nanoreactors optimized in solution were then immobilized on a solid support: aldehyde groups exposed at the compartment external surface reacted *via* an aldehyde-amino reaction with glass surfaces chemically modified with amino groups.

RESULTS: Despite the artificial surroundings, and the thickness of the copolymer membrane, functionality of reconstituted GlpF was preserved, and allowed selective conduct of sugar alcohols to the inner cavity of the polymersome, where encapsulated RDH enzymes served as biosensing entities. After immobilizing the polymer nanoreactors onto modified glass surfaces, the nanoreactors preserved their architecture and activity, and represent active biosensing surfaces for selective detection of sugar alcohols, with high sensitivity. As the enzyme reaction produces NADH, which is fluorescent (with an emission wavelength at 445 nm), it was used as a probe to evaluate the activity after immobilization of polymer nanoreactors with inserted GlpF on a solid support by confocal laser scanning microscopy (CLSM) and UV/Vis spectroscopy. CLSM micrographs of the active surface showed a bright fluorescence of the surface (Fig. 1 left). The cross scratch part of the surface, where no nanoreactors were present after rinsing with water, showed no fluorescence. In addition, UV/Vis spectroscopy was used to evaluate the active surface performance: a linear biosensing reply was obtained for milli-molar range of ribitol, in agreement with the performance of our polymer nanoreactores in solution (Fig. 1 right).

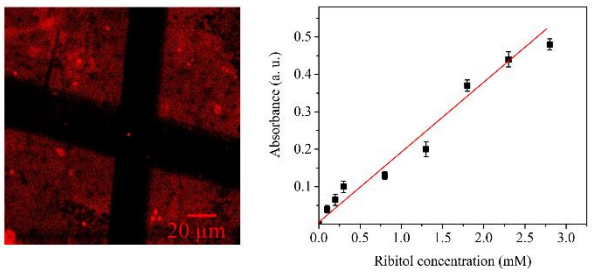

Fig. 1: CLSM micrographs of the active surface with immobilized nanoreactors with inserted GlpF (left) and absorbance intensity of NADH produced by the enzymatic reaction inside nanoreactors with GlpF immobilized on the interior wall of a glass cuvette, in the presence of 0.0 -3.0 mM ribitol (right) (±SD, n=3).

DISCUSSION & CONCLUSIONS: We have developed "active surfaces" for selective biosensing of sugar alcohols with time and space precision based on immobilization of nanoreactors with specific proteins inserted in their membranes, and sugar alcohol sensitive enzymes encapsulated in their cavities. Their immobilization on solid supports provides efficient "active surfaces" due to the rapid change in their fluorescence intensity in the presence of sugar alcohols.

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ACKNOWLEDGEMENTS: We gratefully acknowledge the financial support provided by the Swiss National Science Foundation, the Swiss Nanoscience Institute, and the National Centre of Competence in Research "Molecular Systems Engineering".

USING BIOCATALYTIC POLYMERIZATIONS TO DETECT AND QUANTIFY
Blood-related Diseases

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INTRODUCTION: Blood-related diseases such as malaria and haemolytic anaemia represent major healthcare problems world-wide. Classical diagnostic methods rely either on the direct observation of biological samples, on sophisticated analytical methods or molecular developers such as antibodies, allowing for a wide variety of molecular sensing techniques. These techniques can be, however, time-consuming and often require trained and experienced personnel. More sophisticated methods include bioassays based on PCR. However, these tests involve complex instrumentation and expensive chemicals. As an alternative, portable, low cost and easy to use molecular methods have been developed that allow patients or healthcare workers to carry out diagnostics. Examples are malaria rapid diagnostic tests. However, their sensitivity is often not sufficient for the detection of trace amounts of certain disease-marker molecules, e.g. to detect and quantify malaria at low parasitemia levels or to detect and quantify trace quantities of haemoglobin in body fluids.

RESULTS: We have developed a new assay for the detection and amplification of trace amounts of non-nucleic acid metabolites (e.g. hemozoin from malaria parasites). The assay is based on biocatalytic precipitation polymerizations of N-isopropylacrylamide carried out under conditions of atom transfer radical polymerization (ATRP). In such reactions, the metabolite to be detected acts as the catalyst of the initiation and polymerization reactions. The rate of turbidity formation during the polymerization scales with the concentration of the analyte (Fig. 1).

DISCUSSION & CONCLUSIONS: Hemoglobin quantification could be carried out with high sensitivity e.g. in plasma and urine. Similarly, diagnosis of malaria is possible with the assay at a sensitivity that outperforms conventional malaria rapid diagnostic assays.

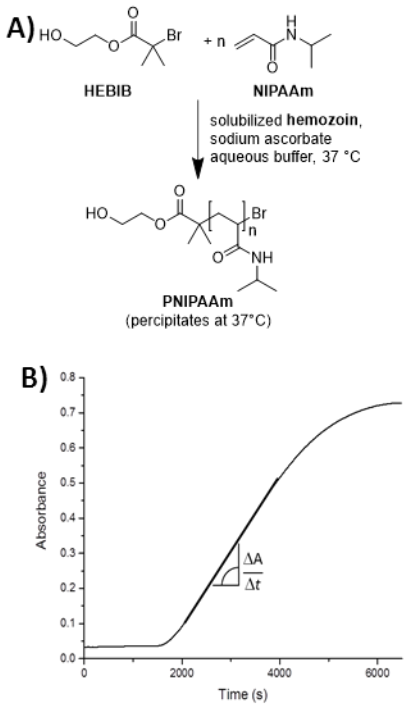

Fig. 1. Hemozoin quantification based on hemozoin-catalyzed polymerization of NIPAAm. a) Reaction scheme for the polymerization. b) Turbidity formation (measured as absorbance at 600 nm) during a polymerization reaction, caused by the precipitation of the temperature-responsive polymer (PNIMAAm). After an initial lag phase the turbidity increases linearly with time. The slope of this section is the readout of the experiment.

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ACKNOWLEDGEMENTS: Financial support by the Swiss National Science Foundation and the Novartis Foundation for Biological-Medical Research is gratefully acknowledged.

CURVATURE-INDUCED WRINKLING PATTERNS AND DEFECT FORMATION IN ELASTIC BILAYER SYSTEMS

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INTRODUCTION: Symmetry-breaking transitions associated with the buckling and folding of curved multilayered surfaces are common to a wide range of systems and processes such as embryogenesis, tissue differentiation and structure formation in heterogeneous thin films or on planetary surfaces. Yet owing to the nonlinearity of the underlying stretching and bending forces, their morphology cannot be reliably predicted by current theoretical models.

RESULTS: Here, we present a generalized Swift–Hohenberg theory that describes the wrinkling morphology and pattern selection in curved elastic bilayer materials. The theory is derived from the covariant formulation of the nonlinear Koiter shell equations by systematic expansion of the elastic energy in the dominant normal component of the displacement field. Using nonlinear stability analysis of the resulting scalar field equation on spherical substrates, we find analytical predictions for the critical curves separating three different morphological phases termed labyrinth, hybrid, and hexagonal phase. These predictions are in quantitative agreement with experiments on spherically shaped PDMS bilayer surfaces, confirming that the curvature of the substrate acts as a symmetry-breaking term that favors hexagonal wrinkling patterns for highly curved substrates.

Our approach builds on general differential-geometry principles and can thus be extended to arbitrarily shaped surfaces. As a particular example of non-spherical substrates, we consider the wrinkling morphologies on toroidal geometries, where the wrinkling pattern are, in general, spatially varying due to the non-constant substrate curvature. Tuning model parameters within experimentally accessible regimes, we derive approximate parameter bounds for the pure hexagonal wrinkling phase on tori. In this regime, the displacement patterns can be interpreted as a model system for two-dimensional crystals. Using numerical simulations, we elucidate how topology, system size, and curvature variations determine the number, location and orientation of crystalline defects. Moreover, these results also suggest that crystals preferentially form along geodesic 'superstructures', suggesting that local crystal growth senses the underlying geometry of the substrate.

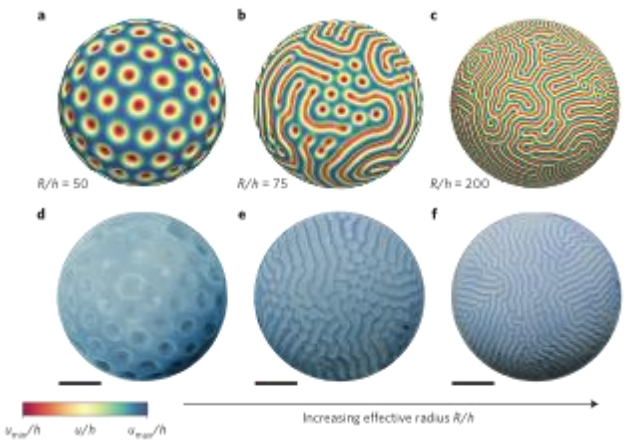

Fig. 1: Wrinkling morphologies of a stiff film of thickness h adhering to a spherical soft substrate of radius R. Top row: Numerical simulations of the generalized Swift-Hohenberg wrinkling equation. Increasing the effective curvature R/h leads to a transition from hexagonal to labyrinth-like patterns. Bottom row: Experimentally observed wrinkling patterns using PDMS bilayers confirm the predicted phase transition.

CONCLUSIONS: The presented effective wrinkling equation explains experimentally observed wrinkling pattern morphologies from micro- to macroscale by a curvature-induced symmetry-breaking transition. Our theoretical results suggests that in the hexagonal wrinkling phase, elastic bilayer systems may offer an accessible and inexpensive way to experimentally study defect formation and localization in curved two-dimensional crystals.

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Surfactant–free Janus Nanoparticles Synthesis and Their Application in Pickering Emulsions

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INTRODUCTION: In contrast to homogeneous particles, Janus nanoparticles (JNPs) are anisotropic. Their anisotropy originates from the shape, chemical composition, polarity contrast between surface regions, etc. JNPs could be exploited in various applications such as solid surfactants, supra-assemblies, sensors and catalyst carriers. Here, we are reporting a surfactant-free seeded emulsion polymerization method for synthesizing gram-scale amounts of polymeric JNPs. By tuning initiator type (AIBN or APS) and amount of second monomer used during seeded emulsion polymerization, one can effectively control the aspect ratio and the amphiphilic contrast between the lobes. It turns out that these parameters determine the emulsification ability of the JNPs and their orientation at the oil/water interface.

METHODS: First, polystyrene (PS) nanoparticles with diameters from 180 nm to 400 nm were synthesized by surfactant-free emulsion polymerization. Then, polymeric JNPs (PS-P(3-TSPM)) were synthesized by surfactant-free seeded emulsion polymerization in present of two types of initiators, AIBN (water insoluble) and APS (water soluble). The synthesized JNPs were characterized by SEM, EDAX, FTIR and zeta potential. After purification, the JNPs were tested for their ability to emulsify heptane/water mixtures. The emulsions were characterized by fluorescence microscopy. Finally, the orientations of JNPs at the wax/water interface were also investigated by low vacuum SEM.

RESULTS: Figures 1A and 1B show the starting PS seed nanoparticles with 200 nm diameter and snowman-like JNPs, respectively. Noteworthy, the yield of snowman-like JNPs after polymerization is over 95% (Figure 1B). In addition, the EDAX mapping (Figure 1C) of the elements clearly shows that silicon only appears on the newly formed lobe, i.e. from P(3-TSPM), which demonstrates the full phase separation between PS and P(3-TSPM) during the seeded emulsion polymerization.

The types of emulsion (o/w or w/o) obtained by emulsification of heptane with JNPs aqueous solution is generally determined by the overall polarity balance and aspect ratio between the two lobes. For example, Figures 2A and 2B show w/o and o/w emulsions, which are stabilized by AIBN-JNPs (2mL 3-TSPM) and APS-JNPs (4mL 3-TSPM), respectively. In some cases, JNPs at interface of wax/water have interesting orientation. For example, AIBN-JNPs (4mL 3-TSPM) are present at the interface with PS lobe facing to the non-polar wax and P(3-TSPM) lobe facing to water, see inset of Figure 2C.

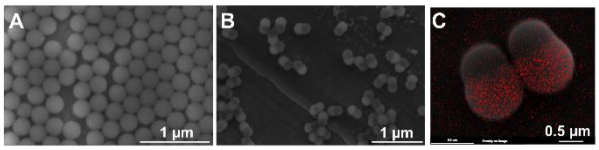

Fig. 1: SEM image of PS nanoparticles (A), SEM image of snowman-like JNPs (B) and EDAX mapping image of JNPs of Si (C).

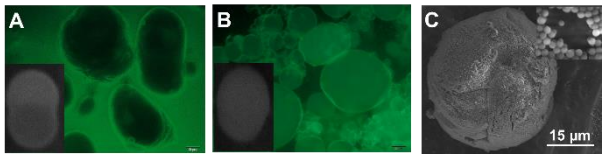

Fig. 2: W/O emulsion stabilized by AIBN-JNPs (2mL 3-TSPM)(A), O/W emulsion stabilized by AIBN-JNPs (3mL 3-TSPM)(B) and SEM image of wax-colloidosome stabilized by JNPs (C).

DISCUSSION & CONCLUSIONS: We successfully synthesized gram-scale JNPs by surfactant-free seeded emulsion polymerization. Additionally, the types of Pickering emulsions that can be obtained are determined by the overall polarity balance and the aspect ratio of the JNPs, which can be adjusted by the initiator type used and amount of 3-TSPM monomer during seeded polymerization.

REFERENCES : [1]D Wu et al, *Langmuir*, ***2016***, 32 (25): 6376-6386.

ACKNOWLEDGEMENTS: We are especially grateful for the financial support of Metrohm Foundation (Herisau, Switzerland) and by ZHAW via Anschubfinanzierung.

Posters

Magnetic Field induced Orientational Ordering of Amyloid-Fe_3O_4 Hybrids

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INTRODUCTION: An external magnetic field is a powerful tool to induce orientational order in suspensions of magneto-responsive rod-like particles, and change the critical value of the dimensionless concentration c at which the isotropic−nematic transition is observed:

$$c = \Phi\, L/D \qquad (1)$$

where Φ is the volume fraction, L the contour length and D the diameter. [l]

By incorporating an external directional field into the Onsager theoretical formalism, Khokhlov and Semenov described that increasing field strength could make the phase transition of rod-like particles occurring at progressively lower critical values of the dimensionless concentration. Furthermore, at the same field strength, suspensions of semi-flexible persistent particles undergo the phase transition at much higher critical values of dimensionless concentration than that of rigid ones. Despite these important predictions, a systematic and comprehensive experimental benchmark of the expected theoretical behavior of rigid and semi-flexible particles in the presence of an external field has yet to be conducted. [2]

METHODS: We prepared magneto-responsive hybrid fibrils by electrostatic complexation of β-lactoglobulin amyloid fibrils of different lengths with Fe_3O_4 nanoparticles. By small angle neutron scattering and optical birefringence measurement, we investigated the effect of concentration, flexibility, and aspect ratio (L/D) on the liquid crystalline ordering in the presence of an external magnetic field, enabling a direct experimental benchmark of theoretical predictions.

RESULTS: Magnetic field remarkably decreased the critical value of dimensionless concentration, inducing a spatially orientational order of amyloid fibrils decorated with spherical Fe_3O_4 nanoparticles at low volume fraction. When changing the aspect ratio and volume fraction at constant dimensionless concentration and magnetic field strength, the stiff fibrils display identical degrees of spatial alignment falling on a single master curve for both order (Fig. 1A) and birefringence (Fig. 1B) curves and field intensity. However, semi-flexible fibrils with equal dimensionless concentration reached much lower orientation levels in comparison with the rigid counterparts as a consequence of their semi-flexible nature (Fig. 1C).

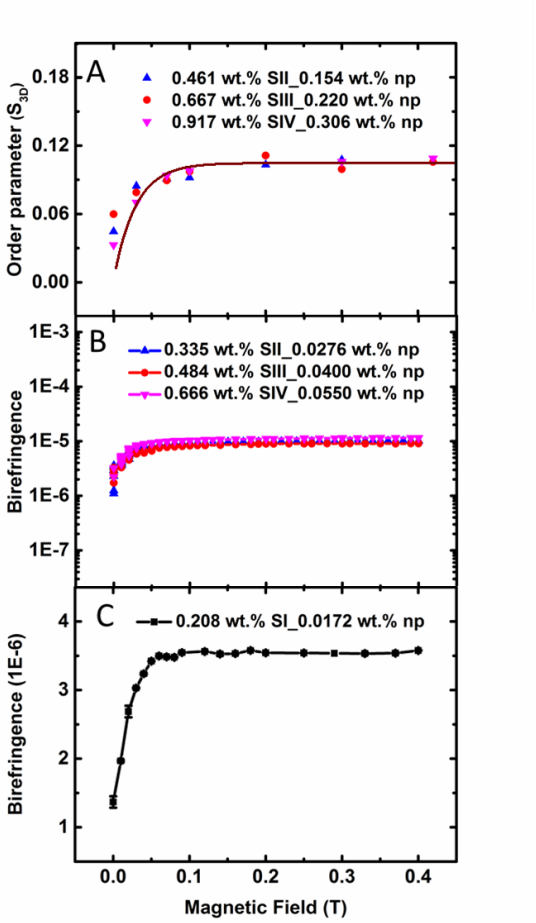

Fig. 1: A. 3D order parameter of stiff hybrid fibrils as a function of magnetic field at constant dimensionless concentration (0.886). Birefringence signal of stiff (B) and semiflexible (C) hybrid fibrils at constant dimensionless concentration (0.643).

DISCUSSION & CONCLUSIONS: These findings are consistent with Khokhlov-Semenov theoretical prediction, which may advance our understanding of field-induced order in fibrous systems and pave the road to design of functional materials with directional features.

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Combining micro-structured Surfaces with antibiotic encapsulated Vesicles to prevent Device Associated Infection

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INTRODUCTION: It is common for medical devices or implants inserted into the body to be prone to bacteria biofilms, causing device associated infections (DAI). DAI are causing high distress for patients and huge socio-economical costs. To stop the infection, the responsible device often has to be removed and if necessary reinserted into the body. The challenge is to prevent biofilm formation and development *in situ*. Passive strategy based on micro- and nano-based surface-patterns has already been reported to create hydrophobic surfaces with self-cleaning and antifouling properties. [1] A first example of an "active surface" able to produce antibiotics on demand for a long period of time has been developed by encapsulating penicillin acylase in nanoreactors. [2, 3] Here, we intend to go one step further in producing a combined effect against bacterial growth by the simultaneous use of nanoreactors and micro- and nano-structured surfaces (Fig. 1).

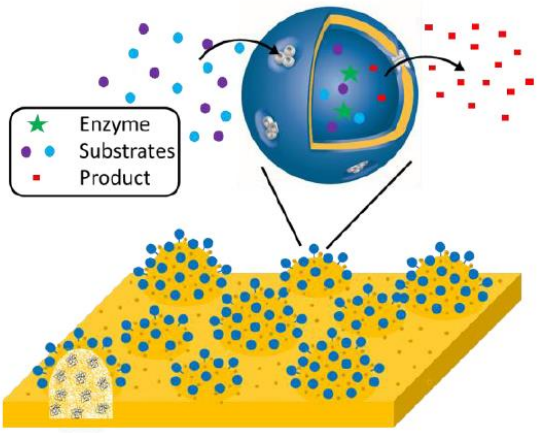

Fig. 1: Microstructured surface is combined with vesicles releasing active compounds "on demand" to prevent DAI.

METHODS: Antimicrobial properties of polypropylene (PP) based surfaces with nanoparticles were studied with the Japanese Industrial Standard (JIS) Test. Poly(2-methyloxazoline)-*block*-poly(dimethylsiloxane)-*block*-poly(2-methyloxazoline) (PMOXA-*b*-PDMS-*b*-PMOXA) based vesicles with partial azide functionalized surfaces were formed by thin film rehydration, and characterized by transmission electron microscopy (TEM), dynamic light scattering (DLS) and Fluorescence correlation Spectroscopy (FCS).

Glass surfaces were functionalized with DBCO groups and studied by contact angle (CA) and laser scanning microscopy (LSM)

RESULTS: PP based surface with silver nanoparticles were effective to kill *E. Coli*. No colonies were observed after overnight incubation on the surface, even in the undiluted 50 µL stripes.

Vesicles with azide groups on their surface were stable and their presence did neither affect the self-assembly process nor the architecture, as characterized by DLS and TEM.

Successful functionalisation of the solid support with DBCO groups has been characterized by CA. An increase of water CA was observed after DBCO surface modification. Furthermore, successful immobilization of an azide dye through a click reaction was shown by LSM.

DISCUSSION & CONCLUSIONS: The immobilization of azide dye molecules has been achieved by reacting overnight the DBCO functionalized surface with the dye. Next, the immobilization of azide modified vesicles on DBCO functionalized surface is studied and optimized. The combination of vesicles with micro- and nano-structured surfaces is expected to serve for an efficient prevention of DAI.

REFERENCES: [1] L Zhang, J. Wu, Y. Wang, Y. Long, N. Zhao, J. Xu (2012) *J. Am. Chem. Soc.* **134:** 9879-9881. [2] K. Langowska, C. G. Palivan, W. Meier (2013) *Chem. Commun.* **49**: 128-130. [3] K. Langowska, J. Kowal, C. G. Palivan, W. Meier (2014) *J. Mater. Chem. B* **2**:4684-4693.

ACKNOWLEDGEMENTS: Financial support was provided by Swiss National Science Foundation (SNSF) which is gratefully acknowledged. We thank Samuel Lörcher for polymer synthesis.

"ACTIVE SURFACES": BIOMOLECULE - POLYMER MEMBRANES FOR EFFICIENT SENSING OF PHENOLS

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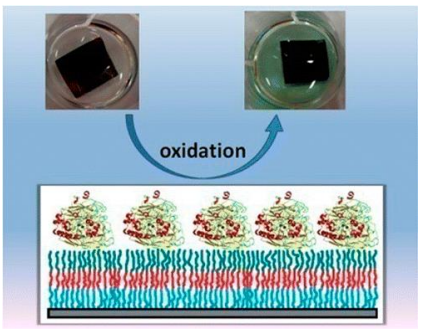

Figure 1: Oxidation of a phenolic compound by the active surface

The design of surfaces that present active compounds at the interface with their environment is on focus today in various domains, such as catalysis, medicine or environmental sciences. An elegant approach is to combine biomolecules (enzymes, proteins, mimics) with synthetic membranes in order to generate a stable and functional hybrid system.[1]

Here we present how two different enzymes are combined with asymmetric membranes and serve for development of "active surfaces" for sensitive detection of specific compounds. Solid supported membranes of PEG_{45}-*b*-$PMCL_x$-*b*-$PDMAEMA_y$ copolymers were prepared by LB-LS methods in different combinations of conditions. Laccase and Tyrosinase, as model enzymes for detection of phenol compounds were immobilized on soft surfaces resulting from polymer films deposition. Interestingly, the enzymes activity and stability varied depending on the film properties, which support further optimization of such active surfaces.

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Key aspects to yield low dispersity of PEO-*b*-PCL diblock copolymers for controlled self-assembly

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INTRODUCTION: Hollow polymeric spheres, or polymersomes, are very advantageous and attractive 3D self-assembled structures due to their ability to keep a desired environment for the encapsulated molecules. When polymersomes with encapsulated molecules are applied as a drug delivery system, the choice of suitable self-assembling polymers severely narrows, as the polymer should be biocompatible and biodegradable. A prominent example of such polymer is amphiphilic poly(ethylene oxide)-*block*-polycaprolactone (PEO-*b*-PCL) diblock copolymer. Under fixed self-assembly conditions PEO-*b*-PCL is able to self-assemble in aqueous solution into polymersomes and other 3D-structures depending on the hydrophilic weight fraction and the block length [1, 2]. Consequently, targeted self-assembly into a certain 3D-structure might require PEO-*b*-PCL with a certain block length and rather narrow dispersity. For example, PEO(2K)-*b*-PCL(9.5K) with $Đ_M$ = 1.14 forms predominantly mesoscale vesicles [1], whereas similar copolymer PEO(2K)-*b*-PCL(9K) but with higher $Đ_M$ = 1.42 forms mostly mesoscale worms [2] under similar self-assembly conditions.

In this work, we describe how to obtain uniform PEO-*b*-PCL ($Đ_M$ < 1.1) by optimizing its well-known synthesis catalyzed by $SnOct_2$. We also discuss the effect of aging of $SnOct_2$ on the overall kinetics of the synthesis. Finally, we compare self-assembled structures formed by uniform PEO-*b*-PCL ($Đ_M$ < 1.1) with its non-uniform analogues ($Đ_M$ > 1.1) to gain insight into the effect of the $Đ_M$ on the self-assembled structures.

METHODS: PEO-*b*-PCL was synthetized via $SnOct_2$ catalyzed coordination-insertion ring-opening polymerization of ε-CL using PEO as a macroinitiator. Self-assembled structures were prepared using film rehydration method. The aggregates were characterized by LSM.

RESULTS: In the present work, PEO-*b*-PCL synthesis was optimized via kinetically controlled polymerization. Purity and degradation of $SnOct_2$ catalyst was studied using ^{119}Sn NMR spectroscopy. Self-assembly behavior of PEO-*b*-PCL with different $Đ_M$ was investigated (Fig. 1).

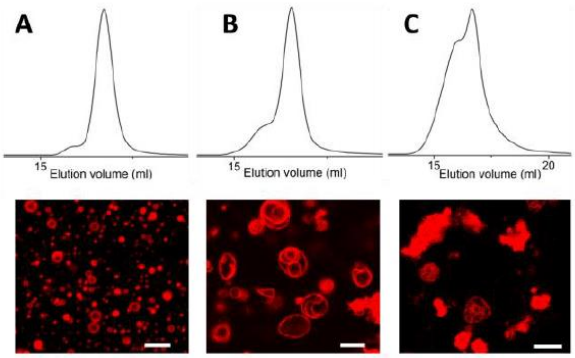

Fig. 1: Elugrams of PEO-b-PCL polymers and LSM images of the corresponding self-assembled structures: A: PEO(2K)-b-PCL(17.4K), $Đ_M$ = 1.08; B: PEO(2K)-b-PCL(16.0K), $Đ_M$ = 1.23; C: PEO(2K)-b-PCL(16.3K), $Đ_M$ = 1.55. Self-assembled structures were stained with Bodipy 630/650 dye. Scale bars are 5 μm.

DISCUSSION & CONCLUSIONS: The optimization of the synthesis of PEO-*b*-PCL diblock copolymers catalyzed by $SnOct_2$ yielded a robust and reproducible strategy to obtain block copolymers with tunable length of the PCL block while maintaining a low dispersity ($Đ_M$ < 1.1). We demonstrated the importance of $SnOct_2$ purification prior to the polymerization, and investigated its degradation during storage under different conditions. The comparison of the mesoscale self-assembled structures formed by PEO-*b*-PCL diblock copolymers suggests that the uniform polymers are beneficial for the self-assembly of distinct and uniform 3D-structures. However, further experimental and theoretical works are required to make this conclusion more solid.

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Surface Rheology of Block Copolymer Stabilized Interfaces: A Combined Experimental, Theoretical and Computational Study

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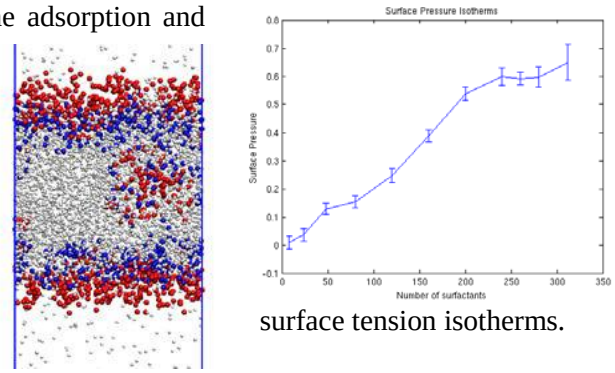
INTRODUCTION: Complex fluid-fluid interfaces are interfaces in which the adsorbed species self-assemble into complex microstructures. These interfaces can be formed by a wide range of surface active components, such as proteins, colloidal particles, polymers, lipids, or mixtures of these components [1]. In this study, our goal is to characterize the microstructure and mechanical properties of fluid-fluid interfaces stabilized by multi-block copolymers, using a multiscale multidisciplinary approach, which integrates state of the art computational methods, surface rheological and interfacial structure evaluation experiments and non-equilibrium thermodynamics. Using Monte Carlo (MC) and non-equilibrium molecular dynamics (NEMD) simulations, surface microstructure, surface rheological properties, and the surface free energy in terms of a set of structural variables can be obtained. Using the results from both experiments and computer simulations as a starting point, we will develop nonlinear coarse-grained constitutive models in the GENERIC [2] framework, able to describe the stress response and structural evolution of the interface as a result of an applied deformation. We will determine the mechanical properties and interfacial structure as a function of surface polymer concentration, chemical structure of the polymers (variation of number, size, and distribution of blocks) and degree of hydrophobicity and rigidity of the sub-blocks.

In this study, we just focus on equilibrium properties of such systems obtained from MC simulation, including adsorption and surface tension isotherms.

MC simulation: Using Monte Carlo simulation, we explore adsorption and surface tension reducing properties of amphiphilic block copolymer at liquid-liquid and liquid-vapor interfaces. The conventional bead-spring model for linear chains is used for block copolymers. All molecules in the system (block copolymer segments and liquid phase particles) are characterized by Lennard-Jones (LJ) potential with different interaction parameters. Configurational bias Monte Carlo (CBMC) method is used for moving block copolymers and Metropolis Monte Carlo move is used for liquid phase molecules. We investigate effects of chain length, block distribution, solvent quality, molecular scale interactions, and copolymer concentration on the adsorption and surface tension isotherms.



(a) (b)

Fig. 1: A liquid-vapour stabilized interface by diblock copolymers (a). Surface pressure isotherms for liquid-liquid interface stabilized by block copolymers (b).

Surface tension is obtained from integrating the difference between normal and tangential pressure tensor components in the direction normal to the interface. Figure 1b represents the surface pressure, i.e. the difference between the surface tension of the bare interface and surfactant stabilized interface with respect to number of surfactants. As we expect, increasing the surface coverage increases the surface pressure until the interface get saturated. After that, increasing the number of surfactants will not increase surface coverage (and hence the surface pressure).

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Controlled radical ring opening polymerisation for smart and degradable polyesters

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[2] *University College London, London, United Kingdom*

Introduction:

Recent years have seen considerable advances in radical and ring-opening polymerisation. One gap however, the development of smart and degradable polymers is only filling slowly.[1] This is mainly due to the fact that ring-opening polymerisation does not tolerate the unprotected amines or alcohols necessary.[2] By using controlled radical ring opening polymerisation (CoRROP) from cyclic ketene acetals (CKAs), this issue can be overcome.

Materials&Methods&Results:

CKAs can be achieved in a 2 step synthesis from the corresponding diol. In a first step the ring is closed using a transacetalisation with chloroacetaldehyd dimethyl acetal. While early reports suggest to accomplish this step using prolonged heating, a cobalt-catalysed method known in organic chemistry proved to be feasible and this step can be accomplished quickly at room temperature.[3] The second step sees the elimination of hydrogen chloride to eventually form the CKA.[4] In order to proof that functional polymers can be formed, amphiphilic block-copolymers from the CKAs will be used for self-assembly into micelles or vesicles.

One of the 3 CKAs targeted is an acetal with 2 methyl units to support the radical formed later (1), another one has a hydroxyl unit in its side chain (2) and the third one contains a tertiary amine (3, all numbers relate to Figure 1). This range is to demonstrate which side-chain moieties can be tolerated by this method.

Conclusion:

Hence, the monomer synthesis can be accomplished also in large quantities, leading to good amounts for the following polymerisation. The resulting material then has the range of properties aimed at and we are certain that this chemistry can fill the gap to form degradable and responsive polyesters.

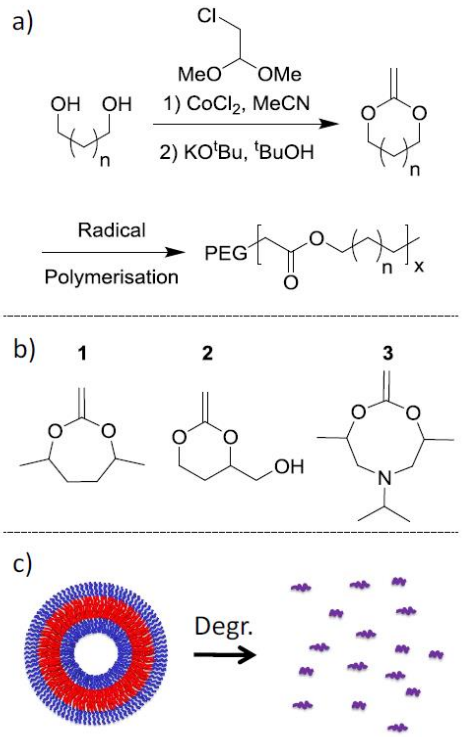

Fig. 1: a) Formation and polymerisation of CKAs starting from a diol, b) CKAs applied in our study and c) application of a polymer from CKAs as degradable polymersome.

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Using proteorhodopsin fusion proteins to direct proton transport across synthetic membranes

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The creation of proto-cells, artificial organelles or similar biomimetic nano-devices is part of the bottom-up approach of synthetic biology. In order to fulfil their desired task, these systems often require substrate and product transport into their reaction compartment. Passive diffusion can be enabled by robust pores, whereas active and controllable transport requires energy. The proton motif force (PMF) is one of the fundamental electrochemical gradients and powers several transport processes. Light-driven proton pumps such as proteorhodopsin (PR) generate a PMF upon illumination. Modern detergent mediated membrane protein reconstitution procedures allow the integration of membrane proteins into synthetic lipid and block copolymer membranes, however they usually lack control over the final orientation of the proteins. In case of directional transporters, this can lead to a loss in functionality. Furthermore, this process relies on the self-assembly of the membrane building blocks and the membrane protein, thus it is not possible to exert direct control on the assembly.[1] Only by changing the starting conditions it is possible to influence the outcome, which in turn requires detailed knowledge about the key parameters.

By fusing green fluorescent protein (GFP) to PR, we bypassed this issue. Due to the hydrophilic nature of GFP, the protein is able to orient itself when reconstituted into preformed vesicles. Design of experiments (DoE) utilizes statistical modelling to identify significant factors and allows their optimization towards a desired outcome in a highly efficient way.[2] For the first time, we applied this methodology in a rational way, finding conditions which lead to proper formation of proteolipo- and proteopolymersomes in the first step.

The fluorescence of GFP allowed us to easily detect PR-GFP in the membrane structure by fluorescence correlation spectroscopy (FCS). It turned out, that lipid and polymer membranes require rather different treatment for successful reconstitution. In a next step, the parameter space was narrowed down further by setting boundary conditions in which the highest pumping activity was assumed. The internal pH of the proteovesicles was measured via fluorescence spectroscopy and the final model was able to predict the activity with a good precision. It allows easy optimization of the process as well as providing the necessary knowledge.

Thus, our approach shows that rational design on a molecular level and assisting the reconstitution process with statistical modelling leads to reliable product.

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Active microrheology in glassy emulsions

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INTRODUCTION: Active microrheology provides many advantages to acquire rheological properties of complex fluids. In active microrheology, an external force is applied to the probe particle to drag it through the host environment. In dense environments such as glassy systems the probe particle is surrounded closely by other particles in the medium, which is defined as a cage. At high volume fractions it takes very long time for the probe to get out of its cage. We are interested in motion of the probe under nonlinear conditions in glassy systems [1].

METHODS: In our experiments, we used shear induced and surfactant stabilized PHMS-in-water 2um sized emulsions with polydispersity of 10% [2]. Those emulsions were then density and refractive index matched, and polystyrene probe particles were added. Later, the emulsions were centrifuged at 4000rpm at 4 °C in order to get over the jamming point. In order to go back to the glass regime and find out the volume fraction of the sample, Light Scattering techniques were used (Figure 1) [3].

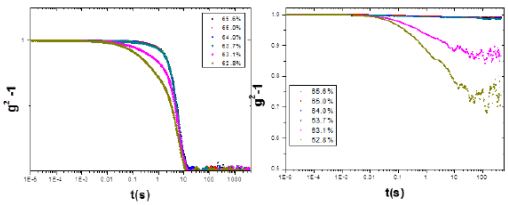

Fig. 1: Comparing light scattering results; Slow rotation method (left) and Pusey-Averaging method.

RESULTS: Using time shared laser tweezer we apply constant force to the probe particle and observe the motion of the probe though the emulsions. Since the emulsions are density and refractive index matched, we can only observe motion of the probe particle. Our preliminary results show that our laser is strong enough to move the probe particle even at high volume fractions but still in glassy regime (~63%). The particle moves within 10 particle diameters

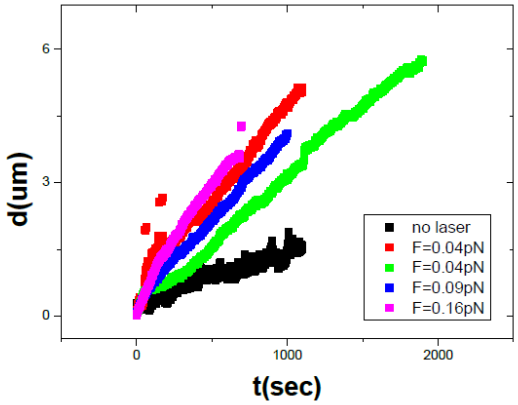

Fig. 2: Displacement of the probe particle under different forces. 0 1000 2000 036 no laser F=0.04pN F=0.04pN F=0.09pN F=0.16pN d(um) t(sec)

DISCUSSION & CONCLUSIONS: Our current results show that with 0.2-0.2pN of applied force the probe particle can be moved out of its cage. This experiment is challenging due to possibility of drag or air bubble in the sample cell. Thus, in our calculations we subtract effect of the flow from the motion of the particle. Our preliminary results are promising and very interesting.

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Characteristics of strong glass formers in charged microgel systems

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For molecular glass formers the dependence of the structural relaxation time τ_α on temperature strongly depends on the type of material used, which has led to the classification of strong and fragile glasses. Fragility expresses here by how much the temperature dependence τ_α deviates from a classical Arrhenius behavior. In strong glasses the system exhibits typical Arrhenius behavior, i.e τ_α increases exponentially upon approach of the glass transition temperature T_g. By contrast, in fragile glasses τ_α diverges critically at T_g. Hard sphere colloidal systems are typical examples of fragile glasses, the parameter governing the glass transition being here the particle concentration.

In this contribution we show that charged permeable colloids can exhibit the characteristics of strong glass formers. This is, however, not due to attractive trapping like in molecular systems, but rather to a repulsive particle interaction that decreases with increasing particle concentration.

Mechanism of the cooperativity in antimicrobial peptides

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INTRODUCTION: Antimicrobial peptides are of great interest due to their ability to inhibit or kill pathogens as the first line of defense. During all these decades after antibiotics discovery, bacteria demonstrated ability to adapt and resist, evolving into our new enemy - the 'superbugs'. In contrast, microbes have not learnt to avoid the lethal attack of AMPs. Therefore AMPs promise a breakthrough in the fight against superbugs.

In this study we aimed to elucidate the fundamental mode of action of antimicrobial cooperativity.

To investigate the mechanism of cooperativity between two different human peptides defensin and cathelicidin we formed artificial lipid bilayers deposited onto highly doped p-type silicon electrodes covered by naturally formed silicon dioxide, as a clean platform to separate peptide-peptide and peptides-bilayer interactions from influence of other components within cell.

METHODS: In this project we used vesicles fusion for bilayer formation from mixture of POPC lipids and NBDPE as a fluorescent agent. The bilayer was formed on highly doped p-type silicon/silicon dioxide wafer with activated surface. For electrochemical properties we used the 3-electrode impedance spectroscopy setup to monitor the barrier function of the bilayer in real time.

Prior to monitoring of barrier function of the bilayer, we performed Fluorescence Recovery After Photobleaching (FRAP) utilizing fluorescent microscopy as a complementary technique to confirm the quality of the formed bilayer, measured as its recovery in the photobleached regions.

RESULTS: *Figure 1* shows impedance spectra before (green line) and after (red line) addition of the vesicle solution. The right- shift in impedance curve indicates bilayer formation as lipids act as superb natural electrical insulator. The obtained spectra were interpreted in terms of equivalent R(RCRC) circuit consisting of simple electrical elements, such as interface and bilayer resistance and capacitance, given in the inset in the figure.

Figure 2 represents images from fluorescent microscopy after photobleaching of region of interest. During 12 minutes lipid bilayer recovered completely thereby proving its fluidity and capability of self-repairing.

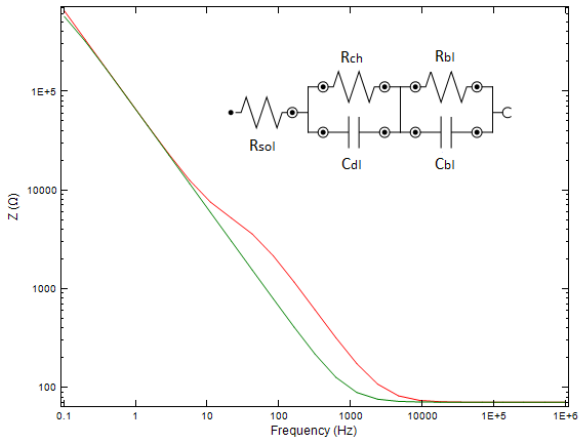

Fig. 1: Impedance spectra simulation of the supported lipid bilayer. Green line represents Si wafer with HEPES buffer before adding vesicle solution and red line corresponds to bilayer formation.

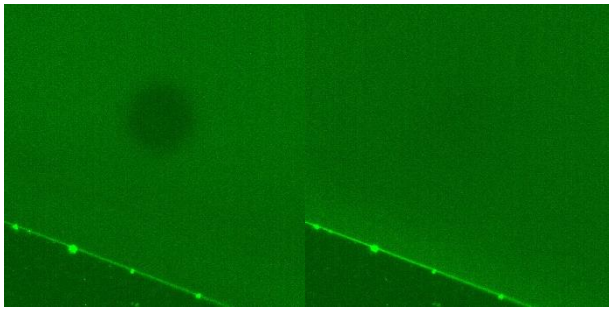

Fig. 2: FRAP images of photobleached region and its recovery after 12 minutes

DISCUSSION & CONCLUSIONS: The obtained resistance of the supported membrane is three times lower than results presented in Purrucker's article hence additional effort are needed to optimize bilayer formation conditions. In the next step we will incorporate different peptides to lipid membrane and we expect to observe decrease in resistance as bilayer's continuity (integrity, barrier function) is disturbed. The milestone goal is to quantify the antimicrobial peptides' cooperativity effect on the bilayer resistance and capacitance.

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Stimulus Dependent Flow through Polymer Modified Filtration Membranes

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INTRODUCTION: Chemically and biologically modified membrane systems are heavily investigated for their expected potential in many applications ranging from drug release to selective filtration. A frequent modification is represented by surface immobilized polymer brushes which can be synthesized by ATRP and can be further functionalized following synthesis[1, 2]. Here we present data on the pH and divalent cation dependent water filtration properties of poly-2- (methacryloyloxy)ethyl succinate (MES) modified cellulose membranes.

METHODS: Cellulose membranes were modified by a "grafting from" approach by performing Atom Transfer Radical Polymerization (ATRP) of MES on bromoisobutyryl bromide modified cellulose membranes. The modified membranes were characterized by FTIR and SEM and functionally analysed by determining the water flow through the membrane at different pH values and concentrations of $CaCl_2$ at a pressure of 0.1 bar.

RESULTS: Liquid flow through poly-MES modified cellulose membranes was heavily dependent on both the pH value and the concentration of divalent cations (Ca^{2+}) (*Fig. 1*).

Investigating the liquid flow of low ionic strength buffers in a pH range between pH 4.5 and pH 8.5 revealed a strong reduction in liquid permeation at higher pH values with the change in flow performance appearing around the pK_a value of MES (pK_a = 5.4). With increasing polymerization time we observed a concomitant influence of the pH value on the liquid flow through modified membranes with a maximal factor of 500:1 of the flow rates at pH 3.2 versus pH 8.5. Similarily, carboxy group interacting divalent cations also influenced the water flow through poly-MES modified membranes. When poly-MES molecules were synthesized on the surface of nanomechanical cantilever sensors and subsequently probed with different concentrations of Ca^{2+} we observed strong bending signals of up to 4 µm, indicative of conformational changes of the immobilized polymers[3].

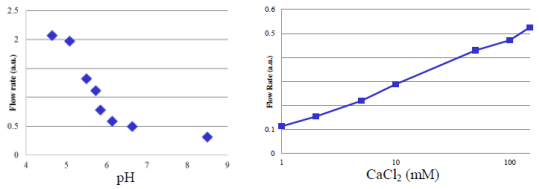

Fig. 1: pH and Ca^{2+} dependent liquid flow through poly-MES modified cellulose membranes.

DISCUSSION & CONCLUSIONS: Here we present data on the pH and divalent cation dependent water flow rates of poly-MES modified cellulose filtration membranes. Further investigations of the mechanism of these effects are necessary. E. g. today most studies on stimulus regulated filtration limit their investigations on water filtration rates. Future work will also have to investigate the membrane diffusion rates for biologically relevant molecules like metabolites, APIs and proteins. Since the carboxy groups of poly-MES modified surfaces can be further functionalized with bio-receptors they represent a suitable system to study the behaviour of functionalized polymer brushes in the presence of specific biomolecules.

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THE MECHANORESPONSIVE NATURE OF METALLOSUPRAMOLECULAR POLYMERS

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INTRODUCTION: The design principles of natural materials are frequently exploited to furnish artificial materials with tailored properties such as responsiveness to mechanical stimuli. In these mechanoresponsive materials, a macroscopic mechanical force is translated into a chemical change on the molecular level.[1] Metallosupramolecular polymers are attractive candidates as mechanoresponsive materials, due to their reversible assembly into dynamic structures and the tunable interaction strength of the binding motif that can be adjusted through a variation of the metal or ligand.[2] We aim to prepare mechanically responsive materials based on metallosupramolecular polymers and investigate their behavior under stress from the macroscopic down to the molecular level.

METHODS: A telechelic polymer with methylbenzimidazolyl pyridine ligands at the two termini was prepared via Mitsunobu reaction with poly(ethylene-*co*-butylene). Additionally, a ligand with an extended p-system was prepared in three steps and amide bond formation with a poly(ethylene-*co*-butylene) dicarboxylate yielded the corresponding telechelic polymers.

RESULTS: Polymeric networks were made by the coordination of metal ions to a telechelic poly(ethylene-*co*-butylene) that was end-capped with methylbenzimidazolyl pyridine ligands.[2,3] Europium- and terbium-based supramolecular polymer films were successfully welded by ultrasonication as a means of mechanical stimulation and the interface of joined pieces was investigated by energy-dispersive X-ray spectroscopy to elucidate the diffusion of metals (Figure 1A). Moreover, telechelic polymers carrying ligands with extended p-systems were prepared to allow better analysis of the (dis)assembly of the metal-ligand complexes upon application of mechanical forces. Upon addition of europium the bright blue fluorescence of the unbound ligands is replaced by the red fluorescence of the europiumbased metal-ligand complexes in solution as well as the solid state (Figure 1B).

DISCUSSION & CONCLUSIONS: Investigating how these supramolecular polymers respond to mechanical stimuli in solution and in the solid state is envisioned to provide a detailed understanding of their mechanochemistry. Application of ultrasound to solutions of the metallosupramolecular polymers resulted in a temporary decrease of the europium fluorescence, indicating the transient disassembly of the metal-ligand complexes.
Investigations of the dynamics in the solid state are currently ongoing.

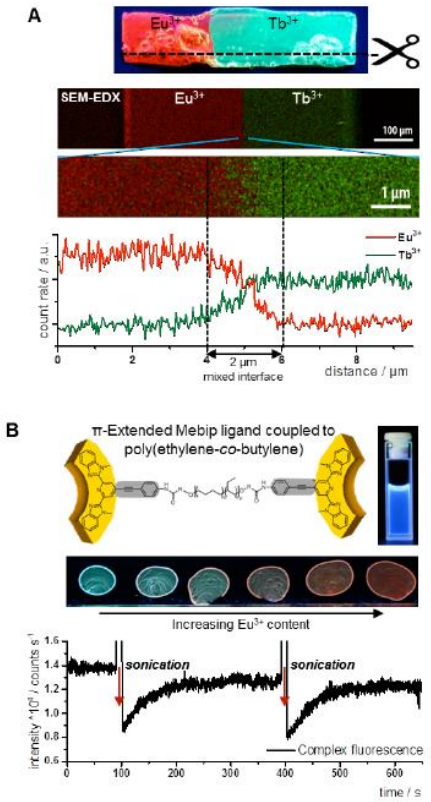

Fig. 1: (A) Mechanochemical welding of europium- and terbium-lap joints and investigation of the welded interface. (B) Fluorescence of a π-extended ligand in solution, the solid state, and upon application of ultrasound.

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