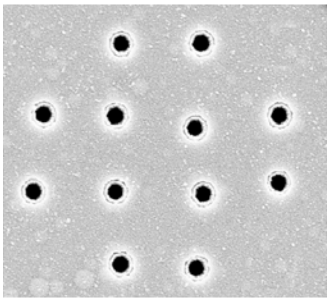
Swiss Soft  Days

3rd WORKSHOP
Fribourg 20.10.2010

Department of Chemistry, University of Fribourg
Chemin du Musée 9, Fribourg

Sponsored by

University of Fribourg

Fribourg Center of Nanomaterials

Local Organizers **Véronique Trappe &**
Frank Scheffold

Soft Matter & Photonics Group
Department of Physics

University of Fribourg

Program

10.15-10.25	*Welcome: Véronique Trappe (U Fribourg)*	
Chair: Giuseppe Foffi (EPFL)		
Plenary		
10.25-11.10	Roland Winkler (Forschungszentrum Jülich) Mesoscale Hydrodynamic Simulations of Polymers in Flow Fields	
Non-Linear Shear (Fundamental and Use)		
11.10-11.25	Brader (U Fribourg)	Rheology of colloidal dispersions
11.25-11.40	Harsche (ETHZ)	Breakage of colloidal aggregates in contracting nozzle
11.40-11.55	Studart (ETHZ)	Predicting droplet sizes in flow-induced dripping
Cell Mechanics		
11.55-12.10	Roduit (EPFL)	Stiffness tomography by AFM
12.10-13.30	**Lunch / Coffee / Posters / Discussion**	
Chair: Cornelia Palivan (U Basel)		
Phenomena at Interfaces		
13.30-13.45	Isa (ETHZ)	Self-Assembly of colloidal particles at liquid-liquid interfaces (SALI) as a route for nanolithography and fabrication of composite materials
13.45-14.00	Fontani (EMPA)	Mechanism of aqueous lubrication with amphiphilic polymers
14.00-14.15	Reynolds (U Zürich)	Aggregation of alpha synuclein on supported lipid bilayers
Single Molecule (Detection and Properties)		
14.15-14.30	Steinwand (U Zürich)	Coherent diffraction imaging with low-energy electrons
14.30-14.45	Benedetti (EPFL)	Single-molecule force spectroscopy of multimodular proteins: a new method to extract kinetic unfolding parameters
14.45-15.00	Maffi (EPFL)	Conformational entropy in macromolecules
15.00-16.00	**Coffee / Posters / Discussion**	
Chair: Emanuela del Gado (ETHZ))		
Potpourri of Concepts		
16.00-16.15	Parker (Firmenich)	Glassy dynamics of gelatine gels
16.15-16.30	Van Gruijthuijsen (U Fribourg)	Depletion interactions in charged, aqueous colloid/polymer-mixtures
16.30-16.45	Tanner (U Basel)	Pseudo-cellular nanoprocessors are able to fight oxidative stress
16.45-17.00	Ferrari (EMPA)	Multi-method approach to characterize super-plasticizers behaviour at the solid-liquid interface
17.00-17.15	Kadau (ETHZ)	From powders to collapsible structures / “living quicksand”: discrete modeling
17.15	*Closing Remarks: Hans-Jörg Limbach (Nestlé)*	

Poster Contributions

Assemblies & Materials		
A1	Schoch (U Basel)	Monitoring structural changes in molecular layers with surface plasmon resonance
A2	Curschellas (ETHZ)	Characteristics of polyglycerol esters at air-water interfaces
A3	Zahn (ETHZ)	Counterion dependent swelling of redox-active polyelectrolyte multilayers
A4	Braun (U Basel)	From polymer vesicles to bulk polymer
A5	Balasubramanian (U Basel)	Polymeric nanoreactors: a new way to improve antioxidant therapy
A6	Chevrier (U Fribourg)	Synthesis of new silver compounds with nicotinic acid derivatives
A7	Balog (PSI)	Relationships between the morphology, water uptake, and proton conductivity of radiation grafted proton exchange membranes
BioPhysics & BioOriented Materials		
B1	Bertseva (EPFL)	Microrheology of healthy and cancerous cells using optical tweezers
B2	Jud (U Fribourg)	Towards a better understanding of eye lens transparency and cataract formation
B3	Gunes (Nestlé)	Scaling of creep-compliance curves of β-lactoglobulin-based systems at pH 2
B4	Toma (U Basel)	DNA interactions with PAMAM6 dendrimers: dynamics and structure evolution in 3D microchannels
B5	Toughraï (U Basel)	Solid-supported biomimetic block copolymers membranes
B6	Baumann (U Basel)	Protein gates allow in situ enzymatic reactions in polymeric nanocompartments: Towards synthetic organelles
Colloids & Polymers		
C1	Sinha (U Genève)	Direct measurement of van der Waals forces between latex particles across aqueous electrolyte solutions
C2	Finessi (U Genève)	Charge reversal of sulfate latex particles by adsorption of cationic polyelectrolytes studied by direct force measurements
C3	Mahmoudi (U Fribourg)	Phase-separation and gelation of protein-poly(ethylene oxide) mixtures: effect of electrostatic interactions
C4	Dietsch (U Fribourg)	Nanoengineering of thermosensitive core-shell particles
C5	Pravaz (U Fribourg)	In-situ polymerization as a route towards polymer-colloid nanocomposites
C6	Fiore (U Fribourg)	Photo-healable metallosupramolecular polymers
C7	Geuss (U Fribourg)	Where are the domains in ferroelectric polymers? A 2D piezoresponse force microscopy answer to an old question.

Abstracts Talks

Plenary Lecture 10.25 – 11.10

Mesoscale Hydrodynamic Simulations of Polymers in Flow Fields

Roland G. Winkler

Institut for Advanced Simulation, Forschungszentrum Jülich

The dynamical behavior of dilute or semidilute solutions of complex fluids is strongly affected or even dominated by hydrodynamic interactions. These interactions are of long-range nature and thus need to be taken into account adequately. For an efficient treatment of such systems by computer simulations, the large length- and time-scale difference between the solvent and the solute degrees of freedom calls for a coarse-grained and simplified description of the solvent dynamics. The multiparticle-collision (MPC) dynamics simulation method is very well suited to study the dynamics of complex fluids. This is demonstrated by our hybrid simulations, which combine the MPC method for the solvent with molecular dynamics simulations for solutes such as polymers and colloids.

In the talk, the MPC method will be briefly outlined and results will be presented for polymers in flow. In shear flow, such molecules undergo large conformational changes and exhibit an intriguing tumbling dynamics. In microchannel flow, their flow properties are determined by an complex interplay of confinement, deformation, orientation, hydrodynamic interactions, and thermal fluctuations. In addition, results will be presented for the bundle formation of helical filaments, a mechanism used by bacteria in their locomotion.

Non-Linear Shear (Fundamental and Use) 11.10 – 11.55

Rheology of colloidal dispersions

J.M. Brader

Department of Physics, University of Fribourg

The ability to control and manipulate the macroscopic flow behaviour (rheology) of a colloidal dispersion by tuning the interactions between the constituents is of vital importance for many applications. However, connecting the macroscopic response to the underlying correlation functions and, ultimately, the microscopic inter-particle interactions is a challenging task for classical statistical mechanics based treatments. We will introduce recent developments in tackling this problem, with particular emphasis on the response of dynamically arrested glassy states to an applied shear flow.

Breakage of colloidal aggregates in contracting nozzle

Yogesh M. Harshe, Miroslav Soos, Marco Lattuada, and Massimo Morbidelli

Department of Chemistry and Applied Biosciences, ETH Zürich

Colloidal aggregates are encountered in polymer processing, pharmaceutical industries, coagulation processes, etc., wherein they are often exposed to different types and magnitudes of flow. The size and morphology of the resulting aggregates is a function of parameters such as their initial structure, the flow strength and flow type, etc, which control the balance between the three main processes occurring in shear dispersions, namely, aggregation, breakage, and cluster restructuring.

Detailed study of interdependence of evolution of the aggregate size and morphology on the imposed flow field and aggregate history, through both modeling and experiments, is missing in the literature. Additionally, a strong link between experiments and modeling has not been established to derive generalized breakage kernels and fragment distribution functions, which could be used in population balance equations-based modeling.

In the present work we have studied the breakage of colloidal aggregates, with two different initial geometries (d_f =1.7 and 2.7) and composed of 90 and 810 nm primary particles, in extensional flow generated by connecting two syringes with a nozzle, over a wide range of applied hydrodynamic stress. To understand the experimentally observed size and structure evolution of aggregates a model based on Stokesian dynamics method and incorporating DLVO and tangential interactions, was developed. The evolution of cluster size and morphology as a function of number of passes through the nozzle, and also their steady-state values as a function of applied stress as obtained with the model are compared with the experimental results. It was found that, using an appropriately defined flow-field for the model, results in qualitative and quantitative agreement with experiments can be deduced, which help explaining the underlying physics.

Predicting droplet sizes in flow-induced dripping

A.R. Studart, R.M. Erb, P.W. Chen, J Studer

Department of Materials, ETH Zürich

We present an encompassing model predicting the size of droplets dripping into an immiscible flowing fluid. The model incorporates the interplay between shear stresses and interfacial tension acting on the droplet. Despite complex flow behavior, we find that break-up relations for isolated droplets can be used to precisely predict the size of dripping droplets that lie on a universal curve. The model describes data obtained for a wide range of fluid properties and flow conditions including single and multiple dripping events and other experimental data previously reported in the literature

Cell Mechanics 11.55 – 12.10

Stiffness tomography by AFM

C.Roduit [1], B. Saha [2], K. Radotic [3], P. Hornitschek [4], G. Dietler [1], S. Kasas [1]

[1] Institut de Physique des Systèmes Biologiques, EPF Lausanne
[2] National Centre for Cell Science, Ganeshkhind, Pune
[3] Center for Multidisciplinary Studies, University of Belgrade
[4] Center for Integrative Genomics, University of Lausanne

Cell mechanical properties play important roles in a wide variety of cellular functions such as cell adhesion, division or migration and diseases such as cancer or osteoporosis. At the nanometrical scale, the mechanical properties of cellular membrane is important in cell signalling, endo and exocytosis or pathogen adhesion and penetration during infection.

Measuring the cell stiffness at a nanometric resolution is therefore important to understand numerous physiological and pathological phenomena.

The atomic force microscope (AFM) is a very convenient tool to perform mechanical measurements on biological samples in nearly physiological conditions. We recently used this instrument to develop a new imaging technique, called stiffness tomography, that permits to reveal structures buried into the bulk of the sample according to their stiffness.

During the presentation we will introduce this new imaging mode as well as its applications to different cell types such as vegetal cells (Arabidopsis thaliana) macrophages and neurons.

Phenomena at Interfaces 13.30 – 14.15

Self-Assembly of colloidal particles at liquid-liquid interfaces (SALI) as a route for nanolithography and fabrication of composite materials

Lucio Isa
Laboratory for Surface Science and Technology, ETH Zürich

Trapping at the interface, combined with lateral mobility and the presence of specific interactions, makes self-assembly of nanometer-sized colloids at liquid-liquid interfaces a process with huge potential for the creation of controlled two-dimensional structures and patterns and of novel ultrathin materials such as membranes and capsules. In this talk I will first show how SALI can be used for the deposition of non-close packed crystalline nanoparticle arrays over mm^2 areas for lithographic masks with independent control on feature size and spacing. Secondly, I will describe how adsorption and self-assembly of oxide core-polymer shell nanoparticles at water/decane interfaces poses intriguing fundamental physical questions and paves the way to the fabrication of composite, functional materials.

Mechanisms of aqueous lubrication with amphiphilic polymers

G.Fontani [1], R.Crockett [1], D.Passerone [1] and N.D.Spencer [2]

[1] Swiss Federal Institute for Materials Science and Technology, EMPA Duebendorf
[2] Laboratory for Surface Science and Technology, ETH Zürich

The adsorption of amphiphilic polymers and their friction behavior has been investigated in order to determine to what extent these polymers can reduce friction and how the latter is affected by the hydrophobic interactions within and between the polymers.
Theoretical studies have been carried out. A polymer chain has been modeled and molecular dynamics simulations have been performed to evaluate the polymer conformation. The model systems used in this project comprise amphiphilic polymers substituted with hydrophobic chains.
In addition studies on the composition and dynamic evolution of solvation shells around the polymer functional groups and on the polymer structure rigidity have been carried out.

Aggregation of Alpha Synuclein on supported lipid bilayers

N.P. Reynolds [1]*, M. Rabe [1], D. Verdes [1], A. Soragni [2], E. Liverani [2],
R. Riek [2], S. Seeger [1]

[1] Institute of Physical Chemistry, University of Zürich
[2] Laboratory of Physical Chemistry, ETH Zürich

Alpha synuclein (aS) is a protein expressed predominantly in neural tissues. Upon aggregation it forms toxic amyloid structures which represent a hallmark pathological feature of Parkinson`s disease. The mechanisms of aggregate formation and its cyto-toxicity are currently unclear, however the cell membrane is thought to play an important role. Here a surface sensitive microscopy technique (super critical angle fluorescence), cryo-SEM and confocol microscopy have been used to study the aggregation of aS with supported lipid bilayers (SLB). The findings suggest a previously unseen mechanism by which the SLB triggers aggregation of aS and the growing aggregates disrupt the bilayer integrity.

Single Molecule (Detection and Properties) 14.15 – 15.00

Coherent diffraction imaging with low-energy electrons

E. Steinwand, J.-N. Longchamp, H.-W. Fink

Physics Institute, University of Zürich

Obtaining structural information from just one single molecule would constitute a big step ahead in structural biology since it would eliminate the need to average over millions of molecules and allow investigating proteins that do not readily crystallize. However, such an imaging method requires radiation with wavelengths in the Ångstrom regime, which additionally allows recording of sufficient elastic scattering events from an individual molecule without destroying it. It has turned out that low-energy electrons, i.e. electrons

with kinetic energies between 50 and 250eV, fulfil these requirements. We have set up an experimental scheme for coherent diffraction imaging with such low-energy electrons. A pivotal component is an electrostatic microlens for collimating the divergent electron beam emitted by the source. First diffraction patterns could be obtained using carbon nanotubes as test-samples.

Single-molecule force spectroscopy of multimodular proteins: a new method to extract kinetic unfolding parameters

F. Benedetti, S. Sekatskii and G. Dietler

Institut de Physique des Systèmes Biologiques, EPF Lausanne

When multimodular proteins composed of a number of identical modules are unfolded in the course of a single-molecule force-spectroscopy experiment, both the probability distribution of the unfolding force and the most probable unfolding force depend on the number of not-yet-unfolded modules in the protein. We show that this unfolding can be described in the framework of a generalized Bell-Evans model, based on certain minimal assumptions concerning the force loading conditions. In particular, our modified equations for the unfolding force predict, for a multimodular protein containing â€œNâ€□ not-yet-unfolded modules, a linear dependence of the most probable unfolding force on â€œln(N)â€□. This conclusion has been confirmed in experiments with GB1 proteins, which verify in addition further consequences of the proposed model, including a modified probability distribution of the unfolding force. Based on these results, we propose a new method for analyzing single-molecule force-spectroscopy data for multimodular proteins, which enables one to extract kinetic unfolding parameters from experimental data with no additional assumptions about the unfolding process.

Conformational entropy in macromolecules

C. Maffi and P. de Los Rios

Institut de Théorie des Phénomènes Physiques, EPF Lausanne

Many biological processes occur on a subtle equilibrium among several contributions to the free energy landscape. The conformational entropy origins from internal and fast fluctuations of the atoms positions which are difficult to be detected. Nevertheless a correct evaluation of this contribution would help to better understand the physics of proteins which has implications in many important fields such as in drug industry.

In this work we use a basic model of the coil-to-globule transition of polymers consisting in self avoiding walks on the lattice with attraction between close monomers. To this model we add internal vibrations which will contribute with an entropic term to the free energy, enhancing the all-or-none character of the transition. Although this effect was not been directly measured, we present a comparison with experiments on related quantities.

Potpourri of Concepts 16.00 – 17.15

Glassy dynamics of gelatin gels

Alan Parker and Valéry Normand

Corporate Research, Firmenich SA, Geneva

We measure the elasticity of thermoreversible gelatin gels during slow cool and heat cycles, with and without stops. The responses, which are measures of the system energy, show the same memory and rejuvenation effects as spin glasses and polymer glasses. At constant temperature, all gels reach an aging regime, where their rate of response decreases linearly in log(time), like polymer glasses and spin glasses. The memory effects after upward temperature jumps also share features with those of spin glasses. We suggest that the parallels between these microscopically diverse systems are due to their temperature-dependant, rugged, hierarchical energy landscapes. Gelatin gels act like "colloidal spin glasses", with length and time scales that should allow the relation between mesoscopic dynamics and macroscopic behavior to be measured experimentally.

Depletion interactions in charged, aqueous colloid / polymer-mixtures

Kitty van Gruijthuijsen [1], Najet Mahmoudi [1], Anna Stradner [1], Remco Tuinier [2],
Marc Obiols-Rabasa [3], Peter Schurtenberger [3]

[1] Adolphe Merkle Institute, University of Fribourg
[2] Van't Hoff Laboratory, University of Utrecht
[3] Physical Chemistry, Lund University

Food and materials sciences have started to enormously profit from parallel developments in soft matter physics. Especially insight into model systems without specific interactions, like hard spheres, charged spheres, and ideal polymers, can easily be extrapolated to their industrial counterparts. In mixtures of these model components, steric exclusion of the polymer from an area around the colloid results in an attractive colloid-colloid interaction potential, the so-called depletion attraction. Theoretical models to predict depletion-induced phase behaviour apply both to organic model systems, as well as to mixtures of natural ingredients. In a next step towards reality's complexity we introduce electrostatic repulsions between the colloids, creating an interesting playing field to study phenomena like phase separation, transient gelation, and more exotic cluster formation. Latex colloids that are both sterically and electrostatically stabilized ensure reversibility of the observed phase behaviour, and increase control over the particle stability. We systematically vary salt concentration and polymer size to cover a series of ratios between the ranges of attraction and repulsion. The resulting phase diagrams are compared to the relatively straightforward Generalized Free Volume Theory.

Pseudo-cellular nanoprocessors are able to fight oxidative stress

P. Tanner, O. Onaca, V. Balasubramanian, G. Palivan

Chemistry Department, University of Basel

Oxidative stress, which is due primarily to an imbalance in reactive oxygen species (ROS) such as superoxide radicals, peroxynitrite, or hydrogen peroxide, represents a significant factor both in pathological conditions ranging from arthritis to cancer, and in toxicological conditions related to the presence of inorganic particles at nanometer scale. Here, we introduce a tool the pseudo-cellular nanoprocessor as part of a new concept with potential application in therapy. Combining two enzymes that act in tandem in situ in the cavities of polymeric vesicles that have the architecture and flow-of-information as known from electronics processors allows us to obtain a highly sensitive catalytic system. This represents an effective means to simultaneously detect and combat superoxide radicals, and related H2O2. Superoxide dismutase and lactoperoxidase were selected as a model highlighting the combination of enzymes. These were shown to participate in a cascade reaction and in situ catalysis in the cavities of nanovesicles, transforming superoxide radicals to molecular oxygen and water and thereby mimicking their natural behavior. The channel protein, outer membrane protein F, facilitated the input/output flow of LPO substrate/products, and dramatically increased the penetration of superoxide radicals through the polymer membrane, as established by nanoprocessor activity assays. The nanoprocessor remaines active after up-take by THP-1 cells, thus exemplifying an effective approach to enzyme therapy. This concept, meets the challenges of simultaneously detecting and detoxifying superoxide radicals and related, harmful peroxides, thus exemplifying an effective approach to enzyme therapy in oxidative stress.

Multi-method approach to characterize superplasticizers behavior at the solid-liquid interface

Lucia Ferrari, Josef Kaufmann, Frank Winnefeld

Laboratory for Concrete/Construction Chemistry, EMPA

Superplasticizers (also known as High-Range Water-Reducing Admixtures) are fundamental cement admix-tures which disperse the colloidal particles in fresh cementitious mixtures. Besides improving the workability properties, this effect drastically improves many other properties and allows the production of high performance concrete or self-consolidating concretes. Despite this wide use of superplasticizers, their fundamental mechanisms of interaction at the particle-water interface are not completely understood yet, leading to unex-pected effects. The main working principles of these admixtures are related to their adsorption on colloidal particles and dispersion nano-forces exerted by superplasticizers, which have high influence on the rheol-ogy. Furthermore, the adsorption process provokes significant alteration of the zeta potential of the particles in the suspension.

A study including all these techniques (adsorption isotherms, force measurements by atomic force micros-copy (AFM), zeta potential, and rheology) is presented here. A comprehensive understanding of superplasti-cizers fundamentals principles is emphasized. Besides fresh cement pastes, synthetic solutions in combina-tion with model inert powders are used to monitor the effect of different ions content. A detailed analysis about polymer architecture and about ionic species in solution is presented in order to illustrate the influence of these parameters on superplasticizer behavior at the solid-liquid interface. Moreover, measurements with AFM clarified the formation of superplasticizer multilayer, and highlighted changes in side chain conformation according to the solution used to monitor it.

From powders to collapsible structures / "living quicksand": discrete modeling

D. Kadau and H. J. Hermann
Institute for building materials, ETH Zürich

The discrete element method constitutes a general class of modeling techniques to simulate the microscopic behavior (i.e., at the particle scale) of granular/soil materials. We present a variant of the contact dynamics method, originally developed to model compact and dry systems with lasting contacts. This variant accounts for the cohesive nature of fine powders and soils. We use a modification of the model to investigate the "quicksand" behavior of a collapsing soil/suspension material. Our model is validated with real data obtained from in situ measurements performed with a specific type of natural quicksand at the shore of drying lagoons (1-2).

The loose granular structures involved in collapsing soils/suspensions and many other materials are investigated in detail. We study the influence of the granular bond number on the density profiles and the generation process of the loose packings, in our case a ballistic deposition process while at the same time the structures collapse due to gravity. This collapse occurs discontinuously in slides which are the basis for developing a model for the final density profiles.

(1) D. Kadau, H.J. Hermann, J.S. Andrade Jr., A.D. Araujo, L.J.C. Bezerra, L.P. Maia, Living Quicksand, Granular Matter 11, 67-71 (2009)

(2) D. Kadau, H.J. Hermann, J.S. Andrade Jr., Collapsing granular suspensions, Europ.Phys.J.E 30, 275-281 (2009)

Abstracts Posters

Assemblies & Materials

A1: Monitoring structural changes in molecular layers with surface plasmon resonance

R.L. Schoch, L.E. Kapinos, R.Y.H. Lim

Biozentrum, Swiss Nanoscience Institute, University of Basel

Here, we have conceived of a method for measuring structural changes in a molecular layer without requiring a direct measurement of the refractive index. As surface plasmon resonance (SPR) is a surface sensitive method, we obtain a value for the excluded volume of a surface tethered molecular layer from the SPR signal arising from non-interacting particles. As a proof of concept, the method was tested by measuring the signal of bovine serum albumin (BSA) atop a polyethylene glycol (PEG) layer. The sensitivity of the method allowed us to distinguish between different length PEGs. Measuring structural changes in a PEG layer might then include the binding of anti-PEG antibodies to a surface tethered PEG layer.

A2: Characteristics of polyglycerol esters at air-water interfaces

C.S. Curschellas [1], D.Z. Gunes [2], H.J. Limbach [2], M.E. Leser [2], E.J. Windhab [1]

[1] Laboratory of Food Process Engineering, ETH Zürich
[2] Nestlé Research Center, Lausanne

The lamellar phase forming surfactant Polyglycerol Ester (PGE) is a good foaming agent. The free fatty acids, which are residues from the production process, play a major role in self-assemblies of this polyglycerol ester (1). This work focuses on the influence of NaCl on the structures formed at the air-water interface. Interfacial shear and dilatational rheology was chosen to further characterize these interfaces. Data acquired at long adsorption times can help in understanding foam stability, and the diffusion rate of the surfactant from the bulk to the air-water interface can help understanding foamability (2).

(1) Duerr-Auster, N., Eisele, T., Wepf, R., Gunde, R., Windhab, E. J. Colloid and Interface Sci. 2008, 327, 446
(2) Rodriguez Patino, J.M., Sanchez, C.C., Rodriguez Nino, M.R. Adv. Colloid Interface Sci. 2008, 140, 95

A3: Counterion dependent swelling of redox-active polyelectrolyte multilayers

R. Zahn [1], F. Boulmedais [2], J. Vörös [1], P. Schaaf [2] and T. Zambelli [1]

[1] Laboratory of Biosensors and Bioelectronics, ETH Zürich
[2] Institut Charles Sadron, CNRS, Strasbourg

Polyelectrolyte Multilayers (PEMs), formed by alternating layer-by-layer (LBL) deposition of polyanions and polycations, have been widely studied during the last decade. The properties of these layers can be tuned by varying the PEM composition, and thus present a promising tool for a wide range of applications.

Here we present a PEM consisting of alternating layers of Poly-L-Glutamic Acid (PGA) and Poly-(Allylamine Hydrochloride) (PAH) containing Ferrocyanide (FC) ions as electrochemically active species. The PEM buildup was monitored in situ using Quartz Crystal Microbalance with Dissipation monitoring (QCM-D). Ferrocyanide (FC) ions were added to the film and cyclic voltammetry was performed to oxidize and reduce the ions.

We found that oxidation and reduction of the incorporated FC ions caused the expansion and contraction of the PEM films. Applying 0.6 V caused the FC ions to be oxidized from [Fe(CN)6]4- to [Fe(CN)6]3-, leading to a thicker and less rigid film. This was observed using electrochemical QCM-D (EC-QCM-D). Subsequent reduction of the ions caused the film to contract back to its original state. Measurements involving different counter ions showed a strong dependency on the anion species, charge, and molarity. No such effects could be shown for different cations. The swelling behavior of the PEM films is also influenced by changes in the pH of the buffer.

The observed swelling behavior is attributed to charge compensation by counter ions. Upon oxidation of the FC, anions diffuse into the PEM and replace the missing negative charge. These ions, and their accompanying hydration shell, cause an increase in the osmotic pressure within the layer, which leads to the observed swelling behavior. Depending on their thermodynamic properties (charge, hydration shell thickness, and hydration entropy), a certain fraction of the counter anions can condensate on the PAH-FC complexes and decrease the swelling. This dependency of the swelling effect on the specific interaction of the polymer to the counter ion is also verified by Isothermal Titration Calorimetry (ITC) and Attenuated Total internal Reflectance Fourier Transformed InfraRed (ATR-FTIR) spectroscopy. High ionic strengths and pH values result in partial Donnan breakdown, which allows water to enter and cations to leave the PEMs.

To conclude, we have investigated a system of electroactive PEMs. Their swelling response can be tuned by choosing different counter ions and pH conditions.

A4: From polymer vesicles to bulk polymer

Jörg Braun and Wolfgang Meier

Department of Chemistry, University of Basel

The formation of polymer vesicles is still not totally understood form the point of view of assembly depending on molecular properties. Our interest is to characterize the different morphologies of amphiphilic block-copolymers in aqueous solutions from dilute polymer systems to the bulk. The phase diagram showing the transition from lyotropic phases to isotropic phases will allow understanding the formation of vesicles in more details.

Four amphiphilic block-copolymers based on poly(ethylene oxide)-co-poly(Î³-methyl-Îµ-caprolactone) - PEO-b-PMCL - with a constant hydrophilic block and varying the hydrophobic Î³-methyl-caprolactone one were synthesized via anionic ring opening polymerization and characterized. Several semi-equilibrium solutions were prepared either by stirring or by shaking and analyzed by dynamic and static light scattering, transmission electron microscopy and small-angle x-ray scattering.

At low concentrations PEO23-b-PMCL32, PEO23-b-PMCL44 and PEO23-b-PMCL54 self-assemble into vesicles with different sizes, the biggest morphology being self-assembled from PEO23-b-PMCL54. Increasing the polymer concentration leads to packing of vesicles, and starting at approximately 20 wt% a lamellar phase is formed. This gets more and more pronounced as the polymer concentration is increasing until 80 wt% where a transition phase occurs, followed by a disordered phase. In the case of the longest polymer chain, just before the transition-phase, an inverse hexagonal phase (H2) is observed. In the case of PEO23-b-PMCL25 besides of vesicles also rod-like micelles are present. Both vesicles and rod-like micelles are packing in a hexagonal manner phase (H1), before the lamellar phase is observed.

These details we obtained from the phase diagram are essential to understand the vesicles formation, and thus control it for future technological applications.

A5: Polymeric nanoreactors: a new way to improve antioxidant therapy

V. Balasubramanian, O. Onaca, F. Axthelm, W. Meier, C. G. Palivan

Department of Chemistry, University of Basel

Many naturally occurring proteins offer promise, particularly in diseases caused by insufficient amounts or inactive variants of those proteins. Unfortunately, this promise has not yet been fulfilled (except insulin and growth factor), largely because of significant barriers to effective bioavailability, as it is the case for antioxidant enzymes, such as superoxide dismutase (SOD), catalase, peroxidases. Even if a variety of approaches have been examined to address these bioavailability limitations, including the synthesis of protein mimics, modified proteins or encapsulation in liposomes/polymeric carriers from where they are delivered, these approaches have various drawbacks, such as instability, loose of activity during the modification, or short circulation lifetime, and release of the compound in non-controlled conditions/biological compartments. Nanoreactors based on encapsulation of active compounds in polymeric vesicles that enable them to act in situ, have recently been developed as a new way for their protection.

Here we present the concept of antioxidant nanoreactors based on the encapsulation of SOD-mimics (SODm) in polymeric vesicles with oxygen permeable membrane. Superoxide anions (O2•–) represent one of the major reactive oxygen species involved in oxidative stress in cells, and known to play an important role in diseases such as asthma, atherosclerosis, neurodegenerative diseases, cancer and AIDS. The antioxidant nanoreactors we develop serve to protect the antioxidant agents and simultaneously to let them act in situ.

First, the antioxidant nanoreactor was formed by the encapsulation of SODm in amphiphilic block copolymer vesicles made of poly-(2-methyloxazoline)-poly (dimethylsiloxane)-poly (2-methyloxazoline). We choose amphiphilic block copolymers because their self-assembly as vesicles allow the simultaneous encapsulation of the active compound in mild conditions that are supposed to not affect the sensitive enzyme. The function of this antioxidant nanoreactor was tested by pulse radiolysis, which demonstrated that SOD remains active inside and detoxifies the superoxide radical in situ. We ensure the system membrane is oxygen permeable and thus fulfills a double role: to shield the antioxidant agent and to enable agent and substrate to react inside the polymeric “cage”. In this way it is possible to improve the bioavailability of an antioxidant agent and to avoid the disadvantages of conventional drug release carriers, such as uncontrolled release of the antioxidant agent. Furthermore, we optimize the antioxidant nanoreactors by encapsulating low molecular weight SODm, by changing the procedure of vesicles formation or by modifying the molecular properties of the vesicles membrane in order to increase their ability to detoxify $O2^{\overline{\ }}$.

The antioxidant nanoreactor was tested for cellular uptake, cytotoxicity in monocytes (THP-1) to assess conditions for possible biomedical applications. Confocal laser scanning microscopy analysis allowed us to see the intracellular uptake and distribution of the nanoreactors. The quantitative uptake was estimated through flow cytometry, and approximately 11% of cells contained internalized nanoreactors even without a functionalized vesicle surface to support a targeted approach. Located mainly in endosome-like compartments, the antioxidant nanoreactors showed minimal cytotoxicity after 24 h incubation compared to free CuIISODm. This clearly points to a unique method to provide SOD-mimic-based antioxidant therapy, whereby encapsulating mimics in polymeric nanoreactors avoids mimic degradation within biological compartments and simultaneously allows SODm to act in situ when concentrated inside nanometer-range polymer cavities.

A6: Synthesis of new silver compounds with nicotinic acid derivatives

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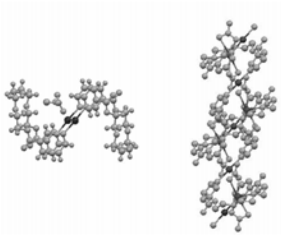
Complexes with silver ions have a great potential for applications in medical uses. Those complexes with ligands derived from nicotinic acid and with polyethylene oxide groups as spacer are synthesized. Upon coordination to silver ions, polymorphism can be observed: metallacycles and, under special conditions, chain structures are obtained.



Finally we propose the development of complexes with longer spacers to study the possible formation of liquid crystals.

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A7: Relationships between the morphology, water uptake, and proton conductivity of radiation grafted proton exchange membranes

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Small-angle neutron scattering contrast-variation study of hydrated proton exchange membranes is presented and the relationships between morphology, water uptake, and proton conductivity are discussed. These polyelectrolyte membranes are synthesized by radiation-induced grafting of poly(ethylene-alt-tetrafluoroethylene), ETFE, with styrene in the presence of a crosslinker (divinylbenzene, DVB) and the polystyrene is sulfonated subsequently.

BioPhysics & BioOriented Materials

B1: Microrheology of healthy and cancerous cells using optical tweezers

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The mechanical properties of cells are critical for their shape, motility and interaction within the tissues which is involved in many physiological and pathological processes including progression of cancer (methastasis). Qualitatively it is known that cancerous cells are softer than normal cells but a detailed characterization is lacking. Here it is proposed to use optical tweezers microrheology (OTM) to measure and to compare two related types of cells: the healthy HCV29 and cancerous T24 epithelial cells of the human urine bladder. OTM is a non-invasive method to evaluate the mechanical parameters of the cells using sub-micrometer tracer particles embedded in the cytoplasm.

The mean-square displacement (MSD) of tracer positions allows to extract such physical quantities as stiffness, compliance, viscosities or dynamic moduli.

The new theoretical equation describing the subdiffusional MSD in the OT confined environment has been used for the data analysis and allowed to extract the viscoelastic moduli in the 1e-5-10 Hz frequency range. In both cell lines the subdiffusion has been observed with very close sub-diffusive exponents: 0.73±0.02 for HCV and 0.68±0.03 for T24, which correspond to the prediction of the model of semi-flexible polymers.

Such a combined analysis will allow a better understanding of the distribution of mechanical response functions between the different components of the cell and their contribution to the cancerogenesis.

B2: Towards a better understanding of eye lens transparency and cataract formation

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For proper vision, an impeccably transparent eye lens is indispensable. This is achieved by a complex architecture and a unique protein composition. Any disturbance of this highly ordered system leads to clouding of the lens. This so called cataract is very frequent in old people and the leading cause for blindness. Understanding interparticle interactions in eye lens proteins is of central importance because of their biological and medical relevance for cataract formation. In addition, due to their easily tunable interaction potentials, the characteristic eye lens proteins gamma- and alpha-crystallin are particularly interesting and ideally suited as well-defined model systems for colloids.

In the present study we investigated the structural and dynamic properties of gamma- and alpha protein solutions up to concentrations corresponding to those found in the eye lens. For this we used small-angle neutron scattering (SANS) combined with light scattering, rheological measurements and molecular dynamics simulations. Our results allow us to interpret the complex protein phase diagrams using simple models from colloid science. We find that the transparency of gamma- and alpha-crystallin mixtures is greatly enhanced by weak, short-range attractions between them at high concentrations comparable to those in the intact eye lens. Provided that these interactions are not too strong, such mutual attractions considerably decrease the critical temperature and corresponding opacity due to light scattering. Hence they are essential for lens transparency.

B3: Scaling of creep-compliance curves of β-lactoglobulin-based systems at pH 2

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In the present work, linear and non-linear interfacial shear rheological properties of β-lactoglobulin based systems were probed at the oil-water interface. Investigations were carried after a long time of adsorption and aging (16.7 hours), at pH 2, where there is a major contribution of repulsive interactions. The systems include native monomers and heat-induced β-lactoglobulin fibers (dialysed or not), of two different lengths. Remarkably, all creep-compliance curves could be superimposed using a single multiplicative factor for each within the window probed 1 - 1000s. They could be fitted to the function $J(t) = J_0 + b.t\text{^}\alpha$, with the same α value and ratio J_0/b for all systems.

B4: DNA interactions with PAMAM6 dendrimers: dynamics and structure evolution in 3D microchannels

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One way of deciphering chromatin organization is to mimic DNA induced condensation in vitro, using less complex systems. To this end, we monitored the self assembly of DNA with PAMAM 6 dendrimers, polycations that are similar in shape and number of charges to the nucleosome core particles. A combination of 3D microfluidics and small angle x-ray scattering (SAXS) is used to investigate the kinetics of this process. Our preliminary SAXS results reveal the evolution of assembly process along the channel length. Thus, providing dynamic structural information of a complex assembly process, discarding kinetically trapped reaction intermediates which are obtained with conventional bulk techniques.

B5: Solid-supported biomimetic block copolymers membranes

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The functionalization of surfaces through biomimetic block copolymer membranes aims at developing smart surfaces for biotechnological applications such as biosensing.

Amphiphilic block copolymer membranes were chosen instead of lipid membranes as mimics of biological membranes because of their properties, such as thickness, chemical and mechanical stability, lower permeability, fluidity, mobility, etc. Upon insertion of membrane proteins, these systems could allow for the preparation of mechanically and chemically robust and air-stable biosensor devices.

B6: Protein gates allow in situ enzymatic reactions in polymeric nanocompartments: Towards synthetic organelles

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In natural conditions, enzymes are exposed to different harmful influences as for example the presence of proteases. By encapsulating them inside polymeric vesicles the enzymes are protected, but also shielded from substrates that prevent their in situ use for development of medical applications. We have developed a model system where we can both encapsulate enzymes and simultaneously assure an exchange of substrate across the vesicle membrane, by insertion of channel proteins (1).

Our system consists of triblock copolymers, channel proteins and the encapsulated enzyme(s) and serves as an enzymatic nanoreactor. PMOXA-PDMS-PMOXA polymers form by self assembly mechanically stable vesicle structures and simultaneously encapsulate horseradish peroxidase and acid phosphatase and reconstitute channel proteins, respectively. The key point is represented by the outer membrane protein F (OmpF) from E. coli, a channel protein allowing passage of molecules with a molecular weight lower than 600 Da, which serves as a gate for the nanoreactor (2).

This system has been tested with several enzyme specific substrates. The conversion of substrates inside nanoreactors was followed by fluorescence and absorbance methods. Furthermore functionalized and active surfaces were obtained via a biotin streptavidin binding (3). Such surface functionalized nanoreactors are model systems for future applications in the development of biosensors, in analytics, microfluidics or single molecule spectroscopy.

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Colloids & Polymers

C1: Direct measurement of van der Waals forces between latex particles across aqueous electrolyte solutions

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Direct force measurements between amidine latex particles immersed in KCl solutions of varying ionic strength are carried out using colloidal probe technique based upon atomic force microscopy. Force profiles of positively charged amidine latex particles are fitted with the solutions to the non linear Poisson-Boltzmann equation using constant regulation parameter. Both the repulsive double layer and the attractive van der Waals components are found to decrease with increasing salt concentration. The latter effect is also verified by measuring van der Waals forces between neutral amidine latex particles immersed in the same KCl solutions of varying ionic strength.

C2: Charge reversal of sulfate latex particles by adsorption of cationic polyelectrolytes studied by direct force measurements

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Forces between negatively charged colloidal latex particles in the presence of adsorbed oppositely charged linear poly(ethylene imine) (LPEI) are studied with the colloidal probe technique based on atomic force microscopy (AFM). With increasing adsorbed amount of LPEI, the repulsive forces are weakened, subsequently become attractive, and finally they become repulsive again. At higher adsorbed amount, the surface saturates and the strength of the interactions remains constant. Comparison with electrophoretic mobility measurements one finds that the attractive forces occur at the isoelectric point (IEP), where the electrophoretic mobility vanishes. At the IEP, the attractive forces can be explained by van der Waals attractions.

C3: Phase-separation and gelation of protein-poly(ethylene oxide) mixtures: effect of electrostatic interactions

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Analogies between model systems in colloid physics and much more complex food systems have started to be utilized to advance our knowledge of colloidal food systems (1,2). Important recent developments in soft matter research are linked to interparticle interactions and stability of self-assembled particles, phase separation, and dynamical arrest. Of particular importance is the ability of suspensions of particles interacting via a short range attraction to form gels as a result of a competition between spinodal decomposition and dynamical arrest (3,4). The fact that such an arrested spinodal decomposition process and the structural and mechanical properties of the resulting solid-like material can be tuned via the interparticle interaction strength and range as well as the initial colloid volume fraction holds a high potential both from a fundamental soft matter physics as well as from an applied food science point of view. In food science, it opens routes to achieve gels at intermediate volume fraction without the necessity of heat treatment or pH variation. As most food systems contain food colloids with residual charges, the relevant interparticle interactions that drive phase separation and arrest are therefore not only dominated by hard core repulsion and a short range attraction, but also contain an additional contribution from a screened Coulomb repulsion that can also result in additional phenomena such as the formation of equilibrium clusters (5). We have thus investigated the phase diagram of casein-poly(ethylene oxide) mixtures and the resulting equilibrium and non-equilibrium structures with a special emphasis on the effect of electrostatic interactions on phase separation and gelation using diffusing wave spectroscopy and confocal laser scanning microscopy. We compare the experimental coexistence curves with the calculated curves predicted by the Generalized Free Volume Theory (6,7).

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C4: Nanoengineering of thermosensitive core-shell particles

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We report the synthesis of micrometric Polystyrene/Poly(N-isopropylmethacrylamide) composite core-shell microgels and their post processing following different methods recently employed for pure polystyrene latices. The different approaches are either mechanical or rely on phase separation in different solvent mixtures. Ellipsoidal, faceted or hollow hemispheroidal core-shell particles have been successfully obtained. The influence of the crosslinked shell on the resulting morphologies and its thermoresponsivity are discussed. Hereby we show that the extension of the different processes to a core-shell geometry offers new complex and tunable systems for the physics of colloids.

C5: In-situ polymerization as a route towards polymer-colloid nanocomposites

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Combining nanoparticles (NPs) and a polymer matrix in order to obtain nanocomposite (NC) systems in a controlled way remains a challenge. Any attempt to create NCs with novel tailored properties thus requires a sound understanding of the relevant parameters that allow a control of the size, anisotropy, volume fraction, orientation, dispersion and distribution of the NPs within the desired polymeric matrix. In the present work, we study the in-situ polymerization pathway toward NCs with a control over the quality of dispersion and distribution of the desired incorporated particles. As a starting point, Stöber silica NPs [1] are integrated into a polymethylmethacrylate matrix (PMMA). In order to achieve it, NPs are either surface modified using 3-(trimethoxysilyl) propylmethacrylate (TPM, 98 %) silane agent or not, followed by their dispersion within a MMA monomer either by successive centrifugations or drying steps. The thus obtained hybrid monomers [2] are further polymerized by a free radical thermally initiated polymerization. The overall process is depicted in the figure 1a. After the establishment of the NCs, characterization in terms of quality of dispersion and distribution of the particles within the polymeric matrix is performed using scattering methods (SAXS & SANS) and Transmitted Electron Microscopy (TEM). Synthetic pathways involving a control of the surface chemistry using TPM appear to increase significantly the homogeneity in terms of dispersion and distribution of the incorporated NPs [2]. Distributions for incorporated particles without any surface treatment appear to be strongly dependant on the transfer method of NPs within the MMA monomer. Finally an overview and perspectives on how we can tune morphology, concentration and orientation of the integrated NPs in order to create adaptive new polymer colloid nanomaterials, with an emphasis on the incorporation of ellipsoidal magnetically orientable NPs, will be given.

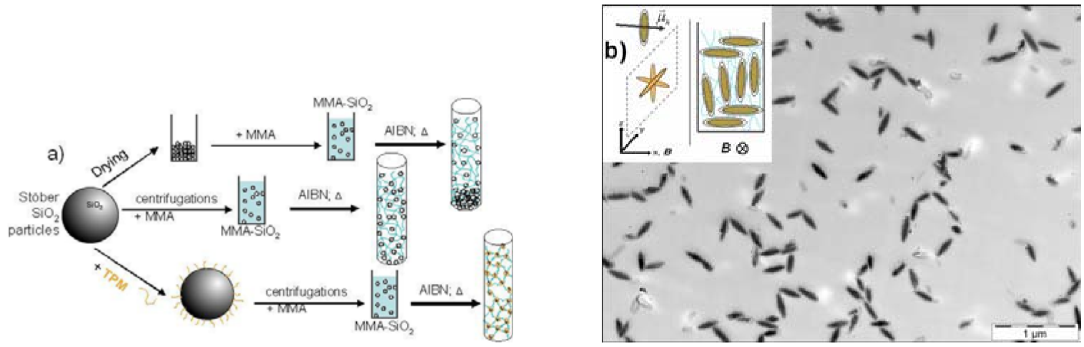

Figure 1: a) Schematic representation of the different methods of incorporation of nanoparticles into a polymer matrix using in-situ polymerization, b) TEM image of a 80nm microtomed nanocomposite of ellipsoidal nanoparticles within a PMMA matrix; as an inset, cartoons depicting oriented anisotropic particles within the polymeric matrix illustrate the TEM image.

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C6: Photo-healable metallosupramolecular polymers

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Metallopolymers are organic-inorganic hybrid materials that combine the unique properties of metal complexes (e.g., optical, magnetic, electronic) with the advantages of polymers at low cost, ease of processing, good mechanical properties. One important subset of these materials is the class of metallosupramolecular materials, where the metal-ligand interaction is dynamic in nature and thus acts as the supramolecular motif. Optically healable metallosupramolecular materials based on a poly(ethylene-co-butylene) polymer end-capped with 2,6-bis(1'-methylbenzimidazolyl)-44-ethynylpyridine (Mebip) ligands (BKB) and metal salts (Zn2+, La3+) were developed. Upon exposure of damaged polymers films of Zn2+-BKB to appropriate stimuli (e.g., light, heat), certain supramolecular motifs can easily disengage, causing a controlled local decrease of the polymer's molecular weight concomitant with an increase of the crack-healing rate. Films of Zn2+-BKB with deliberately introduced defects were shown to heal within minutes and with high efficiency upon exposure to ultraviolet light. One attractive feature of using light as a stimulus for dissociation of the supramolecular motifs is that it can be applied locally, i.e. at the defect site only, which allows one to heal a damaged object under load. Metallosupramolecular polymer films were also reinforced with cellulose nanofibers, or ‘whiskers’ that can form a percolating network and improve the mechanical properties of a material. The combination of Zn2+-BKB polymers and cellulose nanowhiskers gave rise to materials with an increased mechanical strength in comparison to neat polymer films. Metallosupramolecular nanocomposite films were deliberately damaged and then exposed to UV light. These materials healed in a similar fashion to Zn2+-BKB films and the original mechanical properties of the healed materials were recovered. These results demonstrate that metallsupramolecular polymers cannot only be reinforced by cellulose nanowhiskers, but that these materials can also heal efficiently. An overview on the parameters that govern the mechanical properties, light responsiveness, and healing ability of metallosupramolecular polymers will be presented.

C7: Where are the domains in ferroelectric polymers? A 2D piezoresponse force microscopy answer to an old question.

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Ferroelectric polymers offer a broad variety of applications from data storage devices to energy harvesting devices based on their genuine ferro-/piezoelectric properties. These physical properties are strongly determined by the local ferroelectric domain structure. However, compared to ferroelectrics like PZT the understanding of domains in polymers is still in its infancy. Here we report that only 2D PFM is able to reveal pristine domain structures in as-crystallized poly(vinylidene-stat- trifluorethylene) thin films and nanorods. We further proofed that with local and reversible domain switching it is possible to address four out of six possible domain orientations up to the µm scale.