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Physical Chemistry **453**

Beyond nature: a surface that resists wetting by water and oils

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Inspired by nature, many artificial surfaces have been developed in the last decade that mimic the so called Lotus Effect® [1]. On such so called superhydrophobic surfaces, drops of water remain almost spherical and easily roll off. Unknown in nature is a similar effect for oils. In fact, all natural and most artificial superhydrophobic surfaces face the problem of oily contamination. We have recently developed a coating [2] which, applied to textiles, results in fabrics that are completely non wettable by both water and oils. Even drops of non polar liquids with surface tensions below 30mN/m roll off these surfaces on slight inclination. To date, such surfaces are extremely rare and previously required complicated coating procedures or intricate surface manufacturing [3].

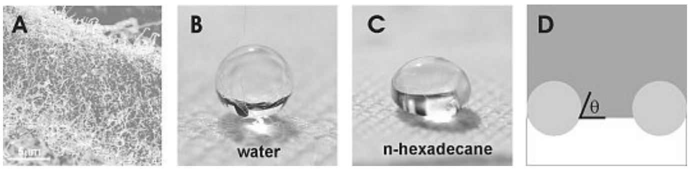

Figure 1. A: A polyester (PET) fiber coated with fluorosilane modified silicone nanofilaments (FSN). B/C: A drop of water/hexadecane on a PET fabric coated with FSN. D: Due to their special surface geometry, fibrous structures are able to resist wetting even though θ < 90° (adapted from [3])

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Physical Chemistry **454**

Corannulene on Cu(111) surface structure and properties using a first-principle theoretical approach

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The interaction of curved aromatic carbon surfaces with metal atoms represents an area of broad interest in organometallic chemistry. The intriguing question regarding the preference for metal binding to the concave versus convex side has been investigated both theoretically and experimentally, most notably focusing on Corannulene $C_{20}H_{10}$ (COR) [1] as prototype system. In this work, we study the specific case of COR adsorption on Cu(111) surface. The interest in this system stems from recent STM experimental findings, [2] which show that CORs adsorbed on Cu(111) gives rise to very peculiar supramolecular aggregation processes. In order to elucidate the fundamental mechanism involved, we adopt a first-principle electronic structure approach by adding to a standard DFT-GGA framework a semiempirical term taking into account dispersion contributions [3], employing the SIESTA code [4]. Structural relaxations of a single COR molecule located on top of the high symmetry sites of a six-layer Cu(111) slab are performed. Subsequently, electronic structure analysis is performed based on projected density of states (PDOS) and work function modification, upon molecule adsorption. Investigations of the Cu(100) slab are also of interest. Ultimately, we will extend structural analysis to the case of several molecules on the copper substrate so to distinguish the contributions of molecule-substrate interaction and molecule-molecule interaction to the adsorption mechanism.

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Polymer Science **455**

Soft Nanotechnology - from Colloid Chemistry to Nanostructured Functional Materials

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I will demonstrate how we can tune the size, shape, surface functionality and properties of nanoparticles and use them as ideal model systems for fundamental investigations as well as for materials applications. These particles can be used in order to study the equilibrium and non-equilibrium phase behavior of colloidal suspensions with different interaction potentials, and I will demonstrate how they allow us to investigate various phase transitions such as crystallization or gel and glass formation. While different nanoparticles are vital for fundamental studies of soft condensed matter, they also offer fascinating possibilities in materials science, and I will thus also demonstrate how we can create nanostructured materials with novel properties using functionalized and responsive particles.

Polymer Science **456**

Electrostatic Interactions between Colloidal Particles Covered with Oppositely Charged Dendrimers

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The colloidal probe technique, based on the atomic force microscope (AFM), is used to measure interaction forces between sulfate polystyrene latex particles covered with poly(amidoamine) (PAMAM) dendrimers, with different coverage, ionic strength and dendrimer generation. When increasing the adsorbed amount of dendrimer, the overall charge can be tuned from negative to positive values, through the isoelectric point (IEP). The potentials obtained from the direct force measurements between the colloidal particles agree well with the ones measured with electrophoresis. At the IEP, a long-range attraction is measured. Ionic strength and dendrimer generation influence the range of this interaction. The attractive force can be explained by the surface charge heterogeneities resulting from the lateral distribution of positively charged dendrimers onto negatively charged particles.

Polymer Science **457**

Preparation of Organic Nano-Particles for Electrophoretic Displays (E-Paper)

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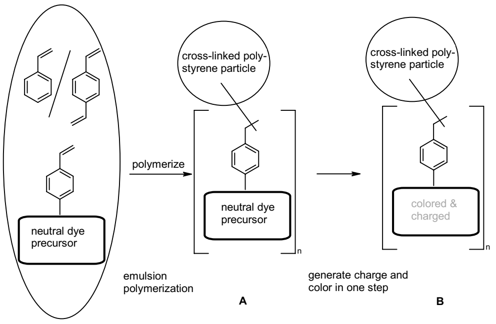
Wide-spread electronic displays suffer from various drawbacks (e.g. permanent energy consumption, fatiguing flickering etc.). Novel display systems based on the movement of charged nano-particles in an electrical field may overcome some of these drawbacks. Although this principle has been verified in the black&white mode, colored e-paper devices are still in their research/prototype state.

One of our approaches to tackle this issue is the use of appropriately functionalized organic nano-particles. Key to our novel synthesis scheme is the step-wise and therefore versatile preparation of the desired particles via an emulsion polymerization. That way the particle size and their appropriate color strength & charge can be controlled. We present the preparation of blue, yellow and magenta particles according this scheme. First application results are encouraging for further development of the concept.



Polymer Science **458**

Monomers Designed for Periodical, Lateral Growth Towards 2D Polymers

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Organic synthesis of 2D polymer remains an unexplored territory with great potential. Like graphene, a representative from Nature, a 2D polymer is a covalent macromolecular network being not only laterally infinite and one monomer unit thick, but also having periodical long-range order [1]. We are currently tackling this challenge head-on. Our strategy is based on rational monomer design which allows the growth only in lateral directions concurrently forming a periodically patterned network as well. Several anthracene-based cyclic monomers have been designed accordingly, where the anthrylene units are embedded in the cyclic skeleton at defined positions and in fixed orientations. By exploiting UV-induced [4+4] cycloadditon among the anthrylene units, these monomers are expected to polymerize in a "programmed" manner towards 2D polymers (Figure 1).

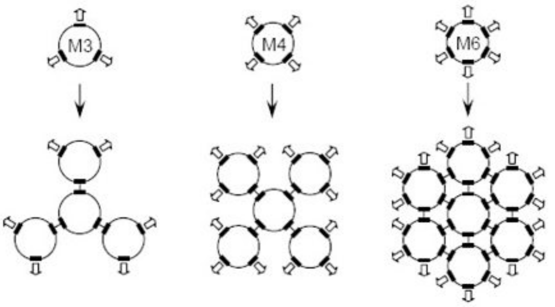

Figure 1. "Programmed" 2D Polymerization

[1] J. Sakamoto, J. van Heijst, O. Lukin, A. D. Schlüter, review submitted.

Polymer Science **459**

Improved Gilch Synthesis of Poly(*p*-phenylene vinylenes) for OLED Applications

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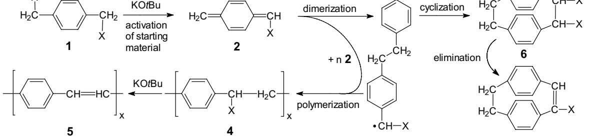
Poly(*p*-phenylene vinylenes) (PPVs) are fascinating semiconductor materials which are applied in devices such as organic light-emitting diodes (OLEDs). A powerful method for PPV preparation is the Gilch route: simple treatment of 1,4-bis(halomethylene)benzene derivatives **1** with tert.-butoxides leads to high-molecular-weight PPVs **5** in yields of 70-80%. The negative side of this process is formation of constitutional defects in the PPV, gelation of the reaction mixture, and difficult control of molar mass and chain architecture.

In order to improve this elegant access to PPVs systematically, profound understanding of the reaction mechanism is urgently required. With this in mind, we proved that α-halo-*p*-quinodimethane **2** represents the real monomer here. It is formed by single 1,6-dehydrohalogenation of starting material **1**. The subsequent chain propagation is initiated by diradicals **3** which appear via spontaneous dimerization of a small fraction of monomers **2**. This is also why [2.2]paracyclophanes such as **6** and **7** represent the dominant side products of this PPV synthesis. The poly(*p*-xylylene) derivative **4** formed by radical chain growth converts into PPV **5** via a final macromolecular dehydrohalogenation cascade.



In our contribution, further experimental evidence of the above conclusions will be presented. Also, we show that it is possible to develop improved reaction conditions based on these findings, allowing minimization of critical defects, proper control of molar masses, and reliable prevention of gel formation. The PPVs obtained via the "best procedure" recipe are comparable – in light intensity as well as in ageing and fatiguing behavior – with commercial materials such as Covion's "super yellow".

Polymer Science **460**

Block Copolymers at Interfaces

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Membrane-forming amphiphilic block copolymers trigger much scientific interest, focusing mainly on self-assembly properties and possible applications of such macromolecules. Biomimicking polymer membranes are often preferred over lipid bilayers for e.g. drug delivery studies, protein immobilization or sensor development, due to their robustness and long-term stability.

Polymer vesicles are good candidates as drug delivery vehicles, since long circulation times in the bloodstream and slow drug release rates can be expected. As a drug-loaded vesicle will sooner or later meet a cell membrane, it is interesting to study interactions between block copolymers and components of natural membranes. Influence of the material properties (polymer chemistry, size, charges) on mixing thermodynamics will be discussed for Langmuir monolayers at the air-water interface as a model system. In particular, we used poly(2-methyloxasoline)-poly(dimethylsiloxane)-poly(2-methyloxasoline) triblock copolymers of different sizes and studied their influence on lipid (dipalmitoylphosphatidylcholine, DPPC) monolayers by surface pressure-area isotherms and Brewster angle microscopy.

Since a certain degree of phase separation is expected in such mixtures, this phenomenon will be exploited further to allow control over miscibility by tuning the polymer properties and the mixture composition. Controlled phase-separated assemblies are especially interesting for applications in life sciences. For example, superstructures into which proteins are to be inserted, can offer a particular compartmentalization to best accommodate the protein and to assure its highest activity. Here, we will focus on physical chemistry of such multiphase systems in order to explain thermodynamic contributions to different morphologies and sizes of 2D aggregates.

Polymer Science **461**

Polymethine Dyes - From Photography to Solar Cells

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Polymethine dyes and polymers are interesting organic semiconductors for photovoltaic applications due to their particularly high absorption in the visible and near-infrared domain [1]. They own favourable redox potentials making it possible to use these materials as electron donors and acceptors. Furthermore, cationic polymethine dyes (cyanine dyes) offer the possibility to select a specific counter-anion in order to exploit the mobility of these ions under the influence of an electric field. Ionic space charge can be formed at organic heterointerfaces which allows controlling the electron transfer process [2]. Up to now, low charge carrier mobility has been a bottleneck for competing with those semiconductors presenting the highest organic solar cell efficiencies so far. Recently, it was shown that doping of the cyanine dye film greatly improves photovoltaic performance [3] demonstrating the full potential of this class of semiconductors.

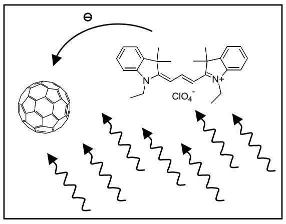

Figure: Schematic representation of the photo-induced electron transfer reaction between a cyanine dye and fullerene C_{60}.

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Polymer Science **462**

Synthesis of hydrophilic/hydrophobic patterned peptide and oligoester foldamers

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Hydrophobic effects have been proven to be prime contributors to the folding behavior and stabilization of proteins [1]. In particular, the distribution of polar (P) and non-polar (NP) residues along the protein's primary sequence plays a decisive role in the nature of the secondary and tertiary structure the protein will adopt [2, 3]. For instance, heptad repeat sequences of the type NP-P-P-NP-NP-P-NP have been shown to promote α-helical folding, while alternating P-NP sequences have been shown to produce β-sheet folding [1].

In order to investigate the influence of the nature of the hydrophilic side chains of polar amino acid residues arranged in α-helical and β-sheet promoting sequence, we have prepared 4 model peptides (**1**-**4**) containing L-allylglycine ($\mathrm{G_{allyl}}$) and L-leucine (L) amino acids. The allyl residues were transformed into groups bearing carboxylic acid, hydroxyl and amino groups in a post-modification procedure based on the radical addition of thiol reagents onto the allyl groups on the allylglycine residues. This method has been shown to be highly functional group tolerant, which will facilitate the production libraries of functional peptides based on the scaffold peptides **1**-**4**.

1: ($\mathrm{G_{allyl}}$-L-$\mathrm{G_{allyl}}$-L-$\mathrm{G_{allyl}}$-L-$\mathrm{G_{allyl}}$-L)$_1$
2: ($\mathrm{G_{allyl}}$-L-$\mathrm{G_{allyl}}$-L-$\mathrm{G_{allyl}}$-L-$\mathrm{G_{allyl}}$-L)$_2$
3: (L-$\mathrm{G_{allyl}}$-$\mathrm{G_{allyl}}$-L-L-$\mathrm{G_{allyl}}$-L)$_1$
4: (L-$\mathrm{G_{allyl}}$-$\mathrm{G_{allyl}}$-L-L-$\mathrm{G_{allyl}}$-L)$_2$

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Polymer Science **463**

Bioinspired structure formation with polyoxazolines

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Bioinspired functional polymers are raising more and more attention as advanced materials for key applications in materials science or biomedicine.[1] Especially interesting are polyoxazolines as non-toxic, biocompatible, and degradable polyamides or pseudopeptides. Well-defined polyoxazolines are available through a combination of controlled cationic polymerization and subsequent chemical modification techniques.[2]

Two polyoxazoline systems will be discussed regarding bioinspired structure formation:

(i) Poly(2-isopropyl-2-oxazoline), a structural isomer to polyleucine and to poly(*N*-isopropylacrylamide), is a thermo-responsive polymer and able to crystallize from dilute aqueous solution. Structures produced exhibit hierarchical ordering from the nanometer length scale up to several microns.[3,4]

(ii) Glycosylated poly(2-oxazoline), made by thio-click photoaddition of a thio-glucose derivative onto poly[2-(3-butenyl)-2-oxazoline],[5] can assemble into vesicles or nanotubes in aqueous solution. The formation of nanotubes is driven by intermolecular hydrogen bonding between the polyamide backbone (H-accepting) and pendant glucose units (H-donating) and not by hydrophobic interactions.[6]

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Polymer Science **464**

Weak polyelectrolyte adsorption on latex particles

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The adsorption of poly acrylic acid on positively charged amidine latex particles as a function of pH is investigated. Electrophoretic mobility measurements are used to characterize the behavior of latex particles as function of polymer dose in different ionic strengths. By means of light scattering techniques we characterize the thickness of adsorbed layer and the corresponding particle colloidal stability. Such measurements enable us to study the effect of the polyelectrolyte charge variation on the adsorption process.

Polymer Science **465**

Colloidal Stability in the Presence of Counterion Association

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The methodology of a generalized model [1] for colloidal stability has been validated against experimentally measured values of the stability ratio and critical coagulation concentrations (CCC), for electrolytes with mono or divalent cation. Besides the classical DLVO theory the generalized model accounts for association of cations with the particles surface charge groups. The model parameters are purely based on information available in the literature. For the monovalent salt the predictions agree well with the experimental data from literature [2], forecasting both the CCC values and stability ratios quantitatively. For the divalent salt the predictions for large values of the stability ratio tend to deviate from the experimental results, but it is noted that the onset of stability, i.e., the CCC, and small stability ratios are correctly forecast. Moreover, a comparison of the above results with those neglecting the effect of counterion association with the particles surface charge groups indicates that the latter substantially overestimates stability ratios in the presence of high salt concentrations, in the case of the monovalent salt, and leads to unrealistic large values of the CCC for the divalent salt.

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Polymer Science **466**

Dependence of Cluster-Cluster Aggregation Effciency on Shear Rate

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When producing or processing sub-micron particles, e.g., in the case of waste-water treatment, emulsion polymerization, or nano-particle precipitation, knowledge of the dependence of the aggregation kinetics on the shear rate is required for process design and control. In addition, the correct description of the aggregation kinetics is a key to properly model and simulate such processes, as well as the basis for the development and validation of any breakage model. Therefore, the aggregation kinetics of model colloidal systems consisting of polymer latexes with different primary particle diameter (120nm, 420nm, and 810nm) were investigated under turbulent conditions in stirred tank. It was found that the integral quantities of the cluster population measured by small-angle static light scattering, i.e., the root-mean-square radius of gyration, the zero-angle intensity of scattered light, and obscuration (extinction) all scale with a dimensionless time of the form $\alpha_{exp}\times\langle G\rangle\times\varphi\times t$. The prefactor represents an experimentally obtained aggregation efficiency which according to experimental data follows a power law of the form $\alpha_{exp}=(N_f)^n$, where N_f is a characteristic dimensionless group, which is proportional to the ratio of the Hamaker constant normalized by thermal energy over the Péclet number, and the value of the exponent n is negative and dependent on the primary particle size. The broad range of primary particles sizes allowed investigating the transition from diffusion-affected aggregation to purely shear-induced aggregation. The results of this work together with literature data provide a relation between the experimentally obtained n and N_f.

Polymer Science **467**

Liquid-Crystalline Dendrimers : Versatile Synthetic Plateforms for the Design of Supramolecular Functional Materials

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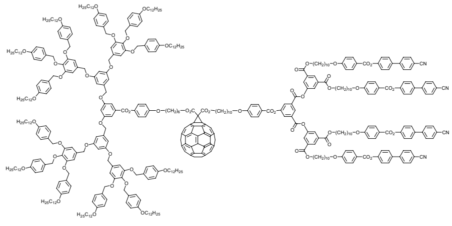
Convergent-type dendrimers are interesting macromolecules which possess a well-defined structure. Furthermore, their size, shape and functionality can be modulated by synthesis, generation by generation.

We have demonstrated that grafting liquid-crystalline dendrimers onto C_{60} by cycloaddition reaction[1] is an effective and elegant way to control the unfavorable effect (on the mesomorphic behavior) of the isotropic C_{60} unit (non mesomorphic dopant).[2]

We demonstrate here the precise role played by dendrimers for the design of fullerene-containing liquid crystals with tailor-made properties.



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Polymer Science **468**

Preparation and Characterization of Novel Ppolymer-Magnetic Colloids

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Magnetic nanocrystals are used in many fields, such as biomedicine, separation technology and material science. For many applications, the nanocrystals need to be encapsulated into a polymeric matrix, thus generating composite nanoparticles. These particles are composed of a core of magnetite nanocrystals and a shell of polymer. It is of utmost importance to control both the amount of nanocrystals encapsulated and the final particle size and size distribution.

In this work, we have prepared novel magnetic colloids having small size (<100nm) and uniform size distribution trough miniemulsion polymerization and have characterized their size distribution and the extent of magnetite incorporation via light scattering and TEM analysis. Magnetite nanocrystals have been prepared through coprecipitation reaction of mixtures of iron salts in presence of ammonium hydroxide and a ligand. Solubility of the nanocrystals in various organic solvents can be tuned by changing the ligands that are anchored on their surface. It was found that the optimization of magnetite encapsulation is strongly dependent upon the chemical nature of ligands, and this motivated us to test different ligands. Our results indicate that, if the ligand is not compatible with the polymer matrix, phase separation occurs and the magnetite accumulates at the surface of the polymer leading to colloidal instability of the dispersion. Ligands carrying reactive polymerizable moieties turned out be particularly effective to achieve a uniform dispersion of magnetite into the polymer matrix.

In order to increase the amount of magnetite incorporated in the magnetic colloid we have added a low boiling point solvent to the monomer before the emulsification. The solvent facilitates the emulsification by reducing the fraction of magnetic material in the droplets, and is subsequently evaporated during and/or after the polymerization reaction. In this manner, it is possible

Polymer Science **469**

Evidence and Relevance of a structural barrier for coalescence of soft sphere colloids

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Coalescence phenomena are widely known for heterogeneous systems with liquid-gas and liquid-liquid interfaces. Coalescence of particles is much less intuitive but occurs for elastomer colloids under certain conditions. As for bubbles or droplets, coalescence of elastomer particles depends in general on bulk viscosity and surface tension (surface energy) as driving force. Thus, prerequisite for coalescence is high polymer chain mobility as can be found for elastomer materials at temperatures sufficiently above so called glass transition temperature. In cases of very fast extrusion or immediate coalescence, drainage of liquid film between approaching surfaces and rupture of the liquid film will control the coalescence event. Our investigations aim at contributing to the fundamental understanding of elastomer particle coalescence on the basis of aggregation studies of soft elastomer particles.

Coalescence of elastomer particles can be induced by destabilization of the colloid. Modeling of the aggregation kinetics allows determination of the cluster structure where fractal structure is indicated by a low fractal dimension of i.e. 1.7 to 1.8 for perikinetic diffusion limited aggregation [1] and full coalescence by a fractal dimension of 3.0. Both situations have been found for elastomer latex of the same polymeric material but different surface characteristics. At 25°C anionic surfactant stabilized particles always coalesce, forming spherical clusters. Particles carrying fixed polar surface groups in contrast form fractal clusters upon aggregation. The coalescence behavior of the first particle type indicates no limitation with respect to flow ability of bulk material. Since both latexes are of identical bulk material, drainage and rupture of the separating liquid film are assumed to control the coalescence process. Intuitively, one may expect that any physicochemical parameters that reduce the mobility of water molecules in the contact region would restrict coalescence. On the other hand, it is known [2, 3] that any ionic species can affect the water structure at interface. Then, polar surface groups or cationic surfactants at the solid-liquid interface may raise a structural barrier between interacting particles. Such a non-DLVO, short range repulsive interaction is often referred to as hydration interaction. Although understanding of the complex phenomenon 'hydration barrier' is far from complete [2] it is generally accepted that an increase in thermal energy is associated with easier dehydration of ionic surface groups or counter ions respectively a reduced order or smaller extension of hydration layer at the particle surface. In this way the structural barrier between attractive particles should be reduced. Indeed, full coalescence for the particles with fixed polar surface groups can be found above a certain temperature, which might indicate the collapse or sufficient reduction of the structural barrier. Coalescence of the surfactant stabilized latex already at ambient temperature is explained by the mobility of surfactants that might get squeezed out of the interaction gap readily when particles approach.

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Polymer Science **470**

Peroxidase-catalyzed Aniline Polymerization in the Presence of Vesicles

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In the presence of anionic vesicles as templates and horseradish peroxidase/hydrogen peroxide as catalyst, aniline can be polymerized to yield mainly linear polyaniline, similarly to what has been reported previously if anionic micelles [1] or negatively charged polyelectrolytes [2] were used as templates. Following initial findings with vesicles formed from dodecylbenzenesulfonate and decanoic acid (1:1, mol/mol) [3], the polymerization reaction in the presence of vesicles was optimized and the reaction system was studied by a number of methods, including ultraviolet/visible/near infrared and electron spin resonance spectroscopy, as well as by cryo transmission electron microscopy and dynamic light scattering. Since the peroxidase is more stable in the presence of the vesicles, as compared to corresponding micelles, the vesicles are superior templates. Fluorescence measurements allowed proposing a first model for the role of the vesicles during polymerization.

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Polymer Science **471**

Use of Asymmetric Flow-Field Flow Fractionation to Characterize Aggregating Colloidal Dispersions

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The estimation of the size distribution of complex colloidal dispersions, such as dispersions of clusters of nanoparticles is a notoriously challenging task. Common characterization techniques include electron microscopy, light scattering, centrifugation, ultrasound spectroscopy etc.. Most of the above mentioned techniques, as for light scattering, provide some average size of the dispersion, but the reconstruction of the entire size distribution is often an ill-posed problem. Other techniques, like electron microscopy, are capable of analyzing individual particles and clusters, and can provide detailed information about the distribution, but require long times and require to dry the sample, a harsh process which can alter the structure of delicate clusters. Flow Field Flow Fractionation, on the other hand, is a well established chromatographic technique able to fractionate mixtures of colloidal nanoparticles having different sizes under gentle conditions. Usually, the extraction of a size distribution is carried out by analyzing the elution times of the samples and by estimating their hydrodynamic radii using a calibration curve. In this work, we make use of Asymmetric Flow Field Flow Fractionation coupled to a two angle static light scattering detector (SLS) to characterize suspensions of different polymeric colloidal nanoparticles undergoing stagnant aggregation under both diffusion and reaction-limited conditions. Two types of particles have been used: rubbery fluorinated colloids undergoing coalescence and hard polystyrene latexes forming fractal clusters. The cluster size distribution is quantitatively reconstructed not from the elution times, but from the analysis of the SLS data, which requires the knowledge of the scattering properties of both clusters and particles. The estimated cluster size distributions favorably compare with the calculated ones from Population-Balance-Equations. These results prove the effectiveness of AF4 in combination with multi-angle light scattering as a tool to quantitatively characterize the size distribution of complex colloidal dispersions.

Polymer Science **472**

Homo and Copolymerization of Cationic Double Charged Monomers: Kinetic Analysis and Characterization of the Resulting Polymers

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Analyzing the homopolymerization of the two double charged cationic monomers bis-1,3(N,N,N-trimethylammonium)-2-propylmethacrylate dichloride (di-M) and bis-1,3(N,N,N-trimethylammonium)-2-propylacrylate dichloride (di-A) [1] revealed strong non-ideality [2] indicated by an initiator reaction order higher than 0.5 and monomer reaction orders of 4.4 and 3.8 for di-M and di-A, respectively. Moreover, autoacceleration was identified at conversion lower than 10%.

The di-A and di-M polymers behave as strong polyelectrolytes in aqueous solution presenting significant differences of the reduced viscosity in water and in salt solution. Counterion activity coefficients as low as 0.11 confirm the strong polyelectrolyte character. Such low values are in agreement with polyelectrolyte theories for a charge distance of 0.12 nm.

Copolymerization with other cationic but less charged monomers, such as diallyldimethylammonium chloride and acryloyloxyethyltrimethylammonium chloride, and with the neutral acrylamide quantified the influence of the ionic strength on the monomer reactivity and proved the dominant role of the distance between the growing radical position and the charged group, whereas the number of charges per monomeric unit was found to be less important.

Kinetic analysis and polymer characterization are the basis for the mechanistic interpretation of the special features of the homo and copolymerization processes. Monomer association and electrostatic interactions seem to be of comparable importance.

Acknowledgements. The FNS supported this research, grant 200021-107737. Taminco N.V. Gent, Belgium, provided the monomers di-A and di-M.

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[2] R. Losada, C. Wandrey. *Macromol. Rapid Commun.* **2008**, *29*, 252.

Polymer Science **473**

Two-Dimensional Synthetic Polymers from Functionalized Calixarenes

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A most justified synthetic approach to the two-dimensional (2D) polymers, the laterally infinite sheet-like macromolecules with defined repeating fragments, consists in the covalent stabilization of self-assembled monolayers. However, microscopy techniques may fail as a tool to prove the molecular structure of 2D polymers comprising repeating fragments of sub-nanometer size. The nanosized calix[4]- and, especially, calix[6]arene derivatives, such as **1** and **2**, are promising subunits in this regard. They can be self-assembled at the air/water interface and photopolymerized.

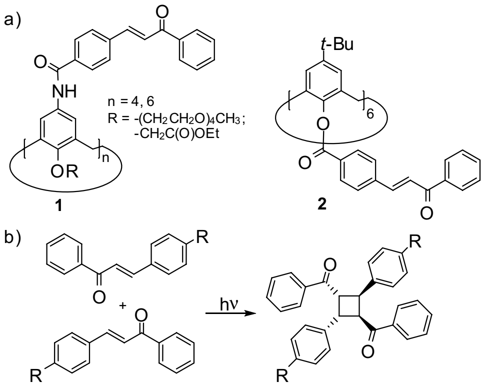

Figure 1. (a) Structures of the monomers for the 2D photopolymerization in Langmuir monolayers. (b) The underlying chemistry: photochemical [2+2] cycloaddition of chalcones.

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Polymer Science **474**

Hydrogel microspheres for cell immobilization - Approaches to improve stability and and biocompatibility

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Sodium alginate, a water-soluble anionic polyelectrolyte, is one of the most widely used polysaccharide for the development of hydrogel materials for cell entrapment. It was demonstrated that hydrogel microspheres, beads and capsules, based on the biopolymer alginate are principally suitable for cell immobilization with final applications in the biomedical field, especially for cells encapsulation with subsequent transplantation [1]. Nevertheless, for the majority of biomedical applications such microspheres are still suffering from insufficient mechanical stability and/or they provoke undesired cell and tissue responses. Using highly purified biopolymers could not yet completely solve all biocompatibility problems.

To overcome both mechanical stability and biocompatibility drawbacks, several approaches are under investigation. These include chemical modification of the alginate by grafting biocompatible or bioactive side chains before hydrogel and microsphere formation, reinforcement of the outer microcapsule hydrogel membrane by non-toxic cross-linking, and modification of the microsphere surface by coating, with or without subsequent covalent cross-linking. Special attention is dedicated to the selection the grafting reactions as well as to the coating material. In addition, physical surface properties governed by the hydrogel components, but also designable by the process conditions of the microsphere formation [2], are subject of the research.

Acknowledgment. The FNS supports this research, grant 205321-116397.

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Polymer Science **475**

Adsorption of poly-l-lysine on silica surfaces

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The adsorption of poly-l-lysine (PLL) on planar silica surfaces was studied as a function of pH and ionic strength. The measurements were performed on a home made fixed angle reflectometer equipped with an impinging jet cell.

In the early stages of the experiment, the adsorption process is transport limited and of first order with respect to the PLL concentration in solution. The adsorption rate increases with increasing ionic strength. Inversely, it decreases with increasing solution pH. These effects can be attributed to changes in the polymer's hydrodynamic radius.

Surface saturation is observed when the reflectometry signal reaches a plateau. The maximum adsorbed amount, obtained from this plateau, increases with pH and ionic strength. Our results are compared to other adsorption experiments carried out with poly(amido amine) dendrimers [1] as well as poly(diallyldimethyl ammonium chloride) [2] adsorbing on silica.

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Polymer Science **476**

Towards a living polymerization by a metallocene coordination catalyst at high temperature

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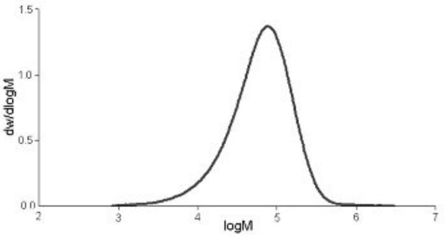
Our group introduced a method based on electrospray mass spectrometry to perform mechanistic studies of Ziegler-Natta polymerization[1]. The method allows to quickly determine if β-hydride transfer, β-hydride elimination or chain transfer to aluminum are important for a specific catalyst. We discovered a simple catalyst where no significant termination processes or chain transfer to aluminum occurs. Therefore, the catalyst has all the required properties to suggest a living polymerization.

To confirm the living nature of our polymerization, we analyzed the distribution of the polymer chains by gel permeation chromatography. The consumption of ethylene remains constant during the polymerization process, the molecular weight of the polymer increases linearly with time and the polydispersity narrows down to 1.5. Moreover, the catalyst has an activity comparable to other metallocene catalysts. Our preliminary results suggest that the metallocene catalyst has the capacity to perform living polymerization at rather high temperature (up to 60°C).



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Polymer Science **477**

Amphiphilic Diblock Copolymers for Molecular Recognition: Metal-NTA Functionalized Vesicles

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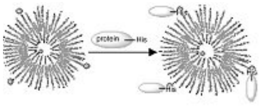
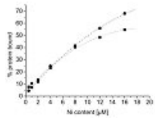
We present the formation and characterization of vesicles composed of metal-functionalized amphiphilic diblock copolymers for selective binding to histidine tagged proteins. We compare them to the metal-functionalised liposomes, previously obtained by [1], in order to show which are the advantages in using synthetic block copolymers. Mixtures of the metal functionalised copolymers with the respective non-functionalized block self assemble in aqueous solution in vesicular structures with a controlled density of the metal complex end-groups on their surface.





The vesicles in solution were characterized by light scattering techniques (DLS & SLS), optical microscopy, and TEM, while the metal centers we investigated by EPR. Fluorescence correlation spectroscopy clearly showed a significant and selective binding of His$_6$-EGFP to the vesicle surface. Vesicles and vesicles + His tagged proteins have been immobilized on surfaces and studied by AFM and QCM techniques.

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Polymer Science **478**

Dendronized Polymers with Crown Ether as Branching Units

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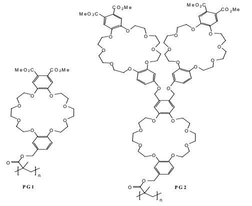
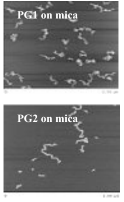
Dendronized polymers (denpols) are well defined, cylindrically shaped molecules and their nano-sized structure received considerable attention in the recent years. They have numerous potential applications ranging from catalyst supports to light-harvesting systems [1]. Crown ethers are fascinating units for phase transfer catalyst, ionic recognition, superstructure formation and nano-devices construction [2]. There have been some successful examples where crown ether unit was utilized as core unit or periphery decoration in dendron/dendrimer synthesis [3] and the interesting properties of crown ether decorated polymers are well studied. Here we describe the synthesis of a dendronized polymer with crown ether as branching unit and methacrylate as the backbone via macromonomer route in the first and second generation. These novel dendronized polymers should exhibit switchable properties upon complexation/decomplexation with metal ions at different solution pH values [4] without varying their aspect ratio. The change in the polymers conformation upon complexation with (in)organic salts in solution, i.e. the stiffness together with their contour lengths and helix induction with chiral guests, on the first and second generation polymers are currently under investigation and will be presented.





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H. Frauenrath, *Prog. Polym. Sci.,* **2005**, *30*, 325–384

Polymer Science **479**

Antioxidant nanoreactors: A way to fight superoxide radicals in oxidative stress

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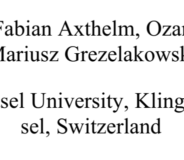
Reactive oxygen species, such as the superoxide radical anion (O_2^-), are responsible for oxidative stress in cells, which has been implicated in various cardiovascular and pulmonary diseases. Superoxide dismutase (SOD), a natural enzyme that converts O_2 into H_2O_2, cannot be a candidate drug for reinforcing antioxidant defenses, as it is quickly eliminated from the bloodstream.

We introduce here the concept of antioxidant nanoreactors in which the antioxidant agents (enzyme, or mimics) are encapsulated in amphiphilic copolymer nanovesicles whose membranes are oxygen permeable (Figure).



First we designed and tested an antioxidant nanoreactor based on encapsulation of SOD in amphiphilic copolymer nanovesicles made of poly-(2-methyloxazoline)-poly (dimethylsiloxane)-poly (2-methyloxazoline). To obtain a complete antioxidant nanoreactor we co-encapsulated a second enzyme, horseradish peroxidase, which is acting in combination with SOD, and give water and molecular oxygen, as final step of the detoxification cascade reaction. In this way the antioxidant agents are both active inside the polymerosomes and protected from the proteolytic attack, which is improving their biodisponibility.

Polymer Science **480**

Interaction between Colloidal Particles and Modified Electrodes: Long-Range Forces and Adhesion

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The adhesion of colloidal particles to solid surfaces represents a ubiquitous phenomenon in many industrial or natural processes. Despite the increased interest in electrowetting during the last years, only little attention has been paid to the applicability of this approach in order to tune the adhesion of colloidal particles. In this study we determine the changes in long-range forces and the adhesion between a colloidal particle and an electrode modified by a self-assembled monolayer (SAM) as function of the potential applied to the electrode. The force profiles and the pull-off forces are measured by atomic force microscopy. The interaction forces were not only determined in function of the applied potential but also for different surface terminations of the SAM. The long-range forces are of electrostatic origins and can be described quantitatively by fitting the interaction force profiles to the solutions of the non-linear Poisson-Boltzmann equation [1]. The pull-off forces provide a measure for the adhesion between colloidal particle and modified electrode. The different mechanisms contributing to the observed adhesion are discussed. These mechanisms include the variation of the interfacial tension according to the Lippmann equation as well as long-range forces of electrostatic origin. Furthermore, for a quantitative description the surface roughness has to be taken into account. We propose various strategies to optimize this approach for different applications such as nanomanipulation.

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Polymer Science **481**

Immobilization and hybridization of Oligonucleotides onto a conductive polymer thin film

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Shared expectations in gene therapy to cure genetically acquired diseases hold the attention and drives major research efforts to establish the human genomic map for instance. Tool-boxes like DNA microarrays, or DNA-chips, and biosensors have been used to reach a higher level of understanding of the genetic machinery and the underlying hybridization process. Even though the devices are on the market for the last 10 years, continuous research on their design and analysis is constantly undertaken to understand and optimize aspects which are still unclear.

Our approach is based on the immobilization of the oligonucleotide probes onto a conductive polymer thin film to study the hybridization process in label-free condition with surface-techniques such as the quartz-crystal microbalance, cyclic voltametry and impedance spectroscopy.

These studies enable the quantification of the immobilized probes and subsequent efficiency of hybridization to gain a deeper understanding of the mechanism occurring at surfaces. Aware of the current knowledge reached in polymer adsorption onto surfaces, assumptions can be made to further comprehend the mechanism of hybridization onto surfaces, which is necessary prior to improve the design and analysis of DNA microarrays.

Polymer Science **482**

Suzuki Polycondensation to "Stacking Polymerization"

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Suzuki polycondensation (SPC) has been developed into a key synthetic access to structurally defined polyarylenes [1]. By using this robust SPC protocol, we are developing "stacking polymerization" to create new polymer sequences in which discotic (or cyclic) molecules are covalently connected by rigid "clips" so as to be forced into a face-to-face, parallel and equidistant stack (Figure 1). Scheme 1 describes an example that was already performed [2]. Anthracene-1,8-ditriflate (**1**) [3] was employed as "clip" monomer and copolymerized with *p*-phenylene diborate (**2**) as simplest discotic monomer. A great variety of "*para*"-bifunctionalizable discotic compounds including porphyrins and polycyclic aromatic hydrocarbons would be applicative as latent monomers. Potential applications might be found in molecular wires as well as electronic devices such as field-effect transistors.

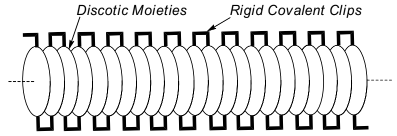
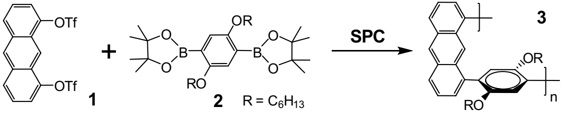


Figure 1. Face-to-Face Alignment of Discs Connected by Rigid Covalent Clips



Scheme 1. An Example of "Stacking Polymerization" by SPC

[1] A. D. Schlüter, *J. Polym. Sci. Part A: Polym. Chem.* **2001**, *39*, 1533. [2] Y. S. Sangvikar, J. Sakamoto, A. D. Schlüter, manuscript in preparation. [3] P. Kissel, F. Weibel, L. Federer, J. Sakamoto, A. D. Schlüter, *Synlett*, in press.

Polymer Science **483**

Anthracene-Based Cyclic Monomers for 2D Polymer Synthesis

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Synthesis of artificial 2D polymers consisting of a one monomer unit thick, covalently bonded periodical network remains uncharted territory in science [1]. We are challenging this problem by organic chemistry approaches. A cyclic monomer has been synthesized where three anthrylene units are embedded in the skeleton at defined positions and in defined orientations (Figure 1a). Its synthesis was carried out by using 1,8-diethynylanthracene [2] and terphenylene-based bridging units as key building blocks. By exploiting UV-induced [4+4] cycloaddition of anthracenes (Figure 1b), this monomer is expected to grow laterally so as to ideally form a 2D periodical network structure (Figure 2).

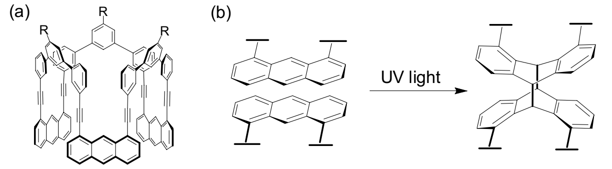

Figure 1. An Example of Anthracene-based Cyclic Monomers

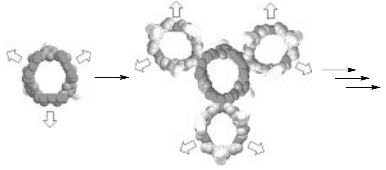

Figure 2. Expected Periodical, Lateral Growth of the Monomer

[1] J. Sakamoto, J. van Heijst, O. Lukin, A. D. Schlüter, review submitted. [2] P. Kissel, F. Weibel, L. Federer, J. Sakamoto, A. D. Schlüter, *Synlett*, in press.

Polymer Science **484**

Fate of Hydrophilic Nanocarriers in Biological Environments

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Hydrophilic nanocarriers, primarily formed by electrostatic interaction between their components, are recognized as nanoparticles having an enormous potential as vectors in biomedical and pharmaceutical applications. However, comprehensive information about the fate of such hydrophilic and potentially degradable nanoparticles is lacking. Due to the size, nanoparticles are, in general, able to enter and transport molecules into cells via pathways forbidden for bigger structures of even identical chemical nature. Understanding particle - cell interactions is crucial to develop safe and successful nanocarriers for biomedical and pharmaceutical applications [1].

Our system of interest is based on chitosan, a biocompatible and biodegradable polysaccharide. Chitosan nanoparticles are formed in an entirely water-based process by electrostatic interactions with other biocompatible molecules [2]. As a prerequisite for the fate investigations, comprehensive characterization and stability studies of the nanoparticles served to identify quantitatively the impact of the raw material characteristics and preparation conditions on the nanoparticles characteristics. Methods include IR and 1H NMR spectroscopy, dilution viscometry, particle size measurements and electron microscopy. Preliminary cytotoxicity and cell uptake experiments *in vitro* were performed to investigate the correlation with nanoparticle characteristics. Cytotoxicity was assessed by the MTT survival test and uptake was monitored in fluorescent microscopy using labeled polymers and/or loaded nanoparticles.

Acknowledgments. The research is supported by the FNS, grant 404740-117323, and Medipol SA.

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[2] WO/2007/031812