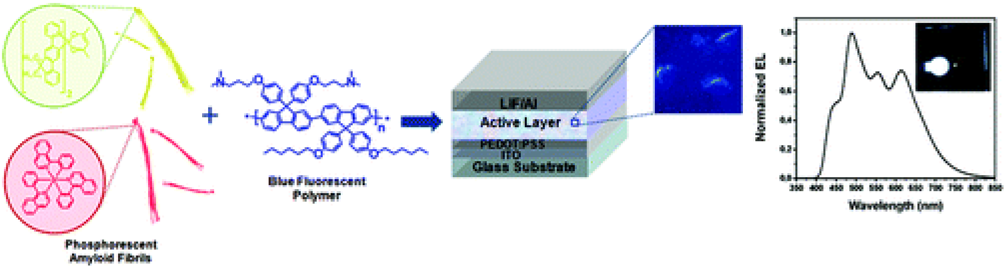
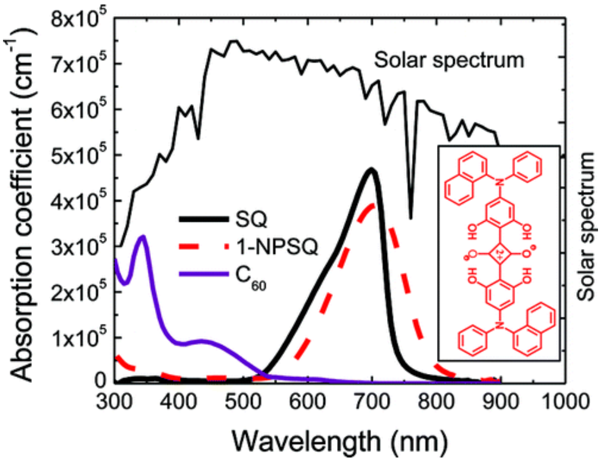
- White Light with Phosphorescent Protein Fibrils in OLEDs
  Rizzo, A.; Solin, N.; Lindgren, L. J.; Andersson, M. R.; Inganas, O. *Nano Lett.* **2010** *10*, 2225-2230.
  Abstract:



Red and yellow phosphorescent insulin amyloid fibrils are used as guest-emitting species within a blue-emitting polyfluorene matrix in light-emitting diodes. The integration of the phosphorescent Ir-complex into the amyloid structures strongly improves the triplet exciton confinement and allows the fabrication of white-emitting device with a very low loading of phosphorescent complex. The overall performances of the devices are improved in comparison with the corresponding bare Ir-complexes. This approach opens a way to explore novel device architectures and to understand the exciton/charge transfer dynamics in phosphorescent light emitting diodes.

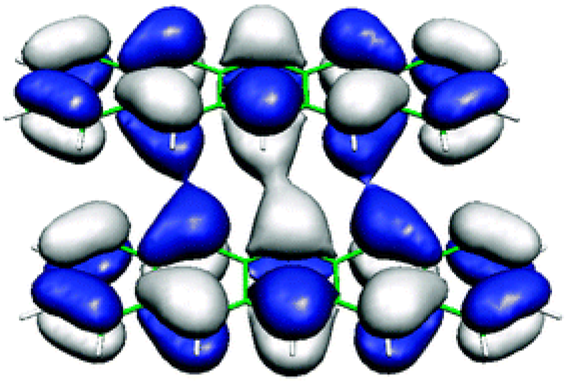
- Arylamine-Based Squaraine Donors for Use in Organic Solar Cells
  Wei, G.; Xiao, X.; Wang, S.; Zimmerman, J. D.; Sun, K.; Diev, V. V.; Thompson, M. E.; Forrest, S. R. *Nano Lett.* **2011**, *11*, 4261-4264.
  Abstract:



Squaraine (SQ) dyes are notable for their exceptionally high absorption coefficients extending from the green to the near-infrared. In this work, we utilize the functionalized SQ donor: 2,4-bis [4-(N-phenyl-1-naphthylamino)-2,6-dihydroxyphenyl] squaraine (1-NPSQ) by substitution of isobutylamines in the common "parent SQ" with arylamines to improve stacking and hence exciton and charge transport. The strong electron-withdrawing arylamine group results in a highest occupied molecular orbital energy of 5.3 eV, compared to 5.1 eV for the parent SQ, making 1-NPSQ a suitable donor when used with a C60 acceptor in an organic photovoltaic cell. Optimized and thermally annealed, nanocrystalline heterojunction 1-NPSQ/C60/bathocuproine solar cells with an open circuit voltage of 0.90(0.01 V, fill factor of 0.64(0.01, and short circuit current of 10.0 (1.1 mA/cm2 at 1 sun, AM1.5G illumination (solar spectrally corrected) result in a power conversion efficiency of 5.7 (0.6%.

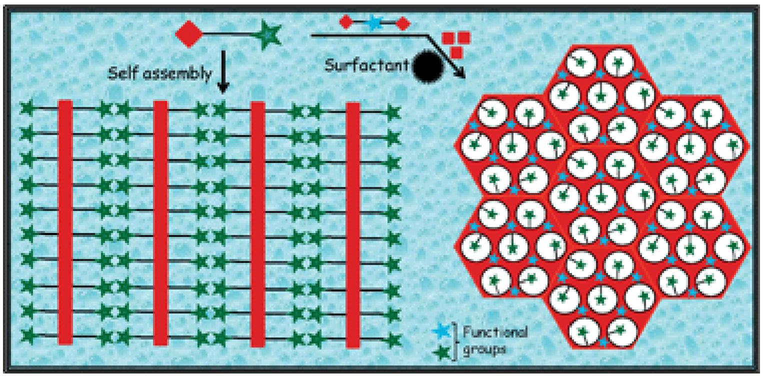
Crystallograpnic data suggest that the intermolecular stacking of 1-NPSQ molecules is closer than that of the parent SQ, thereby reducing the device series resistance and increasing its fill factor.

- Charge transport in high mobility molecular semiconductors: classical models and new theories
  Troisi, A. *Chem. Soc. Rev.* **2011**, *40*, 2347-2358.
  Abstract:



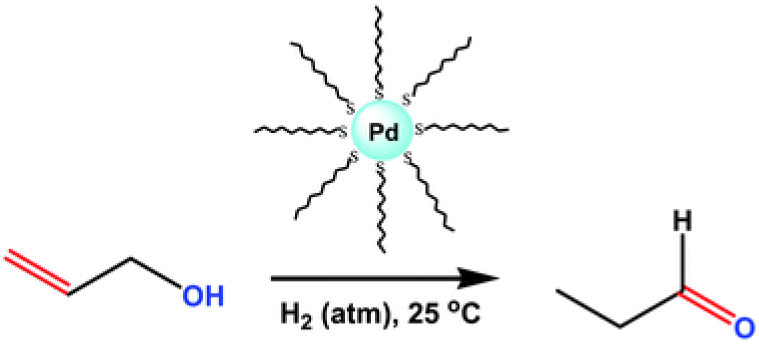
The theories developed since the fifties to describe charge transport in molecular crystals proved to be inadequate for the most promising classes of high mobility molecular semiconductors identified in the recent years, including for example pentacene and rubrene. After reviewing at an elementary level the classical theories, which still provide the language for the understanding of charge transport in these systems, this *tutorial review* outlines the recent experimental and computational evidence that prompted the development of new theories of charge transport in molecular crystals. A critical discussion will illustrate how very rarely it is possible to assume a charge hopping mechanism for high mobility organic crystals at any temperature. Recent models based on the effect of non-local electron–phonon coupling, dynamic disorder, coexistence of localized and delocalized states are reviewed. Additionally, a few more recent avenues of theoretical investigation, including the study of defect states, are discussed.

- From molecular chemistry to hybrid nanomaterials. Design and functionalization
  Mehdi, A.; Reye, C.; Corriu, R. *Chem. Soc. Rev.* **2011**, *40*, 563-574.
  Abstract:



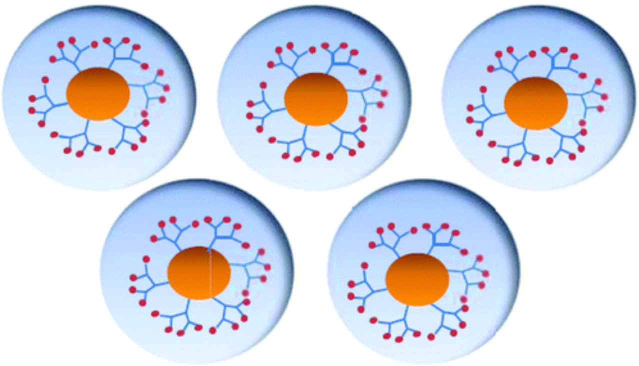
This *tutorial review* reports upon the organisation and functionalization of two families of hybrid organic–inorganic materials. We attempted to show in both cases the best ways permitting the organisation of materials in terms of properties at the nanometric scale. The first family concerns mesoporous hybrid organic–inorganic materials prepared in the presence of a structure-directing agent. We describe the functionalization of the channel pores of ordered mesoporous silica, that of the silica framework, as well as the functionalization of both of them simultaneously. This family is currently one of the best supports for exploring polyfunctional materials, which can provide a route to interactive materials. The second family concerns lamellar hybrid organic–inorganic materials which is a new class of nanostructured materials. These materials were first obtained by self-assembly, as a result of van der Waals interactions of bridged organosilica precursors containing long alkylene chains during the sol–gel process, without any structure directing agent. This methodology has been extended to functional materials. It is also shown that such materials can be obtained from monosilylated precursors.

- Synthesis and catalytic properties of alkanethiolate-capped Pd nanoparticles generated from sodium *S*-dodecylthiosulfate
  Sadeghmoghaddam, E.; Lam, C.; Choi, D.; Shon, Y.-S. *J. Mater. Chem.* **2011**, *21*, 307-312.
  Abstract:



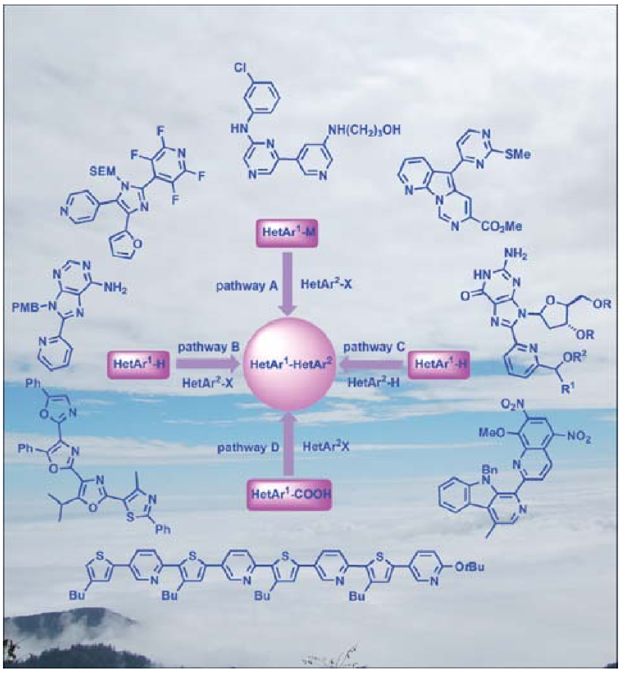
This article presents a synthetic method for alkanethiolate-functionalized Pd nanoparticles that are efficient catalysts for the isomerization of allyl alcohol to propanal. Pd nanoparticles are produced by the borohydride reduction of K_2PdCl_4 in toluene/H_2O using sodium *S*-dodecylthiosulfate as a source for the stabilizing ligands. The nanoparticles are characterized by ^{1}H NMR, Fourier transform infrared (FTIR) spectroscopy, transmission electron microscopy (TEM), UV-vis absorption spectroscopy, and thermogravimetric analysis (TGA). These analyses suggest that the monolayer capped Pd nanoparticles from sodium *S*-dodecylthiosulfate are quite comparable in composition (dodecylthiolate) and core size from those previously prepared from dodecanethiol. However, the catalytic activity of Pd nanoparticles generated from*S*-dodecylthiosulfate is found to be much greater than that of Pd nanoparticles prepared from dodecanethiol. The increased catalytic activity of Pd nanoparticles is likely to be due to the lower ligand density (organic weight fraction) of Pd nanoparticles generated from *S*-dodecylthiosulfate. The catalytic activity of PdAu nanoparticles on the isomerization of allyl alcohol is also demonstrated.

- Hybrid Semiconductor Nanoparticles: π-Conjugated Ligands and Nanostructured Films
  Park, Y.; Advincula, R. C. *Chem. Mater.* **2011**, *23*, 4273–4294.
  Abstract:



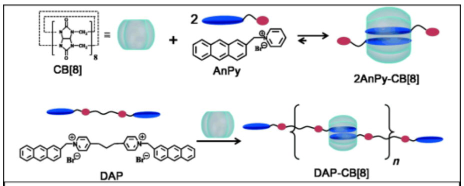
Hybrid inorganic–organic colloidal nanoparticles can be designed to achieve specific and complementary optoelectronic properties different from their sole organic and inorganic counterparts. The efficient coupling between organic and inorganic moieties facilitates optimization of these optoelectronic properties as single particles. Simply dispersing inorganic nanoparticles in an organic (polymer) matrix permits nanocomposite formation, but are usually prone to phase segregation. To achieve more-efficient energy transfer, charge carrier transport, and correspondence between energy levels of the inorganic and organic moieties, direct coupling is necessary. Ligand exchange with highly π-conjugated organic ligands or polymerization of optoelectronically active organic polymer on the surface of the inorganic nanoparticles, results in core–shell-like structures. This increases the surface-area-to-volume ratio contact between organic and inorganic moieties. Because of this advantage, energy transfer mechanism in hybrids can be tuned more efficiently for radiative or nonradiative decay. Recombination of excitons (bound electron–hole pairs) or the isolation of electrons (modulating charge transport) by controlling the conduction band-valence band (highest occupied molecular orbital–lowest unoccupied molecular orbital (HOMO–LUMO)) level becomes more tunable in donor–acceptor materials systems. Such optoelectronic property fine-tuning in a hybrid colloidal system can also be applied toward ultrathin film preparation and two-dimensional patterning (e.g., photovoltaic systems, light-emitting diode materials, sensors, and patterned arrays). A compatible organic shell facilitates greater solubility and dispersion of the inorganic core in a host polymer matrix. The use of a dendronic ligand provides an interesting method for facilitating surface functionalization, nanoparticle solubility, and electrochemical reactivity. By focusing on chalcogenide and semiconductor nanocrystals (NCs) or quantum dots (QDs), it is possible to limit these properties directly to charge carrier transport and energy transfer mechanisms. Each hybrid nanoparticle in essence carries the same properties replicated or dispersed within a film or a pattern. This review focuses on the design, preparation, and properties of such nanomaterial systems.

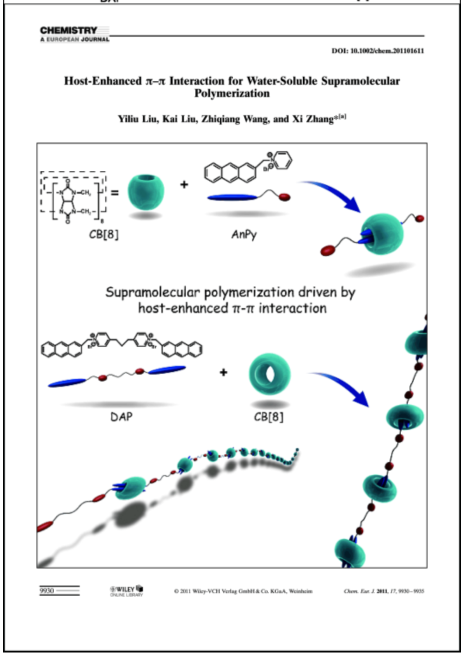
- Recent Progress in Coupling of Two Heteroarenes
  Zhao, D.; You, J.; Hu, C. *Chem. Eur. J.* **2011**, *17*, 5466-5492.
  Abstract:



The biheteroaryl structural motif is prevalent in polymers, advanced materials, liquid crystals, ligands, molecules of medicinal interest, and natural products. Many types of synthetic transformations have been known for the construction of heteroaryl–heteroaryl linkages. Coupling reactions provide one of the most efficient ways to achieve these biheterocyclic structures. In this review, four types of coupling reactions are discussed: 1) transition-metal-catalyzed coupling reactions of heteroaryl halides or surrogates with heteroarylmetals; 2) direct inter- and intramolecular heteroarylations of CH bonds of heteroarenes with heteroaryl halides or pseudohalides; 3) oxidative CH/CH homo- and cross-couplings of two unpreactivated heteroarenes; and 4) transition-metal-catalyzed decarboxylative cross-coupling reactions between haloheteroarenes or heteroarenes and heteroarenecarboxylic acids. The general purpose of this review is to give an exhaustive and clear picture in heteroaryl–heteroaryl bond formation as well as its application in the synthesis of natural products, pharmaceuticals, catalyst ligands, and materials.

- Host-Enhanced π–π Interaction for Water-Soluble Supramolecular Polymerization
  Liu, Y.; Liu, K.; Wang, Z.; Zhang, X. *Chem. Eur. J.* **2011**, *17*, 9930-9935.
  Abstract:





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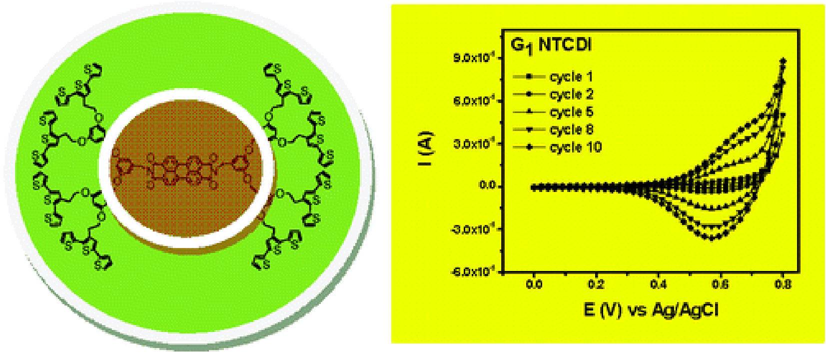
Host-Enhanced π–π Interaction for Water-Soluble Supramolecular Polymerization

Yiliu Liu, Kai Liu, Zhiqiang Wang, and Xi Zhang*[a]

9930 © 2011 Wiley-VCH Verlag GmbH & Co. KGaA, Weinheim *Chem. Eur. J.* **2011**, *17*, 9930–9935

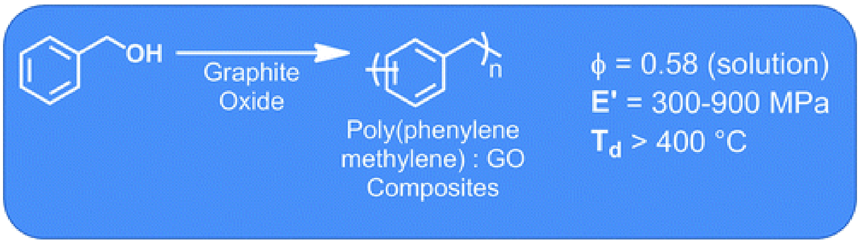
Host-enhanced π–π interaction based on anthracene derivatives and cucurbit[8]uril can be used as the driving force for constructing water-soluble supramolecular polymers. For this purpose, two anthracene moieties were encapsulated into one cucurbit[8]uril cavity, forming a ternary complex. After encapsulation in the host, the distance between the two anthracene moieties was shortened, and the π–π interaction between them was enhanced significantly. To realize supramolecular polymerization, a bifunctional monomer consisting of two anthracene moieties and a short linker in between was carefully designed. Cyclization was avoided in this way. Thus, host-enhanced π–π interaction can function as a new driving force for supramolecular polymerization.

- Electropolymerizable Terthiophene-Terminated Poly(aryl ether) Dendrimers with Naphthalene and Perylene Cores
  Ponnapati, R.; Felipe, M. J.; Advincula, R. *Macromolecules* **2011**, *44*, 7530–7537.
  Abstract:



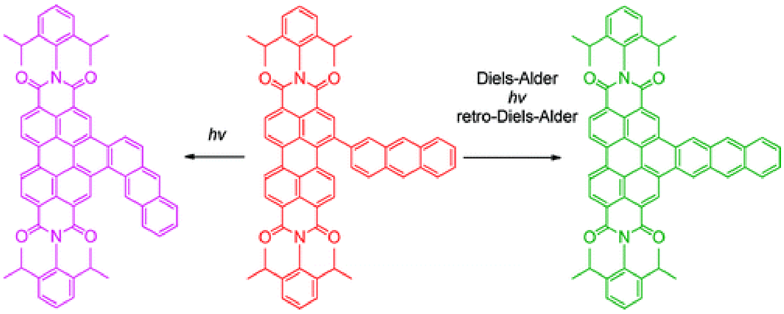
We describe the synthesis and characterization of a new series of poly(aryl ether) dendrimers with both donor and acceptor moieties. By incorporating p-type terthiophene and n-type perylene and naphthalene diimides into one molecule, we have achieved a dual electrochromic material. These dendrimers were designed to act as electrochemically active precursor polymers, which bear the electroactive terthiophene group that can be cross-linked electrochemically to form conjugated polymer network (CPN) films. Characterization of the optical and redox properties shows that it is possible to incorporate donor and acceptor moieties in the same molecule with retention of doping properties. The films made by electrochemical polymerization are highly active in both the p- and n-doping processes and have been investigated by spectroelectrochemical methods.

- Graphite Oxide as a Dehydrative Polymerization Catalyst: A One-Step Synthesis of Carbon-Reinforced Poly(phenylene methylene) Composites
  Dreyer, D. R.; Jarvis, K. A.; Ferreira, P. J.; Bielawski, C. W. *Macromolecules* **2011,** *44*, 7659–7667.
  Abstract:



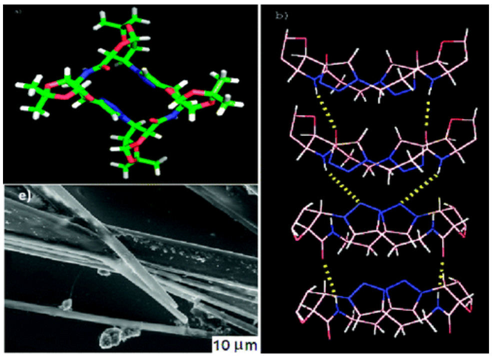
The synthesis and characterization of poly(phenylene methylene) (PPM) and carbon composites thereof are described. The materials were prepared using graphite oxide (GO), which was discovered to function in two distinct roles. First, the GO was found to facilitate the dehydrative polymerization of benzyl alcohol (BnOH) to form PPM. Second, the residual carbon from the GO catalyst, having undergone thermal deoxygenation during the polymerization reaction, served as a graphene-like additive in the resulting composite. While pure (i.e., additive-free) PPM was found to be mechanically compliant (E′ = 40 MPa), inclusion of 0.1 wt % GO in the starting reaction mixture improved the material's mechanical properties significantly (E′ = 320 MPa). Homogeneous dispersion of the additive in the matrix was confirmed by powder X-ray diffraction (PXRD) analysis, Raman spectroscopy, and transmission electron microscopy (TEM). The carbon additive was separated from the PPM via trituration in dichloromethane, and the GO starting material (C:O ratio = 2.0:1; σ = 4.45 × 10–5 S m–1) was found to have undergone significant reduction during the polymerization reaction (C:O ratio = 12.3:1; σ = 801 S m–1). Moreover, the recovered carbon did not assemble into graphite-like aggregates, as determined by PXRD, Raman spectroscopy, and TEM, which indicated that the PPM matrix was able to effectively disperse the additive.

- Anthraceno-Perylene Bisimides: The Precursor of a New Acene
  Li, Y.; Xu, L.; Liu, T.; Yu, Y.; Liu, H.; Li, Y.; Zhu, D. *Org. Lett.* **2011**, *13*, 5692–5695.
  Abstract:



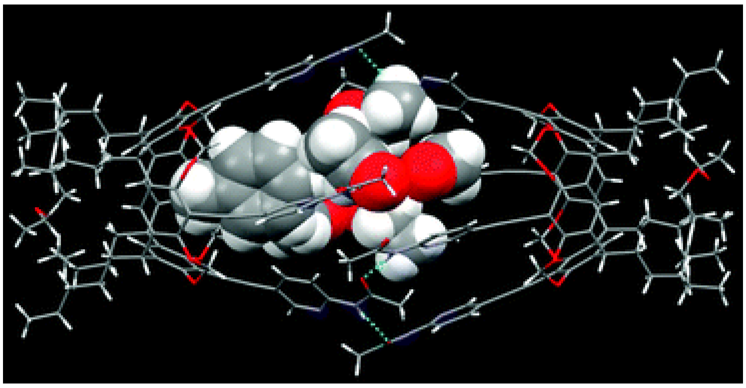
A controlled synthesis strategy for a anthracene-fused perylene bisimide was developed from the cyclization of an anthracene unit pendant to a perylene diimide scaffold. The direct cyclization led to a zigzag molecule, while a Diels–Alder strategy influenced the regiochemistry of cyclization to afford the linear precursor of a new acene.

- Simultaneous Parallel and Antiparallel Self-Assembly in a Triazole/Amide Macrocycle Conformationally Homologous to D-,L-α-Amino Acid Based Cyclic Peptides: NMR and Molecular Modeling Study
  Ghorai, A.; Gayen, A.; Kulsi, G.; Padmanaban, E.; Laskar, A.; Achari, B.; Mukhopadhyay, C.; Chattopadhyay, P. *Org. Lett.* **2011**, *13*, 5512–5515.
  Abstract:



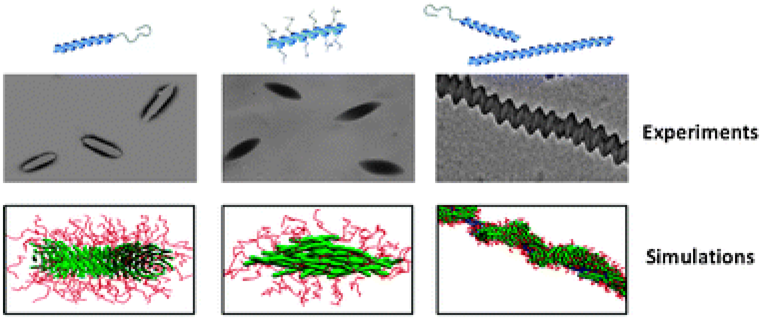
A 1,4-linked triazole/amide based peptidomimetic macrocycle, synthesized from a triazole amide oligomer of *cis*-furanoid sugar triazole amino acids, possesses a conformation resembling the D-,L-α-amino acid based cyclic peptides despite having uniform backbone chirality. It undergoes a unique mode of self-assembly through an antiparallel backbone to backbone intermolecular H-bonding involving amide NH and triazole N2/N3 as well as parallel stacking via amide NH and carbonyl oxygen H-bonding, leading to the formation of a tubular nanostructure.

- Hydrogen-bond driven assembly of a molecular capsule facilitated by supramolecular chelation
  Aakeröy, C. B.; Rajbanshi, A.; Desper, J. *Chem. Commun.* **2011**, *47*, 11411-11413.
  Abstract:



Resorcinarene-based cavitands functionalized with acetamido groups capable of self-complementary hydrogen-bond interactions, were synthesized in order to construct supramolecular capsules. The 1,3-bifunctionalized cavitand produced a polymeric assembly, whereas the tetra-functionalized analogue yielded a discrete capsule held together *via* N–H⋯O hydrogen bonds. The ethynyl species attached to the rim of these host molecules deepen each cavitand and expands the volume of the resulting capsule.

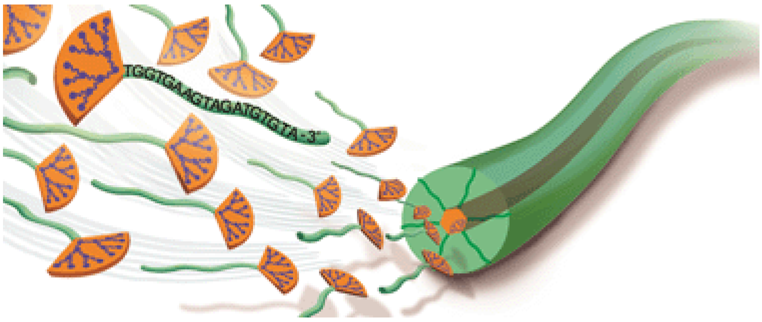
- Self-assembly of polypeptide-based copolymers into diverse aggregates
  Cai, C.; Wang, L.; Lin, J. *Chem. Commun.* **2011**, *47*, 11189-11203.
  Abstract:



Self-Assembly of Polypeptide-Based Copolymers

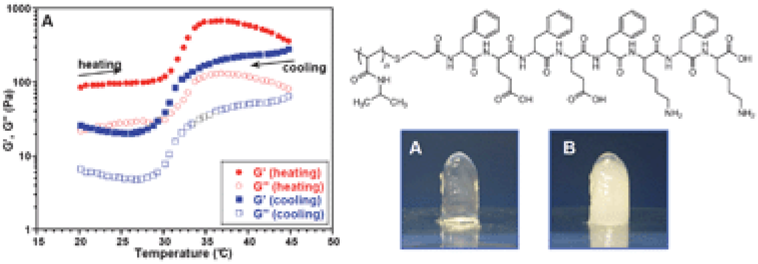
Recently, increasing attention has been given to the self-assembly behavior of polypeptide-based copolymers. Polypeptides can serve as either shell-forming or core-forming blocks in the formation of various aggregates. The solubility and rigidity of polypeptide blocks have been found to have a profound effect on the self-assembly behavior of polypeptide-based copolymers. Polypeptide graft copolymers combine the advantages of a grafting strategy and the characteristics of polypeptide chains and their self-assembly behavior can be easily adjusted by choosing different polymer chains and copolymer architectures. Fabricating hierarchical structures is one of the attractive topics of self-assembly research of polypeptide copolymers. These hierarchical structures are promising for use in preparing functional materials and, thus, attract increasing attention. Computer simulations have emerged as powerful tools to investigate the self-assembly behavior of polymers, such as polypeptides. These simulations not only support the experimental results, but also provide information that cannot be directly obtained from experiments. In this feature article, recent advances in both experimental and simulation studies for the self-assembly behavior of polypeptide-based copolymers are reviewed.

- Amphiphilic DNA-dendron hybrid: a new building block for functional assemblies
  Wang, L.; Feng, Y.; Sun, Y.; Li, Z.; Yang, Z.; He, Y.-M.; Fan, Q.-H.; Liu, D. *Soft Matter* **2011**, *7*, 7187-7190.
  Abstract:



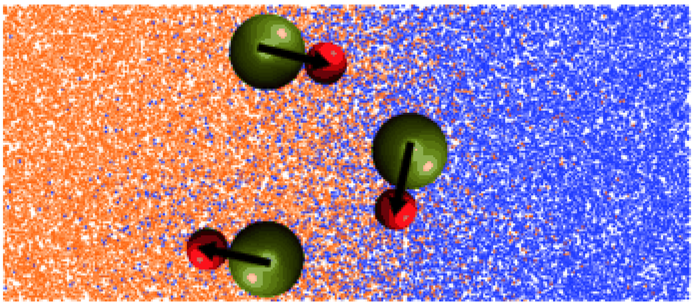
A new kind of amphiphilic DNA-dendron hybrid consisting of a highly hydrophobic dendron and a single stranded DNA is synthesized. The hybrid could assemble into long nanofibers in aqueous phase. The hybridization property of DNA at the shell of fibers associated with the encapsulation ability of dendron at the inner core enable further functionalization, offering a unique class of supramolecular building block.

- Peptide–PNIPAAm conjugate based hydrogels: synthesis and characterisation
  Maslovskis, A.; Tirelli, N.; Saiani, A.; Miller, A. F. *Soft Matter* **2011**, *7*, 6025-6033.
  Abstract:



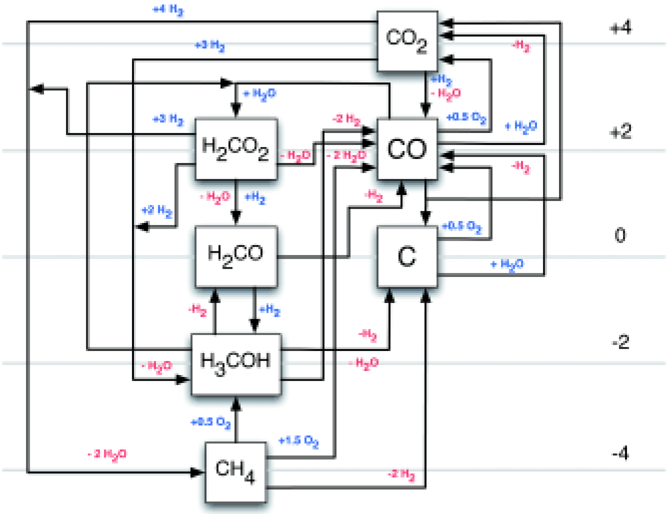
Thermo-responsive PNIPAAm rich hydrogels with controllable properties have been prepared by introducing small quantities of peptide–PNIPAAm bioconjugates where the peptide self-assembled into a β-sheet rich fibrillar network. This was achieved in a one pot synthesis using a thiol-modified peptide as the chain-transfer agent in the free radical polymerisation of NIPAAm. Varying the quantity of modified peptide offered control over the composition of the polymer/conjugate mixture; the higher the peptide concentration, the higher the fraction of conjugate present within the mixture and the greater the degree of aggregation of the PNIPAAm formulations. Once above a critical concentration transparent, doubly thermo-responsive hydrogels formed and the elastic modulus was proportional to the concentration of peptide present. When heated, the hydrogels exhibited an LCST transition due to the PNIPAAm and the temperature was not influenced significantly by the conjugate present, nor the gelled state of the sample. Materials with such controllable thermal and mechanical properties may have great potential to be used in numerous regenerative medical applications.

- Interaction of a Chemically Propelled Nanomotor with a Chemical Wave
  Thakur, S.; Chen, J.; Kapral, R. *Angew. Chem., Int. Ed.* **2011**, *50,* 10165–10169.
  Abstract:



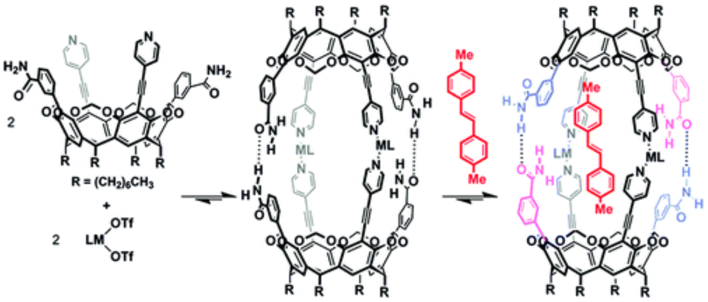
A chemically powered nanodimer motor interacting with a chemical wave results in the deflection of the nanomotor (see picture). Such an effect provides a possible mechanism for the control of nanomotor motion.

- CO Oxidation as a Prototypical Reaction for Heterogeneous Processes
  Freund, H.; Meijer, G.; Scheffler, M.; Schlögl, R.; Wolf, M. *Angew. Chem., Int. Ed.* **2011**, *50,* 10064–10094.
  Abstract:



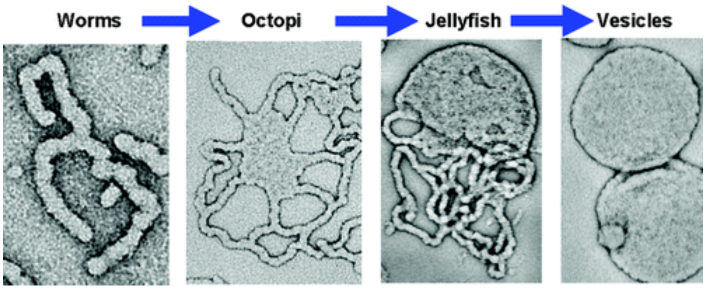
CO oxidation, although seemingly a simple chemical reaction, provides us with a panacea that reveals the richness and beauty of heterogeneous catalysis. The Fritz Haber Institute is a place where a multidisciplinary approach to study the course of such a heterogeneous reaction can be generated in house. Research at the institute is primarily curiosity driven, which is reflected in the five sections comprising this Review. We use an approach based on microscopic concepts to study the interaction of simple molecules with well-defined materials, such as clusters in the gas phase or solid surfaces. This approach often asks for the development of new methods, tools, and materials to prove them, and it is exactly this aspect, both, with respect to experiment and theory, that is a trade mark of our institute.

- Structural Alteration of Hybrid Supramolecular Capsule Induced by Guest Encapsulation
  Yamanaka, M.; Kawaharada, M.; Nito, Y.; Takaya, H.; Kobayashi, K. *J. Am. Chem. Soc.* **2011** *133*, 16650-16656.
  Abstract:



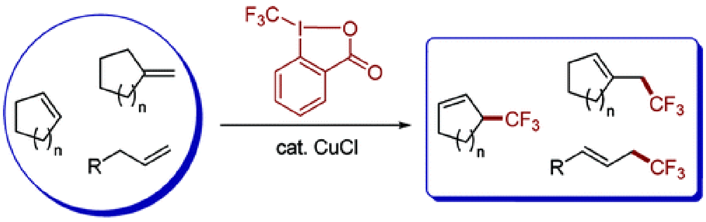
Heterofunctionalized C_{2v} symmetrical cavitand **1** with 4-pyridylethynyl and 3-carbamoylphenyl groups in alternating arrangement was designed and synthesized. A 1:1 mixture of the cavitand **1** and a *cis*-coordinated palladium(II) or platinum(II) complex self-assembled into a hybrid supramolecular capsule via both metal–ligand coordination bonds and hydrogen bonds. Formation of the capsular assembly was confirmed by NMR spectroscopy and mass spectrometry. The hybrid capsule encapsulated the appropriate guest, the molecular sizes of which fit the size of the capsular cavity. Structural alteration of the hybrid capsule was induced by the guest encapsulation. A C_{2h} structure for the encapsulation complex was assigned by 2D NMR spectra analysis. Thermodynamic and kinetic properties of the guest encapsulation were investigated. The kinetics of in/out guest exchange was strongly influenced by hydrogen bonding in the hybrid capsule.

- Mechanistic Insights for Block Copolymer Morphologies: How Do Worms Form Vesicles?
  Blanazs, A.; Madsen, J.; Battaglia, G.; Ryan, A. J.; Armes, S. P. *J. Am. Chem. Soc.* **2011**, *133*, 16581-16587.
  Abstract:



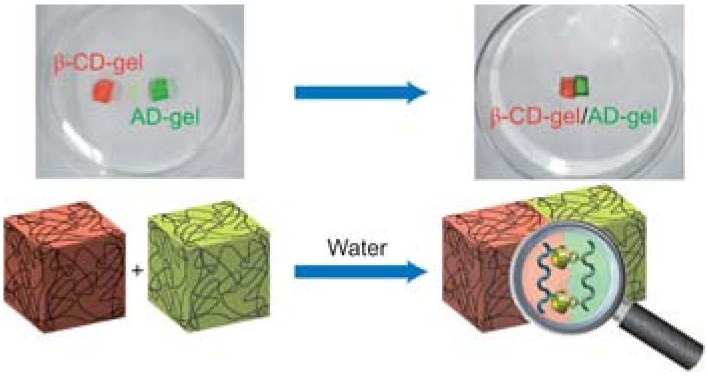
Amphiphilic diblock copolymers composed of two covalently linked, chemically distinct chains can be considered to be biological mimics of cell membrane-forming lipid molecules, but with typically more than an order of magnitude increase in molecular weight. These macromolecular amphiphiles are known to form a wide range of nanostructures (spheres, worms, vesicles, etc.) in solvents that are selective for one of the blocks. However, such self-assembly is usually limited to dilute copolymer solutions (<1%), which is a significant disadvantage for potential commercial applications such as drug delivery and coatings. In principle, this problem can be circumvented by polymerization-induced block copolymer self-assembly. Here we detail the synthesis and subsequent in situ self-assembly of amphiphilic AB diblock copolymers in a one pot concentrated aqueous dispersion polymerization formulation. We show that spherical micelles, wormlike micelles, and vesicles can be predictably and efficiently obtained (within 2 h of polymerization, >99% monomer conversion) at relatively high solids in purely aqueous solution. Furthermore, careful monitoring of the in situ polymerization by transmission electron microscopy reveals various novel intermediate structures (including branched worms, partially coalesced worms, nascent bilayers, "octopi", "jellyfish", and finally pure vesicles) that provide important mechanistic insights regarding the evolution of the particle morphology during the sphere-to-worm and worm-to-vesicle transitions. This environmentally benign approach (which involves no toxic solvents, is conducted at relatively high solids, and requires no additional processing) is readily amenable to industrial scale-up, since it is based on commercially available starting materials.

- Copper-Catalyzed C(sp^3)–C(sp^3) Bond Formation Using a Hypervalent Iodine Reagent: An Efficient Allylic Trifluoromethylation
  Wang, X.; Ye, Y.; Zhang, S.; Feng, J.; Xu, Y.; Zhang, Y.; Wang, J. *J. Am. Chem. Soc.* **2011**, *133*, 16410–16413.
  Abstract:



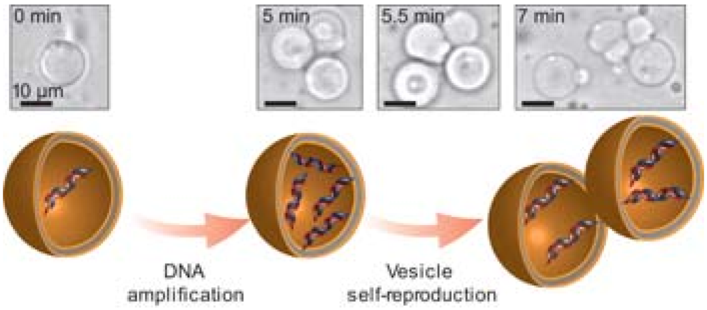
An efficient copper-catalyzed allylic trifluoromethylation reaction has been developed. This reaction provides a general and straightforward way to synthesize allylic trifluoromethylated compounds under mild conditions.

- Macroscopic self-assembly through molecular Recognition
  Harada, A.; Kobayashi, R.; Takashima, Y.; Hashidzume, A.; Yamaguchi, H. *Nature Chem.* **2011**, *3*, 34-37.
  Abstract:



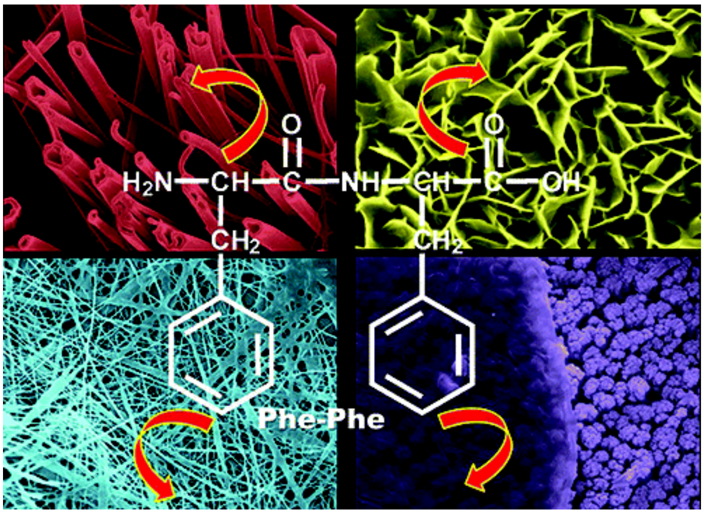
Molecular recognition plays an important role in nature, with perhaps the best known example being the complementarity exhibited by pairs of nucleobases in DNA. Studies of self-assembling and self-organizing systems based on molecular recognition are often performed at the molecular level, however, and any macroscopic implications of these processes are usually far removed from the specific molecular interactions. Here, we demonstrate that well-defined molecular-recognition events can be used to direct the assembly of macroscopic objects into larger aggregated structures. Acrylamide-based gels functionalized with either host (cyclodextrin) rings or small hydrocarbon-group guest moieties were synthesized. Pieces of host and guest gels are shown to adhere to one another through the mutual molecular recognition of the cyclodextrins and hydrocarbon groups on their surfaces. By changing the size and shape of the host and guest units, different gels can be selectively assembled and sorted into distinct macroscopic structures that are on the order of millimetres to centimetres in size.

- Self-reproduction of supramolecular giant vesicles combined with the amplification of encapsulated DNA
  Kurihara, K.; Tamura, M.; Shohda, K.; Toyota, T.; Suzuki, K.; Sugawara, T. *Nature Chem.* **2011**, *3*, 775-781.
  Abstract:



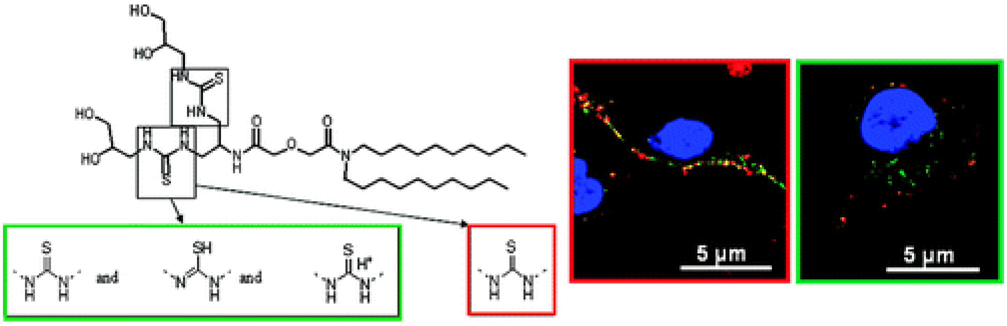
The construction of a protocell from a materials point of view is important in understanding the origin of life. Both self-reproduction of a compartment and self-replication of an informational substance have been studied extensively, but these processes have typically been carried out independently, rather than linked to one another. Here, we demonstrate the amplification of DNA (encapsulated guest) within a self-reproducible cationic giant vesicle (host). With the addition of a vesicular membrane precursor, we observe the growth and spontaneous division of the giant vesicles, accompanied by distribution of the DNA to the daughter giant vesicles. In particular, amplification of the DNA accelerated the division of the giant vesicles. This means that self-replication of an informational substance has been linked to self-reproduction of a compartment through the interplay between polyanionic DNA and the cationic vesicular membrane. Our self-reproducing giant vesicle system therefore represents a step forward in the construction of an advanced model protocell.

- Surface-Induced Self-Assembly of Dipeptides onto Nanotextured Surfaces
  Demirel, G.; Buyukserin, F. *Langmuir* **2011**, *27*, 12533-12538.
  Abstract:



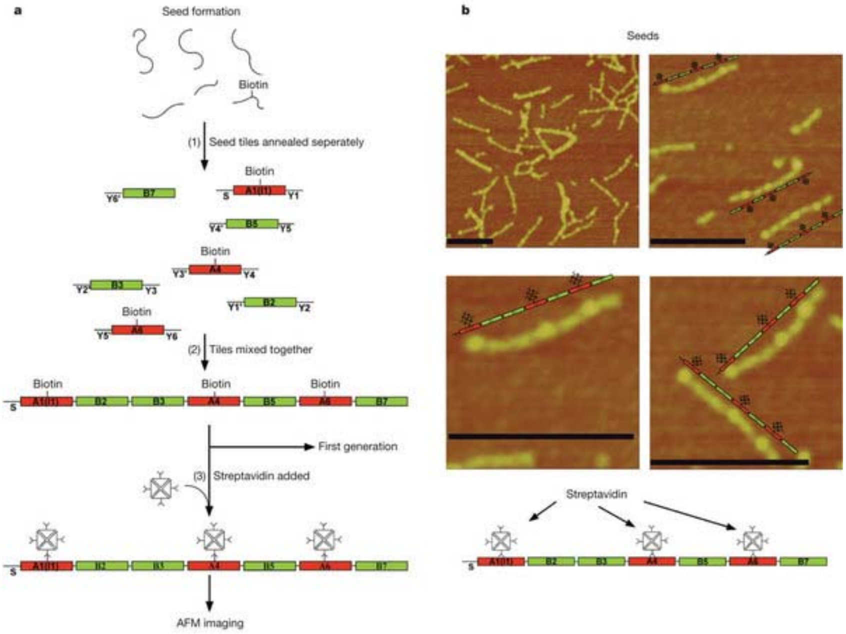
There is an increasing interest for the utilization of biomolecules for fabricating novel nanostructures due to their ability for specific molecular recognition, biocompatibility, and ease of availability. Among these molecules, diphenylalanine (Phe-Phe) dipeptide is considered as one of the simplest molecules that can generate a family of self-assembly based nanostructures. The properties of the substrate surface, on which the self-assembly process of these peptides occurs, play a critical role. Herein, we demonstrated the influence of surface texture and functionality on the self-assembly of Phe-Phe dipeptides using smooth silicon surfaces, anodized aluminum oxide (AAO) membranes, and poly(chloro-*p*-xylylene) (PPX) films having columnar and helical morphologies. We found that helical PPX films, AAO, and silicon surfaces induce similar self-assembly processes and the surface hydrophobicity has a direct influence for the final dipeptide structure whether being in an aggregated tubular form or creating a thin film that covers the substrate surface. Moreover, the dye staining data indicates that the surface charge properties and hence the mechanism of the self-assembly process are different for tubular structures as opposed to the peptidic film. We believe that our results may contribute to the control of surface-induced self-assembly of peptide molecules and this control can potentially allow the fabrication of novel peptide based materials with desired morphologies and unique functionalities for different technological applications.

- Protonation of Lipids Impacts the Supramolecular and Biological Properties of Their Self-Assembly
  Breton, M.; Berret, J.-F.; Bourgaux, C.; Kral, T.; Hof, M.; Pichon, C.; Bessodes, M.; Scherman, D.; Mignet, N. *Langmuir* **2011**, *27*, 12336-12345.
  Abstract:



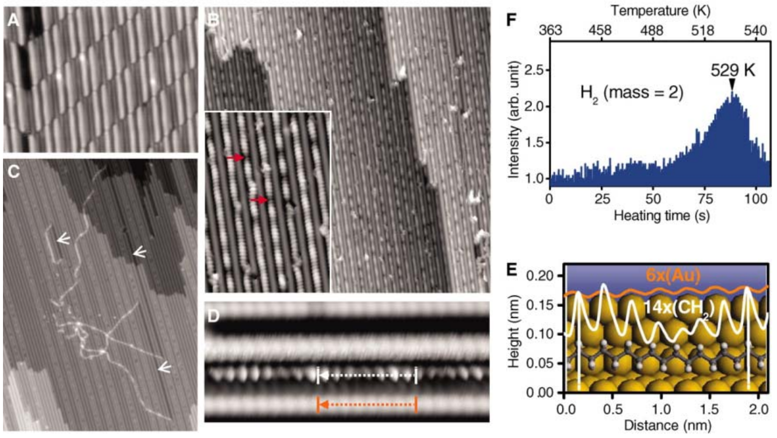
We assessed in this work how a chemical structure difference could influence a supramolecular organization and then its biological properties. In our case study, we considered two amphiphilic lipidic gene vectors. The chemical difference was situated on their hydrophilic part which was either a pure neutral thiourea head or a mixture of three thiourea function derivatives, thiourea, iminothiol, and charged iminothiol. This small difference was obtained thanks to the last chemical deprotection conditions of the polar head hydroxyl groups. Light, neutron, and X-ray scattering techniques have been used to investigate the spatial structure of the liposomes and lipoplexes formed by the lipids. The chemical structure difference impacts the supramolecular assemblies of the lipids and with DNA as shown by fluorescence correlation spectroscopy (FCS), X-ray, and neutron scattering. Hence the structures formed were found to be highly different in terms of liposomes to DNA ratio and size and polydispersity of the aggregates. Finally, the transfection and internalization results proved that the differences in the structure of the lipid aggregates fully affect the biological properties of the lipopolythiourea compounds. The lipid containing three functions is a better gene transfection agent than the lipid which only contains one thiourea moiety. As a conclusion, we showed that the conditions of the last chemical step can influence the lipidic supramolecular structure which in turn strongly impacts their biological properties.

- Self-replication of information-bearing nanoscale patterns
  Wang, T.; Sha, R.; Dreyfus, R.; Leunissen, M. E.; Maass, C.; Pine, D. J.; Chaikin, P. M.; Seeman, N. C. *Nature* **2011**, *478*, 225–228.
  <u>Abstract:</u>



DNA molecules provide what is probably the most iconic example of self-replication—the ability of a system to replicate, or make copies of, itself. In living cells the process is mediated by enzymes and occurs autonomously, with the number of replicas increasing exponentially over time without the need for external manipulation. Self-replication has also been implemented with synthetic systems, including RNA enzymes designed to undergo self-sustained exponential amplification. An exciting next step would be to use self-replication in materials fabrication, which requires robust and general systems capable of copying and amplifying functional materials or structures. Here we report a first development in this direction, using DNA tile motifs that can recognize and bind complementary tiles in a pre-programmed fashion. We first design tile motifs so they form a seven-tile seed sequence; then use the seeds to instruct the formation of a first generation of complementary seven-tile daughter sequences; and finally use the daughters to instruct the formation of seven-tile granddaughter sequences that are identical to the initial seed sequences. Considering that DNA is a functional material that can organize itself and other molecules into useful structures, our findings raise the tantalizing prospect that we may one day be able to realize self-replicating materials with various patterns or useful functions.

- Linear Alkane Polymerization on a Gold Surface
  Zhong, D.; Franke, J.-H.; Podiyanachari, S. K.; Blömker, T.; Zhang, H.; Kehr, G.; Erker, G.; Fuchs, H.; Chi, L. *Science* **2011**, *334*, 213-216.
  Abstract:



In contrast to the many methods of selectively coupling olefins, few protocols catenate saturated hydrocarbons in a predictable manner. We report here the highly selective carbon-hydrogen (C–H) activation and subsequent dehydrogenative C–C coupling reaction of long-chain (>C_{20}) linear alkanes on an anisotropic gold(110) surface, which undergoes an appropriate reconstruction by adsorption of the molecules and subsequent mild annealing, resulting in nanometer-sized channels (1.22 nanometers in width). Owing to the orientational constraint of the reactant molecules in these one-dimensional channels, the reaction takes place exclusively at specific sites (terminal CH_3 or penultimate CH_2 groups) in the chains at intermediate temperatures (420 to 470 kelvin) and selects for aliphatic over aromatic C–H activation.