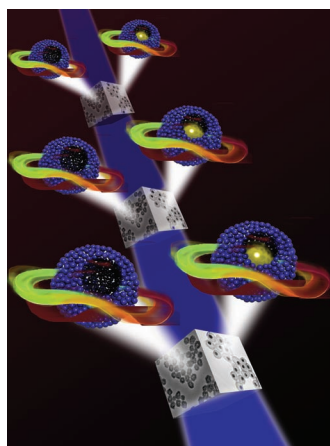


# ADVANCED FUNCTIONAL MATERIALS

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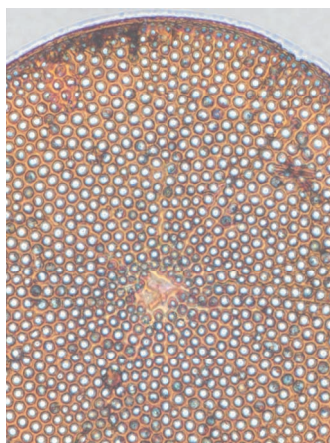


## Hybrid Materials

A general strategy to fabricate monodisperse hollow supraparticles is described by Zhiyong Tang and Yunsheng Xia on page 2585. Core/shell supraparticles are obtained using an in situ assembly method and subsequent selective oxidation of cores results in formation of monodisperse hollow supraparticles. This work highlights that the strategy of combining self-assembly with chemical reaction is promising to tailor both the structures and properties of hollow nanomaterials.

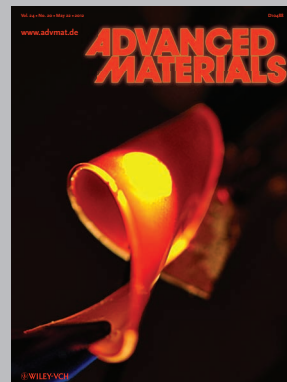
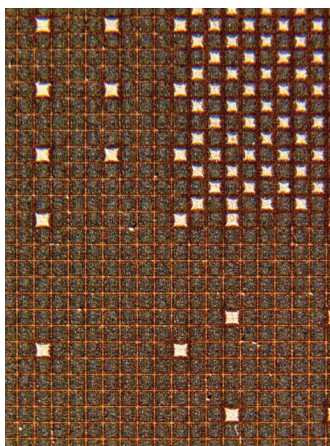
## Biotechnology

On page 2503 Alexander Nesterov-Mueller, Frank Breitling, and co-workers present a microarray system based on a microelectronic chip that allows for a versatile combinatorial molecule synthesis with very high density and, due to the intrinsic alignment, high and reproducible precision. Patterning the chip surface with different microparticle types that embed different monomers allows simultaneous elongation of several thousand different molecule types layer-by-layer.



## Gold Nanostructures

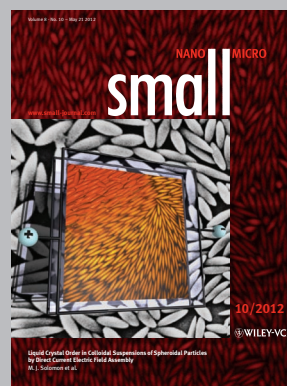
The optical image shows a freestanding (silica-free) three-dimensional nanocrystalline gold replica of the valve of a *Coscinodiscus asteromphalus* (CA) diatom. On page 2550, Joseph W. Perry, Kenneth H. Sandhage, and co-workers report the generation of such gold replicas via the use of a highly conformal and scalable wet chemical coating process. The quasi-periodic hexagonal hole pattern inherited from the native CA diatom valve enables the gold replica to exhibit plasmon-mediated extraordinary infrared transmission.



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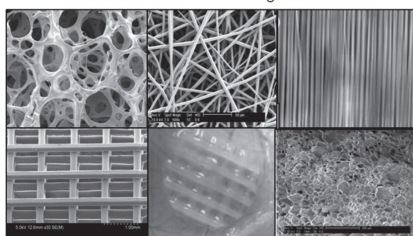
## FEATURE ARTICLE

## Biomimetics

T. G. Kim, H. Shin,\*  
D. W. Lim\* .....2446–2468

### Biomimetic Scaffolds for Tissue Engineering

Extracellular matrix mimicking architectures



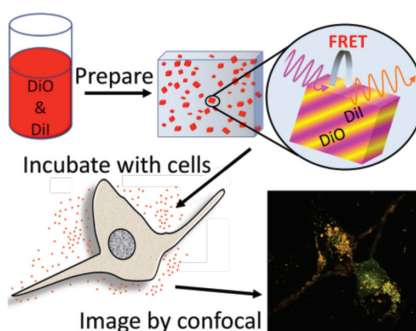
**Extracellular matrix (ECM)-mimicking architecture** should be an underlying concept in the rational design of the biomimetic scaffolds for tissue engineering. Structural and functional engineering of 3D ECM-like scaffolds is demonstrated and the creation of biologically inspired materials that serve as injectable biomaterials is discussed.

## FULL PAPERS

## Organic Nanoparticles

T. O. McDonald, P. Martin,  
J. P. Patterson, D. Smith, M. Giardiello,  
M. Marcello, V. See, R. K. O'Reilly,  
A. Owen,\* S. Rannard\* .....2469–2478

### Multicomponent Organic Nanoparticles for Fluorescence Studies in Biological Systems

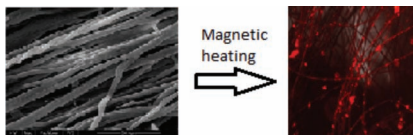


**Dual-component organic nanoparticles** are prepared by a modified emulsion-templated freeze-drying approach to give an aqueous nanosuspension showing fluorescence (Förster) resonance energy transfer (FRET) from within a distribution of single nanoparticles; dissolution of these particles results in loss of this FRET signal. Incubation of the particles with cells and analysis by confocal microscopy allows spatial and physical monitoring of the particles after internalization.

## Electrospun Fibers

C. Huang, S. J. Soenen, J. Rejman,  
J. Trekker, L. Chengxun, L. Lagae,  
W. Ceelen, C. Wilhelm, J. Demeester,  
S. C. De Smedt\* .....2479–2486

### Magnetic Electrospun Fibers for Cancer Therapy

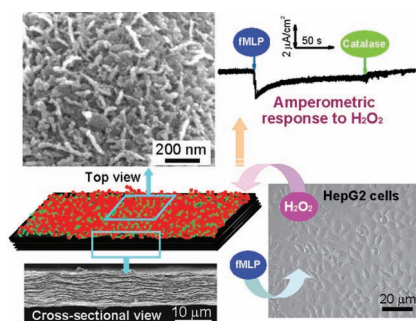


**Magnetic iron oxide particles-containing polystyrene fibers are electrospun** and functionalized to allow binding of cancer cells. The fibers are found to have a high and reproducible heating capacity and result in a significant enhancement of cancer cell killing compared to water bath treatment.

## Biosensors

F. Xiao, Y. Li, X. Zan, K. Liao, R. Xu,  
H. Duan\* .....2487–2494

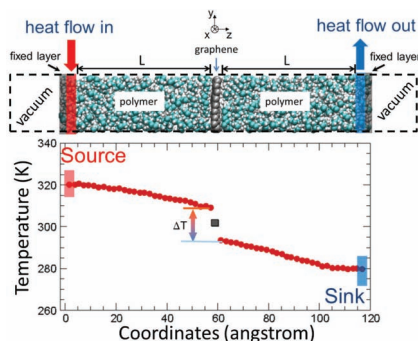
### Growth of Metal–Metal Oxide Nanostructures on Freestanding Graphene Paper for Flexible Biosensors



**A new type of flexible electrochemical sensor with structurally integrated free-standing graphene paper, MnO<sub>2</sub> nanowire networks, and small Pt nanoparticles deposited via ultrasonic electrodeposition** is reported. The sensitivity and selectivity that the flexible electrode demonstrate for the non-enzymatic detection of H<sub>2</sub>O<sub>2</sub> enables its use for monitoring H<sub>2</sub>O<sub>2</sub> secretion by live cells.

## FULL PAPERS

**Understanding thermal transport in polymeric nanocomposite materials** is important to the engineering of better thermally conductive polymer composites. It is demonstrated that thermal energy transport across graphene/graphite-polymer interfaces can be enhanced by increasing the polymer density and graphene size or forming covalent bonds between the graphite edges and polymer molecules.



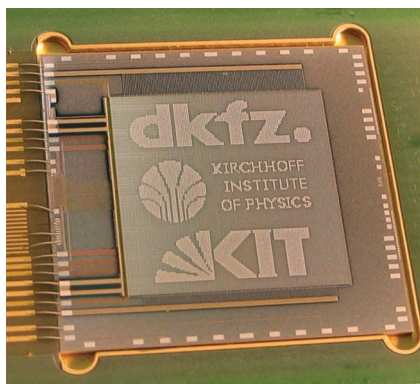
## Nanocomposites

T. Luo,\* J. R. Lloyd\* .....2495–2502

**Enhancement of Thermal Energy Transport Across Graphene/Graphite and Polymer Interfaces: A Molecular Dynamics Study**



**A microarray system based on a micro-electronic chip** that allows for versatile combinatorial molecule synthesis with very high density, and due to the intrinsic alignment, high and reproducible precision, is presented. Patterning the chip surface with different micro-particle types, which imbed different monomers, several thousand different molecule types can be simultaneously elongated layer-by-layer.

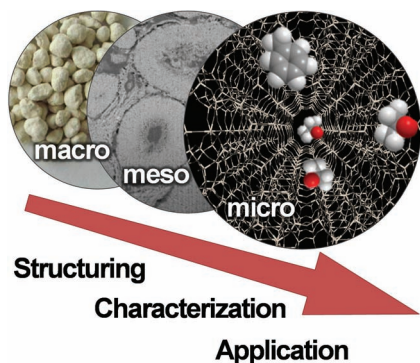


## Biotechnology

F. Loeffler, C. Schirwitz, J. Wagner, K. Koenig, F. Maerkle, G. Torralba, M. Hausmann, F. R. Bischoff, A. Nesterov-Mueller,\*  
F. Breitling\* ..... 2503–2508

**Biomolecule Arrays Using Functional Combinatorial Particle Patterning on Microchips**

**Mesoporous ZSM-5 zeolite powders are structured by granulation with an attapulgite binder.** The influence of shaping and binder addition on the zeolite properties is characterized by multiple techniques, demonstrating the truly hierarchical structure of interconnected micro-, meso-, and macropores. In technical form, mesoporous zeolites preserve their superior functionalities and possess enhanced adsorption capacities with respect to their conventional counterparts.

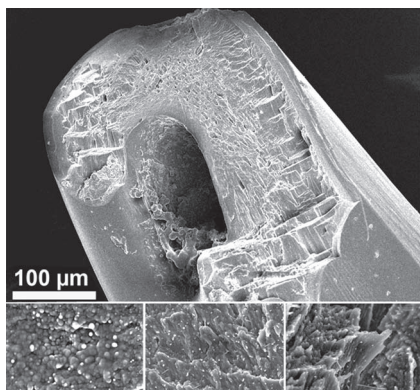


## Zeolites

N.-L. Michels, S. Mitchell, M. Milina, K. Kunze, F. Krumeich, F. Marone, M. Erdmann, N. Marti, J. Pérez-Ramírez\* .....2509–2518

**Hierarchically Structured Zeolite Bodies: Assembling Micro-, Meso-, and Macroporosity Levels in Complex Materials with Enhanced Properties**

**Spiders use their fangs to inject venom into their prey.** Structural and chemical modifications of chitin within the fangs of the spider *Cupiennius salei* influence the fang mechanical properties, allowing them to function as a multi-use injection needle. The needle is able to puncture the prey cuticle, which is made essentially of the same material, a chitin-protein composite.



## Bioinspired Materials

Y. Politi,\* M. Priewasser, E. Pippel, P. Zaslansky, J. Hartmann, S. Siegel, C. Li, F. G. Barth, P. Fratzl .....2519–2528

**A Spider's Fang: How to Design an Injection Needle Using Chitin-Based Composite Material**

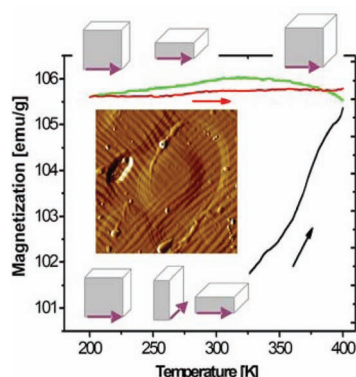


## FULL PAPERS

## Magnetic Materials

Y. Ma, A. Setzer, J. W. Gerlach,  
F. Frost, P. Esquinazi,  
S. G. Mayr\* .....2529–2534

### Freestanding Single Crystalline Fe–Pd Ferromagnetic Shape Memory Membranes – Role of Mechanical and Magnetic Constraints Across the Martensite Transition

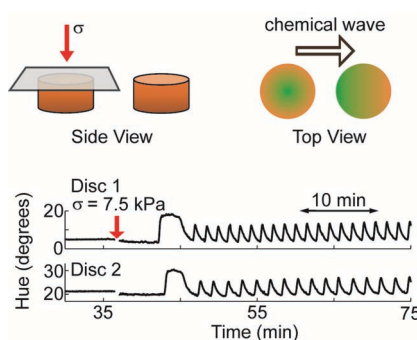


Substrate-attached and freestanding single crystalline Fe<sub>70</sub>Pd<sub>30</sub> ferromagnetic shape memory alloy membranes show fundamentally different magnetic fingerprints, which can be understood in terms of magnetic domain structure and substrate constraints, which pose a bias on the austenite → fct-martensite phase transition. This suggests thermomagnetic actuation as an alternative where only moderate magnetic fields are feasible, but moderate temperature changes are possible.

## Polymer Gels

I. C. Chen, O. Kuksenok,  
V. V. Yashin, A. C. Balazs,  
K. J. Van Vliet\* .....2535–2541

### Mechanical Resuscitation of Chemical Oscillations in Belousov–Zhabotinsky Gels



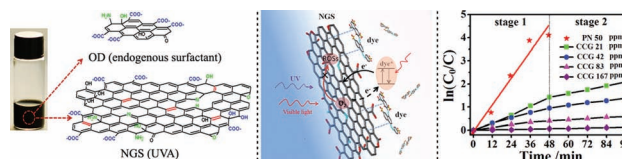
Polymer gels undergoing the Belousov–Zhabotinsky (BZ) reaction exhibit colorful self-oscillations when chemical reagent concentrations are sufficient. It is demonstrated that when the reagents are depleted and the system reaches a steady-state, the oscillations can be resuscitated by applying macroscopic compression, thereby extending the useful lifetime of these materials. Pressure sensor applications are shown in which chemical signals are transmitted away from the deformation site.

## Graphene Oxide

W. He, L. Lu\* .....2542–2549

### Revisiting the Structure of Graphene Oxide for Preparing New-Style Graphene-Based Ultraviolet Absorbers

The structure of graphene oxide (GO) is revisited by elucidating its reduction process solely in the presence of ammonia. This contributes to settling some arguments in recent models of GO and is inspirational in investigating the performance of nitrogen-doped graphene sheets, for the first time, as a new-style, multifunctional UV absorber for new-generation inkjet inks.



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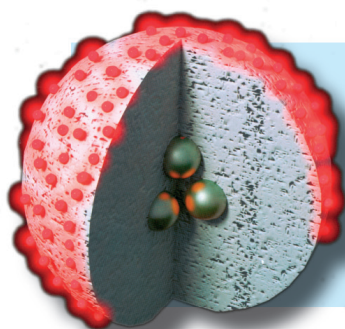
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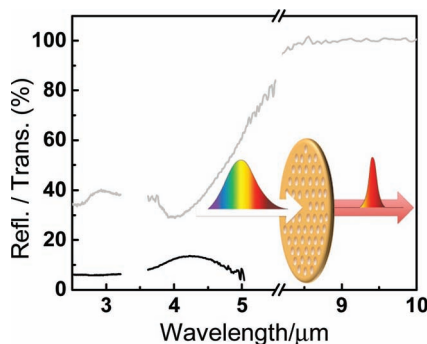
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## FULL PAPERS

The valves of *Coscinodiscus asteromphalus* (CA) diatoms, which possess local hexagonal (quasi-periodic) hole patterns, are converted by a scalable wet chemical process into three-dimensional gold replicas that exhibit extraordinary infrared transmission. Simulated and measured transmission spectra indicate that the infrared transmission of these gold frustule replicas is enabled by the generation and transmission of surface plasmons.

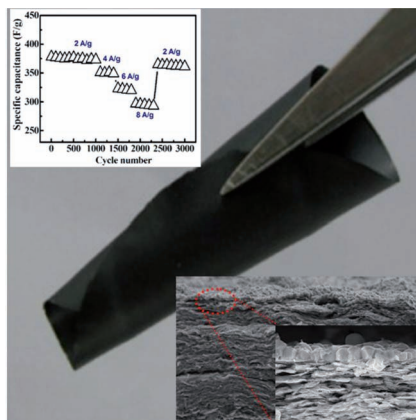


## Hierarchical Structures

Y. Fang, V. W. Chen, Y. Cai,  
J. D. Berrigan, S. R. Marder, J. W. Perry,\*  
K. H. Sandhage\* .....2550–2559

**Biologically Enabled Syntheses of Freestanding Metallic Structures Possessing Subwavelength Pore Arrays for Extraordinary (Surface Plasmon-Mediated) Infrared Transmission**

Flexible and free-standing  $\text{Co}_3\text{O}_4$ /reduced graphene oxide/carbon nanotube (CNT) hybrid paper delivers high specific capacitance with excellent electrochemical stability even at high current densities.

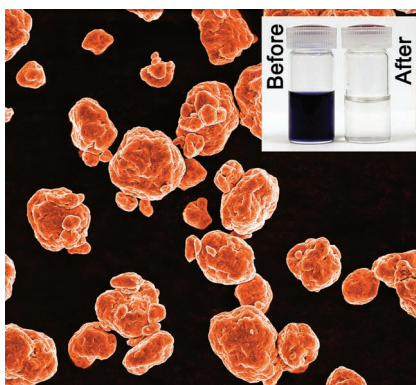


## Hybrid Materials

C. Yuan,\* L. Yang, L. Hou, J. Li, Y. Sun,  
X. Zhang,\* L. Shen, X. Lu, S. Xiong,\*  
X. W. (D.) Lou\* .....2560–2566

**Flexible Hybrid Paper Made of Monolayer  $\text{Co}_3\text{O}_4$  Microsphere Arrays on rGO/CNTs and Their Application in Electrochemical Capacitors**

Fe-based metallic glass powders show outstanding efficiency in degrading organic contaminants due to their far-from-equilibrium thermodynamic nature. The  $-\text{N}=\text{N}-$  bond of the dye can be completely decomposed in a short time. Higher degradation efficiency can be achieved by tailoring the particle morphology. The findings are expected to open new areas for the applications of metallic glasses.

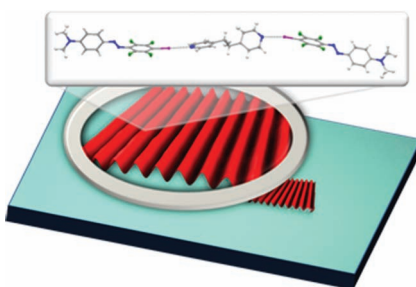


## Waste-Management Materials

J. Q. Wang,\* Y. H. Liu,\* M. W. Chen,  
G. Q. Xie, D. V. Louzguine-Luzgin,  
A. Inoue, J. H. Perepezko .....2567–2570

**Rapid Degradation of Azo Dye by Fe-Based Metallic Glass Powder**

Because of its high directionality and tuneable interaction strength, halogen bonding is a viable tool for designing high-performance supramolecular polymers for holographic inscription of surface relief gratings (SRG). Light-induced surface patterning, a unique phenomenon that occurs in azobenzene-containing polymers, can be more efficient in halogen-bonded polymer–azobenzene complexes than in the analogous hydrogen-bonded complexes.



## Optically Active Materials

A. Priimagi,\* G. Cavallo,  
A. Forni, M. Gorynsztejn–Leben,  
M. Kaivola, P. Metrangolo,\* R. Milani,  
A. Shishido, T. Pilati, G. Resnati,\*  
G. Terraneo .....2572–2579

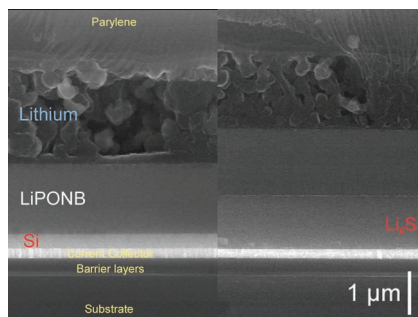
**Halogen Bonding versus Hydrogen Bonding in Driving Self-Assembly and Performance of Light-Responsive Supramolecular Polymers**

## FULL PAPERS

## Batteries

V. P. Phan, B. Pecquenard,\*  
F. Le Cras\* .....2580–2584

### High-Performance All-Solid-State Cells Fabricated With Silicon Electrodes



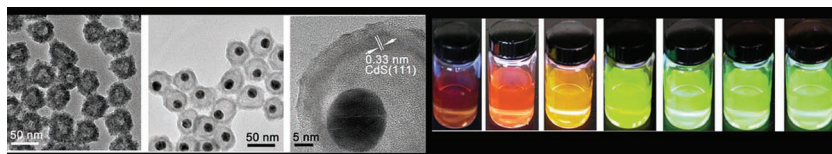
**All-solid-state microbatteries including a silicon thin-film electrode** are prepared by sputtering and evaporation on silicon wafer substrates using a shadow-masking technique. The use of a vitreous solid electrolyte leads to a fully reversible insertion of lithium in the silicon electrode. Scanning electron microscopy cross-sections confirm that the huge volume change of the  $\text{Li}_x\text{Si}$  electrode during cycling does not deteriorate the electrochemical cell in this stacked thin-films architecture.

## Hybrid Materials

Y. Xia, Z. Tang\* .....2585–2593

### Monodisperse Hollow Supraparticles via Selective Oxidation

**Simple hollow supraparticles (SPs) or rattle-type SPs with narrow size distributions** are fabricated by a combination of in situ assembly and selective oxidization. The unique structures and formation processes create products with special optical properties.

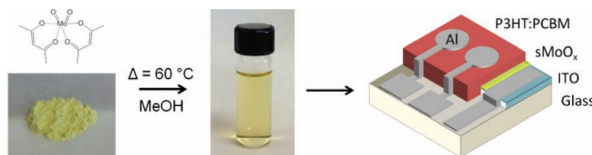


## Solar Cells

J. J. Jasieniak, J. Seifter, J. Jo, T. Mates,  
A. J. Heeger\* .....2594–2605

### A Solution-Processed $\text{MoO}_x$ Anode Interlayer for Use within Organic Photovoltaic Devices

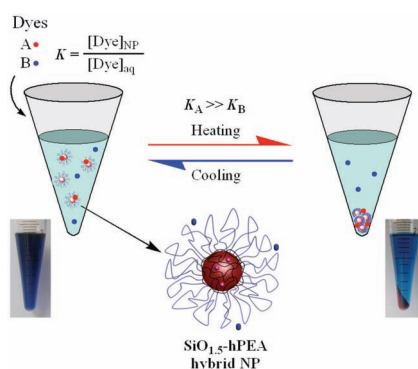
**A solution-processed method to deposit  $\text{MoO}_x$  layers using oxomolybdate precursors is developed.** The chemical, morphological, and electrical properties of these films are found to be suitable for use as anode modifying layers in bulk-heterojunction solar cell devices. To achieve high performance in such devices, the density of  $\text{Mo}^{5+}$  sub-gap states within the  $\text{MoO}_x$  has to be controlled.



## Hybrid Nanoparticles

R. Wang, B. Yu, X. S. Jiang,\*  
J. Yin .....2606–2616

### Understanding the Host–Guest Interaction Between Responsive Core-Crosslinked Hybrid Nanoparticles of Hyperbranched Poly(ether amine) and Dyes: The Selective Adsorption and Smart Separation of Dyes in Water

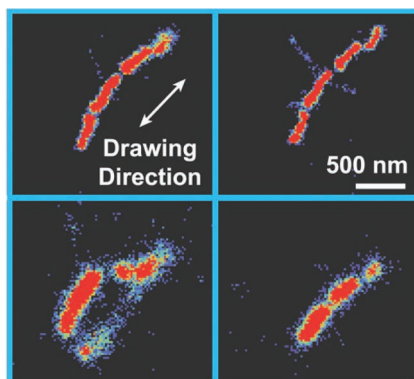


**Responsive hybrid nanoparticles (NPs) of hyperbranched poly(ether amine) (hPEA) ( $\text{SiO}_{1.5}$ -hPEA NPs)** exhibit selective adsorption to hydrophilic dyes and the host–guest interaction between  $\text{SiO}_{1.5}$ -hPEA NPs and dyes is investigated in detail. Based on the established distribution coefficient for the partitioning of dyes between  $\text{SiO}_{1.5}$ -hPEA NPs and water, a methodology is demonstrated for the smart separation of a mixture of dyes in water.



## FULL PAPERS

**Surface occupancy maps of fibrinogen on nanostructured, melt-drawn high-density poly(ethylene) (HDPE) films** are shown. At high surface coverage of fibrinogen, an ordered protein layer forms that restricts diffusion in the direction perpendicular to the drawing direction and increases protein surface residence time. Several examples of the elongated protein occupancy regions that are observed as a result of this restriction are shown.

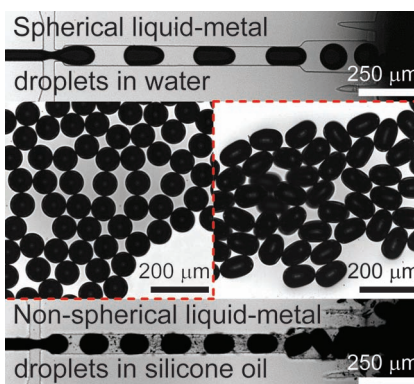


### Surface Nanostructures

M. Kastantin, T. F. Keller, K. D. Jandt, D. K. Schwartz\* .....2617–2623

### Single-Molecule Tracking of Fibrinogen Dynamics on Nanostructured Poly(ethylene) Films

**Liquid metals are interesting materials for optical, electronic, and micro-mechanical applications.** Microfluidic techniques are employed to generate eutectic gallium-indium liquid-metal microdroplets with well-controlled morphologies. Depending on the oxygen content of the carrier fluid, both monodisperse spherical droplets and droplets retaining non-spherical geometries can be produced due to the instantaneous formation of an oxide layer.

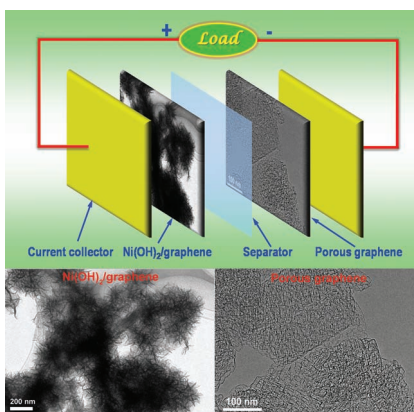


### Microfluidics

T. Hutter, W.-A. C. Bauer, S. R. Elliott,\* W. T. S. Huck\* .....2624–2631

### Formation of Spherical and Non-Spherical Eutectic Gallium-Indium Liquid-Metal Microdroplets in Microfluidic Channels at Room Temperature

**Hierarchical flowerlike Ni(OH)<sub>2</sub> particles on graphene sheets** are prepared by a facile and cost-effective microwave-assisted method. The as-fabricated Ni(OH)<sub>2</sub>/graphene//porous graphene asymmetric supercapacitor exhibits high specific capacitance, enhanced rate capability, excellent electrochemical stability, and high energy density and power density because of the positive synergistic effects of the two electrodes.

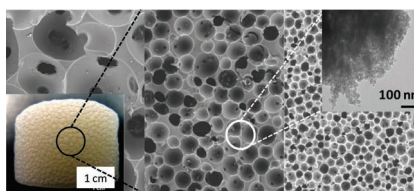


### Supercapacitors

J. Yan, Z. J. Fan,\* W. Sun, G. Q. Ning, T. Wei, Q. Zhang, R. F. Zhang, L. J. Zhi,\* F. Wei\* .....2632–2641

### Advanced Asymmetric Supercapacitors Based on Ni(OH)<sub>2</sub>/Graphene and Porous Graphene Electrodes with High Energy Density

**Limited coalescence-based Pickering high internal phase emulsion** is employed to foster rational design of silica macrocellular foams. Concentrated emulsions with calibrated drop size are produced, leading to the synthesis of monolithic foams with nearly monodisperse macroscopic voids. Such a strategy allows the macrocellular void diameters and the diameter of the windows connecting adjacent cells to be independently tuned.



### Porous Materials

M. Destribats, B. Faure, M. Birot, O. Babot, V. Schmitt,\* R. Backov\* .....2642–2654

### Tailored Silica Macrocellular Foams: Combining Limited Coalescence-Based Pickering Emulsion and Sol–Gel Process