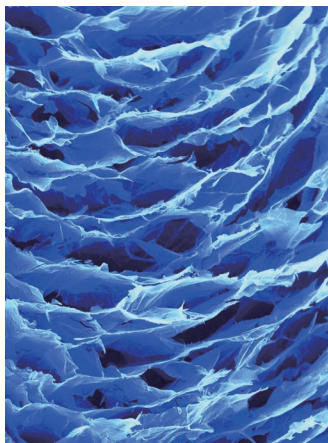


# ADVANCED FUNCTIONAL MATERIALS

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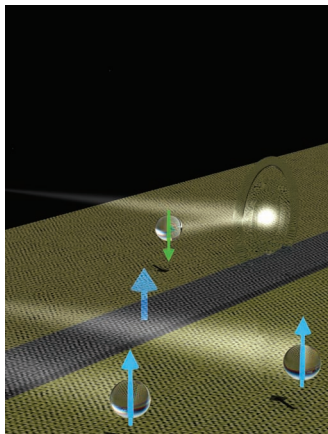
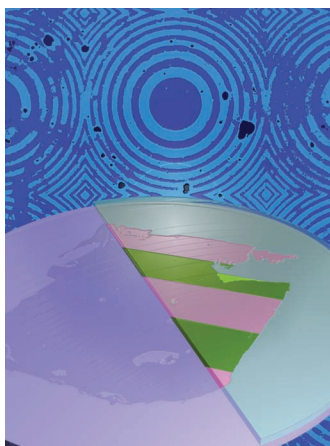


## Graphene

Spongy graphene (SG), a nanoporous material with a high surface area, is used as a versatile and recyclable sorber. On page 4421, Litao Sun, Rodney S. Ruoff, and co-workers report that SG shows highly efficient absorption of petroleum products, fats, and toxic solvents and can be regenerated by heat treatment, yielding almost full release of the adsorbates. SG shows a widespread potential for applications in industry and environmental protection.

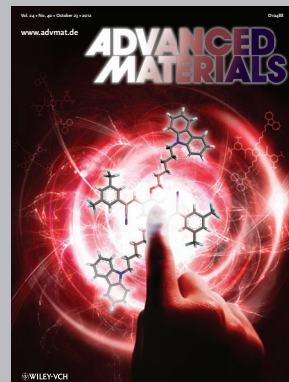
## Surface Characterization

On page 4519, Mihrimah Ozkan, Cengiz S. Ozkan, and co-workers report a new fluorescence quenching microscopy metrology technique that allows the identification of graphene layers and doped/undoped regions across a large graphene landscape by utilizing the fact that undoped regions of graphene quench fluorescence more than the doped regions through resonant energy transfer. Contrast differences in fluorescence across the graphene sheet reveal the complex ring-patterned doping. This metrology technique is well-suited for industrial, large-scale, pristine, and modified graphene sheet surface characterization.



## Spintronics

A spin-polarized-current generator is a fundamental device in spintronics. As reported on page 4471, Isao Ohkubo and co-workers have developed a high-performance spin-filter tunnel junction by growing an atomic-layer-controlled oxide multilayer. In this device, up-spin electrons can selectively tunnel through a nanometer-thick ferromagnetic insulator, while down-spin electrons are reflected. The image shows a false-colored cross-sectional transmission electron microscope image of the device.



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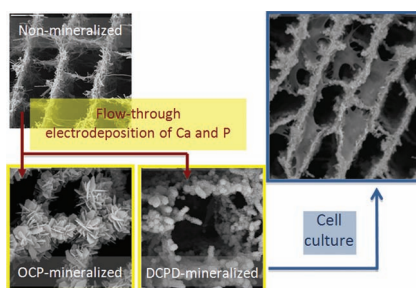
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## FULL PAPERS

## Scaffolds

S. Nardecchia,  
M. C. Serrano,\* M. C. Gutiérrez,  
M. T. Portolés, M. L. Ferrer,\*  
F. del Monte .....4411–4420

### Osteoconductive Performance of Carbon Nanotube Scaffolds Homogeneously Mineralized by Flow-Through Electrodeposition

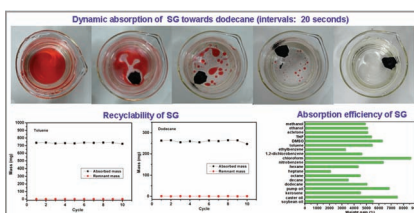


Coating the entire internal surface of “true” 3D scaffolds with a homogeneous mineral layer is by no means a trivial issue. This problem is overcome by the application of a “flow-through” electrodeposition process to scaffolds composed of multiwall carbon nanotubes and chitosan. The scaffolds resulting after mineralization exhibit a remarkable biocompatibility and superior osteoconductive performance.

## Graphene

H. C. Bi, X. Xie, K. B. Yin, Y. Zhou,  
S. Wan, L. B. He, F. Xu, F. Banhart,  
L. T. Sun,\* R. S. Ruoff\* .....4421–4425

### Spongy Graphene as a Highly Efficient and Recyclable Sorbent for Oils and Organic Solvents



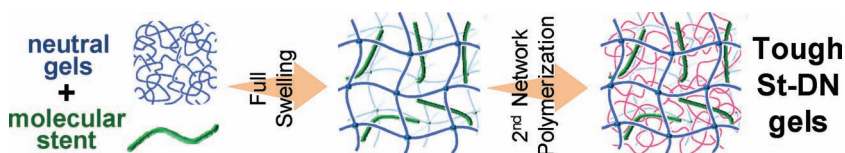
Spongy graphene (SG) shows highly efficient absorption of not only petroleum products and fats but also common toxic solvents such as toluene and chloroform, taking in up to 86 times its own weight. This is several to tens of times higher than that of conventional absorbers. Moreover, the SG can be regenerated more than ten times by heat treatment yielding the full release of adsorbates.

## Hydrogels

T. Nakajima, H. Sato, Y. Zhao,  
S. Kawahara, T. Kurokawa,  
K. Sugahara, J. P. Gong\* .....4426–4432

### A Universal Molecular Stent Method to Toughen any Hydrogels Based on Double Network Concept

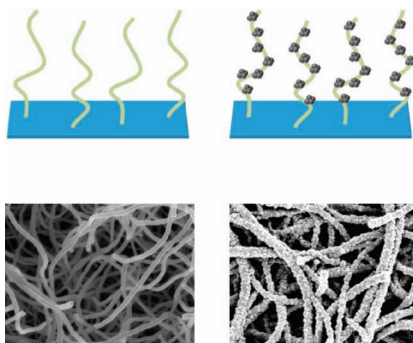
Tough hydrogels can be synthesized from any hydrophilic polymer by the double-network (DN) concept using a molecular stent. The synthesized St-DN gels have excellent strength and toughness comparable to those of conventional tough DN gels and industrial rubbers. This is a promising method for strengthening various functional hydrogels and increasing the scope of their application in various fields.



## Catalysis

G. R. Meseck, R. Kontic,  
G. R. Patzke,\* S. Seeger\* .....4433–4438

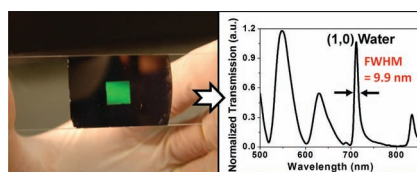
### Photocatalytic Composites of Silicone Nanofilaments and TiO<sub>2</sub> Nanoparticles



A new strategy for the immobilization of nanoscale TiO<sub>2</sub> photocatalysts is presented via their growth on carpets of silicone nanofilaments. These nanofilaments are novel and flexible carrier prototypes for the safe and recyclable application of TiO<sub>2</sub> nanocatalysts. Their compatibility with various surfaces and their functionalization options open up new avenues for photocatalyst design.

## FULL PAPERS

**An extraordinary optical transmission peak in water** with a linewidth of 9.9 nm and a Q factor of 71 is demonstrated via template-stripped metallic nanohole arrays with a surface roughness below 1 nm and a precisely tuned hole size. The nanostructured films exhibit extremely uniform optical properties across a centimeter-sized area and enable a series of practical applications in biosensing and spectroscopy.

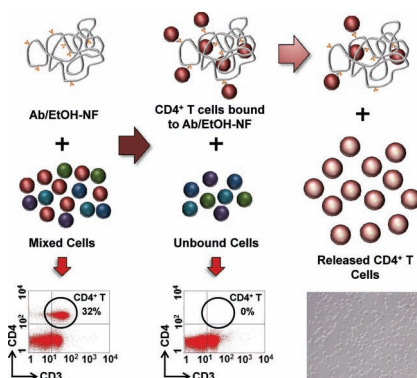


### Plasmonics

S. H. Lee, T. W. Johnson, N. C. Lindquist, H. Im, D. J. Norris, S.-H. Oh\* .....4439–4446

### Linewidth-Optimized Extraordinary Optical Transmission in Water with Template-Stripped Metallic Nanohole Arrays

**Electrospun and alcohol-dispersed PS-PSMA nanofibers**, allowing for facile conjugation of antibodies, could be used as an innovative cell isolation/enrichment and support/release platform. This work opens up a new potential for an innovative immune cell therapy, in which specific immune cells are isolated by antibody-conjugated nanofibers and directly delivered to the target disease sites under the controlled and sustained release of immune cells via in vivo activation.

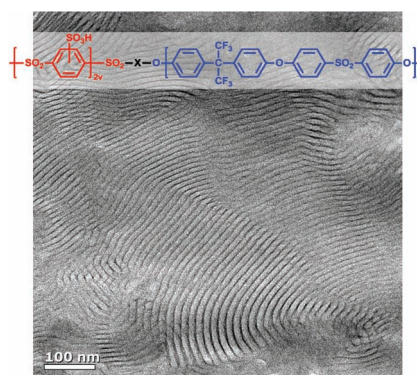


### Cell Delivery

S.-H. Jun, K. Kim, H. J. An, B. C. Kim, C. H. Sonn, M. Kim, J. Doh, C. Yee, K.-M. Lee,\* J. Kim\* .....4448–4455

### Ethanol-Dispersed Polymer Nanofibers as a Highly Selective Cell Isolation and Release Platform for CD4<sup>+</sup> T Lymphocytes

**Phase-separated multiblock copolymers** are quite ordinary, but this is not so if they are based on fully sulfonated polysulfones as the hydrophilic segments. The aromatic sulfone structure gives rise to high chemical and thermal stability, while the hydrophilic domain structure allows for high proton conductivity with low hydrodynamic water transport. Such membranes have the potential to substitute traditional perfluorosulfonic acid (PFSA) membranes in fuel cells.

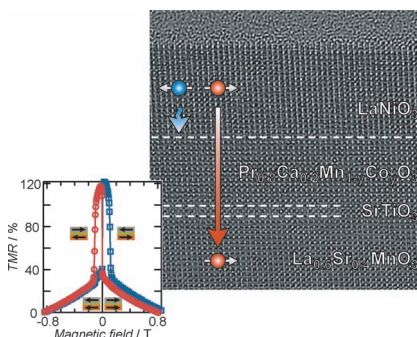


### Fuel Cells

G. Titvinidze, K.-D. Kreuer, M. Schuster, C. C. de Araujo, J. P. Melchior, W. H. Meyer\* .....4456–4470

### Proton Conducting Phase-Separated Multiblock Copolymers with Sulfonated Poly(phenylene sulfone) Blocks for Electrochemical Applications: Preparation, Morphology, Hydration Behavior, and Transport

**An epitaxial oxide spin-filter tunnel junction** is fabricated by pulsed laser deposition with atomic-layer precision. By cutting magnetic coupling at a spin filter/spin detector interface, clear tunnel magnetoresistance exceeding 120% is obtained. The composition-controlled magnetic hardness of a  $\text{Pr}_{0.8}\text{Ca}_{0.2}\text{Mn}_{1-y}\text{Co}_y\text{O}_3$  spin filter layer makes it possible to develop an epitaxial double spin filter tunnel junction.



### Spintronics

T. Harada, I. Ohkubo,\* M. Lippmaa, Y. Sakurai, Y. Matsumoto, S. Muto, H. Koinuma, M. Oshima .....4471–4475

### Large Tunnel Magnetoresistance in Epitaxial Oxide Spin-Filter Tunnel Junctions

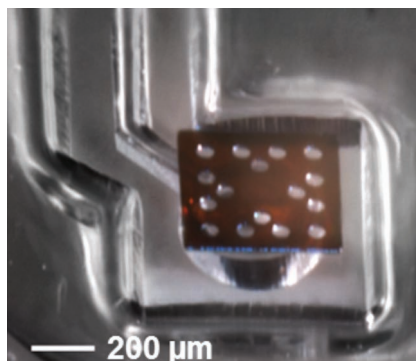


## FULL PAPERS

## Elastomeric Materials

A. Carlson, S. Wang, P. Elvikis,  
P. M. Ferreira, Y. Huang,\*  
J. A. Rogers\* .....4476–4484

**Active, Programmable Elastomeric Surfaces with Tunable Adhesion for Deterministic Assembly by Transfer Printing**



**Active, programmable control of interfacial adhesion** is an important, desired feature of many existing and envisioned systems, including medical tapes, releasable joints, and stamps for transfer printing. A design for an elastomeric surface that offers tunable adhesion strength through a combination of peel-rate dependent effects and actuation of subsurface fluid chambers is presented. When implemented in stamps for transfer printing, these structures provide utility in deterministic assembly of micro- and nanoscale materials onto diverse types of substrates including plastic, paper, and even the living surfaces of leaves.

## Battery Materials

W. Weng, Z. Zhang,\* A. Abouimrane,  
P. C. Redfern, L. A. Curtiss,  
K. Amine .....4485–4492

**Smart Polymeric Cathode Material with Intrinsic Overcharge Protection Based on a 2,5-Di-*tert*-butyl-1,4-dimethoxybenzene Core Structure**

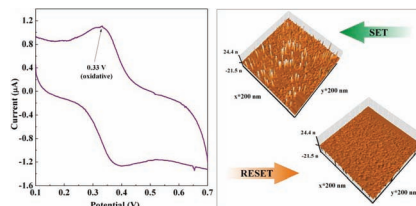


**A new polymeric cathode (I) based on a redox shuttle (RS) core** displays reversible oxidation/reduction at  $\approx 3.96$  V vs Li/Li<sup>+</sup>. At a 1C rate, the polymer I/Li cell shows stable cycling up to 200 cycles between 4.05 and 3.0 V with less than 10% capacity loss. When overcharged, an intrinsic RS mechanism is remarkably activated at 4.2 V.

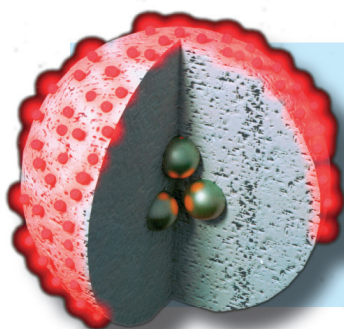
## Biomaterials

M. K. Hota, M. K. Bera,\* B. Kundu,  
S. C. Kundu,\* C. K. Maiti .....4493–4499

**A Natural Silk Fibroin Protein-Based Transparent Bio-Memristor**



**A silk fibroin protein-based biomemristor** demonstrates bipolar memristive switching, which can be explained using the concept of carrier trapping or detrapping in silk fibroin films due to a redox process. The rapid transition between oxidation and reduction states of silk in response to diverse applied electric fields significantly contributes to the memristive switching. This bioelectronics memory device opens up ample opportunities in neuromorphic computing, high-density logic, and memory applications.



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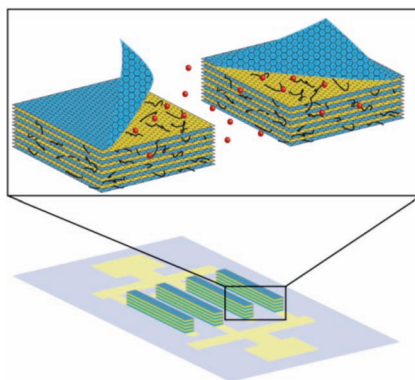
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## FULL PAPERS

**Binder-free micropatterned electrodes of reduced graphene oxide and carbon nanotube composites** are prepared for micro-supercapacitor application by combining electrostatic spray deposition and photolithography lift-off. The high accessibility of the electrode materials and short diffusion path of electrolyte ions results in excellent rate capability and frequency response of the micro-supercapacitors.



## Supercapacitors

M. Beidaghi, C. Wang\* .....4501–4510

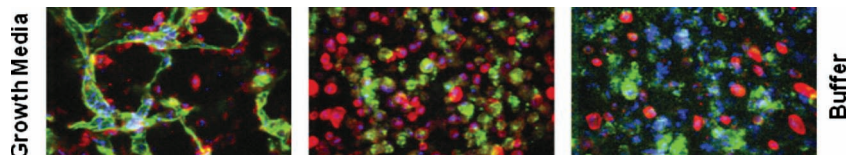
**Micro-Supercapacitors Based on Interdigital Electrodes of Reduced Graphene Oxide and Carbon Nanotube Composites with Ultrahigh Power Handling Performance**

**The most significant tissue engineering barrier** is insufficient vascularization. A micro-fluidic hydrogel is integrated with a self-assembling pro-vasculogenic co-culture to control vessel establishment and create an anastomotic interface for integration with fabricated microchannels. Vessel development is temporally regulated. It is further shown that transport within these regions shifts from diffusion to vessel-supported convective movement.

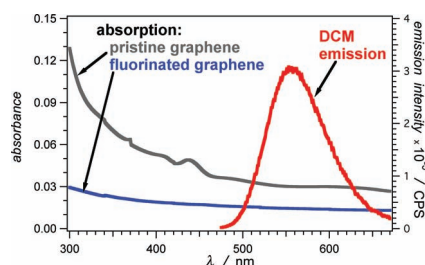
## Tissue Engineering

M. P. Cuchiara, D. J. Gould,  
M. K. McHale, M. E. Dickinson,  
J. L. West\* .....4511–4518

**Integration of Self-Assembled Microvascular Networks with Microfabricated PEG-Based Hydrogels**



**The utilization of fluorescence quenching microscopy (FQM)** for quick visualization of chemical functionalization in relatively large regions of graphene is described. With premeditated choice of the dye, the interaction between fluorophores and graphene provides advantages for probing the chemical structure of functionalized graphene. Segmented FQM can be employed to address the chronic need for microscopy-based graphene metrology.

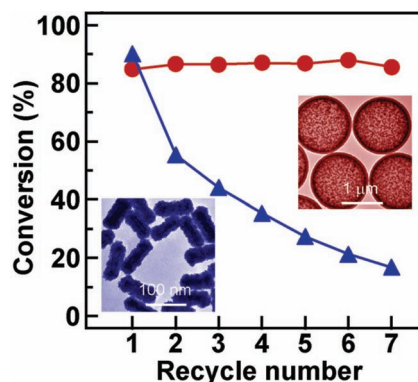


## Functionalized Graphene

M. Ghazinejad, J. R. Kyle, S. Guo,  
D. Pleskot, D. Bao, V. I. Vullev,  
M. Ozkan,\* C. S. Ozkan\* .....4519–4525

**Non-Invasive High-Throughput Metrology of Functionalized Graphene Sheets**

**Different monometallic Au and bimetallic Au-cored nanostructures** are grown using a “ship-in-a-bottle” technique in hollow mesoporous silica microspheres. The microsphere-encapsulated Au nanorod core/Pt shell nanostructures exhibit both high catalytic performance and superior recyclability in the oxidation reaction of *o*-phenylenediamine with hydrogen peroxide, while the conversion percentage in the presence of the corresponding unencapsulated nanostructures decreases rapidly with the recycle number.



## Metal Nanostructures

M. D. Xiao, C. M. Zhao, H. J. Chen,  
B. C. Yang, J. F. Wang\* .....4526–4532

**“Ship-in-a-Bottle” Growth of Noble Metal Nanostructures**

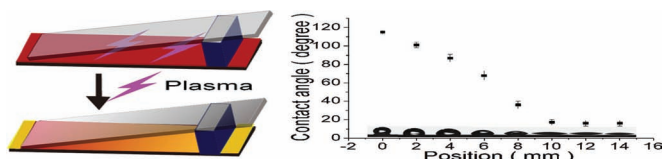
## FULL PAPERS

## Functionalized Surfaces

X. Han,\* L. Wang,  
X. Wang.....4533–4538

### Fabrication of Chemical Gradient Using Space Limited Plasma Oxidation and its Application for Droplet Motion

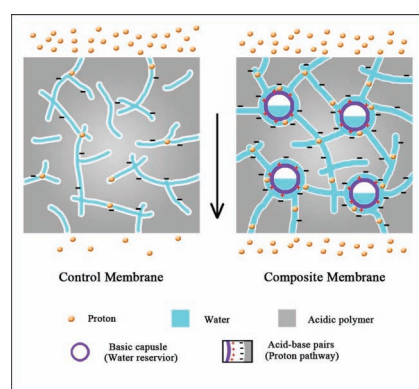
A novel approach, space limited plasma oxidation, is introduced to fabricate chemical gradients on self-assembled monolayer (SAM) surfaces. This method is highly versatile, rapid, and low cost. It provides an easy way to fabricate gradient surfaces which can be either further functionalized by biomolecules (enzymes, antibodies, DNA, etc.) for investigation of biomolecular interactions, cell-adhesion studies and diagnostics, or used for spontaneous droplet motion.



## Composite Materials

J. Wang, X. Yue, Z. Zhang, Z. Yang,  
Y. Li, H. Zhang, X. Yang,  
H. Wu, Z. Jiang\*.....4539–4546

### Enhancement of Proton Conduction at Low Humidity by Incorporating Imidazole Microcapsules into Polymer Electrolyte Membranes

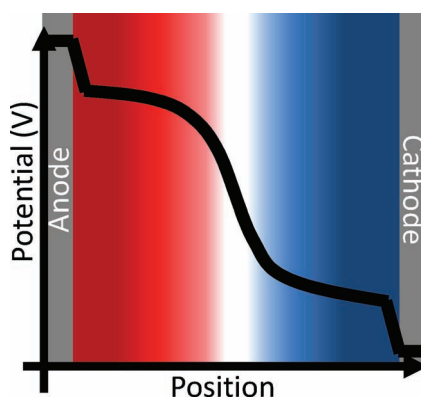


Imidazole microcapsules with high imidazole group loading and large lumen size are synthesized and incorporated into a sulfonated poly(ether ether ketone) (SPEEK) membrane. Due to its superior water retention properties, anhydrous proton transfer feature of embedded imidazole microcapsules, and the low energy barrier pathways from the acid-base pairs, the SPEEK composite membrane displays notably enhanced proton conductivity in particular under low humidity.

## Organic Electronics

S. van Reenen, R. A. J. Janssen,  
M. Kemerink\*.....4547–4556

### Dynamic Processes in Sandwich Polymer Light-Emitting Electrochemical Cells

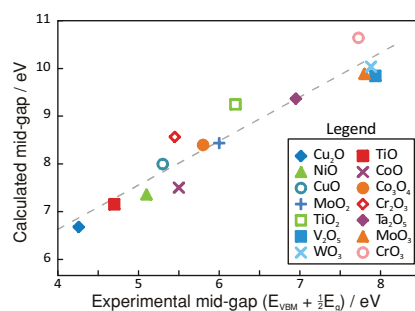


Ion conductivity, electric double layer thickness, and electrochemical doping are identified by numerical modeling of admittance spectroscopy on light-emitting electrochemical cells (LECs). These results confirm the validity of the electrochemical doping model to describe LEC operation and it is shown that in order to significantly decrease the response-time of LECs an electrolyte/salt combination with a minimal ion binding energy must be used.

## Metal Oxides

M. T. Greiner,\* L. Chai, M. G. Helander,  
W.-M. Tang, Z.-H. Lu.....4557–4568

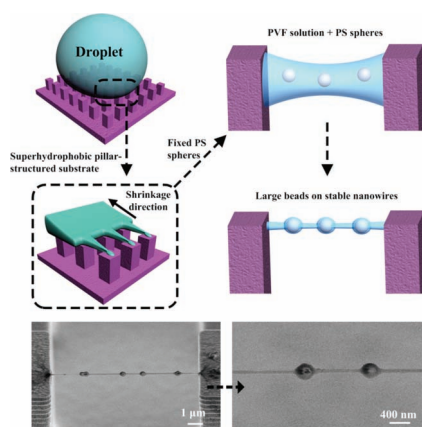
### Transition Metal Oxide Work Functions: The Influence of Cation Oxidation State and Oxygen Vacancies



The work functions of transition-metal oxides are correlated with the oxidation states of their metal cations. Due to the relationship between electronegativity and oxidation state, reduced oxides tend to have lower work functions. The trends revealed provide guidelines for tuning transition metal oxide work functions.

## FULL PAPERS

**Regularly aligning bead-shaped nanowire arrays** are generated, assisted by highly adhesive superhydrophobic pillar-structured silicon substrates. The bead-shaped structures can be introduced by either Rayleigh instability at ultralow polymer concentration or rigid polystyrene spheres serving as bead building blocks. Furthermore, bead-shaped nanowires have the unique ability to directionally drive tiny drops and collect coalesced microdroplets when placed in mist.

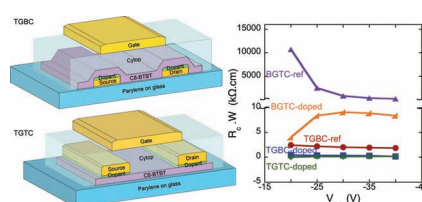


### Nanowires

Y. C. Wu, X. Chen, B. Su,\* Y. L. Song, L. Jiang .....4569–4576

**Elaborately Aligning Bead-Shaped Nanowire Arrays Generated by a Superhydrophobic Micropillar Guiding Strategy**

**An exceptionally low contact resistance** is achieved in top-gated organic field-effect transistors by using coplanar and pseudo-staggered device architectures and introducing a dopant layer. Compared with conventional bottom-gated staggered devices, a remarkably low contact resistance is extracted from the top-gated devices by the modified transfer line method.

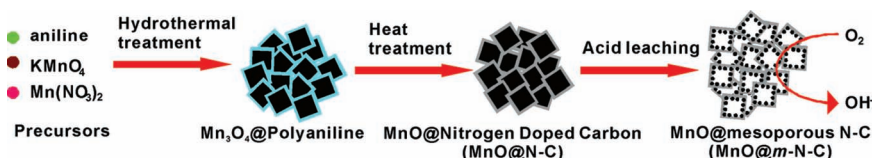


### Field-Effect Transistors

P. Darmawan,\* T. Minari,\* Y. Xu, S.-L. Li, H. Song, M. Chan, K. Tsukagoshi\* .....4577–4583

**Optimal Structure for High-Performance and Low-Contact-Resistance Organic Field-Effect Transistors Using Contact-Doped Coplanar and Pseudo-Staggered Device Architectures**

**A novel MnO-containing mesoporous nitrogen-doped carbon (*m*-N-C) nanocomposite** is facilely synthesized using an in situ manganese oxide template method. The MnO-*m*-N-C composite catalyst shows similar electrocatalytic activity but superior stability and methanol tolerance to a commercial Pt/C catalyst for the four-electron oxygen reduction reaction in alkaline solution.

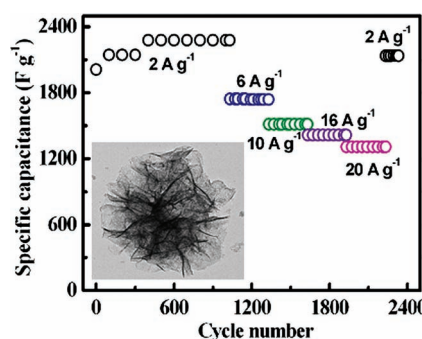


### Catalysis

Y. Tan, C. Xu, G. Chen, X. Fang, N. Zheng,\* Q. Xie .....4584–4591

**Facile Synthesis of Manganese-Oxide-Containing Mesoporous Nitrogen-Doped Carbon for Efficient Oxygen Reduction**

**An advanced 3D self-supported electrode** is fabricated by growing ultrathin mesoporous NiCo<sub>2</sub>O<sub>4</sub> nanosheets on Ni foam with strong adhesion for high-performance electrochemical capacitors. Because of its many advantageous structural features, this self-supported hybrid electrode is able to deliver ultra-high specific capacitance at a very high current density, with excellent cycling stability.



### Electrodes

C. Z. Yuan, J. Y. Li, L. R. Hou, X. G. Zhang, L. F. Shen, X. W. Lou\* .....4592–4597

**Ultrathin Mesoporous NiCo<sub>2</sub>O<sub>4</sub> Nanosheets Supported on Ni Foam as Advanced Electrodes for Supercapacitors**

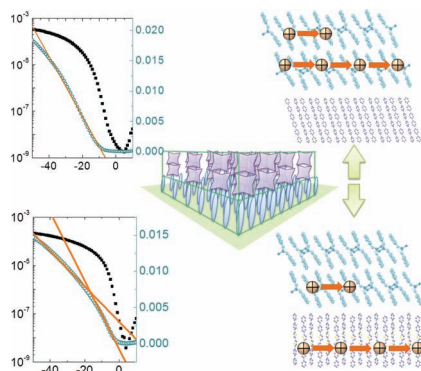


## FULL PAPERS

## Organic Semiconductors

L. Huang, F. Zhu,\* C. Liu, U. Treske,  
M. Grobosch, H. Tian, J. Zhang,  
Y. Geng, M. Knupfer,  
D. Yan\* .....4598–4607

**Crystalline Organic Heterostructures  
Engineering Based on Vanadyl  
Phthalocyanine and Rod-Like  
Conjugated Organic Semiconductors  
with Selected Central Groups**

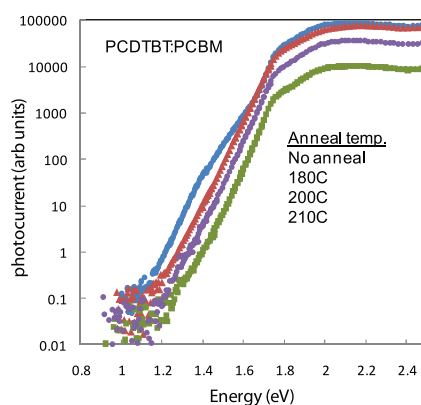


**Highly ordered organic crystals** are constructed from a series rod-like molecules and a disc-like phthalocyanine. The series of rod-like molecules differed in the central groups and present different lattice parameters and electronic structure, which results in distinct morphology and electronic structure in the heterostructure. Transistors based on the crystalline heterostructure present diverse charge transport behavior and field-effect mobilities.

## Solar Cells

R. A. Street,\* A. Krakaris,  
S. R. Cowan .....4608–4619

**Recombination Through Different  
Types of Localized States in Organic  
Solar Cells**



**Recombination in bulk heterojunction solar cells** is explored by observing the results of extended white light illumination, thermal annealing to high temperature, and chemical doping. Measurements of spectral response and other cell characteristics show that recombination is through localized states of different type, depending on the circumstances.