

# ADVANCED FUNCTIONAL MATERIALS

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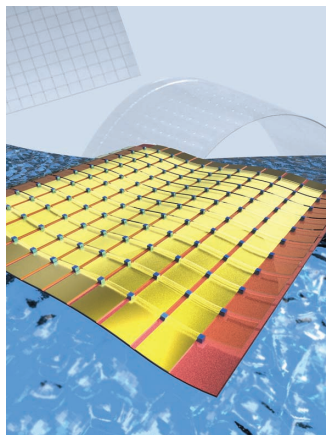


## Biomimetalization

The cover art depicts a row of ultrahard and abrasion-resistant teeth from the giant chiton, *Cryptochiton stelleri*, grinding on a hard surface, which is a rendering of a cross-section of the tooth itself. Chemical and ultrastructural features of the teeth, which enhance fracture mitigation are reported by D. Kisailus and team on page 6093. Lessons obtained from this work can be used towards the development of abrasion resistant materials.

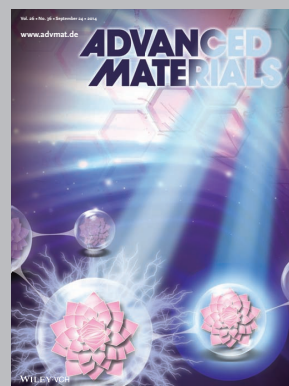
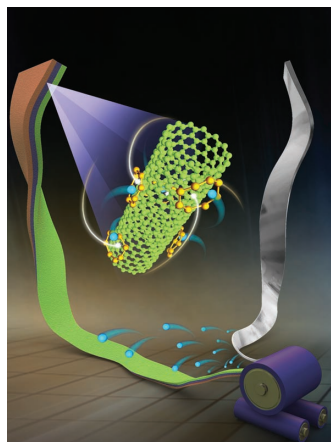
## Flexible Electronics

T. Pan and co-workers present a flexible, transparent, and pressure-sensitive microfluidic film, referred to as microfluidics, for large-area dynamic pressure mapping. On page 6195, utilizing a continuous responsive microfluidic layer as the sensing element, the microfluidic sensors offer an alternative approach to the solid-state counterparts, by offering an unprecedented sensitivity and ultrafast response time in a completely transparent, flexible, and adaptive package.



## Electrodes

On page 6105, J.-Q. Huang, Q. Zhang, and co-workers discuss a hierarchical free-standing carbon nanotube (CNT)/sulfur paper electrode with short CNTs as short-range electrical conductive framework for sulfur accommodation and super long CNTs as both long-range conductive networks and intercrossed mechanical scaffolds. Such rational design of flexible electrode offers the possibility to efficient use of active materials at practical loading.



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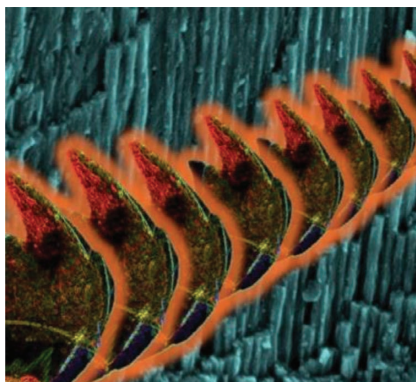
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## FULL PAPERS

## Biom mineralization

L. K. Grunenfelder, E. E. de Obaldia,  
Q. Wang, D. Li, B. Weden, C. Salinas,  
R. Wuhler, P. Zavattieri,  
D. Kisailus\* ..... 6093–6104

**Stress and Damage Mitigation from  
Oriented Nanostructures within the  
Radular Teeth of *Cryptochiton stelleri***

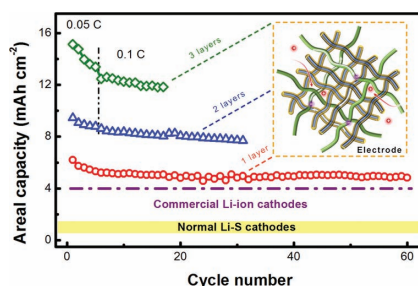


The teeth of the chiton are hard and abrasion resistant. Local and global chemical and ultrastructural features of the teeth, as well as chemical and structural gradients which enhance fracture mitigation, are reported. Lessons obtained can be used towards the development of abrasion resistant materials for tooling and machining applications, as well as coatings for equipment and medical implants.

## Electrodes

Z. Yuan, H.-J. Peng, J.-Q. Huang,\*  
X.-Y. Liu, D.-W. Wang, X.-B. Cheng,  
Q. Zhang\* ..... 6105–6112

**Hierarchical Free-Standing  
Carbon-Nanotube Paper Electrodes  
with Ultrahigh Sulfur-Loading for  
Lithium–Sulfur Batteries**

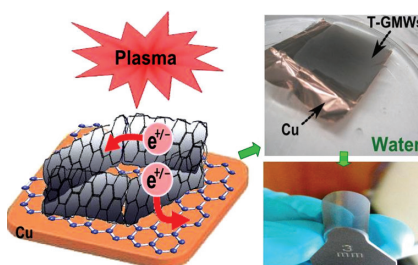


A hierarchical free-standing paper electrode is fabricated using short multi-walled carbon nanotubes (MWCNTs) and super-long CNTs. The MWCNTs function as a short-range electrical conductive framework for sulfur accommodation, while super-long CNTs act as both a long-range conductive network and mechanical scaffold.

## Energy-Efficient Growth

S. Kumar, T. van der Laan, A. E. Rider,  
L. Randeniya, K. Ostrikov\*... 6114–6122

**Multifunctional Three-Dimensional  
T-Junction Graphene Micro-Wells:  
Energy-Efficient, Plasma-Enabled Growth  
and Instant Water-Based Transfer for  
Flexible Device Applications**



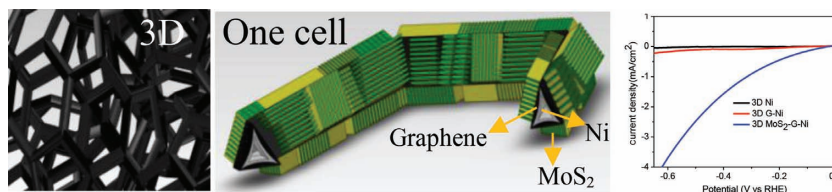
Energy-efficient plasma-enabled chemical vapor deposition (CVD) synthesis is described, as well as a simple instant chemical-free water-assisted decoupling and a transfer process of unique three-dimensional T-junction graphene microwells structures. The structures deposited on a copper foil are then transferred without any significant damage, in de-ionized or tap water at room temperature, into virtually any, including flexible transparent substrates, for their multifunctional device applications.

## Hydrogen Evolution

X. Geng, W. Wu, N. Li, W. Sun,  
J. Armstrong, A. Al-hilo, M. Brozak,  
J. Cui,\* T. p. Chen\* ..... 6123–6129

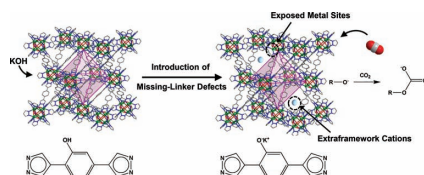
**Three-Dimensional Structures of MoS<sub>2</sub>  
Nanosheets with Ultrahigh Hydrogen  
Evolution Reaction in Water Reduction**

A robust and highly active system is demonstrated for the hydrogen evolution reaction in alkaline solution by developing MoS<sub>2</sub> nanosheet arrays vertically aligned on graphene-mediated 3D Ni networks. The catalytic activity of the 3D MoS<sub>2</sub> nanostructures is found to increase by 2 orders of magnitude as compared to the Ni networks without MoS<sub>2</sub>.



## FULL PAPERS

A boosting of CO<sub>2</sub> capture properties in a series of functional MOF materials can be achieved by post-synthetic treatment with KOH ethanolic solutions, leading to linker removal and the concomitant creation of highly active surface sites for the adsorption of acidic gases.

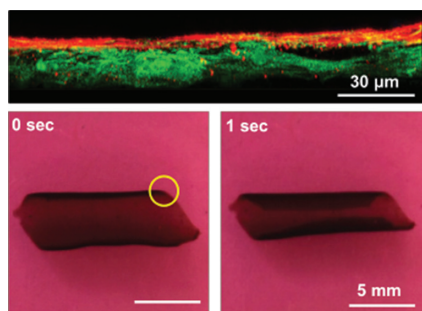


## Metal–Organic Frameworks

E. López-Maya, C. Montoro,  
V. Colombo, E. Barea,  
J. A. R. Navarro\* .....6130–6135

Improved CO<sub>2</sub> Capture from Flue Gas  
by Basic Sites, Charge Gradients, and  
Missing Linker Defects on Nickel Face  
Cubic Centered MOFs

A multilayered cardiac tissue is fabricated by interfacing poly-L-lysine graphene oxide particles on the surface of each cell layer using a layer-by-layer technique. The particles embedded within the multilayer cardiac constructs improve cardiac cell organization, maturation, and cell–cell electrical coupling. Multilayer cardiac tissue shows strong spontaneous beating and frequency-dependent opening/closing actuation under a low external electric field.

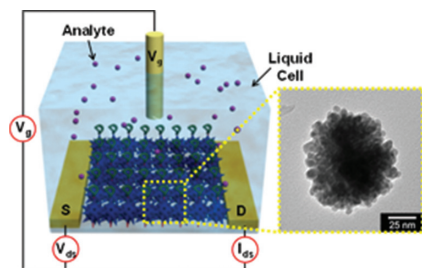


## Tissue Engineering

S. R. Shin, B. Aghaei-Ghareh-Bolagh,  
X. Gao, M. Nikkhah, S. M. Jung,  
A. Dolatshahi-Pirouz, S. B. Kim,  
S. M. Kim, M. R. Dokmeci, X. Tang,\*  
A. Khademhosseini\* .....6136–6144

Layer-by-Layer Assembly of 3D Tissue  
Constructs with Functionalized  
Graphene

A multidimensional system comprising conjugated-polymer nanoparticles, whose surfaces are decorated to increase surface area and allow further polymerization, is fabricated and functionalized with an aptamer. The resulting multidimensional nanoparticles are used in field-effect transistors (FETs) that can act as sensors of the endocrine disruptor bisphenol A (BPA). The aptamer FET sensors exhibit ultrahigh sensitivity and selectivity toward BPA, and their lifetime is expected to exceed that of other FET biosensors.

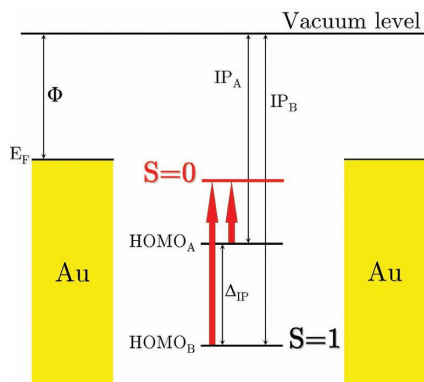


## Sensors

J. S. Lee, S. G. Kim, J. Jun,  
D. H. Shin, J. Jang\* .....6145–6153

Aptamer-Functionalized  
Multidimensional Conducting-Polymer  
Nanoparticles for an Ultrasensitive  
and Selective Field-Effect-Transistor  
Endocrine-Disruptor Sensors

The Fermi level alignment in molecular junctions can be either affected ( $S = 1$ ) or unaffected ( $S = 0$ ) by the modification of the ionization potential of the contacted molecule. Here, it is shown that a transition between these two behaviors is achievable by a simple substitution, while the key role of metal induced gap states (MIGS) in the alignment and orbital polarization processes is further highlighted.



## Molecular Electronics

C. Van Dyck,\* V. Geskin,  
J. Cornil .....6154–6165

Fermi Level Pinning and Orbital  
Polarization Effects in Molecular  
Junctions: The Role of Metal Induced  
Gap States

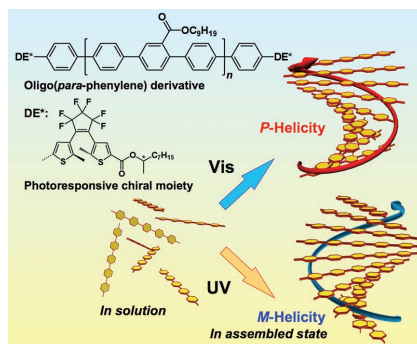


# FULL PAPERS

## Chirality

B. A. San Jose, T. Ashibe, N. Tada,  
S. Yorozya, K. Akagi\*..... 6166–6171

**Helicity Control of  $\pi$ -Stacked Assemblies of Oligo(*para*-phenylene) Derivatives Using Photoresponsive Chiral Moieties at Terminal Sites**

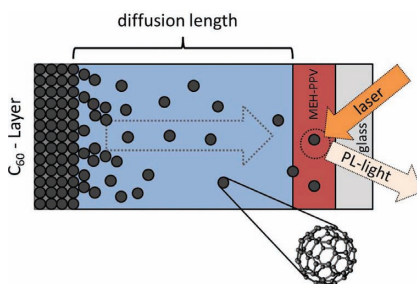


**Oligo(*para*-phenylene) (OPP) derivatives** bearing photoresponsive chiral dithienylethene (DE\*) terminal moieties are synthesized. Photoisomerization of the chiral DE\* terminal moieties prior to assembly, from the open to closed form, changes the handedness of the helically  $\pi$ -stacked structures in the assembled state. Furthermore, the chiral transcription of racemic poly(bithiophenethiophene) by the assembled chiral OPP is demonstrated.

## Molecular Diffusion

F. Fischer, T. Hahn, H. Bässler, I. Bauer,  
P. Strohmriegel, A. Köhler\*..... 6172–6177

**Measuring Reduced  $C_{60}$  Diffusion in Crosslinked Polymer Films by Optical Spectroscopy**

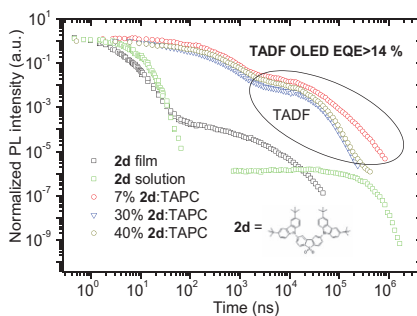


The diffusion of fullerenes such as  $C_{60}$  and PCBM in organic semiconductors can be measured using a novel simple optical method based on photoluminescence quenching. When the mobility of the polymer chains is reduced by chemical crosslinking, the diffusion coefficient of  $C_{60}$  can be reduced by up to three orders of magnitude.

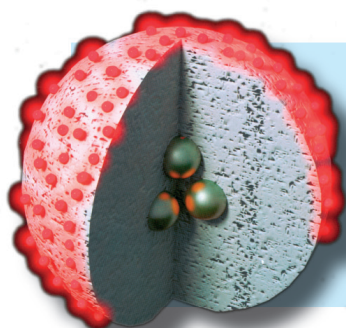
## Fluorescence

V. Jankus,\* P. Data, D. Graves,  
C. McGuinness, J. Santos, M. R. Bryce,  
F. B. Dias, A. P. Monkman... 6178–6186

**Highly Efficient TADF OLEDs: How the Emitter–Host Interaction Controls Both the Excited State Species and Electrical Properties of the Devices to Achieve Near 100% Triplet Harvesting and High Efficiency**



**New emitters harvesting triplets to give 100% internal efficiency** are required to replace Ir based phosphors in OLEDs. Here, it is shown that a D–A molecule in the solid state emits via an intramolecular charge transfer excited state and via exciplex states, and OLEDs based on thermally activated delayed fluorescence achieve >14% external electroluminescence yield and 100% efficient triplet harvesting.



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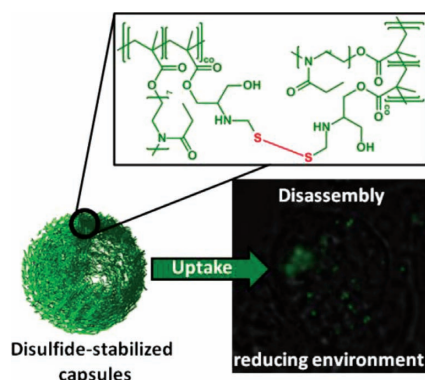
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**Low-fouling, redox-responsive polymer capsules** based on functional poly(2-oxazoline)s are assembled. These capsules are shown to degrade under simulated intracellular conditions as well as intracellularly. Their low-fouling properties make them promising for the delivery and triggered release of therapeutics.

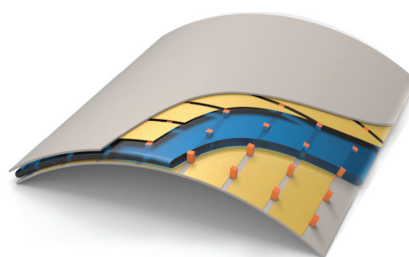


## Polymer Carriers

K. Kempe, S. L. Ng, S. T. Gunawan,  
K. F. Noi, F. Caruso\* .....6187–6194

## Intracellularly Degradable Hydrogen-Bonded Polymer Capsules

**A flexible, transparent, and pressure-sensitive microfluidic film, referred to as a microflotronic,** is reported for large-area dynamic pressure mapping applications. Utilizing a continuous microfluidic layer as the sensing elements, the microflotronic devices offer an alternative approach to the solid-state pressure sensors, by offering an unprecedented sensitivity and ultrafast response time in a completely transparent, flexible, and adaptive platform.

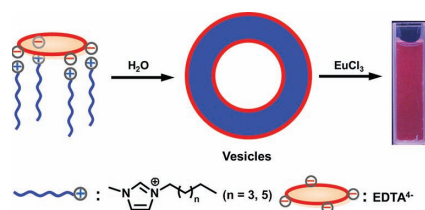


## Flexible Electronics

R. Li, B. Nie, P. Digiglio,  
T. Pan\* ..... 6195–6203

## Microflotronics: A Flexible, Transparent, Pressure-Sensitive Microfluidic Film

**A counterion-directed molecular design strategy** for the spontaneous formation of stable vesicles via readily available imidazolium salts with the EDTA counteranion in aqueous media is proposed. Importantly, the unusual counterion-induced vesicle-like sphere aggregates can further chelate metal ions to enhance the europium-centered emission in aqueous media and make it viable for an optical pH sensor.

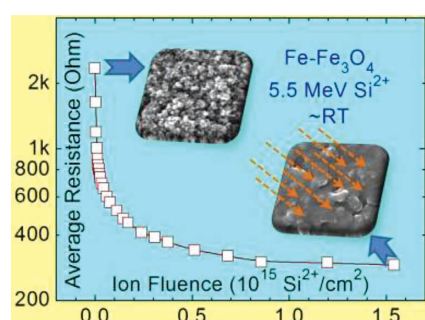


## Counterions

G. Li, S. Zhang,\* N. Wu, Y. Cheng,  
J. You\* ..... 6204–6209

## Spontaneous Counterion-Induced Vesicle Formation: Multivalent Binding to Europium(III) for a Wide-Range Optical pH Sensor

**An in situ study on the nanostructural evolution and electrical resistance variation of  $\text{Fe}_3\text{O}_4$  and  $\text{Fe-Fe}_3\text{O}_4$  core-shell nanocluster films under ion irradiation is presented. Grain growth, phase transition, particle aggregation, and formation of nanowire-like network with nanopores are observed. The electrical resistance exhibits a super-exponential decay with dose. This type of films may have potential as a sensing material for fast neutron monitoring.**



## Nanostructures

W. Jiang,\* J. A. Sundararajan,  
T. Varga, M. E. Bowden, Y. Qiang,  
J. S. McCloy, C. H. Henager Jr.,  
R. O. Montgomery.....6210-6218

## In Situ Study of Nanostructure and Electrical Resistance of Nanocluster Films Irradiated with Ion Beams

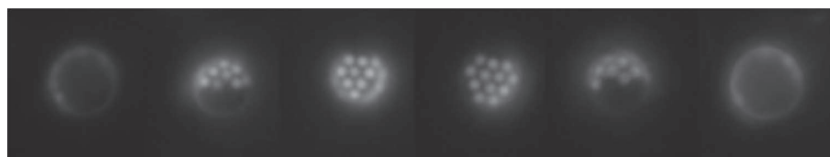
## FULL PAPERS

## Microparticle Synthesis

X. Wang, D. S. Miller, J. J. de Pablo,  
N. L. Abbott\* ..... 6219–6226

**Reversible Switching of Liquid  
Crystalline Order Permits Synthesis of  
Homogeneous Populations of Dipolar  
Patchy Microparticles**

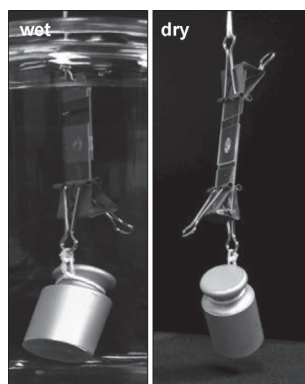
Switching of the internal configurations of micrometer-sized droplets of liquid crystal is used to uniformly position colloids on their surfaces, thus enabling formation of homogeneous populations of patchy microdroplets. The method is generalizable and, when combined with photo-polymerization of the droplets, can be utilized for synthesis of solid “Janus-like” microparticles or functional patchy microparticles with dipolar symmetry, including magnetically responsive systems.



## Adhesives

A. Pena-Francesch, B. Akgun,  
A. Miserez, W. Zhu, H. Gao,  
M. C. Demirel\* ..... 6227–6233

**Pressure Sensitive Adhesion of an  
Elastomeric Protein Complex Extracted  
From Squid Ring Teeth**

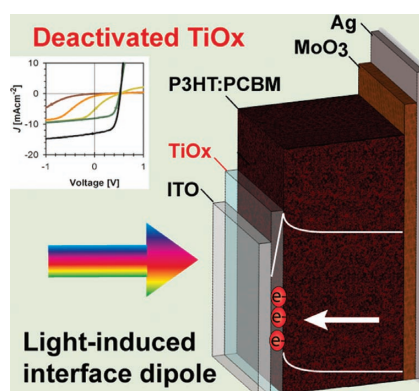


A protein-based adhesives extracted from squid ring teeth is introduced. Under-water adhesion strength of the protein film is approximately 1.5–2.5 MPa. This reusable and recyclable bioelastomers could potentially have a wide range of applications in wet bioadhesive development including dental resins, bandages for wound healing, and surgical sutures in the body, all of which require wet adhesion.

## Interface Dipoles

A. Guerrero, S. Chambon, L. Hirsch,  
G. Garcia-Belmonte\* ..... 6234–6240

**Light-modulated  $\text{TiO}_x$  Interlayer Dipole  
and Contact Activation in Organic Solar  
Cell Cathodes**



Light-modulated dipole (electron accumulation) present at  $\text{TiO}_x$  interlayer accounts for the S-shaped characteristics of organic solar cells. UV-light activation tunes the selectivity of the cathode contact to electrons. Activated devices do not exhibit light-dependent interface dipole. By using capacitance–voltage measurements a complete picture of the contact energetics is drawn.