# ADVANCED FUNCTIONAL MATERIALS

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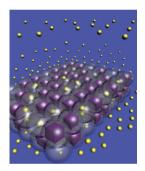


## **Biological Materials**

L. Li and C. Ortiz show on page 3463 that adjacent mineral layers in the laminate composite of a bivalve (Placuna placenta) shell are joined together through characteristic nanoscale screw dislocation-like connection centers. This leads to extensive crack bridging and deflection surrounding macroscopic indentation damage zones, which forms an interconnected network of microcracks, enabling simultaneous damage localization and energy dissipation.

#### Cyclability

Y. Yu and colleagues develop a surfactant-assisted assembly method to fabricate 3D nanoarchitectures in which Sn-based nanoparticles are encapsulated by a porous graphene network. The novel nanostructure design presented on page 3488 offers the necessary void space for the expansion of active particles, preventing them from destroying the 3D conductive graphene framework on cycling, and allowing fast transport of Li-ions via electrolyte filling into the percolating free interstices. As a result, the composites show high reversible capacity and fast rate-capability, as well as ultralong cycle life.



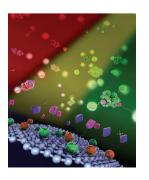
#### Gene Delivery

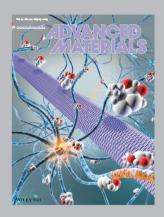
On page 3472, K. Na and co-workers report highly efficient photo-triggered gene expression without damage to the gene from reactive oxygen species by taking advantage of the endo-lysosomal pH-sensitive de-shielding ability of the pH-responsive shielding material incorporated in the gene complexes. Importantly, this newly developed gene delivery system also resolves the tissue penetration issue in drug delivery systems by photo-mediated breaking down the extracellular matrix.

### **Graphene Composites**

Adv. Funct. Mater. 2015, 25, 3447

Two different glycoligands coupled with different fluorophores are co-assembled to a graphene substrate with a quenched fluorescence by X.-P. He and team on page 3483. Each glycoligand-receptor recognition causes the fluorescence recovery of a single fluorescence emission with no interference with each other. Meanwhile, the simultaneous formation of both glycoligand-receptor complexes leads to the production a mixed fluorescence color, making possible the detection of diverse bio-recognition events concurrently with an integrated dual fluorogenic graphene composite material.





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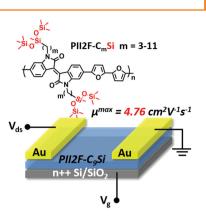


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Alkyl siloxane-terminated side chains equipped isoindigo-based conjugated polymers (PII2F- $C_m$ Si, m = 3-11) are prepared to evaluate the structure-property relationship as the branching point is systematically "moved away" from the conjugated backbone one carbon atom at a time. An improved charge carrier mobility of 4.76 cm<sup>2</sup>  $V^{-1}$  s<sup>-1</sup> (PII2F-C<sub>9</sub>Si) is exhibited.



#### **Charge Transport**

J. Mei, H.-C. Wu, Y. Diao, A. Appleton,

H. Wang, Y. Zhou, W.-Y. Lee,

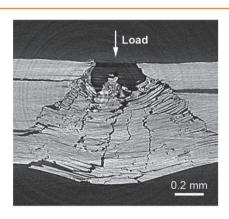
T. Kurosawa, W.-C. Chen.

Z. Bao\*.....3455-3462

Effect of Spacer Length of Siloxane-Terminated Side Chains on Charge Transport in Isoindigo-Based Polymer Semiconductor Thin Films



The remarkable resistance to macroscopic mechanical damage in a highly mineralized shell of the bivalve Placuna placenta originates from a characteristic nanoscale structural motif, that is, screw dislocation-like connection centers, which join adjacent mineral layers together in its laminated structure. This enables the formation of an interconnected network of microcracks within the damage zone, allowing simultaneous efficient energy dissipation and damage localization.

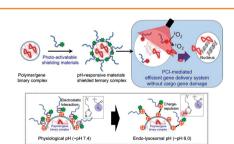


#### **Biological Materials**

L. Li,\* C. Ortiz\*.....3463-3471

A Natural 3D Interconnected Laminated Composite with Enhanced Damage Resistance

A new type of stable light-triggered gene delivery system is designed based on a pH-responsive shielding material incorporated into ternary gene complexes (pH-TCs). The endosomal pH-sensitive de-shielding effect of pH-TCs exhibits highly discriminating light-triggered gene expression both in vitro and in vivo without cargo gene damage.

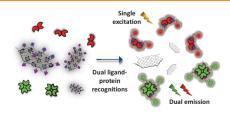


#### **Gene Delivery**

S.-j. Park, W. Park, K. Na\* .....3472-3482

Tumor Intracellular-Environment Responsive Materials Shielded Nano-Complexes for Highly Efficient Light-Triggered Gene Delivery without Cargo Gene Damage

Simultaneous detection of diverse glycoligand-protein recognitions is made possible using a dual-fluorogenic composite material. This multiplex sensing material shows a dual emission, with no interference with each other on a single excitation, upon concurrent interaction with two protein receptors.



#### **Graphene Composites**

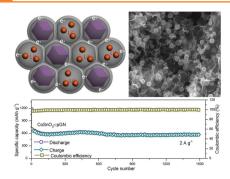
D.-K. Ji, G.-R. Chen, X.-P. He,\* H. Tian ......3483-3487

Simultaneous Detection of Diverse **Glycoligand-Receptor Recognitions** Using a Single-Excitation, Dual-Emission **Graphene Composite** 

#### Cyclability

C. Wu, J. Maier, Y. Yu\*......3488-3496

Sn-Based Nanoparticles Encapsulated in a Porous 3D Graphene Network: Advanced Anodes for High-Rate and Long Life Li-Ion Batteries

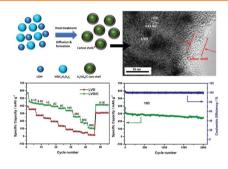


3D porous graphene network-encapsulated Sn-based nanoarchitectures (0D-CoSnO3C 3D-pGN and 0D-Co-Sn⊂3D-pGN) are successfully fabricated by an assembly route. It is the characteristic presence of nanosized pores in the nanostructure that facilitates the access of Li-ions into Sn-based nanoparticles, accommodates the large volume expansion of Sn-based nanoparticles, and preserves the integrity of the 3D graphene-network, leading to excellent electrochemical properties in terms of high rate capability and ultralong cycle life.

#### Lithium-Ion Batteries

C. Zhang, H. Song, C. Liu, Y. Liu, C. Zhang, X. Nan, G. Cao\*...3497-3504

Fast and Reversible Li Ion Insertion in Carbon-Encapsulated Li<sub>3</sub>VO<sub>4</sub> as Anode for Lithium-Ion Battery

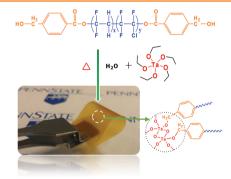


A carbon-encapsulated Li<sub>3</sub>VO<sub>4</sub> composite fabricated by a novel one-step solidstate reaction without external carbon sources presents exceeding rate capability (a reversible capability of 340, 169, and 106 mAh g<sup>-1</sup> at 10 C, 50 C, and 80 C, respectively) and long cyclic performance (80% capacity retention after 2000 cycles at 10 C) as an anode in lithium-ion batteries.

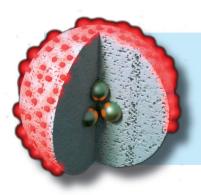
#### **Dielectrics**

K. Han, Q. Li, C. Chanthad, M. R. Gadinski, G. Zhang, Q. Wang\*......3505-3513

A Hybrid Material Approach Toward **Solution-Processable Dielectrics Exhibiting Enhanced Breakdown** Strength and High Energy Density



Ferroelectric polymer-tantalum oxide molecular hybrids exhibit significantly enhanced breakdown strength in comparison with the pristine polymer, leading to higher discharged energy densities. The superior performance is mainly ascribed to the deep traps created in the hybrids, which decrease conduction loss, and the formation of the cross-linked networks that hinder the occurrence of the electromechanical breakdown.



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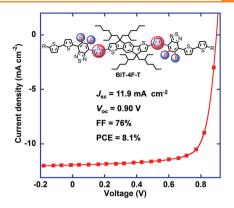
cherth@wiley-vch.de

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(+49) 6201-606-332 Fax: Email: rights@wiley-vch.de Small molecules (BIT-4F-T) based on indacenodithiophene (IDT) and difluorobenzothiadiazole are synthesized for bulk-heterojunction organic solar cells (BHJ-OSCs). Best power conversion efficiency (PCE) of 8.1% and veryhigh FF of 0.76 are achieved, positioning them among the best small-molecule donor materials in simple single-junction organic solar cells. These exciting results verify the significant importance of multiple fluorine substituents of

benzothiadiazole and two thiophene

moieties for high PCE.



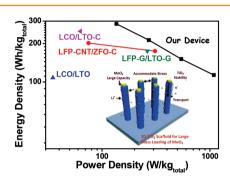
# **FULL PAPERS**

#### **Organic Solar Cells**

J.-L. Wang,\* Q.-R. Yin, J.-S. Miao, Z. Wu, Z.-F. Chang, Y. Cao, R.-B. Zhang,\* J.-Y. Wang,\* H.-B. Wu,\* Y. Cao.....3514-3523

Rational Design of Small Molecular **Donor for Solution-Processed Organic** Photovoltaics with 8.1% Efficiency and High Fill Factor via Multiple Fluorine Substituents and Thiophene Bridge

Synergistic TiO2-MoO3 core-shell nanowire array anode is developed, showing high capacity (ca. 670 mAh g<sup>-1</sup>; 3.986 mAh cm<sup>-2</sup>), excellent cycleability (>200 times), and good rate performance. Ultrahigh energy density (285 Wh  $kg_{total}^{-1}$ ) and power density (1086 W  $kg_{total}^{-1}$ ) are further achieved for a full cell LIB device assembled using the TiO2-MoO3 hybrid array as anode and commercial LiCoO2 film as cathode.

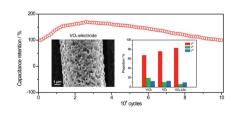


#### **Nanowires**

C. Wang, L. Wu, H. Wang, W. Zuo, Y. Li, J. Liu\*.....3524-3533

Fabrication and Shell Optimization of Synergistic TiO2-MoO2 Core-Shell Nanowire Array Anode for High Energy and Power Density Lithium-Ion Batteries

Aiming at the crucial challenge of poor electrochemical stability for vanadium oxides electrodes, an innovative and effective method is reported to significantly boost their durability and capacitance through tuning the valence state of vanadium.

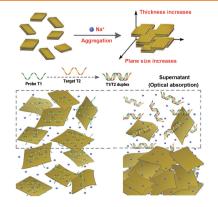


#### **Supercapacitors**

M. Yu, Y. Zeng, Y. Han, X. Cheng, W. Zhao, C. Liang, Y. Tong, H. Tang,\* X. Lu\*.....3534-3540

Valence-Optimized Vanadium Oxide Supercapacitor Electrodes Exhibit **Ultrahigh Capacitance and Super-Long** Cyclic Durability of 100 000 Cycles

Size-dependent optical absorption of semiconductive (2H) layered molybdenum disulfide (MoS2), exhibiting great discriminated abilities to single- and double-stranded DNA, is explored through salt-induced aggregation of 2D nanosheets. Meanwhile, the aggregation behavior of layered MoS2 is remarkably inhibited by the synergistic effects of DNA oligonucleotides, contributing to the developments of biosensors based on optical absorption spectrum of layered  $MoS_2$ .



# **Biosensing**

B. L. Li, H. L. Zou, L. Lu, Y. Yang, J. L. Lei, H. Q. Luo,\* N. B. Li\*...3541-3550

Size-Dependent Optical Absorption of Layered MoS2 and DNA Oligonucleotides Induced Dispersion Behavior for Label-Free Detection of Single-Nucleotide Polymorphism

#### Graphene Oxide

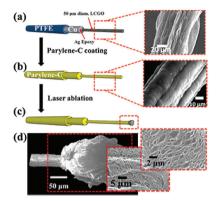
N. V. Apollo, M. I. Maturana, W. Tong, D. A. X. Nayagam,

M. N. Shivdasani, J. Foroughi,

G. G. Wallace, S. Prawer, M. R. Ibbotson.

D. J. Garrett\* ...... 3551-3559

Soft, Flexible Freestanding Neural Stimulation and Recording Electrodes Fabricated from Reduced Graphene Oxide

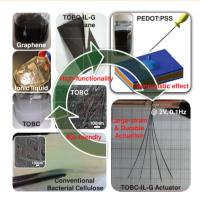


Strong, flexible fibers are wet-spun from a liquid crystalline dispersion of graphene oxide (LCGO), then coated with parylene-C, and laser-excised to create freestanding stimulation electrodes with high charge injection capacity (14 mC cm<sup>-2</sup>). LCGO electrodes stimulate retina in vitro: water-soluble microneedles are utilized to implant the flexible electrodes into cortical tissue enabling acquisition of neural activity.

#### Artificial Muscles

S.-S. Kim, J.-H. Jeon, H.-I. Kim, C. D. Kee, I.-K. Oh\*......3560-6570

**High-Fidelity Bioelectronic Muscular** Actuator Based on Graphene-Mediated and TEMPO-Oxidized Bacterial Cellulose



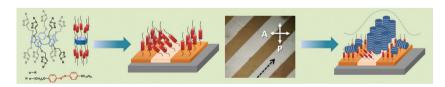
high-fidelity bioelectronic muscular actuator is described that is based 2,2,6,6-tetramethylpiperidine-1-oxyl radical-oxidized bacterial cellulose (TOBC), chemically modified graphene, ionic liquid, and poly(3,4-ethylenedioxythiophene) polystyrene sulfonate. The TOBC-IL-G muscular actuator has biodegradable and biofriendly functionalities and shows exceptionally large static deformation without apparent backrelaxation, much faster response time, and highly durable harmonic actuation compared with conventional biopolymer actuators.

#### **Stimuli-Responsive Materials**

S. Pan, M. F. Ni, B. Mu, Q. Li, X.-Y. Hu, C. Lin, D. Z. Chen,\* L. Y. Wang\*.....3571-3580

Well-Defined Pillararene-Based Azobenzene Liquid Crystalline Photoresponsive Materials and Their Thin Films with Photomodulated Surfaces

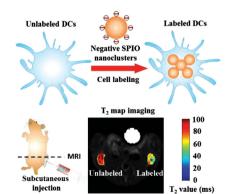
Fascinating photoresponsive thin films are well prepared from the first synthesized pillararene-based macrocyclic azobenzene liquid crystals, which demonstrate rapid reversible photoisomerization and on-off switching property, with further excellent light-triggered modulation of surface free energy, wettability, and even photoalignment-mediated orientation of discotic liquid crystal columnar mesophase. Such welldefined photoresponsive materials provide a promising platform for constructing various novel photoresponsive systems.



#### **Cell Tracking**

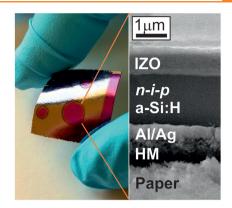
C. Wu, Y. Xu, L. Yang, J. Wu, W. Zhu, D. Li, Z. Cheng, C. Xia, Y. Guo, Q. Gong, B. Song, H. Ai\*......3581-3591

**Negatively Charged Magnetite** Nanoparticle Clusters as Efficient MRI Probes for Dendritic Cell Labeling and In Vivo Tracking



A negatively charged superparamagnetic iron oxide (SPIO) nanocluster is developed. As magnetic resonance imaging (MRI) probe, it has high  $T_2$  relaxivity (335.6 Fe  $mm^{-1}$  s<sup>-1</sup>) and good dendritic cell (DC) labeling efficiency. Labeled DCs with this probe are unaffected in their viability, proliferation, and differentiation capacity. The migration of DCs in vivo can easily be monitored via clinical magnetic resonance imaging scan.

Inkjet printing paper with a cast-coated hydrophilic mesoporous material is proven to be highly suitable to process n-i-p amorphous silicon hydrogenated photovoltaic cells with efficiencies above 3%, opening a new era of energy harvesting, including its seamless integration into ubiquitous formats for a plethora of low-cost flexible and disposable products.

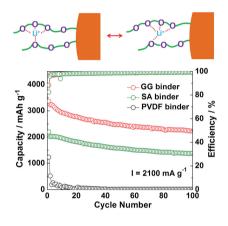


#### **Photovoltaics**

H. Águas,\* T. Mateus, A. Vicente, D. Gaspar, M. J. Mendes, W. A. Schmidt, L. Pereira, E. Fortunato, R. Martins\* ......3592-3598

Thin Film Silicon Photovoltaic Cells on Paper for Flexible Indoor Applications

Guar gum is used as a robust binder for a silicon nanoparticle anode of a lithium-ion battery for the first time. With a large number of polar hydroxyl groups, the guar gum binder can provide effective transport pathways for lithium ions, which significantly improves the electrochemical performance.

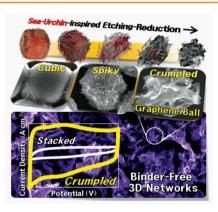


#### **Binders**

J. Liu, Q. Zhang, T. Zhang, J.-T. Li,\* L. Huang, S.-G. Sun\*.....3599-3605

A Robust Ion-Conductive Biopolymer as a Binder for Si Anodes of Lithium-Ion **Batteries** 

Highly crumpled graphene balls with a sea-urchin-shaped template strategy are facilely fabricated in bulk quantities with high yield through simultaneous etching-reduction of graphene oxide-encapsulated iron oxide. The crumpled balls, which exhibit significantly larger surface area and higher water-dispersion stability than those of stacked graphene and other crumpling approaches, and their 3D macroporous networks present superior gravimetric and volumetric capacitance of electrochemical energystorage electrodes.



#### **Supercapacitors**

J. Y. Lee, K.-H. Lee, Y. J. Kim, J. S. Ha, S.-S. Lee,\* J. G. Son\*.....3606-3614

Sea-Urchin-Inspired 3D Crumpled **Graphene Balls Using Simultaneous** Etching and Reduction Process for High-**Density Capacitive Energy Storage**