# ADVANCED FUNCTIONAL MATERIALS

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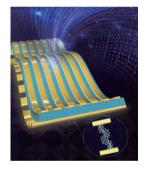


#### **Cancer Nanomedicine**

The front cover illustrates nanoparticle-assisted delivery of siRNA to tumor cells upon systemic administration. After circulating in blood, siRNA-nanoparticles accumulate into the tumor via leaky vasculature while cellular uptake is facilitated by specific interaction between cancer cell receptors and antibodies on nanoparticles. On page 2646, J. W. Gray, W. Yantasee, and colleagues describe the design and synthesis of this mesoporous silica nanoparticle and its application for targeted delivery of siRNA to HER2-positive breast cancer.

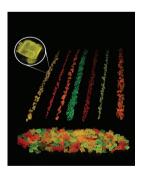
#### Nonvolatile Memory

HKUST-1, a soft metal-organic framework material, is employed by G. Liu, R.-W. Li, and co-workers on page 2677 to develop flexible information storage elements. Uniform and reproducible resistive switching properties are observed in a Au/HKUST-1/Au/PET structure over the wide temperature range of –70 °C to +70 °C, and can be sustained under strain of as high as 2.8%, thus demonstrating great potential for the construction of wearable electronics.



## Semiconductor Nanocrystals

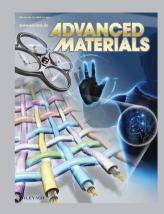
Mixed crystals composed of semiconductor quantum dots are demonstrated as emissive centers and NaCl as host matrix. These composites are prepared using the liquid–liquid diffusion assisted crystallization (LLDC) approach described by N. Gaponik and team on page 2638. These mixed crystals are robust and strong emitters suitable for color conversion applications.



## **Light Extraction**

On page 2660, J. X. Tang and co-workers demonstrate a powerful method for drastically releasing the trapped energy flow in conventional white organic light-emitting diodes by implementing a unique quasi-periodic subwavelength nanofunnel array via soft nanoimprinting lithography. Besides the substantial increase in efficiency over a broad bandwidth without any spectral distortion or angular dependence, this array affords an enticing capacity against scrubbing and the self-cleaning feature, which are critical to the commercial viability in practical applications.





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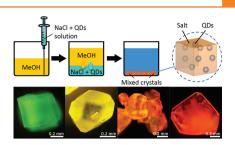


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## The fabrication of robust and processable light emitting composites consisting of semiconductor quantum dots embedded into crystals of ionic salts is demonstrated. The quantum dots from the organic phase may be directly embedded by using the approach in this study. The method is fast and results in crystal powders directly applicable for color conversion and other applications demanding extremely photostable luminescent solids.



## **FULL PAPERS**

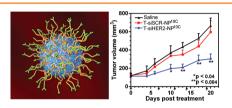
## Semiconductor Nanocrystals

M. Adam, Z. Wang, A. Dubavik, G. M. Stachowski, C. Meerbach, Z. Soran-Erdem, C. Rengers. H. V. Demir, N. Gaponik,\* A. Eychmüller ......2638-2645

Liquid-Liquid Diffusion-Assisted Crystallization: A Fast and Versatile Approach Toward High Quality Mixed Quantum Dot-Salt Crystals



A new nanoparticle platform is engineered for siRNA delivery to tumors and displays high potential for clinical translation. Comprised of a mesoporous silica nanoparticle coated with crosslinked polyethyleneimine-polyethyleneglycol copolymer, carrying siRNA against human epidermal growth factor receptor 2 (HER2), and coupled to therapeutic targeting antibody, it inhibits growth of multidrug-resistant breast tumors in mice upon intravenous administration.

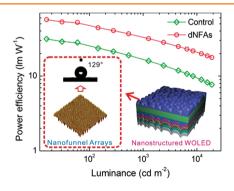


#### Cancer Nanomedicine

W. Ngamcherdtrakul, J. Morry, S. Gu, D. J. Castro, S. M. Goodyear, T. Sangvanich, M. M. Reda, R. Lee, S. A. Mihelic, B. L. Beckman, Z. Hu, J. W. Gray,\* W. Yantasee\* ......2646-2659

**Cationic Polymer Modified Mesoporous** Silica Nanoparticles for Targeted siRNA Delivery to HER2+ Breast Cancer

A powerful method for drastically releasing the trapped energy flow in white organic light-emitting diodes is demonstrated by nanoimprinting multifunctional quasi-periodic nanofunnel arrays. The broadband light extraction is realized without spectral changes and angular dependence, yielding an external quantum efficiency 2.3 times that of the conventional device and improved power efficiency. This extraction nanostructure also affords an enticing capacity against scrubbing and the selfcleaning feature.

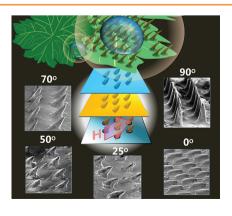


#### **Light Extraction**

L. Zhou, Q.-D. Ou, Y.-Q. Li, H.-Y. Xiang, L.-H. Xu, J.-D. Chen, C. Li, S. Shen, S.-T. Lee, J.-X. Tang\* ......2660-2668

Efficiently Releasing the Trapped Energy Flow in White Organic Light-Emitting **Diodes with Multifunctional Nanofunnel** 

Biomimetic polydimethylsiloxane microcones resembling trichomes of plants are generated by a ferrofluid-molding method to understand the influence of geometry on wettability. First, ferrofluid droplets are arranged and their shape can be changed from right cones into oblique cones by tilting the external magnetic field. Followed by hard molds made with UV-curable tri(propylene glycol) diacrylate, polydimethylsiloxane microcones with different inclination angle are subsequently generated.



## **Biomimetic Surfaces**

C.-Y. Huang, M.-F. Lai, W.-L. Liu, Z.-H. Wei\*.....2670-2676

**Anisotropic Wettability of Biomimetic** Micro/Nano Dual-Scale Inclined Cones Fabricated by Ferrofluid-Molding Method

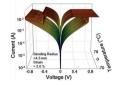
## **FULL PAPERS**

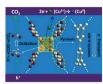
#### **Nonvolatile Memory**

L. Pan, Z. Ji, X. Yi, X. Zhu, X. Chen, J. Shang, G. Liu,\* R.-W. Li\*... 2677-2685

Metal-Organic Framework Nanofilm for Mechanically Flexible Information **Storage Applications** 

With well-defined 3D organic-inorganic frameworks, a HKUST-1 nanofilm is fabricated on flexible substrate, which exhibit uniform and reproducible resistive switching effect under the strain of as high as 2.8%, and over the wide temperature range of -70 to +70 °C.



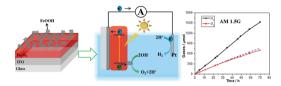


#### Hematite

Q. Yu, X. Meng, T. Wang, P. Li, J. Ye\*......2686-2692

Hematite Films Decorated with Nanostructured Ferric Oxyhydroxide as Photoanodes for Efficient and Stable **Photoelectrochemical Water Splitting** 

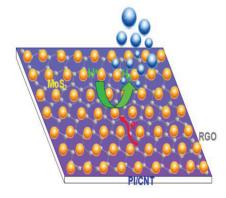
Fe<sub>2</sub>O<sub>3</sub> is decorated with nanostructured FeOOH for the first time. After the decoration of FeOOH, the photoelectrochemical performance of Fe<sub>2</sub>O<sub>3</sub> is greatly enhanced. The Fe<sub>2</sub>O<sub>3</sub>/FeOOH realizes visible-light response, high water oxidation rate, and long-term durability, and shows great potentials as an all-Fe-based photoanode for photoelectrochemical water oxidation.



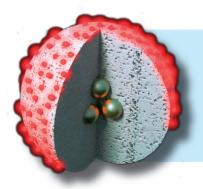
#### **Electrocatalysis**

Y. M. Jiang, X. Li, S. J. Yu, L. P. Jia, X. J. Zhao, C. M. Wang\*......2693-2700

Reduced Graphene Oxide-Modified Carbon Nanotube/Polyimide Film Supported MoS<sub>2</sub> Nanoparticles for **Electrocatalytic Hydrogen Evolution** 



A reduced graphene oxide-modified carbon nanotube/polyimide-MoS<sub>2</sub> film with a small loading of MoS2 exhibits a low overpotential and achieves large current densities at low overpotentials with the synergic effect of MoS2 and reduced graphene oxide-modified carbon nanotube/polyimide film. The excellent hydrogen evolution reaction electrocatalytic activity and good stability suggest the potential application of reduced graphene oxide-modified carbon nanotube/polyimide-MoS<sub>2</sub> film in energy conversion.



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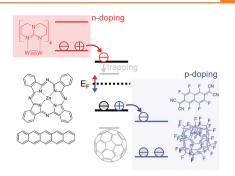
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## **FULL PAPERS**

Fermi level control over the whole electronic gap of an organic semiconductor is demonstrated by applying the concept of molecular p- and n-type doping. In particular, the observation of the dopant saturation and reserve regimes is shown for the first time in organic semiconductors, substantiating the cross-link to classical semiconductor theory. At ultralow concentrations, doping provides filling of trap states.

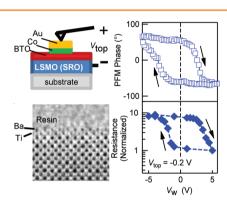


#### **Doping Efficiency**

M. L. Tietze,\* P. Pahner, K. Schmidt, K. Leo.\* B. Lüssem......2701-2707

**Doped Organic Semiconductors:** Trap-Filling, Impurity Saturation, and Reserve Regimes

Surface-termination engineering for a ferroelectric tunnel barrier provides an effective way toward controlling electroresistance of ferroelectric tunnel junctions (FTJs). The fraction of BaOtermination area on the BaTiO<sub>3</sub> surface can be artificially controlled by a combination of epitaxial growth and ex situ surface treatment. The FTIs consisting of BaTiO<sub>3</sub> with dominant BaO-termination exhibit an enhanced resistanceswitching ratio up to 100 000%.

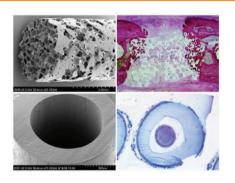


#### **Ferroelectrics**

- H. Yamada, \* A. Tsurumaki-Fukuchi, M. Kobayashi, T. Nagai, Y. Toyosaki,
- H. Kumigashira, A. Sawa......2708-2714

Strong Surface-Termination Effect on **Electroresistance in Ferroelectric Tunnel lunctions** 

By varying the copolymer composition, thermal and mechanical properties, photocrosslinked poly(propylene fumarate)-co-poly( $\varepsilon$ -caprolactone) networks are efficiently controlled from hard and stiff materials to soft and flexible ones, rendering desirable biomaterials for promoting behavior and functions of bone and neuronal cells, and fabricating scaffolds for bone and peripheral nerve regeneration, respectively.

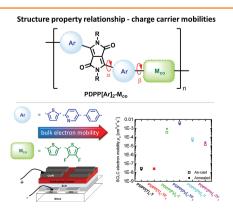


#### Tissue Engineering

S. Wang,\* D. H. R. Kempen, G. C. W. de Ruiter, L. Cai, R. J. Spinner, A. J. Windebank, M. J. Yaszemski, L. Lu\*.....2715-2724

Molecularly Engineered Biodegradable Polymer Networks with a Wide Range of Stiffness for Bone and Peripheral Nerve Regeneration

An excellent bulk electron mobility of  $4.3 \times 10^{-3} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$  is obtained for diketopyrrolo[3,4-c]pyrrole copolymers by systematically tuning the diffusive nonbonding heteroatom interactions and dihedral angles between the aryl flanking units, DPP core and comonomer. Differences in crystalline packing, absorption, energy levels, and charge carrier properties are comparatively studied in a series of copolymers.



#### **Electron Transport**

C. J. Mueller, C. R. Singh, M. Fried, S. Huettner, M. Thelakkat\*....2725-2736

**High Bulk Electron Mobility** Diketopyrrolopyrrole Copolymers with Perfluorothiophene

## **FULL PAPERS**

#### Stem Cells

B. Delalat, A. Mierczynska,

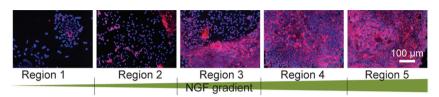
S. Rasi Ghaemi, A. Cavallaro.

F. J. Harding, K. Vasilev,

N. H. Voelcker\*......2737-2744

Materials Displaying Neural Growth Factor Gradients and Applications in Neural Differentiation of Embryoid **Body Cells** 

The critical density of immobilized nerve growth factor required for the neural differentiation of mouse embryonic stem cells is identified using a surface density gradient. This approach allows the optimization of biomaterials to support cell expansion and differentiation, and is highly relevant to efficient cell therapy manufacture.



## **Energy Storage**

J. Zhao, S. Xu, K. Tschulik, R. G. Compton, M. Wei,\* D. O'Hare,\* D. G. Evans, X. Duan ...... 2745-2753

Molecular-Scale Hybridization of Clay Monolayers and Conducting Polymer for Thin-Film Supercapacitors

A molecular-scale superlattice material based on alternating 2D layers of clay monolayers and a conducting polymer is fabricated, which exhibits excellent electrochemical behavior in thin-film supercapacitor devices.



#### **Photoluminescence**

Y. Zuo, J. Cao, S. Feng\*......2754-2762

**Sunlight-Induced Cross-Linked** Luminescent Films Based on Polysiloxanes and D-Limonene via Thiol-ene "Click" Chemistry



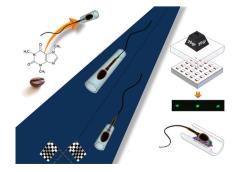
The first example of sunlight-induced cross-linked luminescent films based on polysiloxanes and p-limonene shows potential applications in fabricating colorful UV-light emitting diodes. This study demonstrates how an appropriate synthesis strategy can be applied to terpene monomers while allowing the development of new environmentally benign materials.

#### Microbiorobots

V. Magdanz,\* M. Medina-Sánchez, Y. Chen, M. Guix,

O. G. Schmidt ......2763-2770

**How to Improve Spermbot Performance** 



Improved spermbot performance is demonstrated by biofunctionalization of the inner tube surface, shorter tube design, and caffeine addition. Firstly, spermbot velocity is improved by the use of shorter microtubes; secondly, better coupling efficiency is achieved by binding of fibronectin inside the microtube; and finally, caffeine addition gives a temporary speed boost to the spermbot.

## **FULL PAPER**

Biofunctionalized defect-free few-layer graphene microsheets can be obtained using liquid phase ultrasonic exfoliation of raw graphitic material assisted by the self-assembling fungal hydrophobin Vmh2. This approach enables a highly concentrated and stable exfoliated product. The obtained material is likely to prove valuable for the emerging applications of graphene in the biotechnological field.



#### **Graphene Production**

A. M. Gravagnuolo, E. Morales-Narváez, S. Longobardi, E. T. da Silva, P. Giardina, A. Merkoci\* ......2771–2779

In Situ Production of Biofunctionalized Few-Layer Defect-Free Microsheets of Graphene

