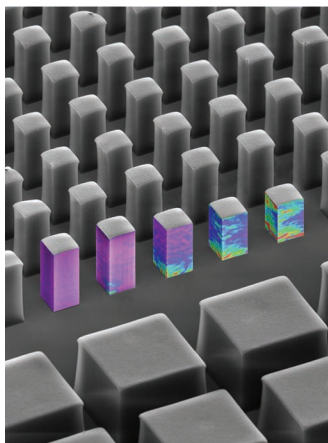


ADVANCED FUNCTIONAL MATERIALS

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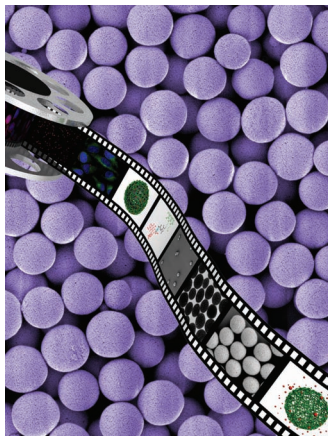
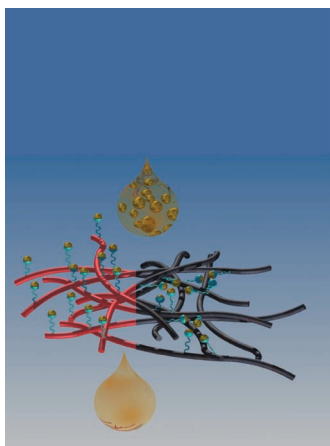


Digital Image Correlation

Digital image correlation is used for the first time to measure the spatially resolved strain evolution of compressed carbon nanotube columns. A consistent local critical strain criterion of 5% is discovered, unifying otherwise disparate column behavior. As reported by Jeffery W. Baur and co-workers on page 4686, this new capability enables the visualization of the inhomogeneous deformation inherent in many nanoscale and microscale materials.

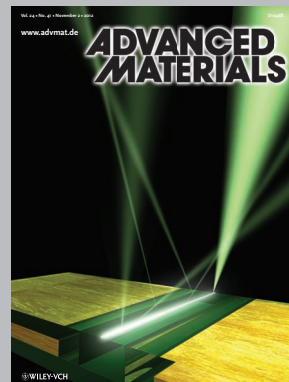
Bionanocomposites

On page 4771, Mady Elbahri and co-workers report on the fabrication of a smart, nanofibrous, plasmonic bio-nanocomposite developed for the first time by bridging the gaps between nanofluid filtration, plasmonics, and bioswitching. The primary bio(protein)-functionalized nanofibrous membrane catches metal nanoparticles through switchable conformational changes of the protein, despite its macroporosity. The metal nanoparticle/biofunctionalized nanofibrous mat can subsequently act as an omnidirectional black absorber.



Drug Delivery

Polymer carriers with finely controlled and responsive properties have received increasing interest for biomedical applications. On page 4718, Frank Caruso and co-workers report a facile and robust approach to prepare pH-labile drug-loaded polymer particles by combining mesoporous silica-templated assembly and polymer-drug conjugates for drug delivery into cancer cells. These particles are a novel and versatile class of stimuli-responsive carriers for controlled drug delivery.



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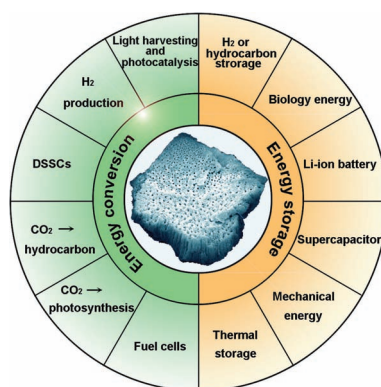
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FEATURE ARTICLE

Porous Materials

Y. Li,* Z.-Y. Fu,
B.-L. Su*4634–4667

Hierarchically Structured Porous Materials for Energy Conversion and Storage



Hierarchically structured porous materials have been heavily involved in newly developed energy storage and conversion systems because of their high surface area, tunable chemical compositions, excellent mechanical and chemical resistance, controllable micro-, meso-, and macroporosities at different length scales, desired multifunctionalities, reduced diffusion effect, light harvesting properties, and confinement effects.

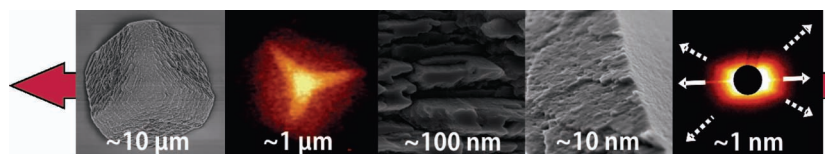
FULL PAPERS

Biomimetics

A. S. Schenk, I. Zlotnikov, B. Pokroy,
N. Gierlinger, A. Masic, P. Zaslansky,
A. N. Fitch, O. Paris, T. H. Metzger,
H. Cölfen, P. Fratzl,*
B. Aichmayer*4668–4676

Hierarchical Calcite Crystals with Occlusions of a Simple Polyelectrolyte Mimic Complex Biomineral Structures

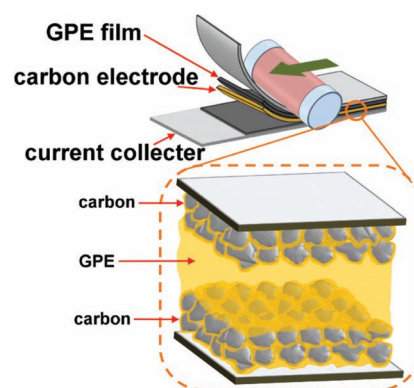
Poly(sodium 4-styrenesulfonate) induces hierarchical structuring from the micrometer down to the nanometer length-scale in calcite, similar to biominerals. The polyelectrolyte molecules are occluded within the mineral crystals, thus generating rough, (104)- and (001)-oriented organic/inorganic interfaces, which prevent brittle cleavage. The hardness and stiffness of the hierarchically structured hybrid crystals depend on the polymer content, which varies in different parts of a single crystal.



Capacitors

C.-W. Huang, C.-A. Wu, S.-S. Hou,
P.-L. Kuo, C.-T. Hsieh,
H. Teng* 4677–4685

Gel Electrolyte Derived from Poly(ethylene glycol) Blending Poly(acrylonitrile) Applicable to Roll-to-Roll Assembly of Electric Double Layer Capacitors

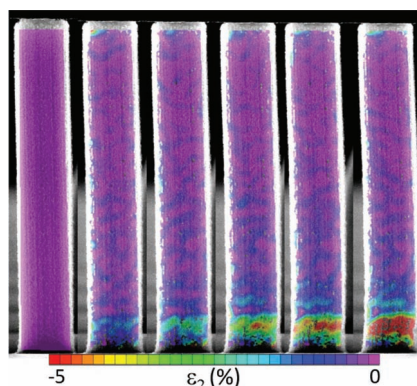


A gel electrolyte derived from swelling a copolymer of poly(ethylene glycol) blending poly(acrylonitrile) with LiClO₄/dimethyl formamide exhibits high ionic conductivity and excellent compatibility with carbon-based supercapacitors. The copolymer substantially reduces resistance to ionic motion across the partition zone and through the carbon/electrolyte interface. The adjustable mechanical integrity of this gel-electrolyte film makes roll-to-roll assembly of supercapacitors scalable to industrial levels.

Carbon Nanotubes

M. R. Maschmann, G. J. Ehlert,
S. J. Park, D. Mollenhauer,
B. Maruyama, A. J. Hart,
J. W. Baur* 4686–4695

Visualizing Strain Evolution and Coordinated Buckling within CNT Arrays by In Situ Digital Image Correlation

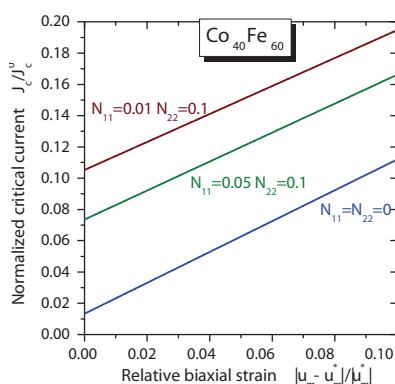


Spatial mapping of deformation fields within CNT array columns is achieved using digital image correlation of in situ scanning electron microscopy (SEM) compression sequences. Strain is highly non-uniform, with buckle formation occurring in localized regions of enhanced strain. A local buckling criterion of 5% principal strain is observed for all columns despite systematic column deformation variation as a function of aspect ratio.



FULL PAPERS

The density of spin-polarized current needed for magnetization switching in spin-transfer torque magnetic random access memories (STT-MRAMs) is predicted to be drastically reduced near spin reorientation transitions driven by lattice strains and/or surface magnetic anisotropy. STT-MRAMs with suitable free layers combine low writing current with very high thermal stability of information storage even at high density.

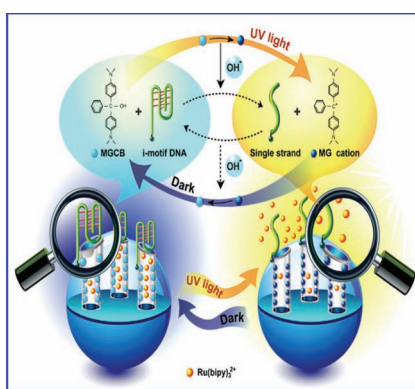


Magnetic Memory

N. A. Pertsev,*
H. Kohlstedt4696–4703

Magnetoresistive Memory with Ultralow Critical Current for Magnetization Switching

A photon-fueled gate-like delivery system is designed by employing malachite green carbinol base (MGCB) as a light-induced hydroxide ion emitter and i-motif DNA as a cap. The photon-fueled MGCB-dependent DNA conformation switch between the i-motif structure and single-strand form the open and closed states of the DNA-gated system, which can be cycled by turning the UV light on and off alternately.

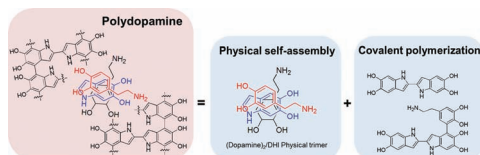


Stimuli-Responsive Materials

D. G. He, X. X. He, K. M. Wang,* J. Cao,
Y. X. Zhao4704–4710

A Photon-Fueled Gate-Like Delivery System Using i-Motif DNA Functionalized Mesoporous Silica Nanoparticles

A physical, self-assembled trimer of (dopamine)₂/5,6-dihydroxyindole (DHI) that exists in polydopamine, which has been known to be formed only by covalent polymerization, is reported. A different perspective of polydopamine formation is revealed where it forms in part by the self-assembly of dopamine and DHI, providing a new clue to understanding the structures of catecholamines such as melanin.

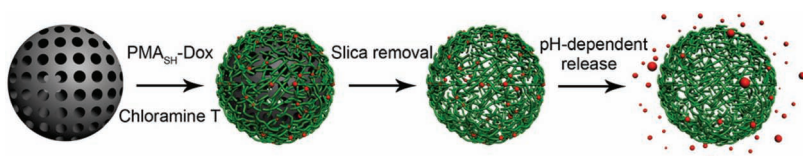


Self-Assembly

S. Hong, Y. S. Na, S. Choi, I. T. Song,
W. Y. Kim,* H. Lee*4711–4717

Non-Covalent Self-Assembly and Covalent Polymerization Co-Contribute to Polydopamine Formation

Polymer-drug particles, prepared using a facile mesoporous silica templating method, are stable at physiological conditions and allow the triggered release of an encapsulated anticancer drug at endosomal/lysosomal pH. Cell viability assays show that the particles result in significant cancer cell death after internalization. The reported particles represent a novel and versatile class of stimuli-responsive carriers for controlled drug delivery.



Drug Delivery

J. Cui, Y. Yan, Y. Wang,
F. Caruso*4718–4723

Templated Assembly of pH-Labile Polymer-Drug Particles for Intracellular Drug Delivery

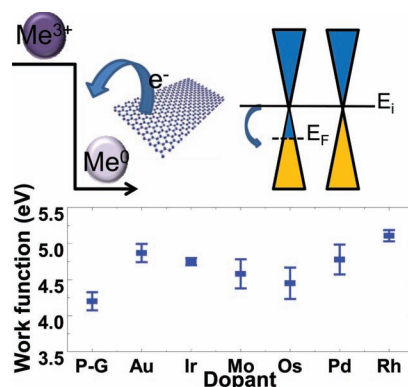


FULL PAPERS

Graphene

K. C. Kwon, K. S. Choi,
S. Y. Kim*4724–4731

Increased Work Function in Few-Layer Graphene Sheets via Metal Chloride Doping

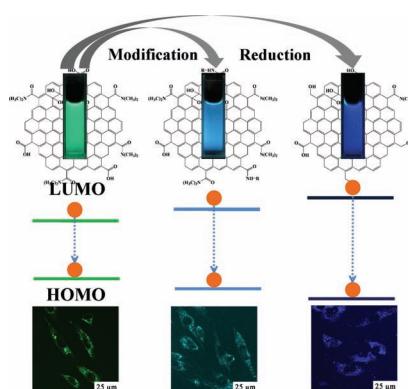


A chemical approach to controlling the work function of graphene is investigated. The work function of graphene increases from 4.2 eV to 5.0, 4.9, 4.8, 4.68, 5.0, and 5.14 eV for the graphene with 20 nm AuCl₃, IrCl₃, MoCl₃, OsCl₃, PdCl₂, and RhCl₃, respectively. Spontaneous charge transfer occurs from graphene to the metal ions, increasing the work function.

Bioimaging

S. Zhu, J. Zhang, S. Tang, C. Qiao,
L. Wang, H. Wang, X. Liu, B. Li, Y. Li,
W. Yu, X. Wang, H. Sun,
B. Yang*4732–4740

Surface Chemistry Routes to Modulate the Photoluminescence of Graphene Quantum Dots: From Fluorescence Mechanism to Up-Conversion Bioimaging Applications

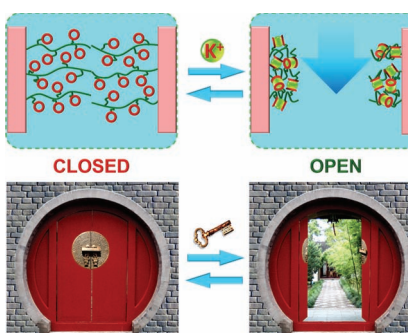


The preparation of controllable fluorescent graphene quantum dots (GQDs) using a new surface chemistry tuning method is reported. The photoluminescence (PL) mechanism is investigated and the competition between both the defect state emission and intrinsic state emission are analyzed in detail. Moreover, the up-conversion PL of GQDs is successfully used in multiphoton luminescent biolabeling under near-IR excitation.

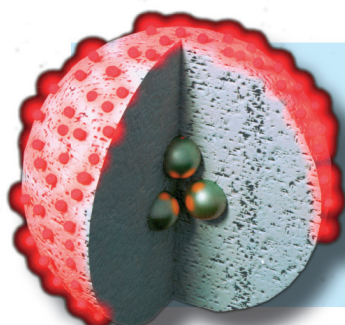
Stimuli-Responsive Materials

Z. Liu, F. Luo, X.-J. Ju,* R. Xie, T. Luo,
Y.-M. Sun, L.-Y. Chu*4742–4750

Positively K⁺-Responsive Membranes with Functional Gates Driven by Host-Guest Molecular Recognition



A unique positively K⁺-responsive membrane is developed to spontaneously “open” the gates in response to the presence of K⁺ ions in the environment. The “opening” of the membrane gates is driven by a cooperative interaction of crown ether-based host-guest complexation and poly(*N*-isopropylacrylamide)-based phase transition and exhibits a satisfactorily rapid, reversible, and reproducible property. The proposed membrane is highly attractive for controlled release, chemical/biomedical separations, tissue engineering, and sensors.



How to contact us:

Editorial Office:

Phone: (+49) 6201-606-235/531
Fax: (+49) 6201-606-500
Email: afm@wiley-vch.de

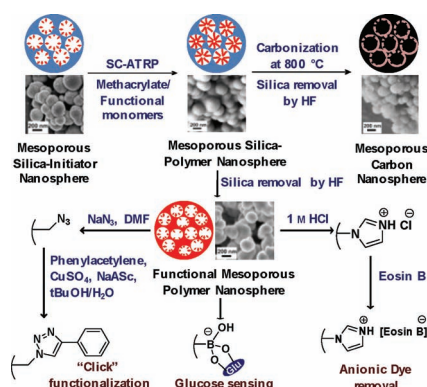
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The synthesis of mesoporous polymer and carbon nanospheres with complete shape retention is demonstrated. The technique allows fabrication of functional mesoporous nanospheres using functional comonomers such as 4-vinylbenzyl chloride, 1-vinylimidazole, and 4-vinylphenylboronic acid with methyl methacrylate. These functionalized mesoporous materials find potential applications in the removal of ionic contaminants, sensing of glucose, and as “clickable” mesoporous materials.

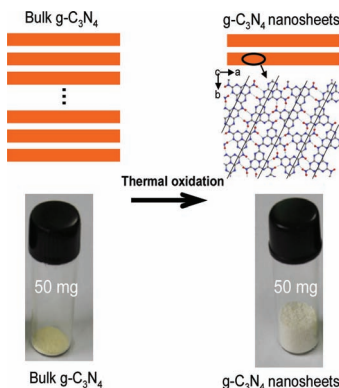


Mesoporous Materials

S. Banerjee, T. K. Paira, A. Kotal, T. K. Mandal*4751–4762

Surface-Confined Atom Transfer Radical Polymerization from Sacrificial Mesoporous Silica Nanospheres for Preparing Mesoporous Polymer/Carbon Nanospheres with Faithful Shape Replication: Functional Mesoporous Materials

Graphene-like C_3N_4 nanosheets produced by direct thermal oxidative etching of bulk “graphitic”- C_3N_4 show superior photocatalytic activities compared to the bulk due to a high specific surface area, a larger bandgap (by 0.2 eV), increased lifetimes of photoexcited charge carriers, and improved electron transport ability.

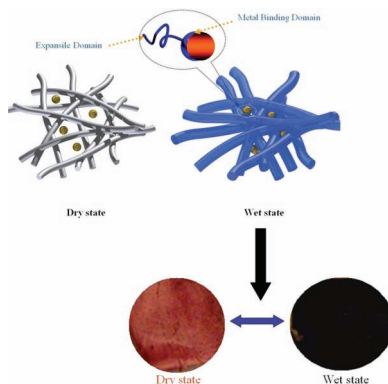


Photocatalytic Nanosheets

P. Niu, L. L. Zhang, G. Liu,* H.-M. Cheng4763–4770

Graphene-Like Carbon Nitride Nanosheets for Improved Photocatalytic Activities

A smart plasmonic absorber is developed based on a metal–polymer bionanocomposite. A novel fabrication approach is adopted for filtration of a metal nanoparticle containing aqueous suspension by a bio(protein)-functionalized nanofibrous membrane. Desirably, nearly all the metal nanoparticles are captured by the protein ligands immobilized onto the nanofibers, creating a metal–polymer bionanocomposite. Conformational changes of the protein upon wetting leads to performance as an omnidirectional black absorber.

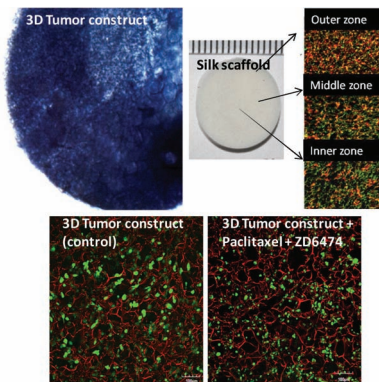


Nanocomposites

M. Elbahri,* Sh. Homaeigohar, R. Abdelaziz, T. Dai, R. Khalil, A. U. Zillohu4771–4777

Smart Metal–Polymer Bionanocomposites as Omnidirectional Plasmonic Black Absorber Formed by Nanofluid Filtration

Cancer cells form a 3D tumor mass, closely contacting and interacting with one another, and are poorly supplied with nutrients and oxygen. This complex tumor microenvironment plays an important role in determining resistance to drugs. The figure shows that the *Antheraea mylitta* silk fibroin scaffold based 3D in vitro tumor model may represent an avascular tumor and help in evaluating anticancer drug efficacy.



Biomedical Applications

S. Talukdar, S. C. Kundu*4778–4788

A Non-Mulberry Silk Fibroin Protein Based 3D In Vitro Tumor Model for Evaluation of Anticancer Drug Activity

FULL PAPERS

Flexible Electronics

G. C. Pidcock,
M. in het Panhuis*.....4790–4800

Extrusion Printing of Flexible Electrically Conducting Carbon Nanotube Networks

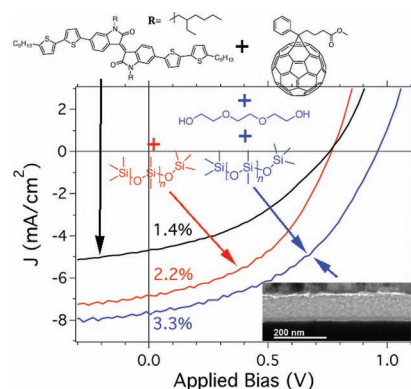


Extrusion printing is used for the controlled deposition of carbon nanotube networks using the biopolymer gellan gum. The biopolymer is an excellent dispersant for carbon nanotubes and thickener for the inks. The actuating and strain gauging ability of printed flexible electrically conducting networks is demonstrated.

Photovoltaic Devices

K. R. Graham, P. M. Wieruszewski,
R. Stalder, M. J. Hartel, J. Mei, F. So,
J. R. Reynolds*.....4801–4813

Improved Performance of Molecular Bulk-Heterojunction Photovoltaic Cells through Predictable Selection of Solvent Additives

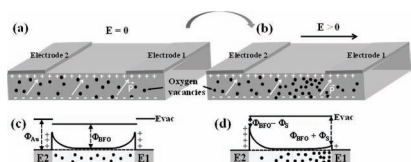


A family of solvent additives is selected based on Hansen solubility parameters and applied to molecular bulk-heterojunction photovoltaic cells. The power conversion efficiency (PCE) and morphology are shown to correlate strongly with the solubility of the donor and acceptor in the solvent additive. By combining solvent additives with complementary effects, the PCE is increased from 1.4% to 3.3%.

Ferroelectric Materials

R. Moubah,* O. Rousseau, D. Colson,
A. Artemenko, M. Maglione,
M. Viret.....4814–4818

Photoelectric Effects in Single Domain BiFeO₃ Crystals

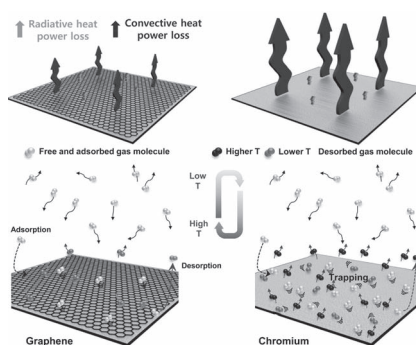


The mechanism leading to the creation of the diodelike structure in BiFeO₃ single crystals is shown. In the virgin state, the distribution of oxygen vacancies is homogenous. Under illumination with an applied electric field, positively charged oxygen vacancies are created and they electromigrate, following the field direction, towards the electrode interface.

Graphene

J. J. Bae, S. C. Lim, G. H. Han, Y. W. Jo,
D. L. Doung, E. S. Kim, S. J. Chae,
T. Q. Huy, N. V. Luan,
Y. H. Lee*.....4819–4826

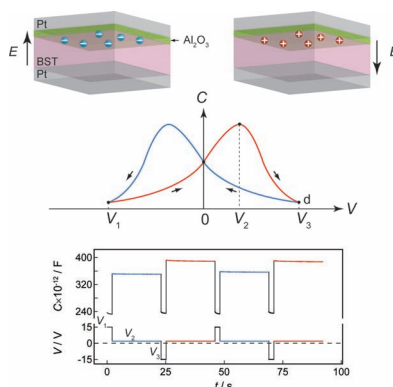
Heat Dissipation of Transparent Graphene Defoggers



A graphene/glass defogger is fabricated and the dynamic response of the temperature as a function of input electrical power is measured. The response time of the graphene/glass defogger is shorter by 44% than that of a Cr/glass defogger. The convective heat-transfer coefficient of graphene is $12.4 \times 10^{-4} \text{ W cm}^{-2} \text{ }^{\circ}\text{C}^{-1}$, similar to that of glass ($11.1 \times 10^{-4} \text{ W cm}^{-2} \text{ }^{\circ}\text{C}^{-1}$) but smaller by 27% than that of chromium ($17.1 \times 10^{-4} \text{ W cm}^{-2} \text{ }^{\circ}\text{C}^{-1}$).

FULL PAPERS

Capacitance hysteresis is introduced in $(\text{Ba,Sr})\text{TiO}_3$ tunable capacitors by insertion of an Al_2O_3 layer. This bilayer capacitor can work as a charge storage device and shows non-volatile programmability at high frequency.

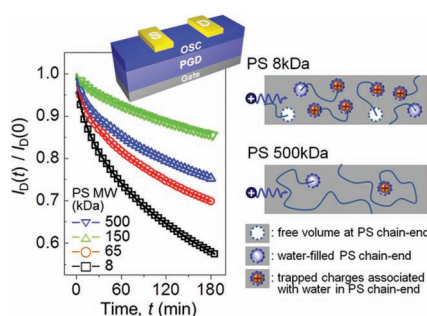


Capacitors

S. Li,* Y. Zheng, R. Jakoby,
A. Klein4827–4832

Electrically Programmable Bistable Capacitor for High-Frequency Applications Based on Charge Storage at the $(\text{Ba,Sr})\text{TiO}_3/\text{Al}_2\text{O}_3$ Interface

The **polymer chain ends of polymer gate-dielectrics (PGDs)** can trap charges in organic field-effect transistors (OFETs). Under bias stress, the drain current decay is found to increase as the molecular weight (MW) of the PGD decreases (MW effect). The free volumes at polymer chain ends are sufficiently large to allow the residence of water molecules, the presence of which significantly increases the density of charge-trap sites.



Organic Transistors

H. H. Choi, W. H. Lee,
K. Cho*4833–4839

Bias-Stress-Induced Charge Trapping at Polymer Chain Ends of Polymer Gate-Dielectrics in Organic Transistors