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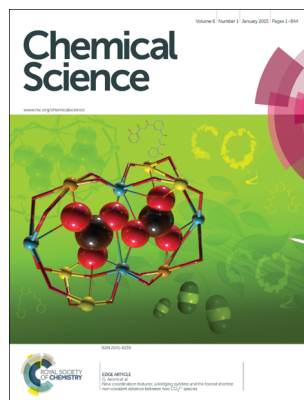
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ISSN 2041-6539 CODEN CSHCBM 6(1) 1–844 (2015)



Cover

See Yong-Gui Gao,
Esther C. Y. Woon *et al.*,
pp. 112–122.
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Sunny Angrom, *Chem. Sci.*,
2015, 6, 112.



Inside cover

See G. Aromí *et al.*,
pp. 123–131.
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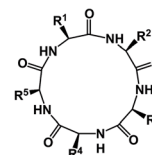
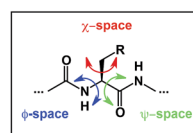
PERSPECTIVE

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Macrocycles: lessons from the distant past, recent developments, and future directions

Andrei K. Yudin*

A noticeable increase in molecular complexity of drug targets has created an unmet need in the therapeutic agents that are larger than traditional small molecules.



ϕ -space possible to control
 ψ -space
 χ -space difficult to control

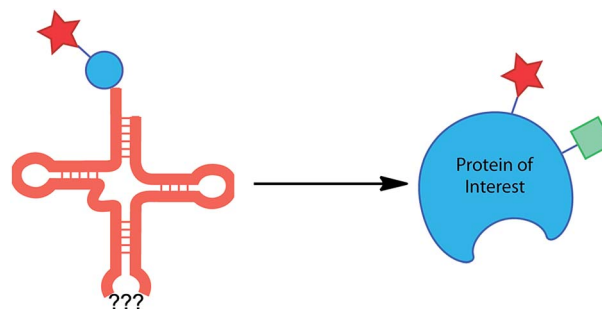
MINIREVIEWS

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Designing logical codon reassignment – Expanding the chemistry in biology

Anaëlle Dumas, Lukas Lercher, Christopher D. Spicer and Benjamin G. Davis*

This review rationalizes the varied designs of systems for incorporation of UAAs into proteins *via* canonical codons.



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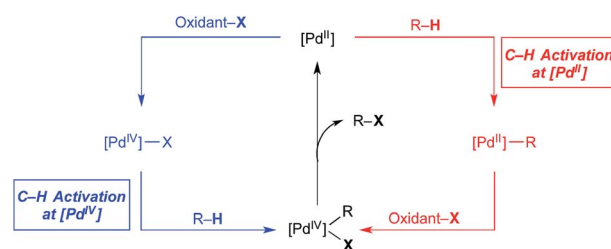
MINIREVIEWS

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Carbon–hydrogen (C–H) bond activation at Pd^{IV}: a Frontier in C–H functionalization catalysis

Joseph J. Topczewski and Melanie S. Sanford*

The direct functionalization of carbon–hydrogen (C–H) bonds has emerged as a versatile strategy for the synthesis and derivatization of organic molecules.

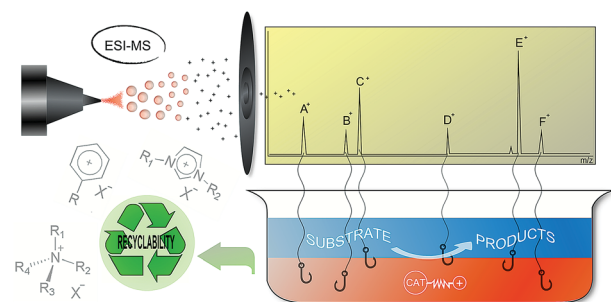


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Charge-tagged ligands: useful tools for immobilising complexes and detecting reaction species during catalysis

Jones Limberger, Bárbara C. Leal, Adriano L. Monteiro and Jairton Dupont*

A critical overview is presented on the use of charged tagged ligands (CTLs) as immobilising agents in organometallic catalysis and as probes for studying mechanisms through electrospray ionisation mass spectrometry (ESI-MS) based on the most recent literature.

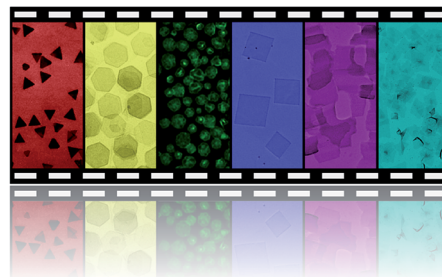


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Thin metal nanostructures: synthesis, properties and applications

Zhanxi Fan, Xiao Huang, Chaoliang Tan and Hua Zhang*

This minireview introduces the recent progress in the synthesis, properties and applications of thin metal nanostructures, especially metal nanoplates and nanosheets.



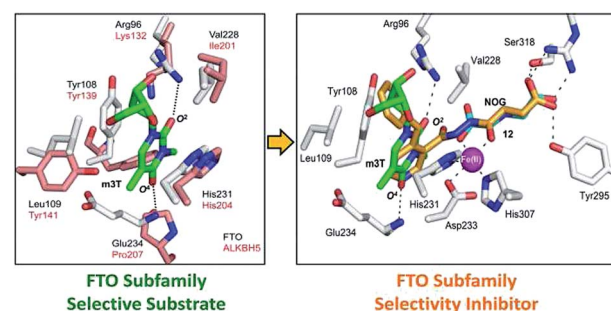
EDGE ARTICLES

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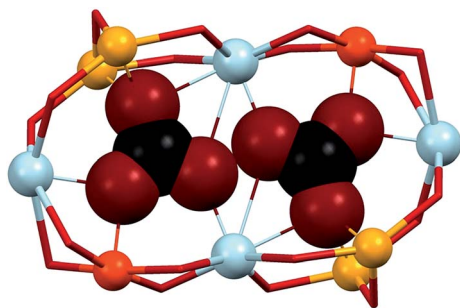
A strategy based on nucleotide specificity leads to a subfamily-selective and cell-active inhibitor of N⁶-methyladenosine demethylase FTO

Joel D. W. Toh, Lingyi Sun, Lisa Z. M. Lau, Jackie Tan, Joanne J. A. Low, Colin W. Q. Tang, Eleanor J. Y. Cheong, Melissa J. H. Tan, Yun Chen, Wanjin Hong, Yong-Gui Gao* and Esther C. Y. Woon*

The AlkB family of nucleic acid demethylases are of intense biological and medical interest. The discovery of a highly selective FTO inhibitor should greatly facilitate the study of these enzymes.



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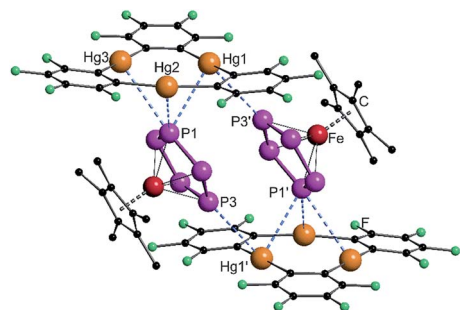


New coordination features; a bridging pyridine and the forced shortest non-covalent distance between two CO₃²⁻ species

V. Velasco, D. Aguilà, L. A. Barrios, I. Borilovic, O. Roubeau, J. Ribas-Ariño, M. Fumanal, S. J. Teat and G. Aromi*

New coordination assemblies furnish a rare crevice pyridine ligand or encapsulation of the closest not bonded CO₃²⁻ species.

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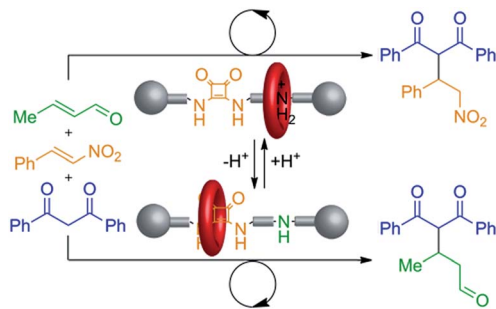


A comparative study of the coordination behavior of *cyclo*-P₅ and *cyclo*-As₅ ligand complexes towards the trinuclear Lewis acid complex (perfluoro-*ortho*-phenylene)mercury

Martin Fleischmann, James S. Jones, François P. Gabbaï and Manfred Scheer*

Reactions of the *cyclo*-E₅ sandwich complexes [Cp*Fe(η⁵-P₅)] (1) and [Cp*Fe(η⁵-As₅)] (2) with the planar Lewis acid [(*o*-C₆F₄Hg)₃] (3) afford compounds that show distinctly different assemblies in the solid state.

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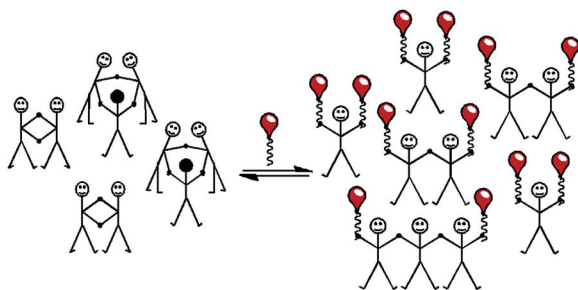


Selecting reactions and reactants using a switchable rotaxane organocatalyst with two different active sites

Jack Beswick, Victor Blanco, Guillaume De Bo, David A. Leigh,* Urszula Lewandowska, Bartosz Lewandowski and Kenji Mishi

The activation mode of a rotaxane-based organocatalyst with both secondary amine and squaramide catalytic units can be switched with acid or base, affording different products from a mixture of three building blocks.

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Applications of dynamic combinatorial chemistry for the determination of effective molarity

Maria Ciaccia, Irene Tosi, Laura Baldini, Roberta Cacciapaglia, Luigi Mandolini, Stefano Di Stefano* and Christopher A. Hunter*

Chain-stoppers give rise to ring-chain equilibria in dynamic libraries allowing determination of thermodynamic effective molarities for macrocyclisation reactions.

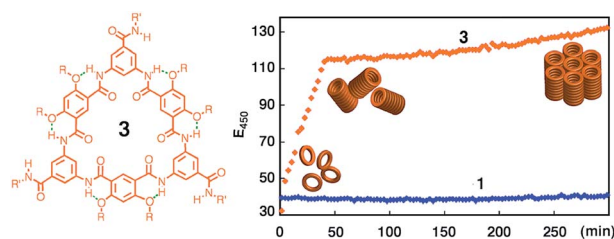


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Extremely strong tubular stacking of aromatic oligoamide macrocycles

Mark A. Kline, Xiaoxi Wei, Ian J. Horner, Rui Liu, Shuang Chen, Si Chen, Ka Yi Yung, Kazuhiro Yamato, Zhonghou Cai, Frank V. Bright, Xiao Cheng Zeng and Bing Gong*

Aromatic oligoamide macrocycles **3** undergo extremely strong stacking in both solution and the solid state, forming tubular assemblies that further aggregate.

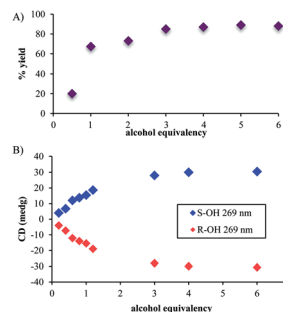


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Mechanistic studies on covalent assemblies of metal-mediated hemi-aminal ethers

Hyun Hwa Jo, Ramakrishna Edupuganti, Lei You, Kevin N. Dalby and Eric V. Anslyn*

The use of reversible covalent bonding in a four-component assembly incorporating chiral alcohols was recently reported to give a method for determining the enantiomeric excess of the alcohols *via* CD spectroscopy.

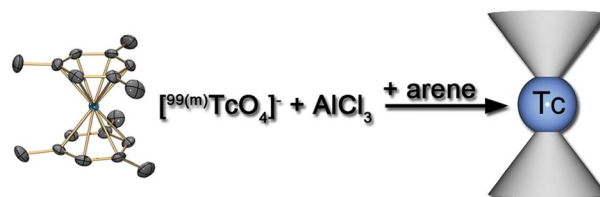


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From Tc^{VII} to Tc^I ; facile syntheses of bis-arene complexes $[^{99(m)}Tc(arene)_2]^+$ from pertechnetate

Michael Benz, Henrik Braband, Paul Schmutz, Jonathan Halter and Roger Alberto*

Bis-arene complexes of technetium represent a fundamental class of organometallic compounds. Detailed insights into their properties are obtained due to novel and convenient synthetic routes.

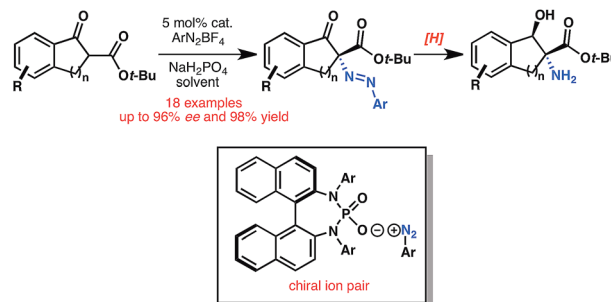


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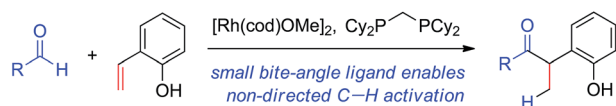
Enantioselective α -amination enabled by a BINAM-derived phase-transfer catalyst

H. M. Nelson, J. S. Patel, H. P. Shunatona and F. D. Toste*

The enantioselective amination of carbonyl derivatives is achieved *via* phase-transfer of aryldiazonium salts by BINAM-derived phosphoric acids.



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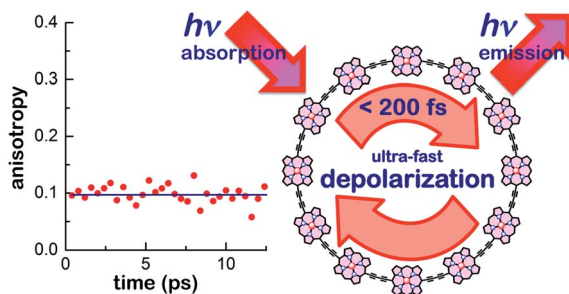


Mechanistic insights into hydroacylation with non-chelating aldehydes

Stephen K. Murphy, Achim Bruch and Vy M. Dong*

Rhodium catalysts with small-bite-angle diphosphines enable branched-selective hydroacylation of 2-vinylphenols via C-H activation of non-chelating aldehydes.

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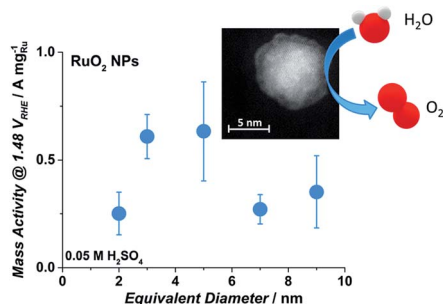


Ultrafast delocalization of excitation in synthetic light-harvesting nanorings

C.-K. Yong, P. Parkinson, D. V. Kondratuk, W.-H. Chen, A. Stannard, A. Summerfield, J. K. Sprafke, M. C. O'Sullivan, P. H. Beton, H. L. Anderson* and L. M. Herz*

When light is absorbed by a nanoring consisting of 6–24 porphyrin units, the excitation delocalizes over the whole molecule within 200 fs. Highly symmetric nanorings exhibit thermally enhanced super-radiance.

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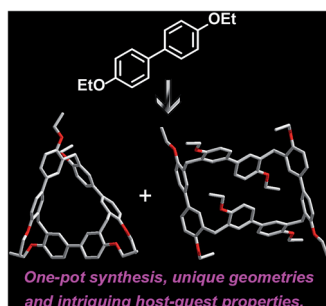


Oxygen evolution on well-characterized mass-selected Ru and RuO_2 nanoparticles

Elisa A. Paoli, Federico Masini, Rasmus Frydendal, Davide Deiana, Christian Schlaup, Mauro Malizia, Thomas W. Hansen, Sebastian Horch, Ifan E. L. Stephens* and Ib Chorkendorff*

Well-defined mass-selected Ru and RuO_2 nanoparticles exhibit an order of magnitude improvement in the oxygen evolution activity, relative to the state-of-the-art, with a maximum at around 3–5 nm.

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Biphen[n]arenes

Huanqing Chen, Jiazeng Fan, Xiaoshi Hu, Junwei Ma, Shilu Wang, Jian Li, Yihua Yu, Xueshun Jia and Chunju Li*

We describe the one-pot synthesis, unique geometries and intriguing host-guest properties of a new class of supramolecular macrocycles – biphen[n]arenes ($n = 3, 4$).

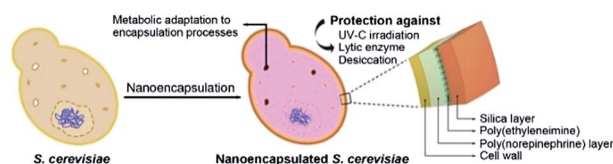


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Organic/inorganic double-layered shells for multiple cytoprotection of individual living cells

Daewha Hong, Hojae Lee, Eun Hyea Ko, Juno Lee, Hyeoncheol Cho, Matthew Park, Sung Ho Yang and Insung S. Choi*

S. cerevisiae encapsulated with a poly(norepinephrine)/silica double-layered shell showed multiple resistance to enzymatic attack, desiccation, and UV-C irradiation. The biochemical response of the encapsulated yeast may also contribute to the UV-C resistance.

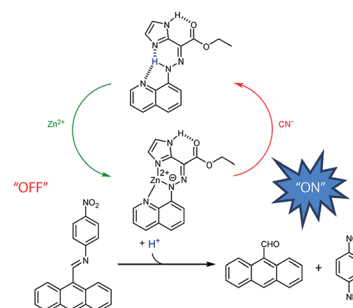


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Regulating signal enhancement with coordination-coupled deprotonation of a hydrazone switch

Justin T. Foy, Debdas Ray and Ivan Aprahamian*

Regulating signal amplification using a switchable catalytic cycle.

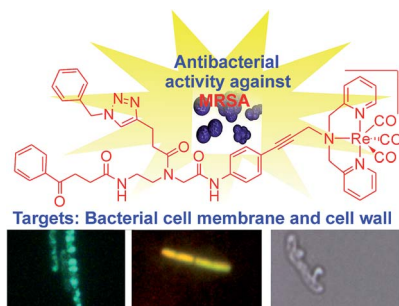


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An organometallic structure-activity relationship study reveals the essential role of a $\text{Re}(\text{CO})_3$ moiety in the activity against gram-positive pathogens including MRSA

M. Patra, M. Wenzel, P. Prochnow, V. Pierroz, G. Gasser, J. E. Bandow* and N. Metzler-Nolte*

A systematic structure activity relationship reveals the contribution of individual organometallic moieties to the potency of a new structural class of hetero-trimetallic antibacterial agents.

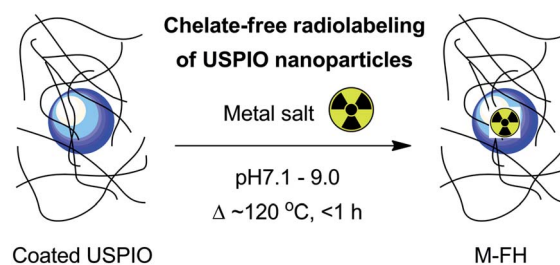


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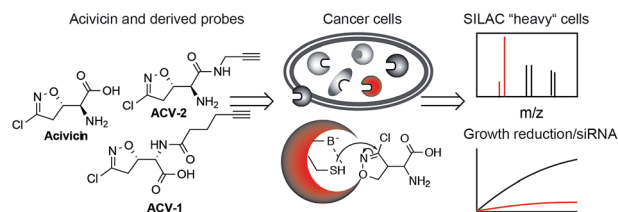
Chelate-free metal ion binding and heat-induced radiolabeling of iron oxide nanoparticles

Eszter Boros, Alice M. Bowen, Lee Josephson, Neil Vasdev and Jason P. Holland*

Holland and co-workers report details of a novel chelate-free reaction for radiolabeling of pre-fabricated nanoparticles using different radionuclides.



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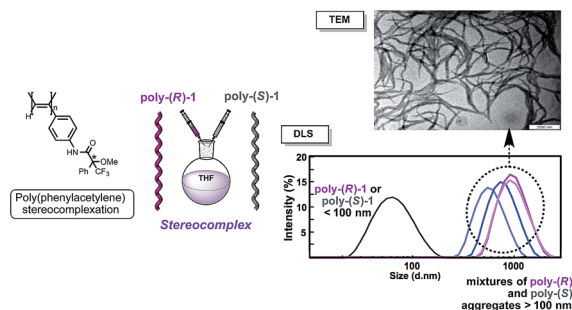


Target discovery of acivicin in cancer cells elucidates its mechanism of growth inhibition

Johannes Kreuzer, Nina C. Bach, Daniel Forler and Stephan A. Sieber*

Using a chemical proteomic strategy we analyzed the targets of acivicin and provided a mechanistic explanation for its inhibition of cancer cell growth.

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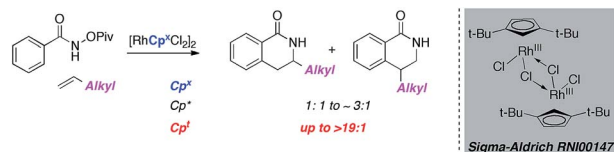


Reversible assembly of enantiomeric helical polymers: from fibers to gels

Seila Leiras, Félix Freire,* Emilio Quiñoá and Ricardo Riguera*

A novel class of stereocomplexes is described by the interaction of helically complementary poly(phenylacetylene)s (PPAs) carrying an α -methoxy- α -trifluoromethylphenylacetamide pendant group.

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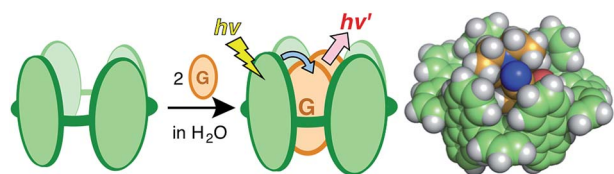


Ligand design for Rh(III)-catalyzed C–H activation: an unsymmetrical cyclopentadienyl group enables a regioselective synthesis of dihydroisoquinolones

Todd K. Hyster, Derek M. Dalton and Tomislav Rovis*

A modified cyclopentadienyl ligand greatly improves regioselectivity in Rh(III) catalyzed alkene insertion/C–H activation.

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An aqueous molecular tube with polyaromatic frameworks capable of binding fluorescent dyes

Keita Hagiwara, Munetaka Akita and Michito Yoshizawa*

An aqueous polyaromatic tube binds two molecules of fluorescent dyes in water and the bound dye dimers exhibit unusual excimer-like emissions through efficient host–guest energy transfer.

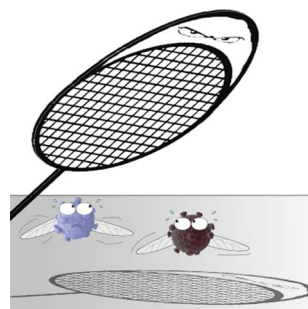


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Macromolecular prodrugs of ribavirin: towards a treatment for co-infection with HIV and HCV

Anton A. A. Smith, Kaja Zuwala, Mille B. L. Kryger, Benjamin M. Wohl, Carlos Guerrero-Sanchez, Martin Tolstrup, Almar Postma and Alexander N. Zelikin*

Macromolecular prodrugs of ribavirin were developed as blood safe formulations with capacity to fight inflammation and human immunodeficiency virus *in vitro*.

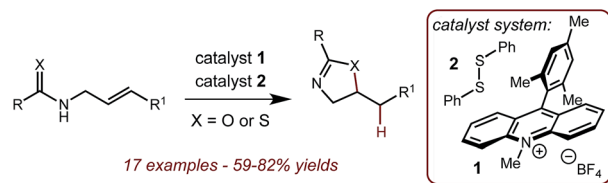


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Divergent regioselectivity in photoredox-catalyzed hydrofunctionalization reactions of unsaturated amides and thioamides

Peter D. Morse and David A. Nicewicz*

A direct method to construct 2-oxazolines and 2-thiazolines from corresponding allylic amides and thioamides is reported.

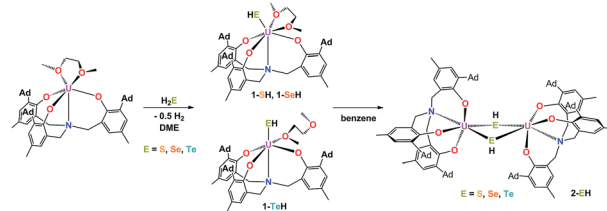


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Reactivity of uranium(III) with H₂E (E = S, Se, Te): synthesis of a series of mononuclear and dinuclear uranium(IV) hydrochalcogenido complexes

Sebastian M. Franke, Michael W. Rosenzweig, Frank W. Heinemann and Karsten Meyer*

Reaction of [(^{Ad}ArO)₃N]U^{III}(DME)] with EH₂ (E = S, Se, Te) yields a complete series of mono- and dinuclear uranium(IV) hydrochalcogenido complexes.

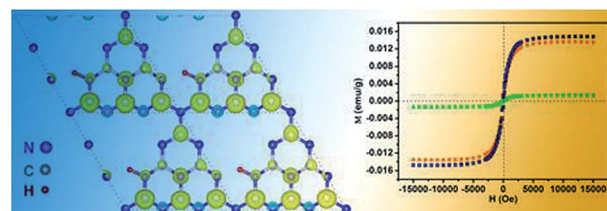


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Hydrogen dangling bonds induce ferromagnetism in two-dimensional metal-free graphitic-C₃N₄ nanosheets

Kun Xu, Xiuling Li, Pengzuo Chen, Dan Zhou, Changzheng Wu,* Yuqiao Guo, Lidong Zhang, Jiyin Zhao, Xiaojun Wu and Yi Xie

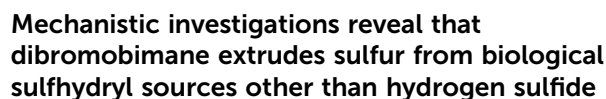
Inducing ferromagnetism in two dimensional metal-free g-C₃N₄ nanosheets has been achieved through the introduction of hydrogen dangling bonds.





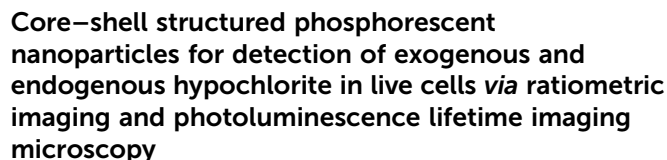
The aryl cycloaurated species are established as reliable models for understanding the gold(III)-catalyzed oxidative cross-couplings between arenes and arylboronic acids.

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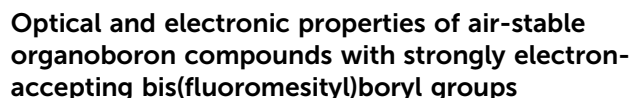
Dibromobimane detects sulfide levels as low as 0.6 pM, but reacts in unexpected ways with thiols, as evidenced by mechanistic investigations.

301



Core-shell phosphorescent nanoparticles were used to detect intracellular ClO^- *via* ratiometric and photoluminescence lifetime imaging.

308



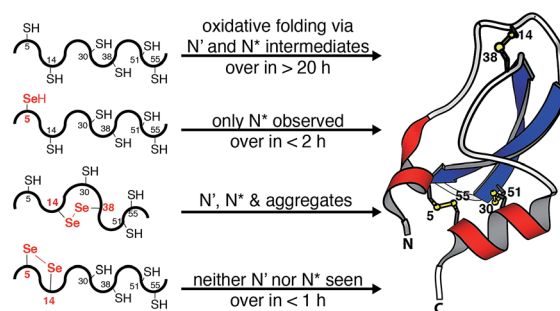
R-Ph-B(FMe)₂ compounds exhibit low reduction potentials, bright emission, a TICT state and unusual long-lived phosphorescence at low temperature.

322

Harnessing selenocysteine reactivity for oxidative protein folding

Norman Metanis and Donald Hilvert*

Turbo-charged folding with selenium: targeted replacement of cysteines in proteins with selenocysteines is a valuable strategy for increasing the rates of oxidative protein folding, altering folding mechanisms, and rescuing kinetically trapped intermediates.

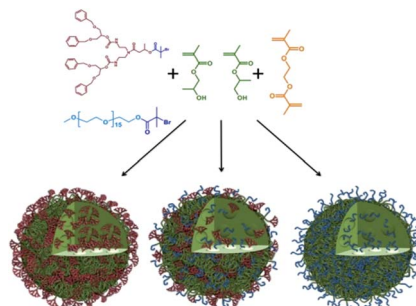


326

Hyperbranched polydendrons: a new nanomaterials platform with tuneable permeation through model gut epithelium

Fiona L. Hatton, Lee M. Tatham, Louise R. Tidbury, Pierre Chambon, Tao He, Andrew Owen* and Steven P. Rannard*

Highly branched vinyl polymers (hyperbranched polydendrons), displaying combinations of dendritic and PEG end groups, have been synthesised using a mixed initiator approach. Nanoprecipitated polydendron particles have exhibited controlled permeation through a gut epithelium model.

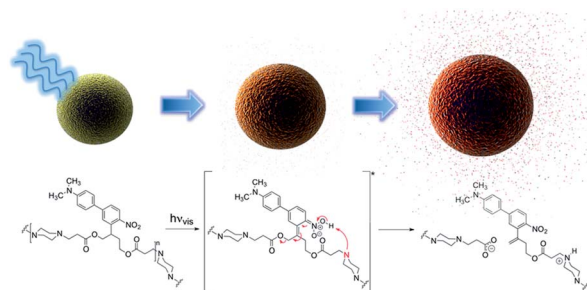


335

In vivo visible light-triggered drug release from an implanted depot

Carl-Johan Carling, Mathieu L. Viger, Viet Anh Nguyen Huu, Arnold V. Garcia and Adah Almutairi*

A visible light-degradable polymer allows on-demand *in vivo* photorelease of encapsulated dexamethasone from a subcutaneously implanted particle depot.

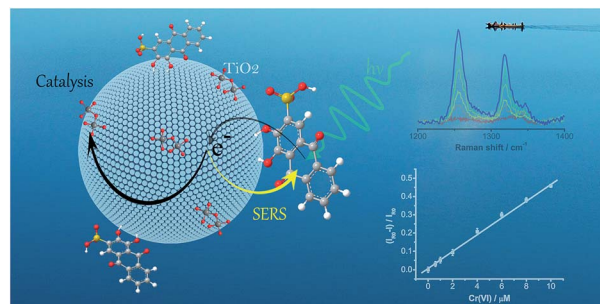


342

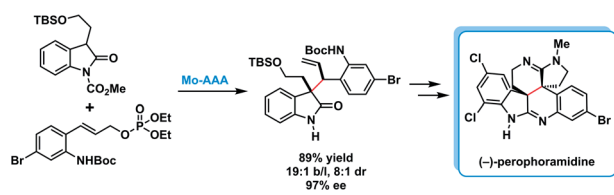
Semiconductor-driven "turn-off" surface-enhanced Raman scattering spectroscopy: application in selective determination of chromium(vi) in water

Wei Ji, Yue Wang, Ichiro Tanabe, Xiaoxia Han, Bing Zhao* and Yukihiro Ozaki*

A novel "turn-off" SERS strategy for the detection of metal ions was demonstrated based on the optical and catalytic properties of semiconductor materials.



349

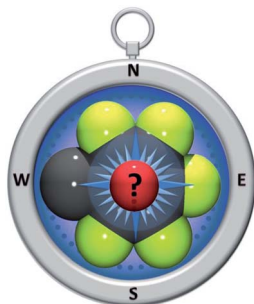


A catalytic asymmetric total synthesis of (–)-perophoramidine

B. M. Trost,* M. Osipov, S. Krüger and Y. Zhang

We describe a catalytic asymmetric total synthesis of the ascidian alkaloid (–)-perophoramidine employing a Mo-catalyzed asymmetric allylic alkylation and unprecedented imino ether allylation as key steps.

354

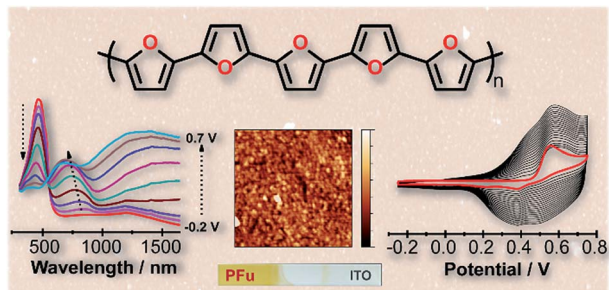


The pentafluorophenyl group as π -acceptor for anions: a case study

Michael Giese, Markus Albrecht,* Arto Valkonen and Kari Rissanen*

A unique structural study investigates the variability of anion– π bonding in the solid state structures of pentafluorophenyl arenes. The hapticity concept is used as tool to describe the structural differences of various anion– π complexes.

360

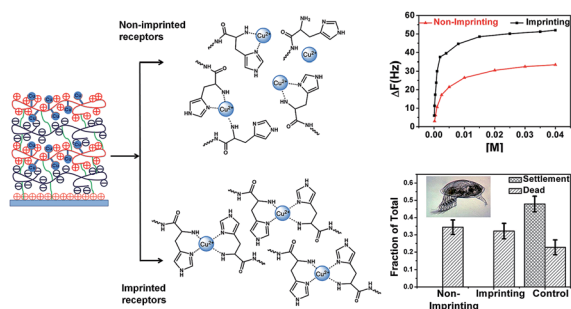


Conducting polyfurans by electropolymerization of oligofurans

Dennis Sheberla,* Snehangshu Patra, Yair H. Wijsboom, Sagar Sharma, Yana Sheynin, Abd-Elrazek Haj-Yahia, Adva Hayoun Barak, Ori Gidron and Michael Bendikov

Polyfuran films produced by electropolymerization of a series of oligofurans substituted with alkyl groups show improved properties, such as good conductivity and stability, well-defined spectroelectrochemistry and smooth morphology.

372



Imprinting of metal receptors into multilayer polyelectrolyte films: fabrication and applications in marine antifouling

Sreenivasa Reddy Puniredd, Dominik Jańczewski,* Dewi Pitrasari Go, Xiaoying Zhu, Shifeng Guo, Serena Lay Ming Teo, Serina Siew Chen Lee and G. Julius Vancso*

Polymeric films constructed using layer-by-layer fabrication were employed as a platform for metal ion immobilization and applied as a marine antifouling coating.

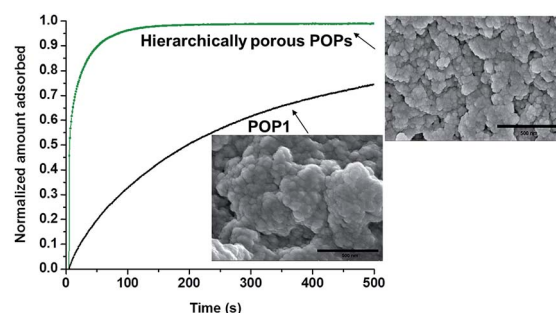


384

Hierarchically porous organic polymers: highly enhanced gas uptake and transport through templated synthesis

Sanjiban Chakraborty, Yamil J. Colón, Randall Q. Snurr and SonBinh T. Nguyen*

Hierarchically porous POPs with highly enhanced gas uptake and transport properties were obtained through polymerization within a monolithic mesoporous template.

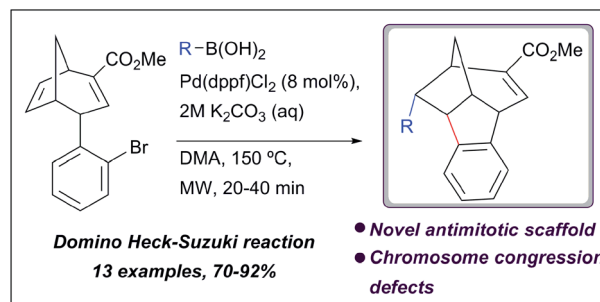


390

Synthesis of a novel polycyclic ring scaffold with antimitotic properties via a selective domino Heck–Suzuki reaction

Esther Alza, Luca Laraia, Brett M. Ibbeson, Súil Collins, Warren R. J. D. Galloway, Jamie E. Stokes, Ashok R. Venkitaraman and David R. Spring*

The synthesis of a previously undescribed sp^3 -rich 6-5-5-6 tetracyclic ring scaffold using a palladium catalysed domino Heck–Suzuki reaction is reported.

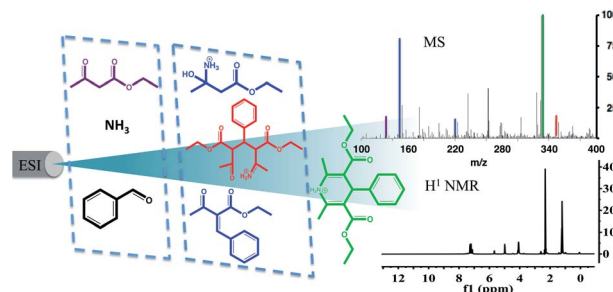


397

Accelerated Hantzsch electro spray synthesis with temporal control of reaction intermediates

Ryan M. Bain, Christopher J. Pulliam and R. Graham Cooks*

Complex reaction sequences can be followed in electrosprayed droplets on the millisecond time scale.

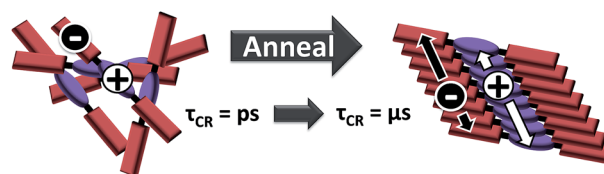


402

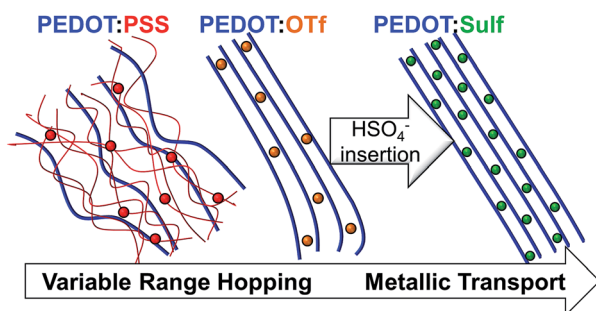
Long-lived charge carrier generation in ordered films of a covalent perylenediimide–diketopyrrolopyrrole–perylenediimide molecule

Patrick E. Hartnett, Scott M. Dyar, Eric A. Margulies, Leah E. Shoer, Andrew W. Cook, Samuel W. Eaton, Tobin J. Marks* and Michael R. Wasielewski*

Self-ordering of covalent electron donor–acceptor building blocks in thin films upon solvent vapor annealing results in a 10^4 increase in photo-generated charge carrier lifetime.



412

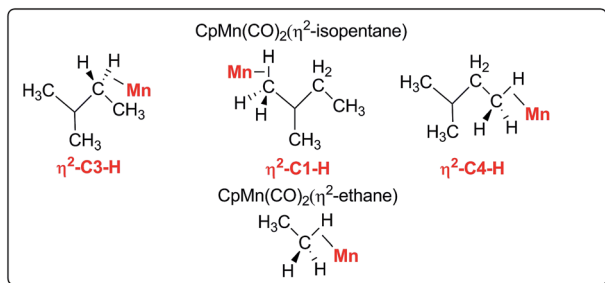


Metallic behaviour of acid doped highly conductive polymers

Nicolas Massonnet, Alexandre Carella,* Arnaud de Geyer, Jérôme Faure-Vincent and Jean-Pierre Simonato*

Polymerizing PEDOT with poorly coordinating counter-ions facilitates their substitution by acids. The resulting materials display a high electrical conductivity and true metallic behaviour.

418

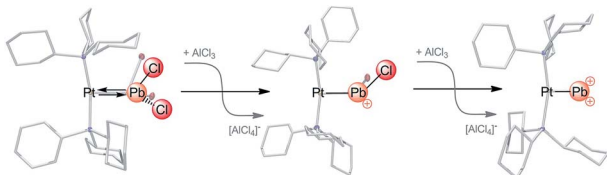


Detection of σ-alkane complexes of manganese by NMR and IR spectroscopy in solution: (η⁵-C₅H₅)-Mn(CO)₂(ethane) and (η⁵-C₅H₅)Mn(CO)₂(isopentane)

Olga Torres, James A. Calladine, Simon B. Duckett, Michael W. George* and Robin N. Perutz*

Irradiation (355 nm) of CpMn(CO)₃ in liquid ethane at 133 K yields CpMn(CO)₂(η²-C-H-ethane) characterised by IR and ¹H NMR spectroscopy with a lifetime of ca. 360 s; the corresponding isopentane complex is formed in propane/isopentane mixtures and exists in three isomeric forms.

425

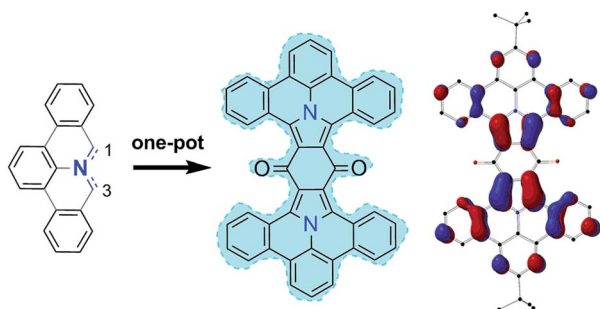


Stepwise isolation of low-valent, low-coordinate Sn and Pb mono- and dications in the coordination sphere of platinum

Holger Braunschweig,* Mehmet Ali Celik, Rian D. Dewhurst, Magdalena Heid, Florian Hupp and Sakya S. Sen

Halide groups are sequentially removed from platinum complexes containing tin and lead dihalide ligands, creating novel mono- and dicationic complexes.

436



Polycyclic aromatic azomethine ylides: a unique entry to extended polycyclic heteroaromatics

Reinhard Berger, Manfred Wagner, Xinliang Feng* and Klaus Müllen*

Based on polycyclic aromatic azomethine ylides, a metal-free "cycloaddition-planarization-sequence" is proposed, providing a unique entry to nitrogen-containing polycyclic aromatic hydrocarbons.

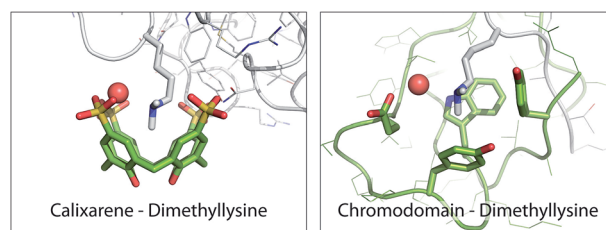


442

Structural study of a small molecule receptor bound to dimethyllysine in lysozyme

Róise E. McGovern, Brendan D. Snarr, Joseph A. Lyons, James McFarlane, Amanda L. Whiting, Irina Paci, Fraser Hof and Peter B. Crowley*

X-ray crystallography reveals how a calixarene can bind to dimethyllysine to form a complex with features similar to the aromatic cage motif of a chromodomain bound to a histone tail.

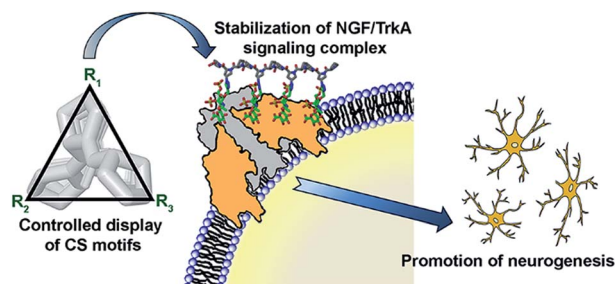


450

Tailored chondroitin sulfate glycomimetics via a tunable multivalent scaffold for potentiating NGF/TrkA-induced neurogenesis

Pei Liu, Liwei Chen, Jerry K. C. Toh, Yi Li Ang, Joo-Eun Jee, Jaehong Lim, Su Seong Lee and Song-Gil Lee*

We have engineered structurally well-defined tunable chondroitin sulfate glycopeptides using a polyproline scaffold to selectively modulate the NGF-mediated neuronal signaling pathway.

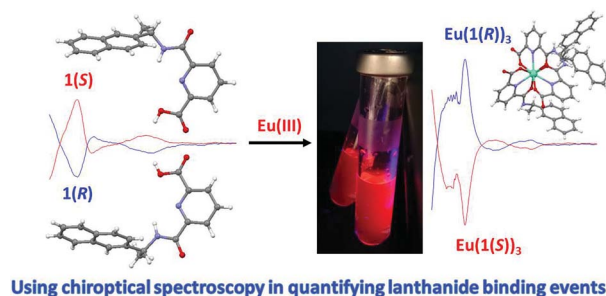


457

The application of chiroptical spectroscopy (circular dichroism) in quantifying binding events in lanthanide directed synthesis of chiral luminescent self-assembly structures

Oxana Kotova,* Salvador Blasco, Brendan Twamley, John O'Brien, Robert D. Peacock, Jonathan A. Kitchen, Miguel Martínez-Calvo and Thorfinnur Gunnlaugsson*

The binding of asymmetrical and optically pure tridentate ligands containing one carboxylic group and 2-naphthyl as an antenna to lanthanide ions was studied in CH₃CN.

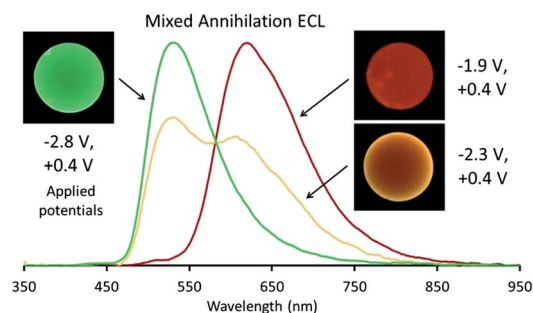


472

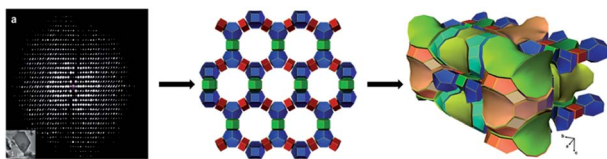
Annihilation electrogenerated chemiluminescence of mixed metal chelates in solution: modulating emission colour by manipulating the energetics

Emily Kerr, Egan H. Doeven, Gregory J. Barbante,* Conor F. Hogan,* David J. Bower, Paul S. Donnelly, Timothy U. Connell and Paul S. Francis*

The emissions of the mixed annihilation ECL of metal complexes can be effectively predicted by estimations of reaction exergonicity.



480



ITQ-54: a multi-dimensional extra-large pore zeolite with $20 \times 14 \times 12$ -ring channels

Jiuxing Jiang, Yifeng Yun, Xiaodong Zou,* Jose Luis Jorda and Avelino Corma*

A silicogermanate zeolite with straight intersecting $20 \times 14 \times 12$ -ring channels has been synthesized and its structure has been solved using the rotation electron diffraction method.

486

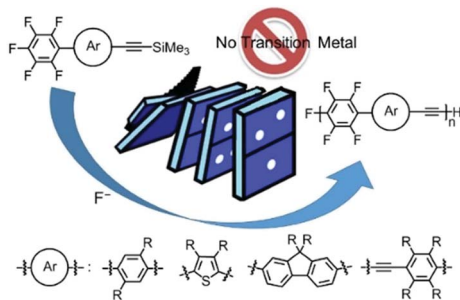


"Self-repairing" nanoshell for cell protection

Nan Jiang, Xiao-Yu Yang,* Guo-Liang Ying, Ling Shen, Jing Liu, Wei Geng, Ling-Jun Dai, Shao-Yin Liu, Jian Cao, Ge Tian, Tao-Lei Sun, Shi-Pu Li and Bao-Lian Su*

Self-repairing biohybrid nanoshells provide living cells with high activity and extended viability in harsh micro-environments.

492

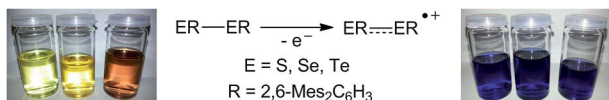


Transition-metal-free controlled polymerization for poly(*p*-aryleneethynylene)s

Takanobu Sanji,* Asahi Motoshige, Hideaki Komiyama, Junko Kakinuma, Rie Ushikubo, Satoru Watanabe and Tomokazu Iyoda

A catalytic amount of fluoride anions promoted the polymerization of 1-pentafluorophenyl-4-[(trimethylsilyl)ethynyl]benzene, providing a high-molecular-weight polymer in a chain-growth-like manner.

497



Diaryldichalcogenide radical cations

Ole Mallow, Monther A. Khanfar, Moritz Malischewski, Pamela Finke, Malte Hesse, Enno Lork, Timo Augenstein, Frank Breher, Jeffrey R. Harmer, Nadezhda V. Vasilieva, Andrey Zibarev, Artem S. Bogomyakov, Konrad Seppelt and Jens Beckmann*

One-electron oxidation of diaryldichalcogenides is the key to the preparation of the corresponding radical cations.

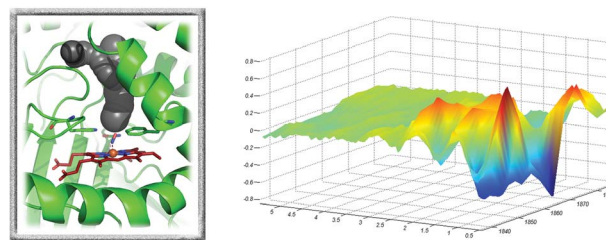


505

Ultrafast infrared spectroscopy reveals water-mediated coherent dynamics in an enzyme active site

Katrin Adamczyk, Niall Simpson, Gregory M. Greetham, Andrea Gumiero, Martin A. Walsh, Michael Towrie, Anthony W. Parker and Neil T. Hunt*

Ultrafast infrared spectroscopy provides insights into the dynamic nature of water in the active sites of catalase and peroxidase enzymes.

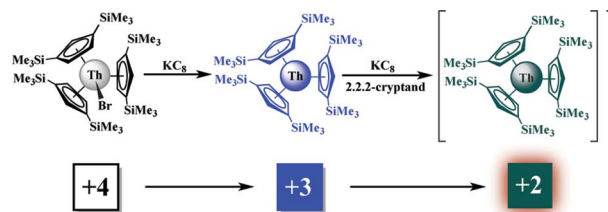


517

Synthesis, structure, and reactivity of crystalline molecular complexes of the $\{[C_5H_3(SiMe_3)_2]_3Th\}^{1-}$ anion containing thorium in the formal +2 oxidation state

Ryan R. Langeslay, Megan E. Fieser, Joseph W. Ziller, Filipp Furche* and William J. Evans*

Structural, spectroscopic, and DFT analysis of the first molecular complexes of Th^{2+} indicate they have a $6d^2$ electron configuration of the type expected for the transactinide ions Rf^{2+} and Db^{3+} .

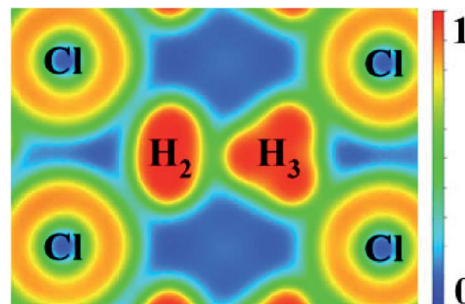


522

Stabilization of H_3^+ in the high pressure crystalline structure of H_nCl ($n = 2-7$)

Ziwei Wang, Hui Wang,* John S. Tse,* Toshiaki Iitaka and Yanming Ma

Using the CALYPSO structure searching method, multicenter bonding H_3^+ ions were stabilized under high pressure in a hydrogen rich H-Cl system.

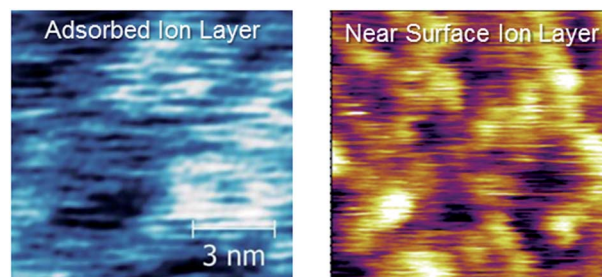


527

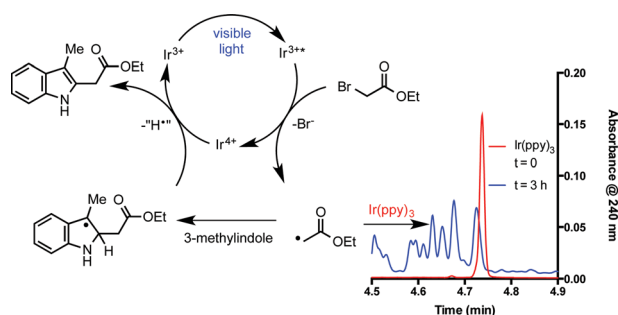
Ion structure controls ionic liquid near-surface and interfacial nanostructure

Aaron Elbourne, Kislun Voitchovsky, Gregory G. Warr and Rob Atkin*

In situ amplitude modulated atomic force microscopy (AM-AFM) has been used to resolve the 3-dimensional nanostructure of five protic ILs at and near the surface of mica.



537

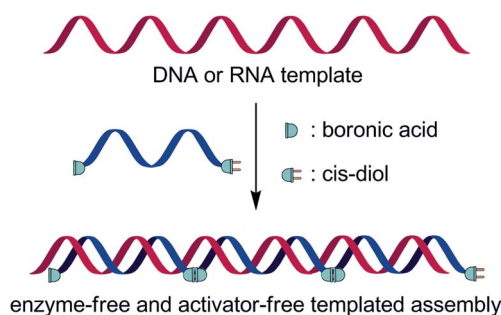


Ligand functionalization as a deactivation pathway in a *fac*-Ir(ppy)₃-mediated radical addition

James J. Devery III, James J. Douglas, John D. Nguyen, Kevin P. Cole, Robert A. Flowers II* and Corey R. J. Stephenson*

Kinetic, synthetic, and spectroscopic evidence demonstrates the instability of *fac*-Ir(ppy)₃ under visible light-mediated photoredox conditions resulting from *in situ* functionalization.

542

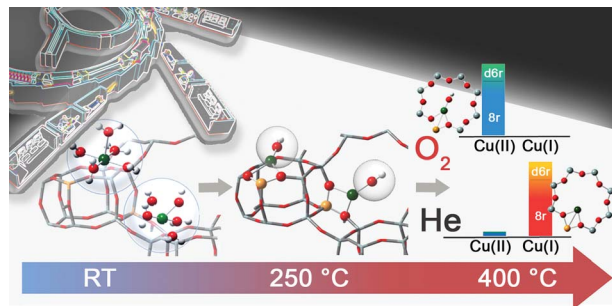


pH-controlled DNA- and RNA-templated assembly of short oligomers

Renaud Barbeyron, Jean-Jacques Vasseur* and Michael Smietana*

A stimuli-responsive enzyme-free and activator-free DNA- and RNA-templated assembly of bifunctional short oligonucleotides is reported.

548

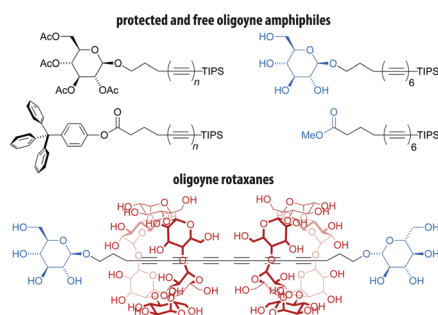


Revisiting the nature of Cu sites in the activated Cu-SSZ-13 catalyst for SCR reaction

E. Borfecchia, K. A. Lomachenko, F. Giordanino, H. Falsig, P. Beato, A. V. Soldatov, S. Bordiga and C. Lamberti*

X-ray absorption and emission spectroscopy, FTIR and DFT unravel the major Cu species in the activated Cu-SSZ-13 catalyst for NH₃-SCR.

564



Facile synthesis of oligoynes and their rotaxanes

Stephen Schrettl, Emmanuel Contal, Tobias N. Hoheisel, Martin Fritzsche, Sandor Balog, Ruth Szilluweit and Holger Frauenrath*

Carbon-rich organic compounds containing a series of conjugated triple bonds (oligoynes) are relevant synthetic targets, but an improved access to oligoynes bearing functional groups would be desirable.

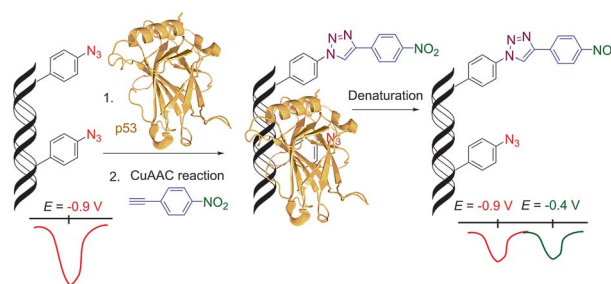


575

Azidophenyl as a click-transformable redox label of DNA suitable for electrochemical detection of DNA–protein interactions

Jana Balintová, Jan Špaček, Radek Pohl, Marie Brázdová, Luděk Havran, Miroslav Fojta* and Michal Hocek*

A new azido-based DNA redox label which can be transformed into nitrophenyltriazole by a CuAAC click reaction was developed. It was used for the mapping of DNA–protein interactions with electrochemical detection.

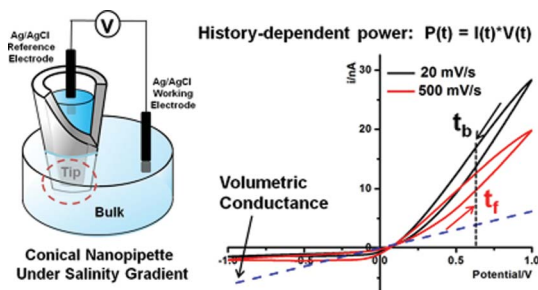


588

History-dependent ion transport through conical nanopipettes and the implications in energy conversion dynamics at nanoscale interfaces

Yan Li, Dengchao Wang, Maksim M. Kvetny, Warren Brown, Juan Liu and Gangli Wang*

We report striking time-dependent ion transport characteristics at nanoscale interfaces in current–potential (I – V) measurements and theoretical analyses.

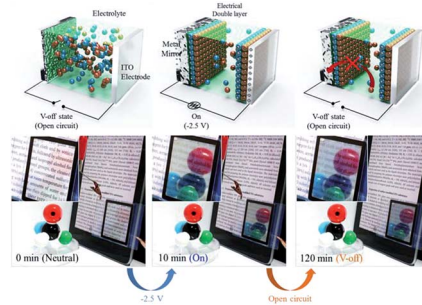


596

Switchable silver mirrors with long memory effects

Chihyun Park, Seogjae Seo, Haijin Shin, Bhimrao D. Sarwade, Jongbeom Na and Eunkyong Kim*

An electrochemically stable and bistable switchable mirror is achieved for the first time by introducing a thiol-modified ITO electrode to stabilize the metallic film and ionic liquids as an anion-blocking layer, to achieve a long memory effect.

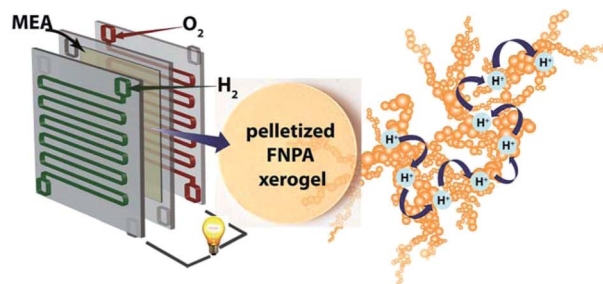


603

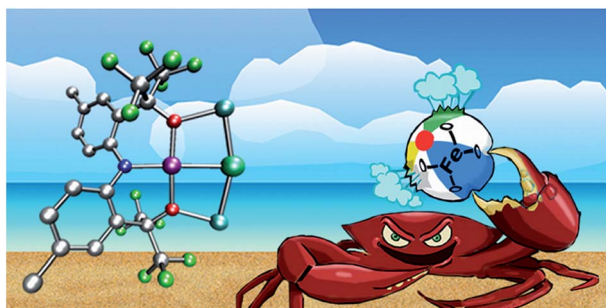
Fe(III) phytate metallogel as a prototype anhydrous, intermediate temperature proton conductor

Harshitha Barike Aiyappa, Subhadeep Saha, Pritish Wadge, Rahul Banerjee* and Sreekumar Kurungot*

Protogenic phytic acid is immobilized by its gelation with iron nitrate in DMF. The resulting pelletized xerogel is observed to show a high proton conductivity of $2.4 \times 10^{-2} \text{ S cm}^{-1}$ at 120°C and is tried as solid electrolyte for dry H_2/O_2 fuel cell operation.



608

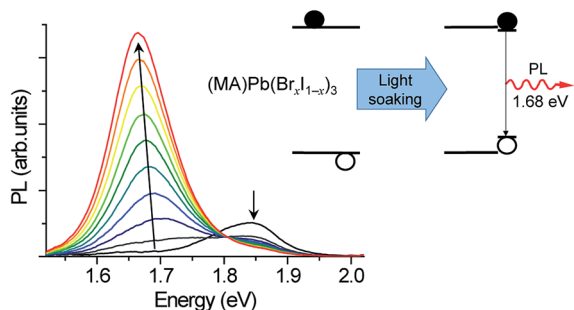


A high-spin square-planar Fe(II) complex stabilized by a trianionic pincer-type ligand and conclusive evidence for retention of geometry and spin state in solution

M. E. Pascualini, N. V. Di Russo, A. E. Thuijs, A. Ozarowski, S. A. Stoian, K. A. Abboud, G. Christou and A. S. Veige*

Extensive spectroscopic evaluation of a novel ONO^{3-} -trianionic pincer Fe(II) complex indicates the rare square-planar geometry and $S = 2$ spin state are retained in solution.

613

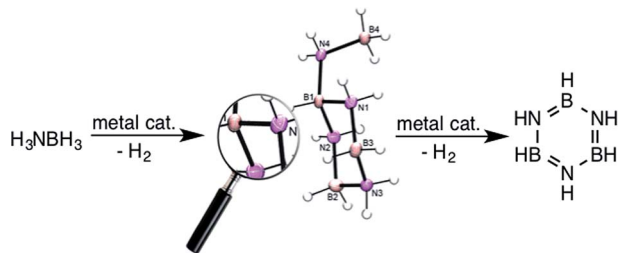


Reversible photo-induced trap formation in mixed-halide hybrid perovskites for photovoltaics

Eric T. Hoke, Daniel J. Slotcavage, Emma R. Dohner, Andrea R. Bowring, Hemamala I. Karunadasa* and Michael D. McGehee*

A reversible photo-induced instability has been found in mixed-halide photovoltaic perovskites that limits the open circuit voltage in solar cells.

618

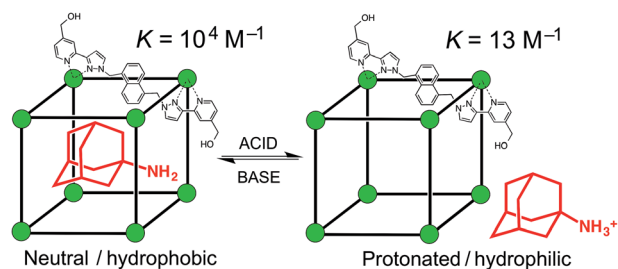


Probing the second dehydrogenation step in ammonia-borane dehydrocoupling: characterization and reactivity of the key intermediate, B-(cyclotriborazanyl)amine-borane

Hassan A. Kalviri, Felix Gärtner, Gang Ye, Ilia Korobkov and R. Tom Baker*

The key intermediate in metal-catalyzed dehydrogenation of ammonia-borane to borazine is shown to be the BN analog of ethylcyclohexane.

625



pH-dependent binding of guests in the cavity of a polyhedral coordination cage: reversible uptake and release of drug molecules

William Cullen, Simon Turega, Christopher A. Hunter* and Michael D. Ward*

Binding of organic guests containing acidic or basic groups inside a water-soluble coordination cage host shows strong pH dependence.

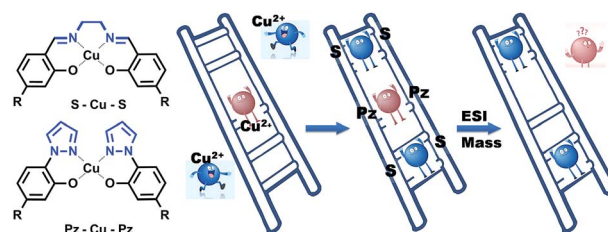


632

DNA based multi-copper ions assembly using combined pyrazole and salen ligandosides

Meng Su, María Tomás-Gamasa and Thomas Carell*

The pyrazole and salen ligandosides, when combined, are able to create stable multi-copper ion complexing DNA duplex structures in a cooperative fashion.

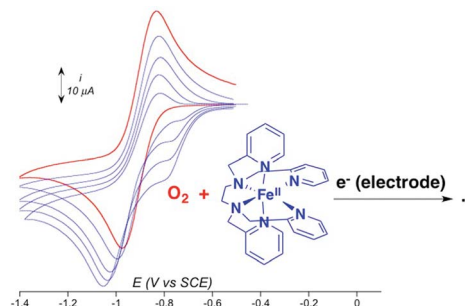


639

Electrochemical study of a nonheme Fe(II) complex in the presence of dioxygen. Insights into the reductive activation of O₂ at Fe(II) centers

N. Ségaud, E. Anxolabéhère-Mallart,* K. Sénéchal-David, L. Acosta-Rueda, M. Robert and F. Banse*

An original mechanistic study of the reaction of [(L)Fe^{II}]²⁺ (L = TPEN) with dioxygen has been carried out by cyclic voltammetry. Electrochemical data of intermediates [(L)Fe^{IV}(O)]²⁺, [(L)Fe^{III}(OOH)]²⁺ and [(L)Fe^{III}(OO)]⁺ are reported. Reaction mechanism between this Fe^{II} complex and O₂ under reductive conditions is determined.

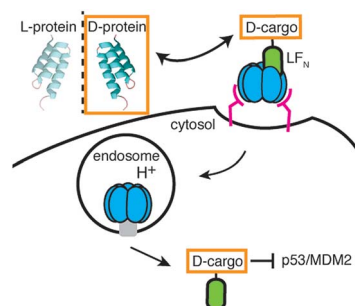


648

Delivery of mirror image polypeptides into cells

Amy E. Rabideau, Xiaoli Liao and Bradley L. Pentelute*

Mirror image peptides have unique stability and immunogenic properties in mammals, making them attractive agents to investigate.

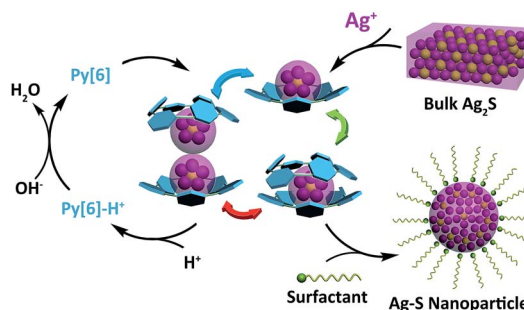


654

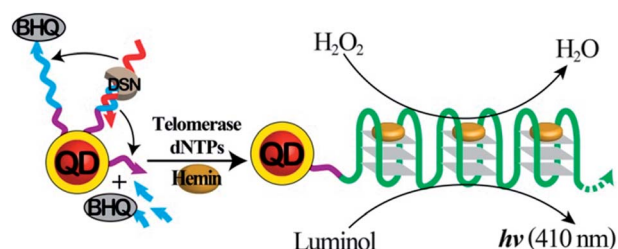
A macrocycle-assisted nanoparticlization process for bulk Ag₂S

Xin He, Yuechao Wang, Cai-Yan Gao, Hong Jiang and Liang Zhao*

A new nanoparticlization process for the bulk-to-nano transformation of Ag₂S by incorporating both top-down and bottom-up approaches is reported.



659

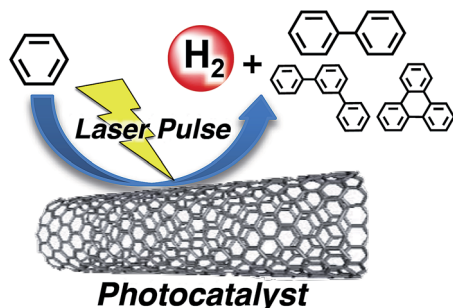


Diagnosing the miR-141 prostate cancer biomarker using nucleic acid-functionalized CdSe/ZnS QDs and telomerase

Amily Fang-ju Jou, Chun-Hua Lu, Yen-Chuan Ou,^{*} Shian-Shiang Wang, Shih-Lan Hsu, Itamar Willner^{*} and Ja-an Annie Ho^{*}

The miR-141 prostate cancer biomarker is optically detected by a two-step analytical platform that includes telomerase and semiconductor quantum dots.

666

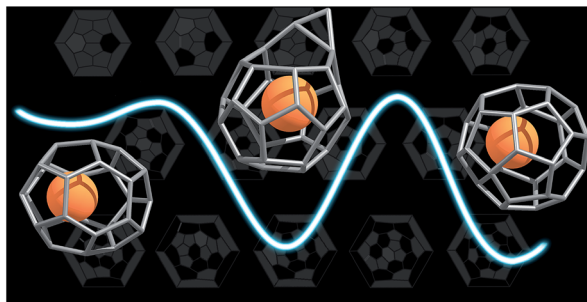


Laser-induced pinpoint hydrogen evolution from benzene and water using metal free single-walled carbon nanotubes with high quantum yields

Kei Ohkubo, Naoki Kohno, Yusuke Yamada and Shunichi Fukuzumi^{*}

Metal-free photocatalytic hydrogen evolution occurred efficiently in benzene containing SWCNTs under laser irradiation with an extremely high turnover number of 2 000 000 and a high quantum yield of 130%.

675



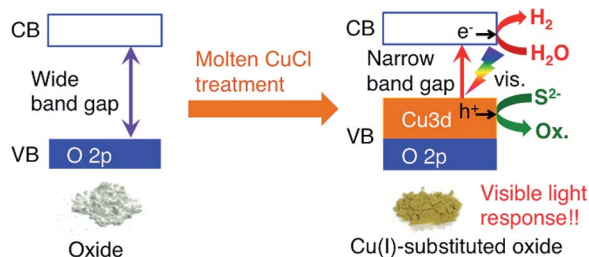
Small endohedral metallofullerenes: exploration of the structure and growth mechanism in the Ti@C_{2n} (2n = 26–50) family

Marc Mulet-Gas, Laura Abella, Paul W. Dunk, Antonio Rodríguez-Fortea,^{*} Harold W. Kroto^{*} and Josep M. Poblet^{*}

Analysis of the structure and the bottom-up growth mechanism in the family of small endohedral metallofullerenes Ti@C_{2n} (2n = 26–50).

687

Development of visible-light-driven photocatalyst for H₂ evolution



Sensitization of wide band gap photocatalysts to visible light by molten CuCl treatment

Katsuya Iwashina, Akihito Iwase and Akihiko Kudo^{*}

Cu(I)-containing metal oxide photocatalysts for hydrogen evolution under visible light irradiation were prepared using molten CuCl treatment.

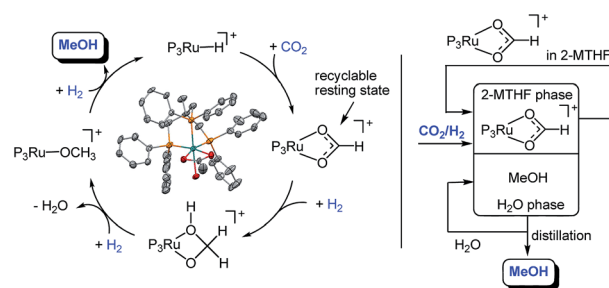


693

Hydrogenation of carbon dioxide to methanol using a homogeneous ruthenium–Triphos catalyst: from mechanistic investigations to multiphase catalysis

Sebastian Wesselbaum, Verena Moha, Markus Meuresch, Sandra Brosinski, Katharina M. Thenert, Jens Kothe, Thorsten vom Stein, Ulli Englert, Markus Hölscher, Jürgen Klankermayer* and Walter Leitner*

The hydrogenation of CO₂ to methanol using a recyclable molecular organometallic catalyst in the absence of an alcohol additive is demonstrated for the first time.

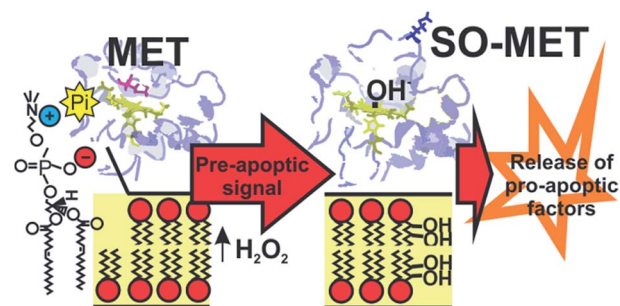


705

Specific methionine oxidation of cytochrome c in complexes with zwitterionic lipids by hydrogen peroxide: potential implications for apoptosis

Daiana A. Capdevila, Waldemar A. Marmisollé, Florencia Tomasina, Verónica Demicheli, Magdalena Portela, Rafael Radi and Daniel H. Murgida*

The rise of H₂O₂ concentration that characterizes the initiation of apoptosis can specifically oxidize Met80 in cytochrome c bound to zwitterionic phospholipids, yielding a stable peroxidase.

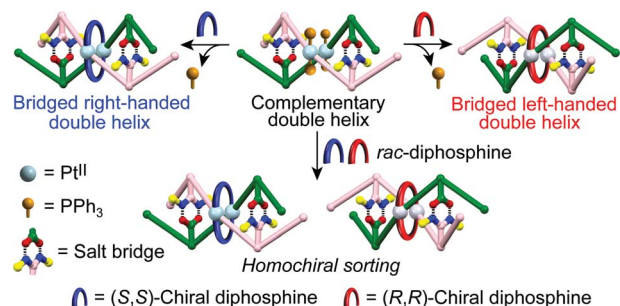


714

Chiral tether-mediated stabilization and helix-sense control of complementary metallo-double helices

Miki Horie, Naoki Ousaka, Daisuke Taura and Eiji Yashima*

Control of the helical sense and stability enhancement of Pt^{II}-linked double helices by interstrand cross-linking using chiral diphosphines and complete homochiral self-sorting is demonstrated.

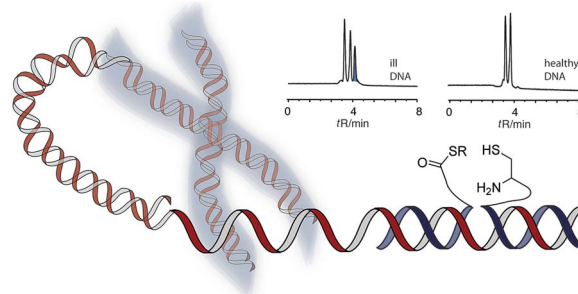


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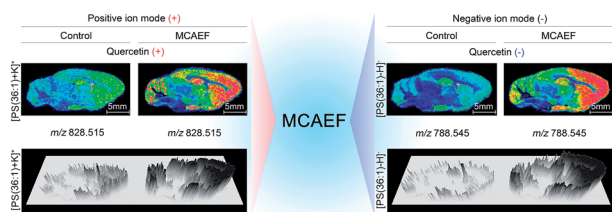
Template-directed ligation on repetitive DNA sequences: a chemical method to probe the length of Huntington DNA

Anika Kern and Oliver Seitz*

Several genomic disorders are caused by an excessive number of DNA triplet repeats.



729

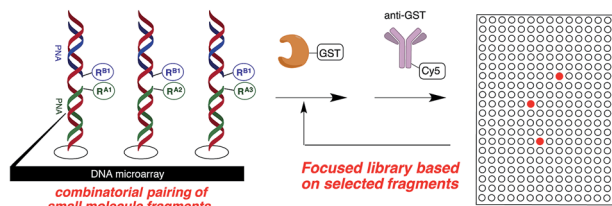


Matrix coating assisted by an electric field (MCAEF) for enhanced tissue imaging by MALDI-MS

Xiaodong Wang, Jun Han, Juncong Yang, Jingxi Pan and Christoph H. Borchers*

A novel technique, termed matrix coating assisted by an electric field (MCAEF) for enhancing tissue imaging by matrix-assisted laser desorption/ionization mass spectrometry (MALDI-MS) was developed in this study.

739

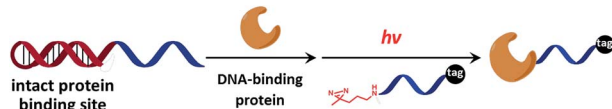


DNA display of fragment pairs as a tool for the discovery of novel biologically active small molecules

J.-P. Daguer, C. Zambaldo, M. Ciobanu, P. Morieux, S. Barluenga and N. Winssinger*

A focused library for Hsp70 was prepared from fragments identified from an array combinatorially pairing two libraries of small molecule fragments. Screening of the focus library yielded high affinity ligand to Hsp70.

745

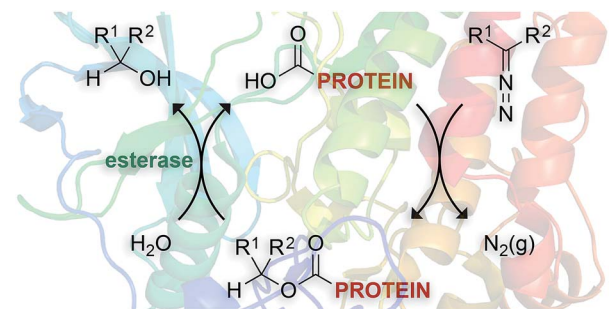


Photoaffinity labeling of transcription factors by DNA-templated crosslinking

Ying Liu, Wenlu Zheng, Wan Zhang, Nan Chen, Yang Liu, Li Chen, Xiaozhou Zhou, Xingshuo Chen, Haifeng Zheng and Xiaoyu Li*

A dual-probe system can specifically capture DNA-binding proteins with an unmodified binding site.

752



Diazo compounds for the bioreversible esterification of proteins

Nicholas A. McGrath, Kristen A. Andersen, Amy K. F. Davis, Jo E. Lomax and Ronald T. Raines*

A diazo compound is shown to convert carboxylic acids to esters efficiently in an aqueous environment.

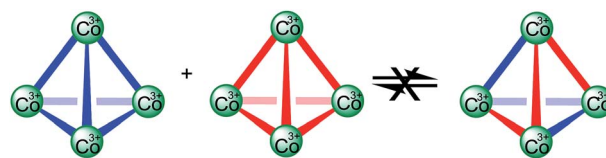


756

Non-equilibrium cobalt(III) "click" capsules

P. R. Symmers, M. J. Burke, D. P. August, P. I. T. Thomson, G. S. Nichol, M. R. Warren, C. J. Campbell and P. J. Lusby*

Constitutionally non-dynamic cobalt(III) tetrahedral capsules have been prepared using an assembly-followed-by-oxidation protocol.

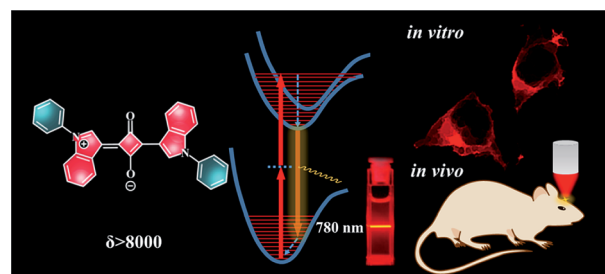


761

Rational design of small indolic squaraine dyes with large two-photon absorption cross section

Chun-Lin Sun, Qing Liao, Ting Li, Jun Li, Jian-Qiao Jiang, Zhen-Zhen Xu, Xue-Dong Wang, Rong Shen, De-Cheng Bai, Qiang Wang, Sheng-Xiang Zhang, Hong-Bing Fu and Hao-Li Zhang*

Assisted by theoretical analysis, we designed a small indolic squaraine with $\delta > 8000$ GM at 780 nm, which is ideal for both *in vitro* and *in vivo* bio-imaging applications.

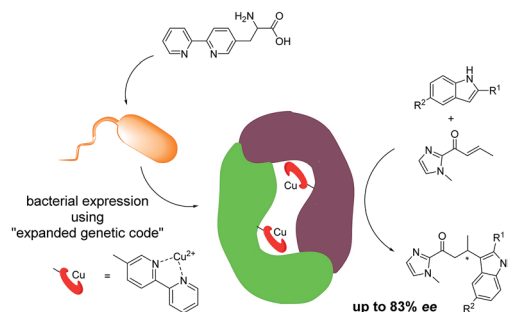


770

Novel artificial metalloenzymes by *in vivo* incorporation of metal-binding unnatural amino acids

Ivana Drienovská, Ana Rioz-Martínez, Apparao Draksharapu and Gerard Roelfes*

Artificial metalloenzymes for asymmetric catalysis were created by *in vivo* incorporation of unnatural metal binding amino acids into LmrR.

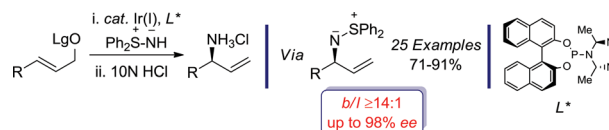


777

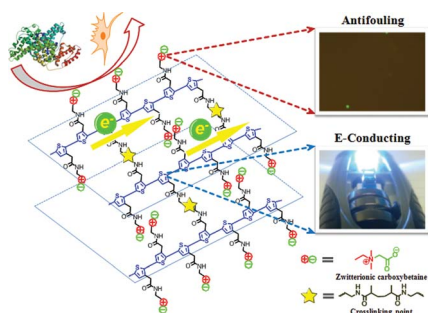
Enantioselective construction of C-chiral allylic sulfilimines via the iridium-catalyzed allylic amination with *S,S*-diphenylsulfilimine: asymmetric synthesis of primary allylic amines

Rebecca L. Grange, Elizabeth A. Clizbe, Emma J. Counsell and P. Andrew Evans*

We have devised a highly regio- and enantioselective iridium-catalyzed allylic amination reaction with the sulfur-stabilized aza-ylide, *S,S*-diphenylsulfilimine.



782

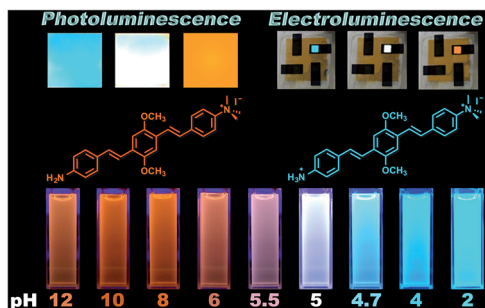


Integrated zwitterionic conjugated poly(carboxybetaine thiophene) as a new biomaterial platform

Bin Cao, Qiong Tang, Linlin Li, Chen-Jung Lee, Hua Wang, Yanqiao Zhang, Homero Castaneda* and Gang Cheng*

An integrated zwitterionic conjugated polymer-based biomaterial platform was designed and studied to address some of the key challenges of conjugated polymers in biomedical applications.

789

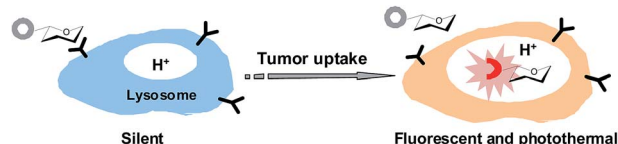


A new pH sensitive fluorescent and white light emissive material through controlled intermolecular charge transfer

Y. I. Park, O. Postupna, A. Zhugayevych, H. Shin, Y.-S. Park, B. Kim, H.-J. Yen, P. Cheruku, J. S. Martinez, J. W. Park, S. Tretiak* and H.-L. Wang*

Fabrication of a unique white light LED from a stimuli-responsive organic molecule is reported. Emission properties are dominated by the pH of the solution through intermolecular charge transfer.

798

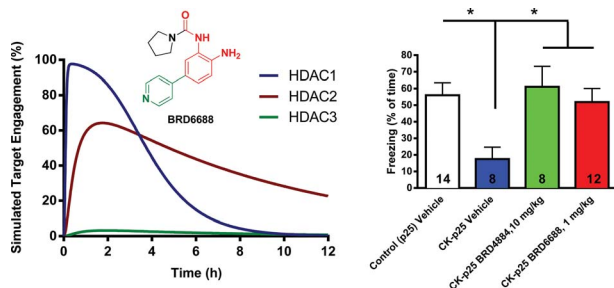


A sialic acid-targeted near-infrared theranostic for signal activation based intraoperative tumor ablation

Xuanjun Wu, Mingzhu Yu, Bijuan Lin, Hongjie Xing, Jiahuai Han and Shoufa Han*

A sialic acid-targeted near-infrared profluorophore with pH-responsive fluorescence and photothermal properties was developed for fluorescence-guided staging and photothermal therapy of viable tumors exposed during surgery.

804



Kinetically selective inhibitors of histone deacetylase 2 (HDAC2) as cognition enhancers

F. F. Wagner, Y.-L. Zhang, D. M. Fass, N. Joseph, J. P. Gale, M. Weiwert, P. McCarren, S. L. Fisher, T. Kaya, W.-N. Zhao, S. A. Reis, K. M. Hennig, M. Thomas, B. C. Lemerrier, M. C. Lewis, J. S. Guan, M. P. Moyer, E. Scolnick, S. J. Haggarty, L.-H. Tsai and E. B. Holson*

Kinetically selective inhibitors of HDAC2 enhanced learning and memory in a CK-p25 mouse model of neurodegeneration.



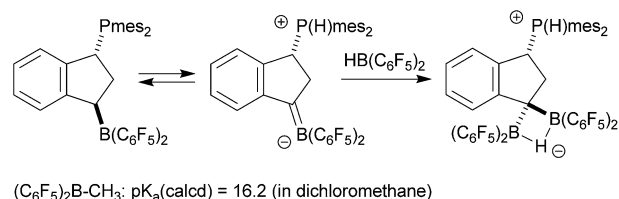
EDGE ARTICLES

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 α -CH acidity of alkyl-B(C₆F₅)₂ compounds – the role of stabilized borata-alkene formation in frustrated Lewis pair chemistry

Philip Moquist, Guo-Qiang Chen,
Christian Mück-Lichtenfeld, Kathrin Bussmann,
Constantin G. Daniliuc, Gerald Kehr and Gerhard Erker*

Alkyl-B(C₆F₅)₂ boranes are markedly α -CH-acidic. Using DFT we have calculated the pK_a -values of a series of examples. Typically, (C₆F₅)₂B-CH₃ [pK_a (calcd) = 18.3 in DMSO, 16.2 in dichloromethane] is almost as CH-acidic as cyclopentadiene.

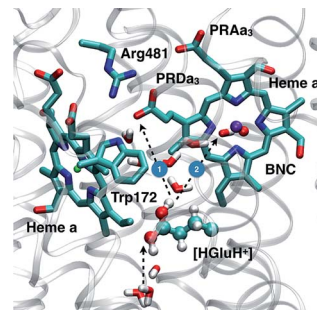


826

Microscopic basis for kinetic gating in cytochrome c oxidase: insights from QM/MM analysis

Puja Goyal, Shuo Yang and Qiang Cui*

Understanding the mechanism of vectorial proton pumping in biomolecules requires establishing the microscopic basis for the regulation of both thermodynamic and kinetic features of the relevant proton transfer steps.



CORRECTION

842

Correction: Pattern-based sensing of aminoglycosides with fluorescent amphiphiles

Ziya Köstereli, Rosario Scopelliti and Kay Severin*

