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ISSN 1359–7345 CODEN CHCOFS (37) 4369–4508 (2008)

Cover
See Holm Petzold and Peter J. Sadler, pp. 4413–4415. The antioxidant glutathione induces formation of this oxidised ruthenium arene complex in water – as unexpected as square ripples! Image reproduced by permission of Holm Petzold and Peter J. Sadler from *Chem. Commun.*, 2008, 4413.

Inside cover
See Adrian J. Mulholland *et al.*, pp. 4425–4427. The trisaccharide NAM-NAG-NAM bound covalently to Asp52 in the active site of hen egg white lysozyme, from QM/MM simulations. Image reproduced by permission of Anna L. Bowman, Ian M. Grant and Adrian J. Mulholland from *Chem. Commun*, 2008, 4425

CHEMICAL BIOLOGY

B73

Drawing together research highlights and news from all RSC publications, *Chemical Biology* provides a 'snapshot' of the latest developments in chemical biology, showcasing newsworthy articles and significant scientific advances.

Chemical Biology

October 2008/Volume 3/Issue 10

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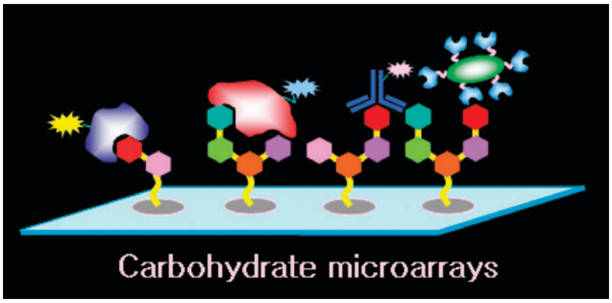
FEATURE ARTICLE

4389

Carbohydrate microarrays as powerful tools in studies of carbohydrate-mediated biological processes

Sungjin Park, Myung-Ryul Lee and Injae Shin*

Methods for the preparation of carbohydrate microarrays and their applications in biological and biomedical research are reviewed.



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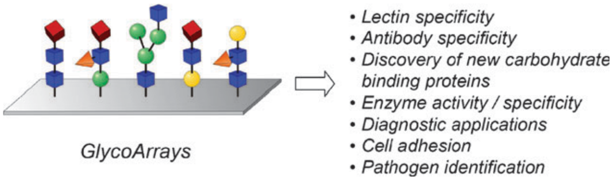
FEATURE ARTICLE

4400

Glycoarrays—tools for determining protein–carbohydrate interactions and glycoenzyme specificity

Nicolas Laurent, Josef Voglmeir and Sabine L. Flitsch*

This Feature Article reviews the recent developments in carbohydrate microarray fabrication and discusses a number of biologically-relevant applications of such tools in glycomics.



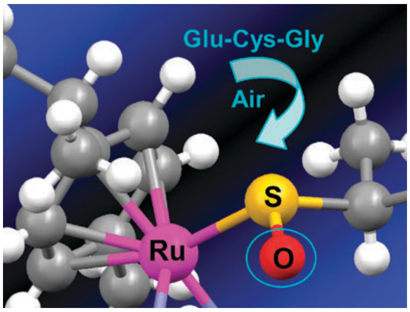
COMMUNICATIONS

4413

Oxidation induced by the antioxidant glutathione (GSH)

Holm Petzold and Peter J. Sadler*

Surprisingly, the well known antioxidant and abundant cellular tripeptide glutathione can promote oxygen atom transfer to organometallic half-sandwich ruthenium arene thiolato complexes of anticancer interest.

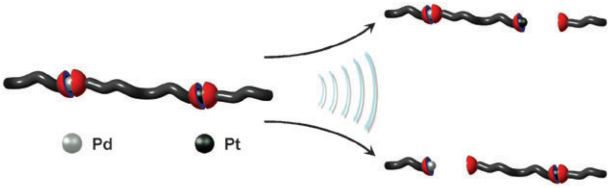


4416

Selectivity of mechanochemical chain scission in mixed palladium(II) and platinum(II) coordination polymers

Jos M. J. Paulusse and Rint P. Sijbesma*

The relative strength of coordinative bonds was studied by ultrasonic scission of coordination polymers containing Pd–phosphine and Pt–phosphine bonds in series.

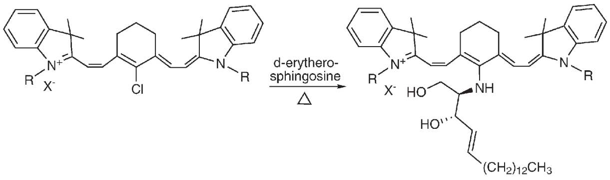


4419

Microwave-assisted synthesis of near-infrared fluorescent sphingosine derivatives

Kumar R. Bhushan, Fangbing Liu, Preeti Misra and John V. Frangioni*

Microwave-assisted synthesis of near-infrared fluorescent sphingosine derivatives is described, along with demonstration of product subcellular localization.



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Sir Harold Kroto, Florida State University
Klaus Müllen, Max-Planck Institute for Polymer Research
Sir J Fraser Stoddart, Northwestern University
Vivian W W Yam, The University of Hong Kong
Richard N Zare, Stanford University

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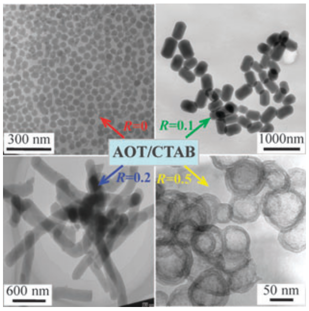
4422

Fine control over the morphology and structure of mesoporous silica nanomaterials by a dual-templating approach

Hongmin Chen and Junhui He*

Mesoporous silicas with varied morphologies and pore structures, including nanospheres, nanoellipsoids, helical nanorods and multi-lamellar nanovesicles, were synthesized by using cetyltrimethylammonium bromide (CTAB) and sodium bis(2-ethylhexyl) sulfosuccinate (AOT) as co-templates.

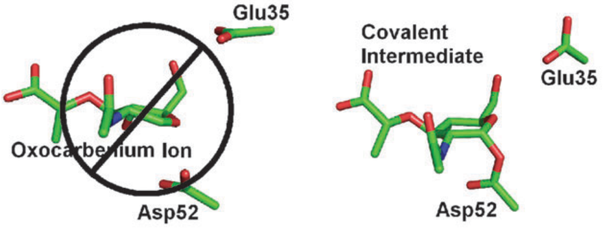


4425

QM/MM simulations predict a covalent intermediate in the hen egg white lysozyme reaction with its natural substrate

Anna L. Bowman, Ian M. Grant and
Adrian J. Mulholland*

Quantum mechanics/molecular mechanics molecular dynamics simulations indicate that the reaction of native HEWL with its natural substrate involves a covalent intermediate, in contrast to the 'textbook' mechanism for this seminal enzyme.

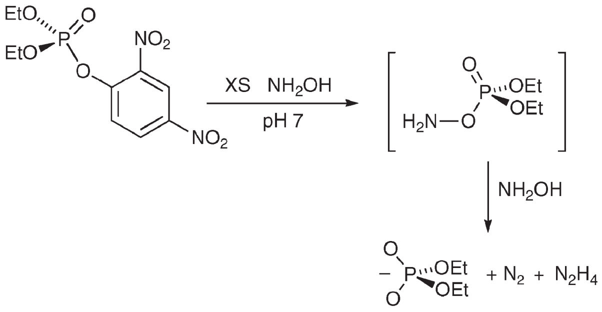


4428

Hydroxylamine as an oxygen nucleophile. Chemical evidence from its reaction with a phosphate triester

Anthony J. Kirby, Bruno S. Souza, Michelle Medeiros, Jacks P. Priebe, Alex M. Manfredi and Faruk Nome*

Hydrazine is produced when hydroxylamine reacts with an activated phosphate triester, providing direct chemical evidence for an intermediate phosphorylated on oxygen.

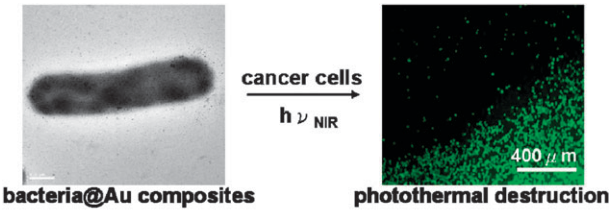


4430

Biocompatible bacteria@Au composites for application in the photothermal destruction of cancer cells

Wen-Shuo Kuo, Ching-Ming Wu, Zih-Syuan Yang, Szu-Yu Chen, Cheng-Ying Chen, Chih-Chia Huang, Wei-Ming Li, Chi-Kuang Sun and Chen-Sheng Yeh*

Live gold nanoshell-coated bacteria (bacteria@Au) served as photothermal therapeutic agents and were conjugated with anti-EGFR antibodies to destroy cancerous cells with a NIR laser.



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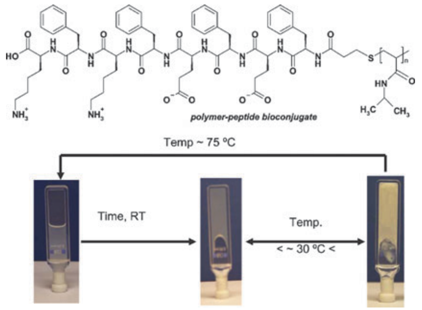
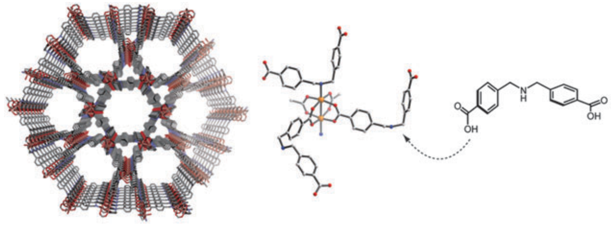


Selective synthesis of double temperature-sensitive polymer–peptide conjugates

Florentina Stoica, Cameron Alexander, Nicola Tirelli, Aline F. Miller and Alberto Saiani*

A novel synthetic route has been developed to couple selectively a modified octa-peptide, that is able to gel at low temperature, to the prototypical thermoresponsive polymer poly(*N*-isopropylacrylamide) to give a bioconjugate that exhibits double thermoresponsiveness.

4436

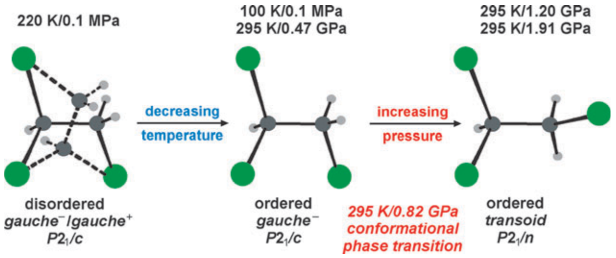


Kagomé type extra-large microporous solid based on a paddle-wheel Cu^{2+} dimer

Satoshi Horike, Shinpei Hasegawa, Daisuke Tanaka, Masakazu Higuchi and Susumu Kitagawa*

A robust extra-large microporous coordination polymer with a Kagomé type structure was synthesized by carboxylate/amine multifunctional ligand with a Cu^{2+} cluster and the hexagonal 1D channels (~15 Å) showed type I and IV adsorption isotherms for gas molecules.

4439

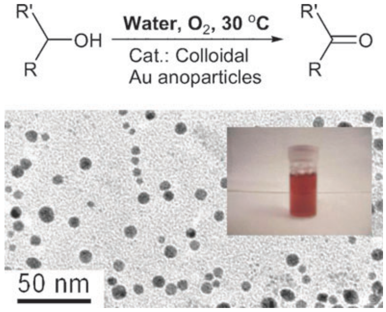


Energetics of conformational conversion between 1,1,2-trichloroethane polymorphs

Maciej Bujak, Marcin Podsiadło and Andrzej Katrusiak*

Pressure-induced transformations between *gauche*$^-$, *gauche*$^+$ and *transoid* conformations have been evidenced by X-ray single-crystal diffraction for 1,1,2-trichloroethane, and the energies of intermolecular interactions, conformational conversion, and the latent heat have been determined.

4442



Amphiphilic block copolymer-stabilized gold nanoparticles for aerobic oxidation of alcohols in aqueous solution

Xueguang Wang, Hajime Kawanami,* Nazrul M. Islam, Maya Chattergee, Toshirou Yokoyama and Yutaka Ikushima

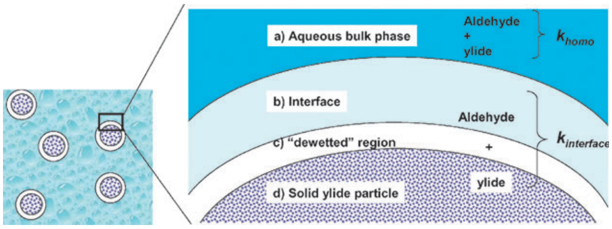
Stable colloidal Au nanoparticles in P123 aqueous solution were synthesized simply and were successfully applied for the oxidation of alcohols using O_2 at 30 °C in aqueous solution.

4445

Unusual temperature dependence of salt effects for "on water" Wittig reaction: hydrophobicity at the interface

Shraeddha Tiwari and Anil Kumar*

An unusual variation with temperature of the salt effects in aqueous Wittig reaction is observed, suggesting hydrophobic acceleration of reactions comprising of "on water" reactants is fundamentally different from that for reactions with small non-polar solutes dissolved in water.

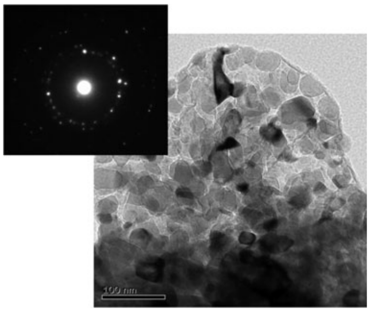


4448

Temporal and spatial imaging of hydrogen storage materials: watching solvent and hydrogen desorption from aluminium hydride by transmission electron microscopy

Shane D. Beattie,* Terry Humphries, Louise Weaver and G. Sean McGrady*

An *in situ* thermal desorption study of solvated aluminum hydride (alane) by transmission electron microscopy and selected area diffraction has permitted characterisation of the structural and morphological changes during desorption of solvent and hydrogen in real-time.

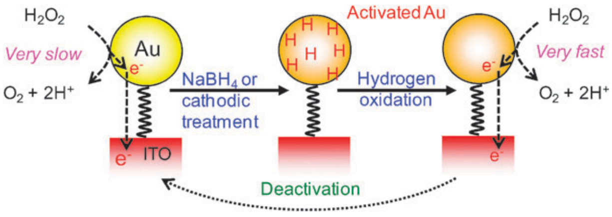


4451

Enhancement of the electrocatalytic activity of gold nanoparticles *via* $NaBH_4$ treatment

Jagotamoy Das, Srikanta Patra and Haesik Yang*

We report on the enhancement of the electrocatalytic activity of Au nanoparticles after $NaBH_4$ treatment and its application to H_2O_2 detection.

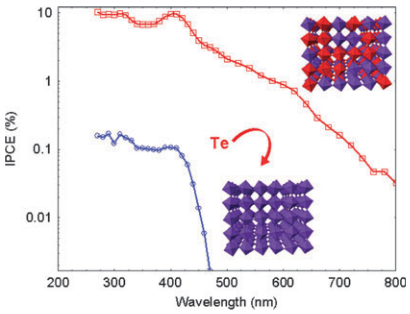


4454

Enhanced long-wavelength transient photoresponsiveness of WO_3 induced by tellurium doping

Bin Yang and Vittorio Luca*

Tungsten trioxide (WO_3) films doped with 0.25 atom% tellurium synthesised by a sol–gel route, show strong transient photocurrents under chopped sub-bandgap illumination ($h\nu$ = 1.8 eV < E_g) at low bias potentials from 0.2 to 0.7 V. Such effects are ascribed to the presence of a localized narrow band (NB) between the VB and the CB in this material.



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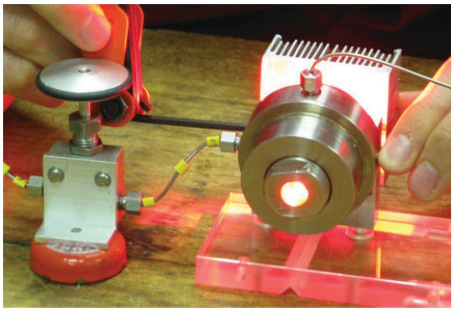
4457

Homogeneous photochemical oxidation *via* singlet O_2 in supercritical CO_2

Richard A. Bourne, Xue Han, Adrian O. Chapman,
Nicholas J. Arrowsmith, Hajime Kawanami,
Martyn Poliakoff and Michael W. George*

Singlet O_2 is generated with a CO_2-philic photosensitiser and used to convert α-terpinene to ascaridole cleanly and rapidly in supercritical CO_2.

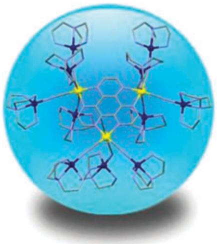


4460

Cyanide-bridged tetradecanuclear $Ru^{II}{}_3M^{II}{}_{11}$ clusters (M^{II} = Zn^{II} and Cu^{II}) based on the high connectivity building block $[Ru_3(HAT)(CN)_{12}]^{6-}$: structural and photophysical properties

Juan Manuel Herrera,* Enrique Colacio,
Corine Mathonière, Duane Choquesillo-Lazarte and
Michael D. Ward*

Synthetic, structural and photophysical properties of two new cyanide-bridged tetradecanuclear $Ru^{II}{}_3M^{II}{}_{11}$ clusters (M^{II} = Zn^{II}, Cu^{II}) are reported.



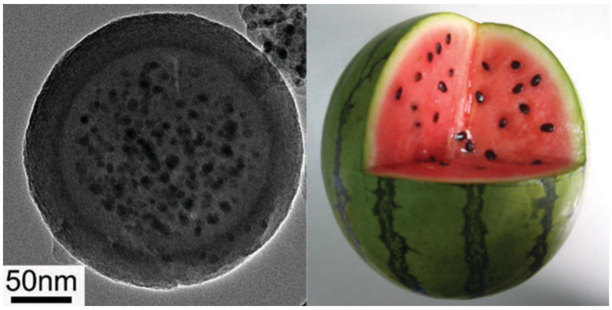
$[\{M^{II}(tren)\text{-}CN\}_{11}Ru_3(HAT)(CN)]^{16+}$
($M = Zn^{II}, Cu^{II}$)

4463

A facile approach to fabricate functionalized superparamagnetic copolymer-silica nanocomposite spheres

Dechao Niu, Yongsheng Li,* Xiaoli Qiao, Liang Li,
Wenru Zhao, Hangrong Chen, Qiaoling Zhao, Zhi Ma and
Jianlin Shi*

Watermelon-structured amino- or thiol-functionalized superparamagnetic copolymer-silica nanospheres (NH_2-SMCSNs/SH-SMCSNs), which consist of a magnetic core and a silica cross-linked block copolymer shell, have been fabricated.

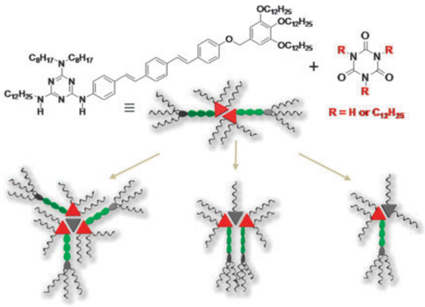


4466

Cyanurate-guided self-assembly of a melamine-capped oligo(*p*-phenylenevinylene)

Shiki Yagai,* Shun Kubota, Kanako Unoike,
Takashi Karatsu and Akihide Kitamura

An oligo(*p*-phenylenevinylene) capped on one end by a monotopic triple hydrogen-bonding module shows distinct optical properties and self-organization behavior upon complexation with cyanurates with different numbers of triple hydrogen-bonding sites.



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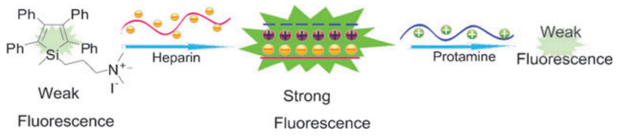
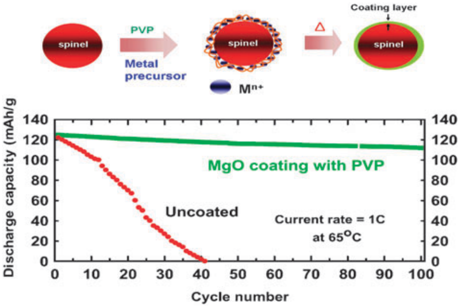


The convenient fluorescence turn-on detection of heparin with a silole derivative featuring an ammonium group

Ming Wang, Deqing Zhang,* Guanxin Zhang and Daoben Zhu*

Linear fluorescence enhancement was observed for a silole with an ammonium group upon addition of heparin, and accordingly the convenient fluorescence detection of heparin was established.

4472

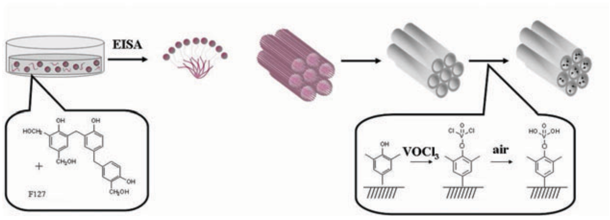


PVP-functionalized nanometre scale metal oxide coatings for cathode materials: successful application to $LiMn_2O_4$ spinel nanoparticles

Sunhye Lim and Jaephil Cho

PVP functionalized metal oxide coatings on spinel nanoparticles demonstrated significantly improved rate characteristics under extensive cycling at 65 °C and exhibited over 100% improved capacity retention compared to the bare counterpart.

4475

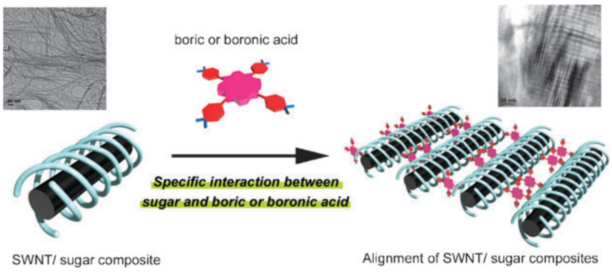


Ultra stable ordered mesoporous phenol/formaldehyde polymers as a heterogeneous support for vanadium oxide

Ilke Muylaert,* Marijke Borgers, Els Bruneel, Joseph Schaubroeck, Francis Verpoort and Pascal Van Der Voort

Ordered mesoporous phenol/formaldehyde polymers are presented as a novel support for vanadium oxide. The high stability of the support, the presence of tetrahedral vanadium oxide monomers on the surface and a strongly reduced leaching behaviour of the catalytic species opens up new and straightforward prospects for heterogeneous liquid phase catalysis.

4478



Hierarchical carbon nanotube assemblies created by sugar–boric or boronic acid interactions

Shingo Tamesue, Munenori Numata, Kenji Kaneko, Tony D. James and Seiji Shinkai*

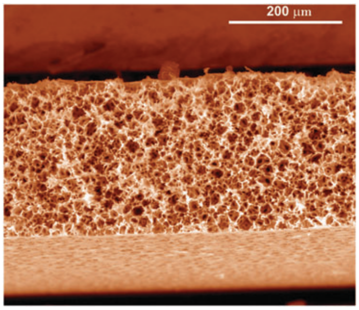
Polysaccharide "schizophyllan (SPG)" can entrap as-grown and cut single-walled carbon nanotubes (SWNTs). We here report that the SWNT–SPG composites can be aligned regularly using the covalent bond formation between boric acid or boronic acid derivatives and the 4,6-dihydroxyl group of the glucose side-chain unit of SPG.

4481

Open cellular reactive porous membranes from high internal phase emulsions

Irena Pulko and Peter Krajnc*

Porous membranes with open cellular architecture and thicknesses between 30 and 500 μm were prepared by applying high internal phase emulsions (HIPEs) incorporating styrene, 4-vinylbenzyl chloride, divinylbenzene and ethylhexyl acrylate.

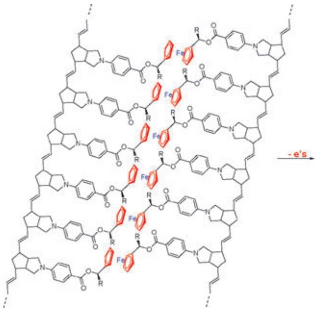


4484

Electrochemical oxidation of double-stranded polybisnorbornenes containing linearly aligned ferrocene linkers

Cheng-Lan Lin, Hui-Chun Yang, Nai-Ti Lin, I-Jui Hsu, Yu Wang* and Tien-Yau Luh*

Seventy percent of the ferrocene moieties in double-stranded polybisnorbornenes are electrochemically oxidised to antiferromagnetic products. Strong interactions between neighbouring units in these polymers are suggested.



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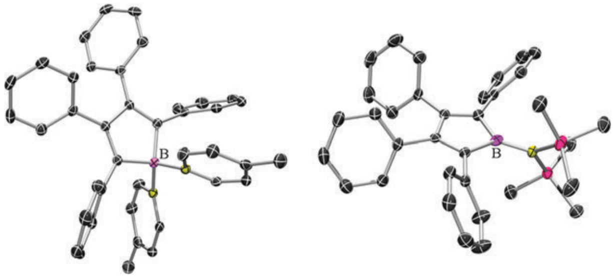
Direct functionalization at the boron center of antiaromatic chloroborole

Holger Braunschweig* and Thomas Kupfer

The direct functionalization of the boron center in antiaromatic chloroborole $ClBC_4Ph_4$ represents a potential new approach toward the introduction of borole moieties into the backbone of organic π-conjugated frameworks.

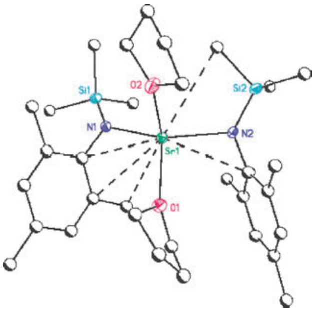


4490

$BiPh_3$—A convenient synthon for heavy alkaline-earth metal amides

Miriam M. Gillett-Kunnath, Jonathan G. MacLellan, Craig M. Forsyth, Philip C. Andrews,* Glen B. Deacon* and Karin Ruhlandt-Senge*

An environmentally benign redox transmetallation ligand exchange route provides a facile synthesis of molecular alkaline-earth metal organoamides.



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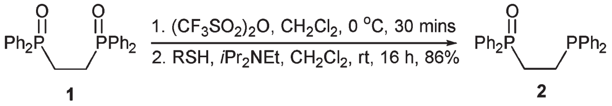
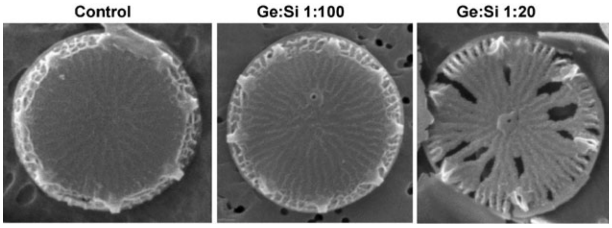


Selective mono reduction of bis-phosphine oxides under mild conditions

Maria J. Petersson, Wendy A. Loughlin* and Ian D. Jenkins*

Bis-phosphine oxides can be selectively reduced to bis-phosphine monoxides under exceptionally mild conditions using triflic anhydride and a thiol.

4495

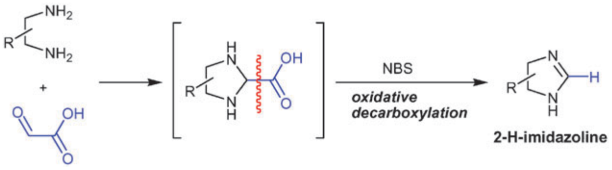


A self-propagating system for Ge incorporation into nanostructured silica

Aubrey K. Davis and Mark Hildebrand*

Technologically-relevant levels of Ge can be incorporated into cell wall silica of the diatom *Thalassiosira pseudonana* with no aberration in structure at low levels, whereas higher levels alter structure.

4498

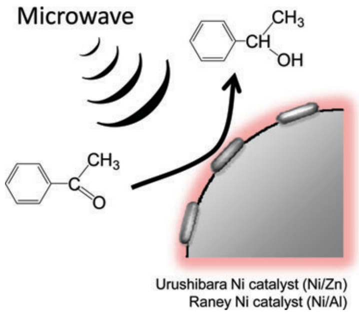


Oxidative decarboxylative synthesis of 2-H-imidazolines from glyoxylic acid and 1,2-diamines

Kenichi Murai, Maiko Morishita, Ryo Nakatani, Hiromichi Fujioka* and Yasuyuki Kita*

A novel method to prepare 2-H-imidazolines from glyoxylic acid monohydrate and 1,2-diamines is described. The key reaction of this method is the oxidative decarboxylation of the 2-carboxy imidazolidines by NBS.

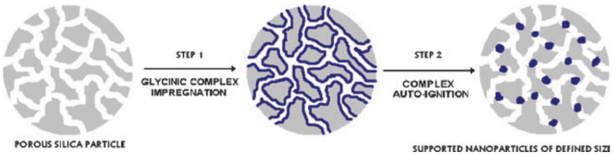
4501



Microwave effect on the surface composition of the Urushibara Ni hydrogenation catalyst and improved reduction of acetophenone

Satoshi Horikoshi,* Junichi Tsuzuki, Futoshi Sakai, Masatsugu Kajitani and Nick Serpone*

The microwave thermal effect for a heterogeneous catalysis reaction was examined for the hydrogenation of acetophenone with Urushibara and Raney-Ni catalysts.


POROUS SILICA PARTICLE
STEP 1
GLYCINIC COMPLEX IMPREGNATION
STEP 2
COMPLEX AUTO-IGNITION
SUPPORTED NANOPARTICLES OF DEFINED SIZE

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Chemical Biology

Synthetic lesions added to DNA to probe its stability

Discovering the secrets of DNA repair

ISTOCKPHOTOS

Fixing damaged DNA: if only it were this simple...

A modified DNA is helping scientists to understand the sophisticated DNA repair mechanisms that allow dormant bacteria to come 'back to life'.

Thomas Carell and Eva Bürckstümmer at the Ludwig Maximilian University of Munich, Germany, have made short DNA strands containing lesions. Carell explains that this is the key to understanding DNA repair. 'So far any study of this enigmatic process has been hampered by a lack of DNA containing this lesion,' he explains.

The lesions are analogues of those triggered when UV light acts on DNA stored in spores such as the *Bacillus* bacteria spore. In nature, these spores can lie dormant for many years, storing DNA, but then return to life, explains Carell. How spores store DNA and how lesion repair occurs are the questions the German duo would like to answer.

Carell and Bürckstümmer made their DNA strands by synthesising two isomers of a dinucleotide lesion analogue and incorporating them into DNA. They found that one DNA was more stable than the other, suggesting that the natural lesion could have a similar structure to the analogue in the more stable DNA. Carell points out that similar lesion analogues are substrates for the spore DNA repair enzyme so that the new strands could help further studies into the enzyme mechanism.

Glen Burley, an expert in DNA nanotechnology at the University of Leicester, UK, says that the work is exciting as it provides a method for investigating how spores repair damaged DNA. 'This is a compelling question as DNA damage processes in spores differ from those in mammals,' he says. 'These methods would likely lead to a greater understanding of how spores can survive for long periods and in hostile conditions – for example hot springs.'

Carell explains that although the repair process in the spore is unique, lesion recognition by enzymes is more general. 'Such enzymes are also operating in our cells,' he says, 'so a deeper understanding of this class of enzymes is desperately needed.' Carell adds that he is particularly interested in learning more about failures in repair. 'These are responsible for mutations which in turn lead to dangerous cellular situations which might produce cancer,' he says. *Katherine Davies*

Reference

E Bürckstümmer and T Carell, *Chem. Commun.*, 2008, 4037 (DOI: 10.1039/b810008j)

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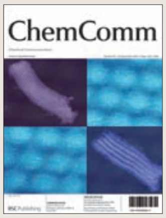
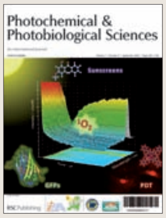
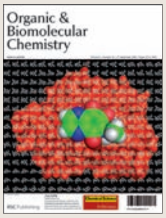
Nature's fruitful chemistry

This month's Instant insight examines why the changing colour of autumn leaves could be good news for your health







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Research highlights

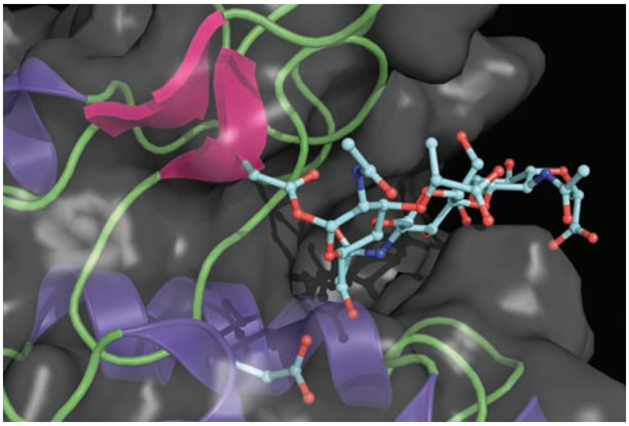
Calculations validate alternative to much-published enzyme mechanism

Rewriting the biochemistry textbooks...

Hen egg white lysozyme is a common example in textbooks discussing enzyme mechanism. But now scientists from the University of Bristol, UK, have used molecular dynamics simulations to show that the traditional mechanism is wrong. 'The textbooks need to be rewritten,' says researcher Adrian Mulholland, who led the team behind the work.

Lysozymes break down polysaccharides in bacterial cell walls, and so play a role in defence against pathogens. The textbook mechanism of hen egg white lysozyme proceeds through an intermediate in which a sugar ring on the substrate interacts ionically with the enzyme. In the revised mechanism the bond is covalent.

'Knowing how reaction intermediates form is central to understanding why enzymes are such efficient catalysts,' says Mulholland. 'This sort of detailed knowledge is also important in designing enzyme inhibitors as drugs,' he adds.



The hen egg white lysozyme mechanism proceeds through a covalent intermediate

Reference
A L Bowman I M Grant and A J Mulholland, *Chem. Commun.*, 2008, DOI: 10.1039/b810099c

A covalent intermediate has been suggested previously, clarifies Mulholland. But the experimental work that led to this proposal relied on modified enzymes and substrates because the wild type enzyme is too efficient for any intermediate to be detected. 'Some people have suggested that the experiments were not relevant to the real target,' Mulholland says.

Mulholland's computational model is based on the wild type enzyme and substrate. Moreover it includes the entire protein as well as its water environment, in contrast to previous smaller models. The evidence from the modelling and the experiments together is enough to confirm the revised mechanism, says Mulholland.

Stephen Withers was involved in the experimental research on hen egg white lysozyme, and is pleased that Mulholland's calculations have confirmed the revised mechanism. The scientist from the University of British Columbia in Vancouver, Canada, welcomes the development of computational studies to supplement experiments. 'It is simply not possible to experimentally probe all proteins,' he says. 'We shall rely on computational approaches increasingly to guide our choice of systems to study experimentally.' *Danièle Gibney*

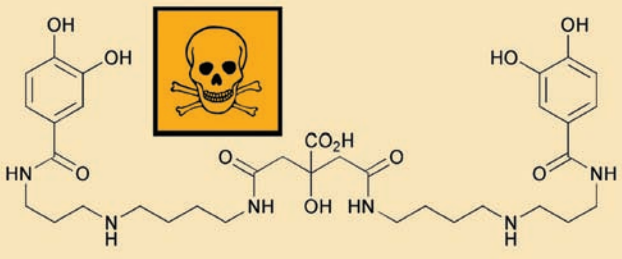
Enzyme study reveals details of potential target in the fight against anthrax

... and revising route to stealthy siderophore

A surprising result has led UK scientists to revise the proposed biosynthetic route to petrobactin – a molecule needed by anthrax-causing bacteria to replicate. Greg Challis at the University of Warwick and colleagues say that their findings may ultimately lead to the development of new antibiotics for the disease.

Petrobactin is an iron scavenger secreted by *Bacillus anthracis* to collect the iron it needs to reproduce. Called a stealth siderophore, it evades the molecule our immune system produces to capture it, contributing to the organism's virulence.

Challis's group has examined the role of the enzyme AsbB in petrobactin's biosynthesis. They first identified possible substrates for AsbB and obtained pure enzyme. Then, by treating the enzyme with different substrate combinations and finding which led to products and how quickly, the researchers showed that a previously proposed intermediate is unlikely to be significant in the pathway.



Petrobactin – the stealth iron scavenger of *Bacillus anthracis*

Reference
D Oves-Costales *et al*, *Chem. Commun.*, 2008, 4034 (DOI: 10.1039/b809353a)

Challis says that he and his team were surprised by the findings, which led them to suggest a revised pathway to petrobactin. They had assumed, based on studies of other siderophore biosynthetic pathways, that the route would be significantly different from the one they now propose.

'Our results provide the basic biochemical knowledge required to screen for inhibitors of AsbB,' says Challis. This could lead to inhibitors for petrobactin biosynthesis and so a potential anthrax treatment, he adds.

The team's results also confirmed earlier computational predictions that enzymes such as AsbB use particular citrate derivatives as substrates, something that Christopher Schofield who investigates biosynthetic enzymes at the University of Oxford, UK, finds interesting. He says that the work 'provides more evidence for the value of chemical insights into bioinformatic analyses' and agrees that it could also one day help provide new antibacterials.

Challis and group member Daniel Oves-Costales were recently awarded a Biotechnology and Biological Sciences Research Council (BBSRC) grant to continue investigating the mechanisms of petrobactin biosynthesis and to search for inhibitors. *Frances Galvin*

Spectroscopic techniques combined for easier metabolomic analysis

Metabolic markers for diabetes

Finding how a trace molecule's concentration varies when it's mixed in a jumble of others is not straightforward but this is just what a metabolomics scientist does every day. Now an international team is combining several analytical techniques to make the process easier and applying its method to study diabetes.

Metabolomics is the study of metabolites – the small molecules formed by specific processes inside a single cell. As variations in levels of some of these may be linked to metabolic disorders such as diabetes, identifying these so-called metabolic markers has the potential to provide diagnostic information and to lead to better therapies.

With this in mind, Geoffrey Gipson at Drexel University, Philadelphia, US, and colleagues in the US and UK, combined spectroscopic methods such as nuclear magnetic resonance (NMR) and liquid chromatography-mass spectrometry (LC-MS) to analyse the metabolites in urine and tissue samples from mice deficient in the hormone protein leptin. Leptin has a key role in regulating metabolism and the leptin-deficient mice display several genetic traits associated with type 2 diabetes. By comparing the metabolite levels with those in control mice, the researchers confirmed that many pathways, including fatty acid metabolism, are altered in 'diabetic' mice. Gipson says that these pathways could potentially be targeted for diabetes treatments.

Leptin-deficient mice (left) show genetic traits associated with diabetes and obesity

ONRL

While Gipson admits that there are limitations in using animals to model human conditions, he explains that metabolism is a fundamental biological process which has been highly conserved over evolutionary time. 'As such, there are many cross-species molecular similarities which allow researchers to gain critical insights into human disease,' he adds. Given this, the biggest challenge, says Gipson, is in applying the new knowledge gained through metabolic profiling research into therapies for patients living with diabetes.

Jules Griffin, an expert in metabolomics at the University of Cambridge, UK, commends the team's integrated approach. 'While NMR has been used widely in mammalian disease models,' says Griffin, 'the use of LC-MS based approaches has lagged behind, in part because of the greater challenges in technical reproducibility. By picking a well-characterised model of type 2 diabetes the team has been able to validate its approach and then extend its analysis to new metabolite changes not previously described.'

Kathleen Too

Reference

G T Gipson *et al*, *Mol. BioSyst.*, 2008, **4**, 1015 (DOI: 10.1039/b807332e)

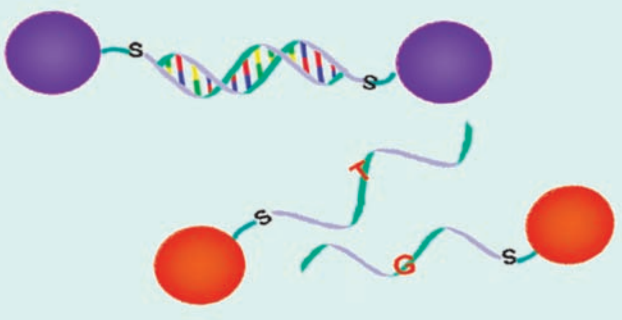
Protein maintains activity at high temperature to identify nucleotide differences

Mismatched DNA analysis by eye

A colourful approach to detecting DNA variations can take the heat. A team led by Changill Ban from Pohang University of Science and Technology and Min Su Han from Chuang-Ang University, Seoul, in South Korea, has tested a technique that reveals mismatched DNA base pairs by a temperature change.

The team's method detects DNA in which one base is paired with any base that is not its complementary partner. Mismatch detection can be used to find single base differences called single nucleotide polymorphisms (SNPs). Rapid SNP detection is essential, says Ban, as these DNA variations can be a marker for genetic disease.

The team bound gold nanoparticles to single stranded DNA which then pairs to form duplex DNA containing mismatches. As the DNA pairs, the system turns from red to purple as the nanoparticles aggregate. On heating, the system reverts to red as the strands unpair or 'melt'. The researchers observed that the colour change occurs at a higher temperature when the system is treated with MutS, a protein that binds selectively to mismatched pairs. They also found that complexes containing different DNA mismatches melt at different temperatures, so it is possible to identify which bases are mismatched.



When gold-bound DNA strands unpair the system changes from purple (top) to red (bottom) as the gold de-aggregates

Ban says that, as a colorimetric assay, the system is 'simpler and more convenient than other methods, allowing detection with the naked eye.' Most current colorimetric detection methods rely on enzyme activity, requiring physiological conditions to work. By using MutS, the team avoids this limitation, as the protein maintains its activity under a range of pHs and at high temperatures.

The next stage will be to adapt the method for microarray technology, says Ban, allowing high throughput screening of DNA samples.

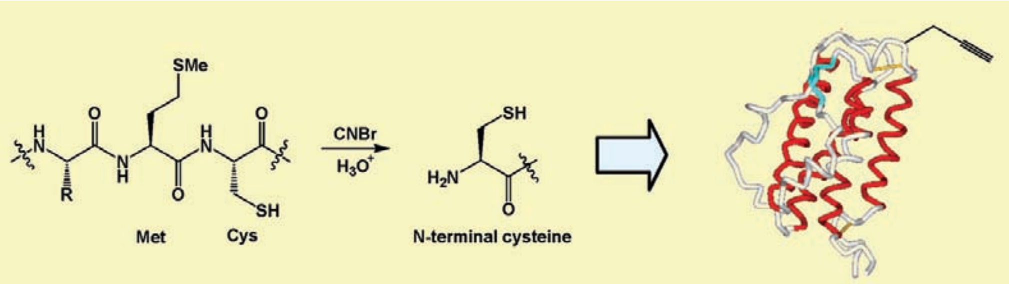
Harriet Brewerton

Reference

M Cho, M S Han and C Ban, *Chem. Commun.*, 2008, DOI: 10.1039/b811346g

Versatile protein synthesis optimised to make biologically active target

Mix and match protein building blocks



Cyanogen bromide is used to cleave a protein next to a methionine residue to give a protein fragment for NCL

An improved procedure for breaking proteins in two is allowing chemists to create new proteins. Derek Macmillan and Jonathan Richardson from University College London, UK, have used their method to make a biologically active protein.

Low reaction yields make creating large proteins by chemical synthesis unfeasible, yet not all proteins can be prepared easily by other routes. Native chemical ligation (NCL) is one technique that chemists can use to create these proteins by joining a small peptide containing a thioester to a protein fragment with an N-terminal cysteine. In this way troublesome proteins can be created without having to synthesise the whole protein from scratch.

The advantages of NCL are its 'versatility and reliability', explains Macmillan. 'The reaction is conducted in aqueous solution with no protecting groups, and the product has a native peptide bond,' he adds.

But whilst the small peptides required for NCL can be made by peptide synthesis, preparing the large protein fragment is often more problematic and poses many of the same problems associated with creating whole proteins. Now, Macmillan and Richardson have optimised their procedure for generating these fragments. The starting, bacterially-derived protein is cleaved next to a methionine to give two fragments, one with the cysteine at its N-terminus. The cysteine-containing fragment is then isolated and combined with a synthetic peptide containing a thioester to create a whole, semi-synthetic protein.

Using their optimised procedure Macmillan and Richardson were able to create a biologically active variant of erythropoietin, a hormone protein involved in red blood cell production which is used in the treatment of anaemia. Commenting on this success, Phil Dawson an expert in synthetic protein chemistry at The Scripps Research Institute, La Jolla, US, says that 'the functional complexity of large proteins can turn even the most straightforward procedure into a daunting challenge. The Macmillan lab has taken an important first step towards establishing a robust semi-synthesis for an important therapeutic target.'

Russell Johnson

Reference

J P Richardson and D Macmillan, *Org. Biomol. Chem.*, 2008, DOI: 10.1039/b811501j

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Imaging of essential and toxic elements in biological tissues by LA-ICP-MS
J Sabine Becker *et al*, *J. Anal. At. Spectrom.*, 2008, **23**, 1275 (DOI: 10.1039/b805228j)

Protein modification for single molecule fluorescence microscopy
Mark S Dillingham and Mark I Wallace, *Org. Biomol. Chem.*, 2008, **6**, 3031 (DOI: 10.1039/b808552h)

The use of gold nanoparticles in diagnostics and detection
Robert Wilson, *Chem. Soc. Rev.*, 2008, **37**, 2028 (DOI: 10.1039/b712179m)

Modeling the reactive properties of tandemly activated tRNAs
Maria Duca *et al*, *Org. Biomol. Chem.*, 2008, **6**, 3292 (DOI: 10.1039/b806790b)

Probing (macro)molecular transport through cell walls
Giona Kilcher *et al*, *Faraday Discuss.*, 2008, **139**, 199 (DOI: 10.1039/b717840a)

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Mapping out success

OBC lecture award winner, Akimitsu Okamoto, talks about chemical probes and daydreaming over maps. Vikki Allen meets him in Pittsburgh to find out more

Akimitsu Okamoto

Akimitsu Okamoto is leader of the Okamoto Initiative Research Unit at RIKEN Advanced Science Institute. He is the 2008 recipient of the *Organic & Biomolecular Chemistry* lecture award. His primary research interests focus on the design and synthesis of biopolymers, and recognising and visualising single components or atoms in biopolymers.

What inspired you to become a chemist?

I grew up with many empty chemical boxes around the house. My father used to work in a trading company that handled chemicals for the porcelain and chinaware that Nagoya is known for and he used to bring the empty boxes home. I became interested in these boxes; I wanted to know what had been inside them, and what the chemicals had looked like.

What motivated you to specialise in bioorganic/biological chemistry?

I started out in graduate school in synthetic organic chemistry, working on natural products, so I made many compounds. Such compounds are often used in biotechnology. Choosing biological chemistry wasn't a conscious decision. The department that I studied at was very small and gave a good basic range of courses, but I found the 'bio' courses the most interesting and they introduced me to DNA. Bioorganic chemistry sounds good, too! And bioorganic chemistry is such an important field, bridging chemistry and biology. Since then, my research has changed gradually towards biological chemistry.

I began my independent career nine years ago, initially in collaboration with Professor Isao Saito, who worked at Kyoto University at the time. The projects that we started then led me into the research that I now do.

What hot project are you working on at the moment?

I want to see living cells working. I want to see DNA and RNA working. So in my laboratory, we make lots of nucleic acid probes – fluorescent probes, chemical probes. There are a number of protein probes but we don't yet have any really good methods to look at RNA and DNA.

We are working on a number of projects to look at living cells – stem cells – and are trying to understand the mechanism by which they work. What is it that causes them to live, and to become skin or hair? What is the 'life' in these cells? What roles do RNA and DNA play? It's quite a fundamental project.

What do you love most about your job?

I enjoy my job. There is a lot of work to do in the DNA and RNA field using our chemical probes. The probes work really well, so I like to know that we can use our probes to look at problems and that they are well designed.

You are this year's recipient of the *OBC* lecture award. How do you feel about the award?

I'm happy, very happy. Of course it is good to know that my work has been appreciated. I would very much like to thank my collaborators and laboratory co-workers. The topic of my award lecture will focus on one particular example of the work we do looking at the use of osmium to investigate cells. It's very interesting.

Which scientist from the past would you most like to meet and why?

This is a really difficult question. I respect all scientists that are dedicated and work hard every day, those who try. Whether you get a really good result depends on if you are lucky or not. I would not be able to choose just one person in particular.

What do you do when you are not working?

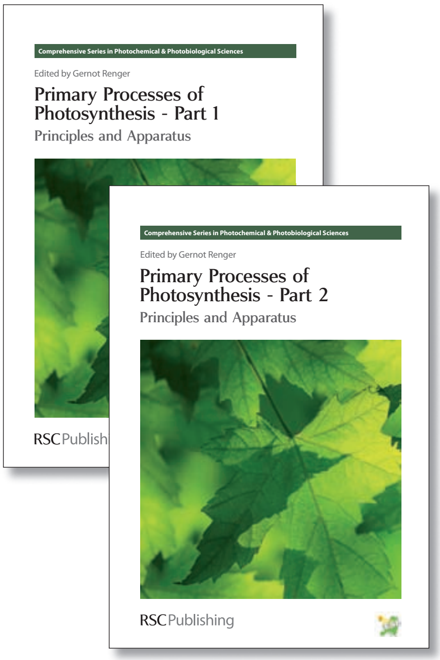
I like going to book stores. And I really love maps. I don't have enough time to travel, so I like to read the maps of places that I would like to visit instead. I always buy maps when I do get to travel. Last week, I was giving talks in the Czech Republic and so I bought a country map. I also recently went to Mexico and have a Mexican map too.

I particularly like the world map. Czech maps and Spanish maps show you the language differences and reflect the different cultures of countries. Sometimes, maps can be funny too. I have an Australian world map that is upside down. The south is at the top of the map and north towards the bottom. I think it's a special map though!

I would love to visit everywhere, but my first ever foreign holiday was to the UK when I was a graduate and I would love to go back. I visited London and Stonehenge.

And finally, if you weren't a scientist what would you be?

When I was an undergraduate, I got a licence to act as a travel agent in Japan. I wanted to be a tour organiser and travel around with groups of people exploring places. I like planning travel, but then I started going into the chemical labs every day and just didn't have the time to do this as well. So, if I didn't do chemistry I would be a tour operator.



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Instant insight

Nature's fruitful chemistry

Bernhard Kräutler and Thomas Müller at the University of Innsbruck in Austria explain why the changing colour of autumn leaves could be good news for your health

How green plant pigments disappear in the autumn, when the colours of the leaves of deciduous trees and shrubs change from green to red and yellow, has been a longstanding puzzle. While chlorophyll biosynthesis has been well studied, how chlorophyll breaks down remained a fascinating enigma until about 17 years ago. This lack of basic knowledge is all the more surprising, as chlorophyll metabolism is probably the most visible manifestation of life on Earth. In fact, it is even seen from outer space, and the total annual turnover of chlorophyll has been estimated to involve more than 1000 million tons.

In 1991 a colorless chlorophyll catabolite from senescent (aging) plant leaves was identified as a linear tetrapyrrole, which turned out to be distantly related to bilirubin and phytobilins, products of heme breakdown.[1] Since then, chlorophyll breakdown products have been identified in a variety of plant leaves, and their structural features revealed. According to these studies, chlorophyll breakdown in higher plants leads first to coloured compounds – as transient, enzyme-bound intermediates only. In a later stage, fluorescent catabolites occur merely fleetingly, and colourless, nonfluorescent tetrapyrrolic catabolites are formed rapidly. The latter accumulate in de-greening leaves and are considered the final products of chlorophyll breakdown, which apparently is a well controlled program for rapid detoxification of the photoactive green plant pigment.

But chlorophyll breakdown is not merely a detoxification process for the plant. It has also been associated with recycling of important nutrients, of reduced nitrogen, in particular. In the case of the catabolites, the four chlorophyll nitrogen atoms remain in the known tetrapyrrolic breakdown products and are thus not available for the plant to re-use. Nevertheless, they will eventually become part of more global recycling, possibly involving lower organisms.

Recently, we addressed the puzzle of chlorophyll breakdown in ripening fruit. In freshly ripe apples and pears the very same linear tetrapyrroles were detected as chlorophyll catabolites. In fact, these breakdown products were also identical to the ones found in the de-greening leaves of the pear tree. Accordingly, chlorophyll breakdown appears to take a common pathway in fruit ripening and leaf senescence. As senescence is considered to accompany programmed cell death – yet ripening, commonly, is not – this finding is remarkable indeed.

In fruit, the tetrapyrrolic chlorophyll catabolites become a part of traditional food and may be a positive component in our diet as they are effective antioxidants. Exploratory studies have pointed to their beneficial health effects in mammals. Thus, the occurrence of such chlorophyll remains in apple peels may give a new twist to the old Welsh saying 'An apple a day, keeps the doctor away'.

140 years after Gregor Mendel used de-greening in peas as part of his experiments to establish the laws of inheritance, the basis for his observation is now known to be genetic control of chlorophyll breakdown. While the process may no longer be a total enigma, however, its wide-reaching benefits remain a field ripe for investigation.[2]

Read more in Bernhard Kräutler's perspective in Photochemical and Photobiological Sciences.

Catabolites formed as fruit and leaves change colour may be valuable nutritional components

References

1 B Kräutler *et al*, *Angew. Chem., Int. Ed.*, 1991, **30**, 1315

2 B Kräutler, *Photochem. Photobiol. Sci.*, 2008, DOI: 10.1039/b802356p

Essential elements

Engineering success

CrystEngComm celebrated its tenth year of publication in style on 28 August with a lunch reception held at the XXI Congress and General Assembly of the International Union of Crystallography in Osaka, Japan. As part of the celebrations, the journal also awarded five poster prizes at the meeting.

Since its launch in 1999, *CrystEngComm* has gone from strength to strength, growing in size by more than a factor of ten. The journal now boasts the fastest publication times and highest immediacy index for a crystal engineering journal, plus an impressive impact factor of 3.47. In his welcome speech, *CrystEngComm* editor Jamie Humphrey outlined the successes of the past decade and extended his thanks: 'This success has been possible only through the support that you and other members of the crystal engineering community have given the journal – your support as authors, referees, readers and in some cases editorial and advisory board members.'

Regular *CrystEngComm* author Pierangelo Metrangolo of Milan, Italy, who attended the lunch reception, cites the journal as one of his favourites for publication of his research. 'In particular,' he says, 'I appreciate the speed at which papers are processed and the very kind co-operation of the editorial staff. What else to say: Happy Birthday *CrystEngComm*..., and keep up the good work!'

A decade since launch and the future for *CrystEngComm* has never looked so bright. Celebrations will continue later this year with an anniversary theme issue, including articles by editorial and advisory board members, and the journal is also heavily involved in the organisation of a crystal engineering symposium as part of the IUPAC Congress next year in Glasgow.

Visit www.crystengcomm.org for updates on these and other exciting events.

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Or paste the following URL into your browser: www.facebook.com/pages/Energy-Environmental-Science/24375018213.
See you there!

A warm reception in Philadelphia

The atmosphere inside the Philadelphia Marriott mirrored the sunny blue sky outside as guests gathered at the RSC Reception. Held on 17 August, it coincided with the 236th American Chemical Society National Meeting and Exposition taking place at the Pennsylvania Convention Center.

Around 200 people listened to RSC president Dave Garner as he welcomed guests, including Nobel prize winner Bob Grubbs from Caltech, a variety of eminent and emerging researchers, plus university librarians and local RSC members. The incoming president of the ACS, Tom Lane, was also there with a number of his society colleagues, indicating the continuing warm friendship between the two chemical societies.

Left to right: Jonathan Sessler (U Texas at Austin), Kate Sear (deputy editor ChemComm, RSC), Kevin Burgess (Texas A&M), Peter Wipf (Pittsburgh)

Guests enjoyed refreshments while catching up with friends old and new, and RSC staff were on hand to describe the latest RSC initiatives, including the hot topics of *Energy & Environmental Science*, *Integrative Biology* and *Metallomics*, the three newest RSC journals.

At the end of a genial evening, everyone was looking forward to meeting again – so see you all in Salt Lake City in spring 2009!

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