# ChemComm

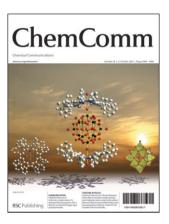
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# IN THIS ISSUE

ISSN 1359-7345 CODEN CHCOFS (39) 3969-4060 (2007)



Cover See Takahiko Kojima et al., page 3997. A saddle-distorted Mo(v)dodecaphenylporphyrin complex and a Keggin-type polyoxometalate react to give a novel molecular conglomerate named as "Porphyrin Hamburger". Image reproduced by permission of Atsutoshi Yokoyama, Takahiko Kojima, Kei Ohkubo and Shunichi Fukuzumi from Chem. Commun., 2007, 3997.

# CHEMICAL SCIENCE

# C73

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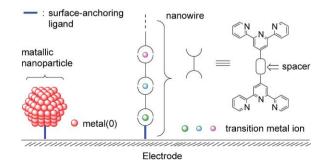
# FEATURE ARTICLES

# 3983

# Assembly of nanosize metallic particles and molecular wires on electrode surfaces

Yoshinori Yamanoi and Hiroshi Nishihara\*

This article highlights recent developments in the assembly of nanosize metallic particles and molecular wires on gold and silicon electrode surfaces. The fabrication, properties, and characteristics of functional nanostructured biointerfaces on electrode surfaces are also described.



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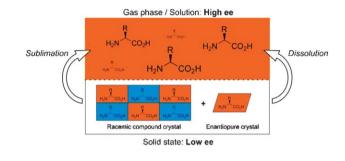
# FEATURE ARTICLES

### 3990

# Spoilt for choice: assessing phase behavior models for the evolution of homochirality

### Donna G. Blackmond\* and Martin Klussmann

Enantioenrichment based on amino acid phase behavior in solution and in sublimation experiments is rationalized by the same underlying fundamental physical chemistry principles in both cases.



# COMMUNICATIONS

## 3997

# A discrete conglomerate of a distorted Mo(V)-porphyrin with a directly coordinated keggin-type polyoxometalate

Atsutoshi Yokoyama, Takahiko Kojima,\* Kei Ohkubo and Shunichi Fukuzumi\*

The reaction of a saddle-distorted Mo(v)dodecaphenylporphyrin complex and a Keggin-type polyoxometalate gives a discrete and nanosized molecule, [{Mo(DPP)(O)}\_2(H\_2SiW\_{12}O\_{40})], which involves direct coordination between the Mo(v) centers and terminal oxo groups of the polyoxometalate and exhibits excellent stability in solution to show reversible multi-redox processes.

### 4000

# A self-assembled receptor for the recognition of phosphate and acetate anions in neutral aqueous solution

Céline Olivier, Zacharias Grote, Euro Solari, Rosario Scopelliti and Kay Severin\*

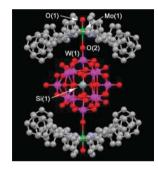
The reaction of [(benzene)RuCl<sub>2</sub>]<sub>2</sub> with a piperazine-bridged bis(dihydroxypyridine) ligand and LiOH leads to the formation of an expanded helicate, which is able to bind phosphate and acetate with mM affinities in aqueous solution at neutral pH.

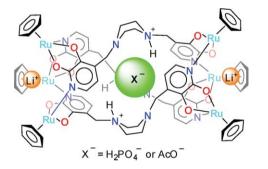
### 4003

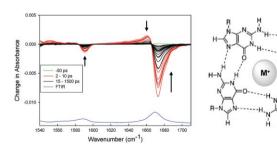
# Examination of intermolecular electronic interactions in the crystal structure of $C_{60}(CF_3)_{12}$ by experimental electron density determination

Lilianna Chęcińska, Sergey I. Troyanov, Stefan Mebs, Christian B. Hübschle and Peter Luger\*

The experimental electron density of  $C_{60}(CF_3)_{12}$  was analyzed to provide information about electronic interactions in a region where hexagons of adjacent molecules approach closely at only 3.3 Å.







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## 4006

# Uranyl stabilized Schiff base complex

Mohan S. Bharara, Stephen A. Tonks and Anne E. V. Gorden\*

A bimetallic uranyl Schiff base complex,  $[(UO_2)_2(Salpro)(OH)(Solvent)_2]$  is characterized by X-ray diffraction and determined to be unusually resistant to nucleophilic addition and transamination, unlike related complexes with transition metals.

4009

# Zwitterionic $\pi$ -radical involving EDT-TTF-imidazole and F<sub>4</sub>TCNQ: redox properties and self-assembled structure by hydrogen-bonds and multiple S $\cdots$ S interactions

T. Murata, Y. Morita,\* Y. Yakiyama, Y. Nishimura, T. Ise, D. Shiomi, K. Sato, T. Takui and K. Nakasuji\*

The reaction between EDT-TTF-imidazole and  $F_4TCNQ$  produced a zwitterionic  $\pi$ -radical, which formed a selfassembled structure by hydrogen-bonding and S…S interactions and exhibited three-step oxidation processes and high electrical conductivity.

# 4012

# Enantioselective catalysis with *tropos* ligands in chiral ionic liquids

Mike Schmitkamp, Dianjun Chen, Walter Leitner, Jürgen Klankermayer\* and Giancarlo Franciò\*

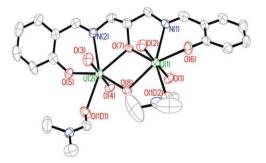
Tropoisomeric biphenylphosphine ligands in combination with chiral ionic liquids allow enantioselective homogeneous rhodium-catalysed hydrogenation and the catalytic system is reusable after extraction with  $scCO_2$ .

# 4015

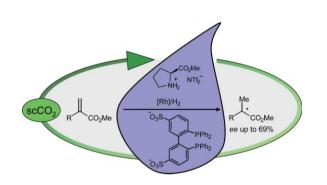
# Sol-gel encapsulation of cells is not limited to silica: long-term viability of bacteria in alumina matrices

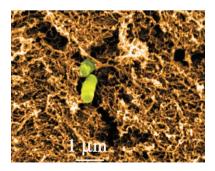
Makhlouf Amoura, Nadine Nassif, Cécile Roux, Jacques Livage and Thibaud Coradin\*

The authors report the first successful encapsulation of *Escherichia coli* bacteria in a non-silica host. A colloidal route was developed that allows the long-term preservation of cell viability in alumina gels.



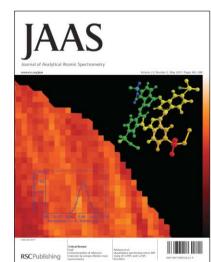








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## 4018



# Area-selective growth of amorphous carbon nanofibers *via* catalytic decomposition of polyimide thin film

Jung Hoon Kim, Nuri Oh, Chang Kyung Kim and Chong Seung Yoon\*

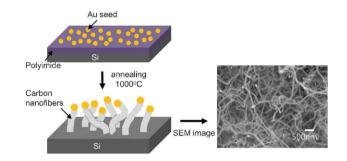
A dense layer of amorphous carbon nanofibers was fabricated by pyrolyzing a thin film of polyimide using a monolayer of gold nanoparticles as a catalyst.

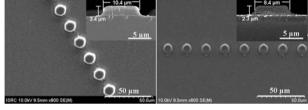


# Inorganic polymer photoresist for direct ceramic patterning by photolithography

Tuan Anh Pham, Pilnam Kim, Moonkyoo Kwak, Kahp Y. Suh and Dong-Pyo Kim\*

A novel negative, inorganic polymer photoresist was demonstrated to be suitable for simple and direct fabrication of tribological SiCN-based ceramic microstructures *via* UV photolithography and subsequent pyrolysis at 800 °C.



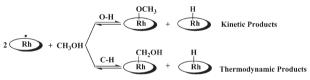


### 4024

# Competitive O–H and C–H oxidative addition of CH<sub>3</sub>OH to rhodium(II) porphyrins

Shan Li, Weihong Cui and Bradford B. Wayland\*

Competitive O–H and C–H oxidative addition reactions of methanol with rhodium(II) complexes of a *m*-xylyl tethered diporphyrin ('Rh(*m*-xylyl)Rh') and tetramesitylporphyrin ((TMP)Rh<sup>II</sup>·) are reported.

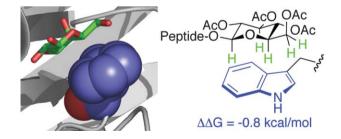


### 4026

Evaluation of a carbohydrate- $\pi$  interaction in a peptide model system

Sarah E. Kiehna, Zachary R. Laughrey and Marcey L. Waters\*

Using a  $\beta$ -hairpin model system, a carbohydrate– $\pi$  interaction was quantified in water. The interaction energy was found to be -0.8 kcal mol<sup>-1</sup>, greater than a  $\pi$ - $\pi$  interaction within the same model system.



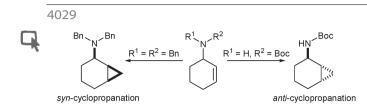


CF<sub>3</sub>SO<sub>3</sub>H

*p*-C<sub>6</sub>H₄Cl<sub>2</sub> 80°C

4032

q



# Diastereoselective Simmons-Smith cyclopropanations of allylic amines and carbamates

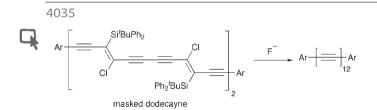
Stephen G. Davies,\* Kenneth B. Ling, Paul M. Roberts, Angela J. Russell and James E. Thomson

A highly diastereoselective, stereodivergent cyclopropanation protocol using  $CF_3CO_2ZnCH_2I$  (Shi's reagent), amenable to the synthesis of either diastereoisomer of the trifluoroacetic acid salt of 2-aminobicyclo[4.1.0]heptane, has been developed.

# Superacid promoted reactions of *N*-acyliminium salts and evidence for the involvement of superelectrophiles

Yiliang Zhang, Daniel J. DeSchepper, Thomas M. Gilbert, Kiran Kumar S. Sai and Douglas A. Klumpp\*

Experimental and theoretical studies suggest the involvement of dicationic, superelectrophilic *N*-acyliminium ions in conversions catalyzed by superacids.

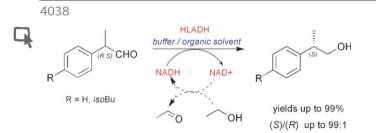


 $\overset{\delta^+ O^-}{\swarrow} \overset{H^- OTf}{\longrightarrow} \overset{\delta^-}{\longrightarrow}$ 

# β-Chlorovinylsilanes as masked alkynes in oligoyne assembly: synthesis of the first aryl-end-capped dodecayne

Simon M. E. Simpkins, Michael D. Weller and Liam R. Cox\*

An aryl-end-capped dodecayne has been prepared using a four-fold fluoride-mediated dechlorosilylation of a masked dodecayne precursor containing four  $\beta$ -chlorovinylsilane residues that serve as masked alkynes; the unstable dodecayne product has been characterised by UV-vis absorption spectroscopy and 'matrix-free' MALDI-TOF mass spectrometry.



## Highly efficient asymmetric reduction of arylpropionic aldehydes by Horse Liver Alcohol Dehydrogenase through dynamic kinetic resolution

Daria Giacomini,\* Paola Galletti, Arianna Quintavalla, Gabriele Gucciardo and Francesca Paradisi

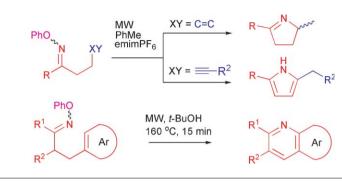
The realization of an efficient enzymatic process to obtain, in quantitative yields, (2S)-aryl propanols *via* a dynamic kinetic resolution (DKR) with high enantiomeric ratios and efficient cofactor recycling.

## 4041

# Microwave-assisted preparations of dihydropyrroles from alkenone *O*-phenyl oximes

Fernando Portela-Cubillo, Jackie S. Scott and John C. Walton\*

Suitably unsaturated *O*-phenyl oximes release iminyl radicals that ring close to dihydropyrroles, pyrroles or pyridines in microwave-assisted reactions.

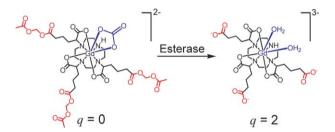


### 4044

### An esterase-activated magnetic resonance contrast agent

Marco Giardiello, Mark P. Lowe\* and Mauro Botta

On enzyme-activation of an acetoxymethyl ester-bearing contrast agent, an 84% increase in relaxivity is observed.



### 4047

## Immobilization of nanofibrous metal oxides on microfibers: A macrostructured catalyst system functionalized with nanoscale fibrous metal oxides

Hitoshi Ogihara,\* Masahiro Sadakane, Qiang Wu, Yoshinobu Nodasaka and Wataru Ueda\*

Nanofibrous LaMnO<sub>3</sub> can be immobilized on macrostructured materials using carbon nanofibers as templates; their application as macro-nanostructured catalysts are also presented.

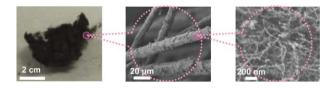
### 4050

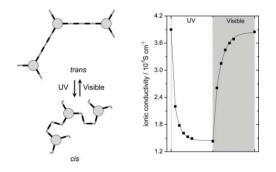
## Control of ionic conductivity of ionic liquid/ photoresponsive poly(amide acid) gels by photoirradiation

Masahiro Tamada, Toshiyuki Watanabe, Kazuyuki Horie and Hiroyuki Ohno\*

1,3-Dibutylimidazolium bromide was soaked into a polymer network prepared by condensation of

4-4'-diaminoazobenzene, pyromellitic dianhydride and 1,3,5-tri(4-aminophenyl)benzene to form photoresponsive ion conductive gels.





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# 4053

## Characterisation of 3-methylorcinaldehyde synthase (MOS) in *Acremonium strictum*: first observation of a reductive release mechanism during polyketide biosynthesis

Andrew M. Bailey, Russell J. Cox,\* Kate Harley, Colin M. Lazarus, Thomas J. Simpson\* and Elizabeth Skellam

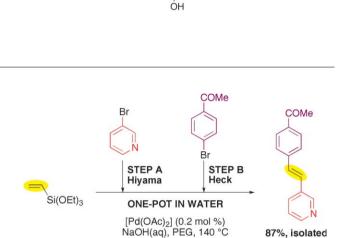
Heterologous expression in *Aspergillus oryzae* of a PKS gene isolated from *Acremonium strictum* produced 3-methylorcinaldehyde demonstrating the role of the terminal reductase in product release.

### 4056

# Consecutive palladium-catalyzed Hiyama–Heck reactions in aqueous media under ligand-free conditions

Álvaro Gordillo, Ernesto de Jesús\* and Carmen López-Mardomingo

(E)-1,2-Diarylethenes are synthesized by consecutive one-pot Hiyama–Heck reactions carried out in aqueous media under air. The only additives required are sodium hydroxide, palladium acetate and poly(ethylene glycol), and the products are isolable in many cases by simple filtration of the water solution.



PT

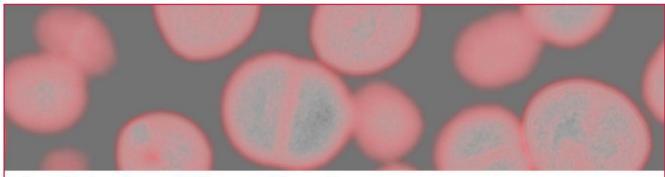
OH

Me

Me

OHC

NT



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# October 2007 / Volume 4 / Issue 10 / ISSN 1478-6524 / CSHCBM / www.rsc.org/chemicalscience

# **Chemical Science**

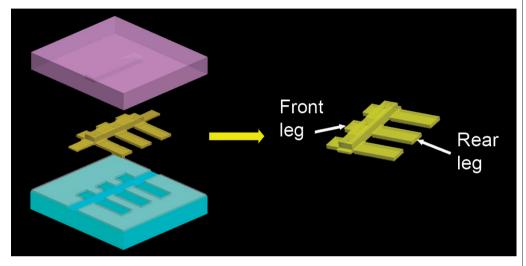
Heart cells power a biocompatible microrobot **Robots with a heart** 

Robots small enough to roam the human body and powered by living heart muscle have been built by scientists in Korea.

Sukho Park at Chonnam National University, Korea, and colleagues have designed a cellpowered microrobot. Park's team made the robot by growing heart muscle tissue from a rat onto tiny robotic skeletons made from polydimethylsiloxane (PDMS). PDMS is a biocompatible polymer making the robot suitable for use in biomedical applications.

What is special about these robots, says Park, is that they do not need an external power supply. Instead it is the relaxing and contracting heart muscle cells, which gain their energy from a glucose culture medium, that provide the power. These selfbeating cells allow the robot to move its six legs.

The robot has three short front legs (400 micrometres long) and three longer back legs (1200 micrometres long), which are all attached to a central rectangular



body. As the heart cells contract, the longer rear legs bend inwards. This creates a difference in friction between the front and rear legs, which pushes the robot forward. The scientists measured the robot's average speed at about 100 micrometres per second.

Park says these crab-like robots could be used inside the body to

Are robots like these soon to be roaming around in our blood vessels? clear blocked tubes or arteries. The robots could travel along the length of a blocked vessel, releasing a dissolving agent to clear the blockage as they go. *Sarah Corcoran* 

Reference J Kim *et al., Lab. Chip.*, 2007, DOI: 10.1039/ b705367c

# **In this issue**

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Sensor to detect sarin with a glow response

# Testing the toxicity of ionic liquids

Ionic liquids have often been touted as the ultimate green solvent, but just how green is green?

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# Instant insight: A bright future

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A snapshot of the latest developments from across the chemical sciences



Green Chemistry





# **Chemical Science**

# **Research highlights**

# A visible warning to the presence of sarin Glowing report for nerve agent detection

A chemiluminescent sensor could be used to detect sarin with a glow response, say US scientists.

Eric Anslyn at the University of Texas at Austin, and colleagues have designed a chemiluminescent system that can detect an analogue of the nerve gas sarin, giving off a blue glow in its presence. The analogue, diisopropyl fluorophosphonate or DFP, is in the same class of compounds as sarin, but is less toxic. The blue glow can be seen with the naked eye, which, Anslyn explained, is an advantage over established methods that require an external light source to view the results.

Anslyn's system is based on the chemiluminescence of oxalate esters. This is a well-known and well-understood phenomenon, where oxalate esters produce chemiluminescence – luminescence as a result of a chemical reaction



DFP (a sarin analgoue shown above) triggers a blue glow

– in the presence of fluorescent dyes such as diphenylanthracene.

Anslyn's team modified this system by adding a supernucleophile called an oximate. This molecule reacts with the oxalate ester in the system, blocking the chemiluminescence reaction. But when the sarin analogue DFP is present the oximate prefers to react with the phosphorous in the toxic molecule instead. This leaves the oxalate esters free to trigger the luminescence.

Work is underway to hone the system for practical use. 'The use of chemiluminescence is fascinating and has many possibilities, and we are just venturing into this area,' said Anslyn. Jon Silversides

### Reference

H S Hewage, K J Wallace and E V Anslyn, *Chem. Commun.*, 2007, 3909 (DOI: 10.1039/b706624d)

# Breast is still best, despite the presence of toxic pollutants Contaminants still present in breast milk

Forty years after the lesson taught by the notorious pesticide DDT, it might be hoped that fat-loving toxins would have been eliminated from human tissues. However, a US study has shown that levels of some flame retardants and organochlorine pesticides in breast milk are still high enough to warrant concern, despite many of them having been phased out of use.

Kurunthachalam Kannan and colleagues from the New York State Department of Health, US, took breast milk samples from 38 mothers in the state of Massachusetts, and analysed them for various flame retardants and organochlorine pesticides, such as DDT and lindane. They found that although levels varied widely between individuals, there was a clear correlation between amounts of different contaminants. This suggests a common source of exposure, which is likely to



be household dust or food, says Kannan.

The most prevalent contaminants in the samples studied, says Kannan, were polybrominated diphenyl ethers. These are flame retardants applied

### Chemicals used as flame retardants can end up in breast milk

### Reference

B Johnson-Restrepo *et al.,* J. Environ. Monit., 2007, DOI: 10.1039/b711409p to household furnishings and electronic goods that are still widely used in many countries. The levels of these chemicals were between ten and 100 times higher than in Asian and European samples, which can be explained by the great market demand for them in the US, said Kannan.

The scientists point out that despite the continued presence of these contaminants, breast milk is still the best food for babies. Gina Solomon, senior scientist at the Natural Resources Defense Council, a US-based non-governmental environmental and health advocacy organization based in New York, agreed. 'The benefits of breast milk still outweigh any harm from these contaminants, and we must control or eliminate the chemicals rather than stop mothers from breast-feeding,' said Solomon. David Barden

# 'Ecosystem in a box' strategy to determine the effects of structure on toxicity **Testing the toxicity of ionic liquids**

Ionic liquids have often been touted as the ultimate green solvent, but just how green is green? Researchers in Germany have come up with a way of testing the ecotoxicology of different ionic liquids using an 'ecosystem in a box'.

**3ASF** 

Bernd Jastorff at the University of Bremen and colleagues have tested the toxicological effects of a variety of ionic liquids across the range of biological complexity – from enzymes to whole organisms, of both aquatic and terrestrial origin. Previous studies on the toxicology of ionic liquids have been performed in vitro or just on single organisms, and some effects of structure on toxicity are well known, but this work represents the development of a general strategy for investigating the toxicity of ionic liquids.

'Suppliers of chemicals are increasingly being held responsible for the effect their products have on



the environment,' said Marianne Matzke, a member of the research team. 'It's important that both the technical properties are optimised and the hazards minimised at the same time in the development of new ionic liquids. A strong collaborative effort between all the interested parties – chemists, biologists and ecologists – is needed to achieve this.' The composition, and hence toxicity, of ionic liquids can vary enormously

**Reference** M Matzke *et al, Green Chem.,* 2007, DOI: 10.1039/b705795d

Ken Seddon, an ionic liquids expert at Queen's University, Belfast, UK, said, 'it's important that sweeping generalisations are not made about the toxicity of ionic liquids. There are literally millions of possible combinations of cations and anions, and this work represents a first and very important step in understanding the effects of structure on toxicity. There are very few centres worldwide capable of achieving this and the collaboration between suppliers of ionic liquids like Merck and the academic community is key.'

The plan is to create a well defined structure activity relationship for the toxicity of ionic liquids. 'Our eventual goal would be to design a totally biodegradable, non-toxic ionic liquid with the properties desired by the chemists,' said Matzke.

Stephen Davey

# Azides found to disintegrate when exposed to light Light-sensitive azides pose problems

Organic azides, which have recently become important compounds in organic synthesis, have been found to unexpectedly decompose under light.

Organic azides consist of a carbon chain attached to a row of three nitrogen atoms, and have in recent years become valuable compounds in organic chemistry, not least due to their versatility in reactions, such as the 'click' reaction of azides and alkenes. They have also been used as biochemical labels and as drug components.

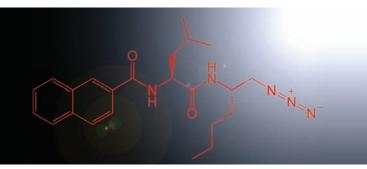
Now, a team of bioscientists have found that certain organic azides can decompose when exposed to light. Previously, such decomposition was only known to occur under strong heating or intense ultraviolet irradiation. This could raise issues for the future use of these azides in chemical and biochemical applications, say the researchers.

David Fairlie and colleagues at the

The azide shown (an enzyme inhibitor) decomposes to an even more biologically active form under light

# Reference

G Abbenante, G T Lee and D P Fairlie, *Chem. Commun.*, 2007, DOI: 10.1039/b708134k



University of Queensland, Brisbane, Australia, left an azide derivative of an amino acid in a clear glass vial exposed to light, and found that it decomposed at the rate of about one percent a day to give an aldehyde and an amine, which they isolated by chromatography. The aldehyde is known to be much more biologically active than the azide, meaning that even trace amounts could have profound effects, said Fairlie. Summarising, he said that this decomposition has 'very important ramifications for the storage, chemical reactivity, and biological properties of azides'.

Stefan Bräse, professor of organic chemistry at the University of Karlsruhe, Germany, is fascinated by the results. 'Organic azides are always good for surprises,' he said, 'but the discovery that laboratory light might convert azides into bioactive compounds poses both problems and opportunities in the future in biochemistry and chemical biology.' David Barden

# **News in brief**

### **Spontaneous superlattices**

Zinc oxide nanocrystals that selfassemble into 'superlattices' are promising luminescent materials, say scientists in China.

### From polymers to porous carbon

Microporous carbon materials with finely tuned pore sizes can be made from hyper-branched polymers.

## A perfect partner for DNA extraction

'DNA-surfactants' can cleanly and easily extract specific DNA sequences, potentially allowing mass production of DNA.

See www.rsc.org/chemicalscience for full versions of these articles

# This month in Chemical Technology

### **Finding fission by-products**

Researchers in Canada have developed a method for the rapid ultra-trace measurement of strontium-90 in environmental samples.

### Wet. not wet. wet

Bored of bath-cleaning? Wearied by window-washing? Thanks to a group of scientists in Israel these chores could soon be consigned to the past.

### Sizing up the danger of volcanic ash

Analysing the grain size of volcanic ash particles might provide a quick and easy way to calculate their potential threat to human health.

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# This month in Chemical Biology

### **Promise for Parkinson's**

Polish researchers explore the link between copper and Parkinson's disease.

### **Cell preservation all wrapped up**

Freezing cells inside glass cages could improve human fertility treatments.

# **Identifying transgenic soya**

A plant's metallic make-up could be used to identify it as genetically modified, say researchers in Brazil.

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# Sol-gel route proves a 'smart' move



A stable and highly conducting electrolyte for electrochromic devices such as 'smart' windows has been made by scientists in Portugal. Electrochromic materials can

respond to electricity by changing colour. For example, smart windows change between transparent and opaque states at the flick of a switch.

Electrochromic devices contain electrodes separated by an ionconducting electrolyte. A burst of electricity triggers a rush of cations through the electrolyte and into the electrochromic material (usually tungsten trioxide) where they find a home within its structure. This electrochemical reaction changes the transparency of the material

The new electrolyte proved its potential in a prototype smart window

Reference

and another burst of electricity can change it back.

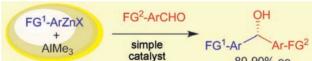
An important part of the set-up is the electrolyte, which is the focus of research being carried out at the University of Trás-os-Montes and Alto Douro. Verónica de Zea Bermudez, who led the research, explained, 'the electrolyte must display, not just high ionic conductivity, but also high electrochemical stability.' And this is exactly what has been achieved by their use of the sol-gel technique to make new electrolytes.

The sol-gel technique is a versatile method of making materials. De Zea Bermudez's team used it to make an organically modified silicate polymer known as a di-ureasil ormolyte. The researchers doped this silica-based hybrid material with potassium triflate (KCF<sub>3</sub>SO<sub>3</sub>) and investigated its effect on potassium ion conductivity.

The researchers said they were surprised by just how high their ion conductivity measurements were. The ormolyte was tested within a prototype smart window device, fully demonstrating its potential. Gavin Armstrong

### S C Nunes et al, J. Mater. Chem., 2007, DOI:10.1039/ h708905h

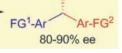
# Zinc gets a reaction



Chemists have expanded the range of reactions that can be carried out with organozinc compounds.

Simon Woodward and colleagues at the University of Nottingham, UK, have shown that normally unreactive organozinc halides (RZnX) can be used as versatile reagents in asymmetric carbonyl addition chemistry. Woodward's team developed a straightforward system for the activation of RZnX in aldehyde additions using only readily-available reagents and a simple ligand.

'Normally for asymmetric carbonyl addition one uses diorganic species [ZnR<sub>2</sub>]. However,



**Activating organozinc** compounds with an aluminium reagent opens up a whole new world of reactivity

Reference

J Shannon et al. Chem.

10.1039/b710681e)

Commun., 2007, 3945 (DOI:

only eight of these are routinely available - which is a bit of a blow if the R-group you want to add is not one of them! The fact that over 200 organozinc halide species are commercially available seemed to us an untapped resource that deserved to be mobilised,' explained Woodward.

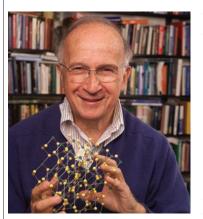
The researchers generated their catalyst from RZnX and a widely available aluminium-based reagent. 'The well-known functional group tolerance of organozinc halides is an additional advantage,' said Woodward.

John Brown, an expert in the catalysis of organic reactions at Oxford University, UK, welcomed the work. 'Woodward's group has discovered a method which is operationally simple and obviously useful,' he said. Michael Spencelayh

# **Interview**

# **Making connections**

Roald Hoffmann talks to Alison Stoddart about chemical bonding and his new playground of high pressure chemistry



# **Roald Hoffmann**

Roald Hoffmann is professor of theoretical chemistry at Cornell University, US. Roald shared the 1981 Nobel Prize in chemistry for the development of theories concerning the course of chemical reactions. His current interests include high pressure chemistry, chemical bonding, poetry and philosophy.

## What prompted you to become a chemist?

As a child, I read two books while I was in a refugee camp – the biography of Marie Curie by her daughter and the story of a black American agricultural chemist, George Washington Carver. These accounts formed a saintly image of chemistry. So the science interest was there early on, but there was pressure on me to become a doctor. I decided not to follow this path, but only in graduate school did I commit to chemistry. I liked chemistry; it fitted my abilities and interests. Somehow, I have been intuitively able to latch on to the chemical essence of a question.

## What were your thought processes behind the Woodward–Hoffmann rules?

I had just done calculations on boron hydrides with William Lipscomb and Martin Gouterman. This gave me some respect for experiment and for complexity outside of planar  $\pi$  electron systems. I also developed a method to calculate almost any molecule, in particular organic molecules, so I was ready when Robert Burns Woodward approached me. Woodward knew that the stereochemistry of electrocyclic reactions presented an important problem. But I was young; I only realised that we had explained something worthwhile when I saw the reaction of the community. From this work, I learned to form explanations instead of just doing calculations.

# Why did the Woodward–Hoffmann rules have such a big impact?

They formed a connection between organic and theoretical chemistry. That meeting point had been prepared for but the rules cemented things. Molecular Orbital theory was perceived as useful by organic chemists and not too difficult to learn.

Organic chemists were given a pictorial language of where electrons were in molecules through pictures of orbitals and orbital interactions. This connected to the geometric structural language, prevalent for describing structures, adding an electronic dimension to the stereochemical pictorial language which was already there. When we drew ethylene in three dimensions and added the  $\pi^*$  orbital, it wasn't that far a stretch.

Why are you interested in high pressure chemistry?

The high pressure realm is a playground for bonding ideas. It's so much fun to see carbon dioxide turn into quartz at high pressure – that is, in terms of its bonding properties. When the pressure is removed, carbon dioxide gas bubbles off. The high pressure community needs a valued high pressure synthesis of a desired material (other than diamond). The problem is that after you reach a metastable structure, the return to the ambient pressure form usually does not encounter large barriers because the reactions are symmetry-allowed. I would love to design a reaction that would face high barriers but it's tough.

# What advice would you give to young researchers?

Whether you are young or old, looking for connections is attractive. But you need to be good at one thing (in my case it was theoretical chemistry); that gives you psychological strength. The danger then is to get stuck in that field, the area you're good in, and follow your intellectual fathers. You have to look outside the field and look for connections. So my advice would be to specialise, but don't forget the forest for the trees. Look for the connections between things – that's what makes us human.

### If you weren't a scientist, what would you be?

I could easily have gone off into the arts or humanities. I may have become an art historian. I have no regrets – I have somehow been able to carve out a land of my own between chemistry, poetry and philosophy. Some of the science informs my poetry, and perhaps my writing in science has improved because of my love of literature.

Two metaphors which are important to me are bridges and connections. My Nobel lecture was called 'Building bridges between inorganic and organic chemistry' – it was about the isolobal analogy. I think I have helped to build bridges between sciences, the arts and humanities.

### What achievement are you most proud of?

It's the combination of making connections between different parts of science and being a teacher. Teachers wake up the minds of people – rather than teaching facts, they empower young people to make use of the abilities within them. I am proud of being a good teacher and teaching in many ways.





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# Instant insight A bright future

Andy Benniston at Newcastle University, UK, explains how photocatalysts could solve the planet's energy crisis, and reduce  $CO_2$  emissions while they're at it

The world is going through a major change as countries start to come to grips with the realisation that current sources of energy - coal, oil and gas - are not unlimited. Coupled with the expected growth in population, it is clear that our limited fossil fuels will not be able to meet future energy requirements. And as global warming predictions kick in, the production of the greenhouse gas carbon dioxide (CO<sub>2</sub>) from burning fossil fuels must be minimised on a global scale. It is clear that solving these two problems is a priority, and will require not just a handful of nations taking up the challenge but a real world-wide effort.

Although the picture painted may seem all 'doom and gloom', it should not be forgotten that the fuels we burn today come from carbon sources that were, in a simple sense, 'fixed' by nature many millions of years ago. The current situation is only the consequence that we are now using up these sources at an alarming rate and nature cannot keep up!

So how can we solve the energy crisis and reduce CO2 emissions at the same time? The answer is either to find a fuel that does not produce CO<sub>2</sub>, or to find a process that uses  $CO_2$  in fuel production. If the two processes could be coupled together then the cycle set up would clearly be highly beneficial. This may seem far-fetched, but this cycle is almost the same as that used by plants to covert water into oxygen that we breathe, and CO<sub>2</sub> into carbohydrates that we eat. We breathe out CO<sub>2</sub> completing the cycle. The energy source for all this to operate comes from the sun. which to all intent purposes affords the planet unlimited power.

A major challenge for scientists is to mimic the process that plants successfully use to harness solar

### Nature is way ahead of us when it comes to using sunlight to produce energy

Reference A C Benniston, Phys. Chem. Chem. Phys., 2007, DOI: 10.1039/b708166a energy. We may think that this is a new idea, but back in the late 1970s and 1980s many groups tried to solve the problem, to differing levels of success.

One area of research focussed on the splitting of water to form hydrogen, which is an ideal fuel since its combustion reforms water. In a related manner the oxidation of water to oxygen was also attempted, since by linking the two processes together the overall reaction would be splitting of water into its constituent elements. Photochemical driven reactions were found that could carry out hydrogen and oxygen production separately, but coupling the two reactions together was never satisfactorily solved.

If the above reactions are difficult to achieve then the controlled reduction of CO<sub>2</sub> is even more so, since several different products are feasible depending on the number of added electrons and protons. One particularly interesting outcome is the six electron/proton product, methanol, which again is a fuel that can be burned to afford energy. It can be speculated that an ideal scenario would be coupling of the water splitting reaction to the  $CO_2$  reduction reaction. The greenhouse gas would be converted to a fuel and sunlight would provide the energy source, thus solving all our problems in one go!

What this idyllic story lacks is the science behind how to achieve some of the reactions needed. The real challenge lies in finding photocatalysts that capture sunlight and drive the chemical transformations.

There has been a major research effort to construct artificial model systems using the same type of building blocks used by nature for sunlight capture. One part of the work performed in this area uses porphyrin-based conjugates that incorporate metal-based poly(pyridyl) relays. Careful thought has gone into their design in an attempt to create photocatalysts capable of mimicking the light driven charge separation reaction used by plants to create fuel.

But this is only part of the story as the synthesis of such catalysts is time consuming, and requires controlled build up of the final structure. Even after careful design and synthesis, detailed photophysical studies can often reveal the system does not behave as expected and it's back to the drawing board.

There is a still a long way to go before an 'artificial leaf' is available and a new generation of scientists is needed to take up the challenge now.

Read Andy Benniston's Perspective article 'Porphyrin linked poly(pyridyl)-based conjugates as artificial photosynthetic reaction centre models' in a forthcoming issue of PCCP.

# **Chemical Science**

# **Essential elements**

# And the winner is ...



Left to right - Richard Kidd, Richard Gedye and Bernard Donovan

Months of hard work were rewarded recently as *RSC Project Prospect* was named as winner of the 2007 ALPSP/Charlesworth Award for Publishing Innovation.

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aspect of scholarly publication, the judges described *RSC Project Prospect* as 'the clear winner ... journals incorporate standard metadata within the full text of articles and combine this with an elegant and intuitive on-screen manifestation of the advantages of including this metadata. As a result, sophisticated and effective searching of the literature is greatly improved and the value gained from reading each article is significantly enhanced. It is delightfully simple to use and benefits to authors and readers are immediately obvious.'

Receiving the award at the ALPSP Annual Dinner in London on September 13th, project manager Richard Kidd declared: 'RSC Publishing is proud to win the 2007 award, which is great recognition for the work our publishing staff and academic partners have put into the development and evolution of *Project Prospect.*'

This is the first time that RSC Publishing has received the award for publishing innovation, and staff are understandably delighted.

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