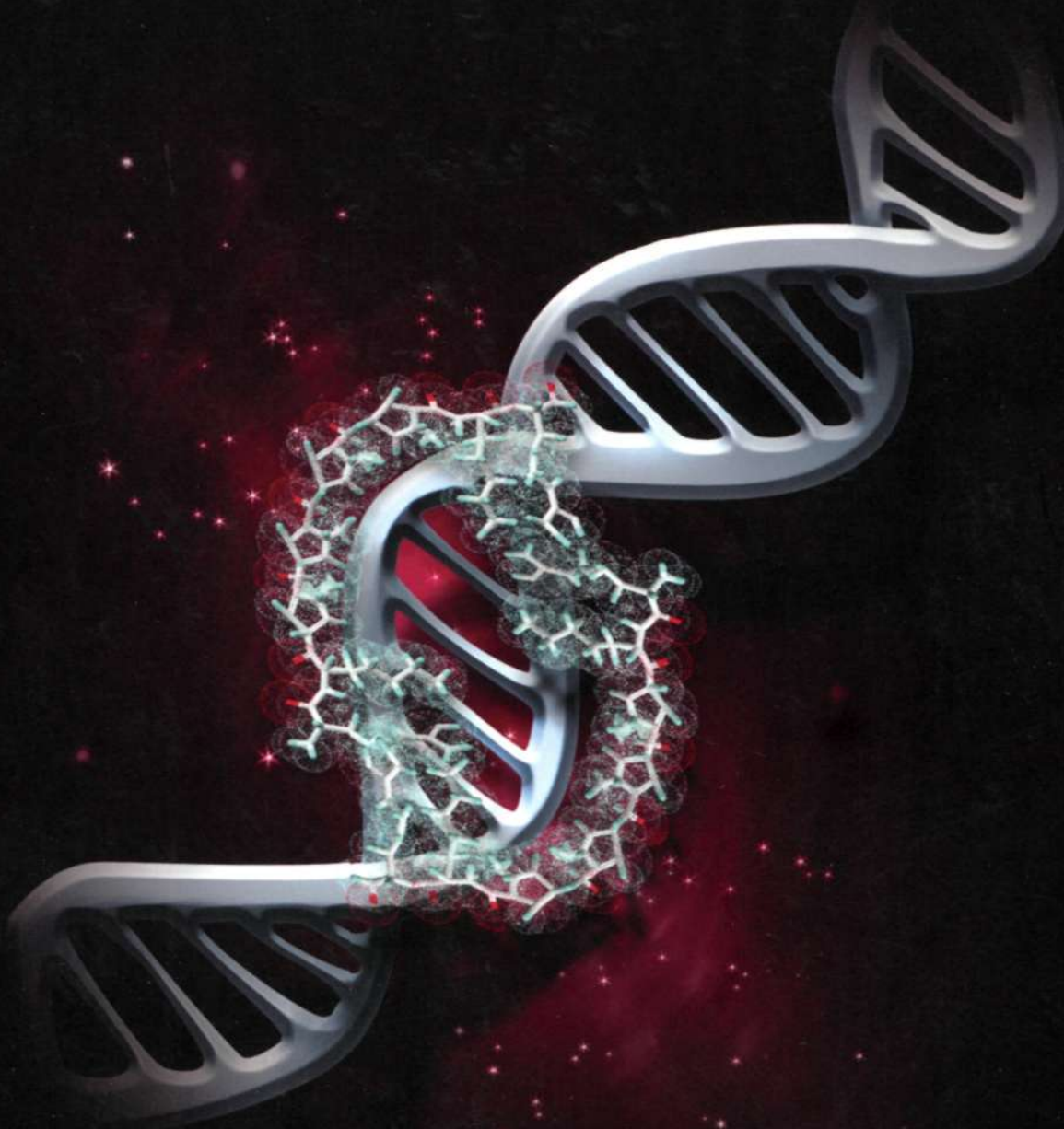


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# Organic & Biomolecular Chemistry

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**PAPER**

Leung Sheh *et al.*

Energetic studies on DNA–peptide interaction in relation to the enthalpy–entropy compensation paradox



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# Organic & Biomolecular Chemistry

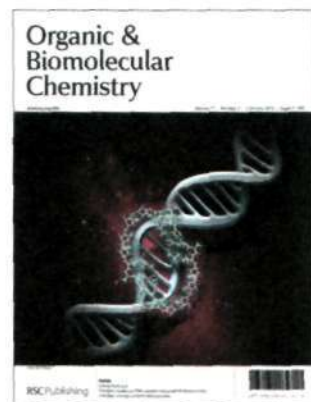
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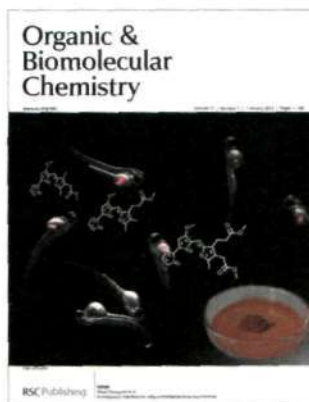


### Cover

See Leung Sheh *et al.*, pp. 48–61.

Energetic basis for molecular recognition of peptides to DNA enlightens the universal EEC paradox.

Image reproduced by permission of Leung Sheh from *Org. Biomol. Chem.*, 2013, **11**, 48.



### Inside cover

See Alison Thompson *et al.*, pp. 62–68.

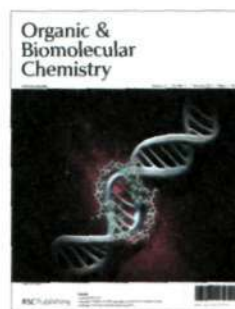
Image reproduced by permission of Alison Thompson from *Org. Biomol. Chem.*, 2013, **11**, 62.

## EDITORIAL

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### Happy New Year from *Organic & Biomolecular Chemistry*

On behalf of the *OBC* Editorial Board and Editorial Team, we welcome you to this first issue of 2013.



## EMERGING AREA

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### Oligonucleotide-templated chemical reactions: pushing the boundaries of a nature-inspired process

Claudia Percivalle, Jean-François Bartolo and Sylvain Ladame\*

Widespread in nature, oligonucleotide-templated reactions of phosphodiester bond formation have inspired chemists who are now applying this elegant strategy to the catalysis of a broad range of otherwise inefficient reactions.



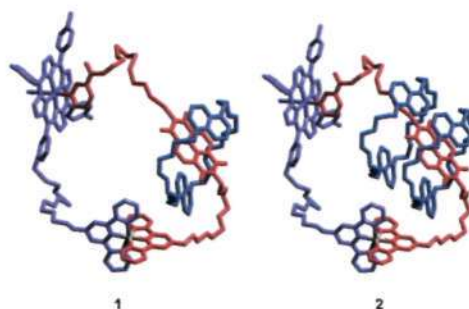
Федеральное государственное  
бюджетное учреждение науки  
Центральная научная библиотека  
Уральского отделения  
Российской академии наук (ЦНБ УрО РАН)

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### Synthesis of a four-component [3]catenane using three distinct noncovalent interactions

Miguel Á. Alemán García and Nick Bampos\*

A multicomponent assembly is described resulting in [2] and [3]catenanes using three flexible components and three distinct noncovalent interactions.

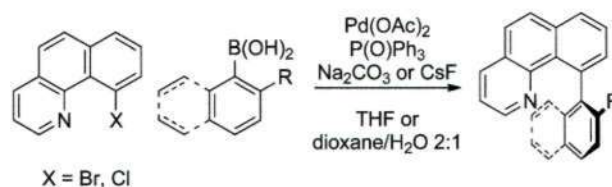


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### Synthesis of sterically encumbered C10-arylated benzo[*h*]quinolines using *ortho*-substituted aryl boronic acids

Marko Weimar and Matthew J. Fuchter\*

The challenging coupling of 10-halobenzo[*h*]quinolines with *ortho*-substituted aryl boronic acids has been achieved using Pd(OAc)<sub>2</sub>/P(O)Ph<sub>3</sub> as the catalytic system.

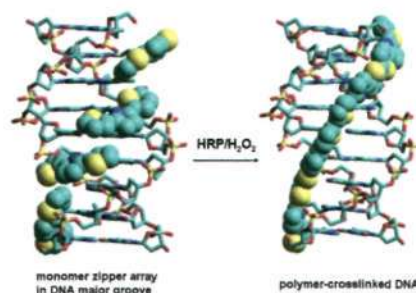


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### Structural stabilization of DNA-templated nanostructures: crosslinking with 2,5-bis(2-thienyl)pyrrole monomers

Wen Chen and Gary B. Schuster\*

Ordered 2,5-bis(2-thienyl)pyrrole (SNS) zipper arrays are formed by hybridization of complementary DNA oligomers each containing covalently bound SNS monomers.

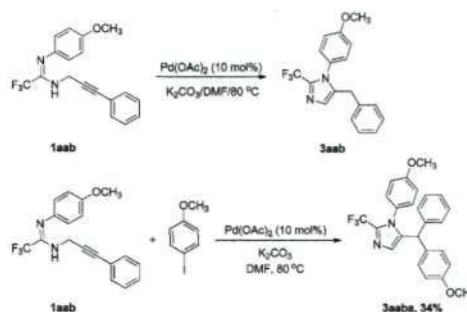


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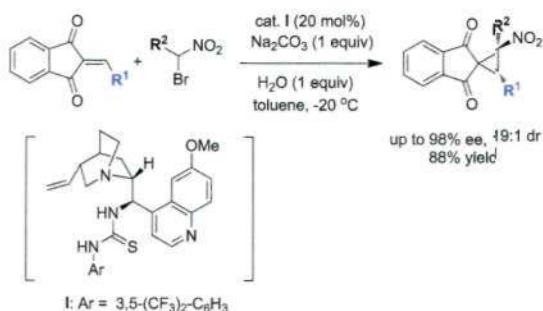
### Pd-catalyzed coupling reaction of fluorinated propargyl amidines with aryl iodides

Shan Li, Yafen Yuan, Yajun Li, Zhengke Li, Lisi Zhang and Yongming Wu\*

Catalyzed by ligand free Pd(OAc)<sub>2</sub>, 2,5-disubstituted imidazole was prepared in good yield by the reaction of fluorinated propargyl amidines with iodoarene.



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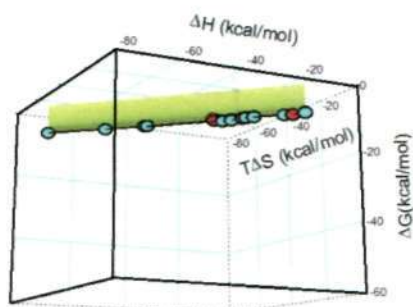
### An efficient organocatalytic enantioselective synthesis of spironitrocyclopropanes

Utpal Das, Yi-Ling Tsai and Wenwei Lin\*

An organocatalytic construction of substituted spironitrocyclopropanes starting from 2-arylidene-1,3-indandiones and bromonitroalkanes has been described.

PAPERS

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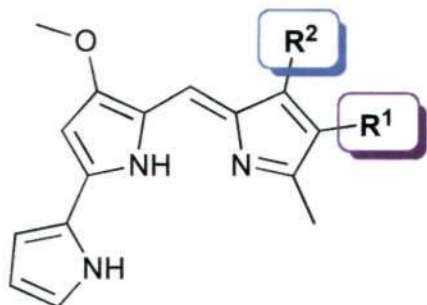


### Energetic studies on DNA–peptide interaction in relation to the enthalpy–entropy compensation paradox

Robin C. K. Yang, Jonathan T. B. Huang, Shih-Chuan Chien, Roy Huang, Kee-Ching G. Jeng, Yen-Chung Chen, Mokai Liao, Jia-Rong Wu, Wei-Kang Hung, Chia-Chun Hung, Yu-Ling Chen, Michael J. Waring and Leung Sheh\*

The energetic basis for molecular recognition of peptides to DNA enlightens the universal EEC paradox.

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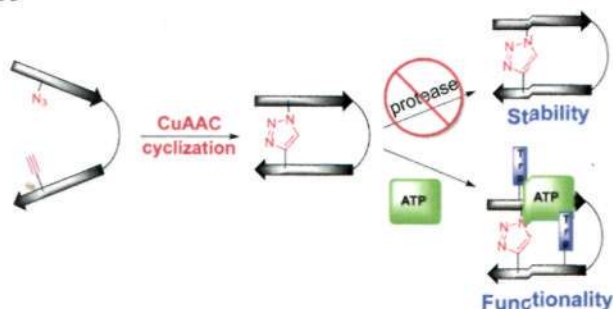


### Investigations regarding the utility of prodigiosenes to treat leukemia

Deborah A. Smithen, A. Michael Forrester, Dale P. Corkery, Graham Dellaire, Julie Colpitts, Sherri A. McFarland, Jason N. Berman and Alison Thompson\*

Structural modification of the C-ring resulted in a series of prodigiosenes that displayed interesting activity *in vitro* and *in vivo*.

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### Positional effects of click cyclization on $\beta$ -hairpin structure, stability, and function

Jessica H. Park and Marcey L. Waters\*

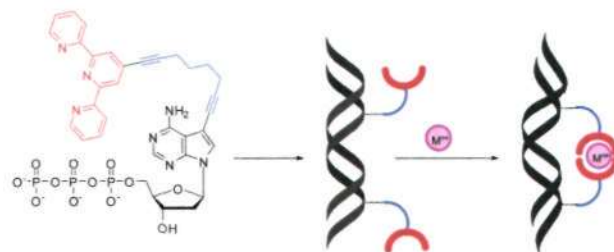
“Clicking” of  $\beta$ -hairpin peptides at several different positions was explored as a method to influence folding, protease resistance, and function.

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### Synthesis of nucleosides and dNTPs bearing oligopyridine ligands linked through an octadiyne tether, their incorporation into DNA and complexation with transition metal cations

Lubica Kalachova, Radek Pohl, Lucie Bednárová, Jindřich Fanfrlík and Michal Hocek\*

Nucleotides bearing terpyridine ligands attached *via* a flexible octadiynyl tether were prepared and enzymatically incorporated into DNA where formation of intra-strand complexes with  $\text{Fe}^{2+}$  and  $\text{Ni}^{2+}$  was observed.

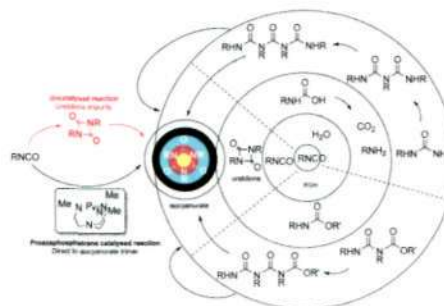


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### The formation of high-purity isocyanurate through proazaphosphatrane-catalysed isocyanate cyclo-trimerisation: computational insights

Jack N. Gibb and Jonathan M. Goodman\*

Polyurethane foams are widely used materials and control of their physical properties is a significant challenge.

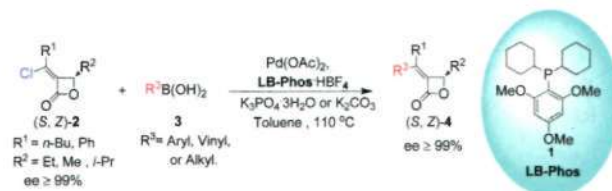


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### Pd-catalyzed Suzuki coupling reaction of chloroalkylidene- $\beta$ -lactones with LB-Phos as the ligand

Pengbin Li, Bo Lü, Chunling Fu and Shengming Ma\*

The LB-Phos-HBF<sub>4</sub> salt was applied for the Pd-catalysed Suzuki coupling reactions of optically active (*Z*)- $\alpha$ -chloroalkylidene- $\beta$ -lactones.

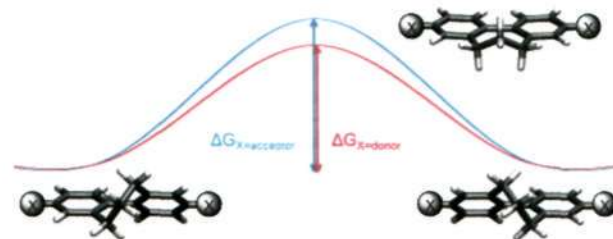


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### Atropisomerization of di-*para*-substituted propyl-bridged biphenyl cyclophanes

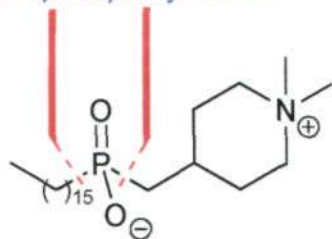
Jürgen Rotzler, Heiko Gsellinger, Angela Bihlmeier, Markus Gantenbein, David Vonlanthen, Daniel Häussinger,\* Wim Klopper\* and Marcel Mayor\*

The influence of electron donors and electron acceptors of variable strength in the 4 and 4' position of 2 and 2' propyl-bridged axial chiral biphenyl cyclophanes on their atropisomerization process was studied.



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## Radical hydrophosphinylation

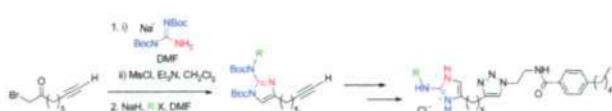


## Synthesis of a phosphinate analogue of the anti-tumour phosphate di-ester perifosine via sequential radical processes

Marios S. Markoulides and Andrew C. Regan\*

A phosphinate analogue of the anti-tumour phosphate di-ester perifosine has been prepared using two sequential radical hydrophosphinylation reactions.

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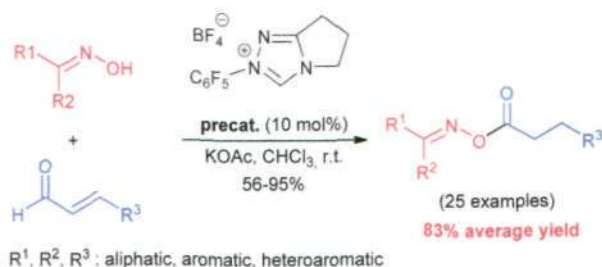


## N-Substituted 2-aminoimidazole inhibitors of MRSA biofilm formation accessed through direct 1,3-bis(tert-butoxycarbonyl)guanidine cyclization

Andrew A. Yeagley, Zhaoming Su, Kára D. McCullough, Roberta J. Worthington and Christian Melander\*

A method to access 2-substituted derivatives of a known 2-aminoimidazole/triazole biofilm modulator was developed. Analogues possessing aliphatic substituents exhibited increased biofilm inhibition activity against MRSA compared to the parent compound. Several analogues also displayed synergy with oxacillin against MRSA.

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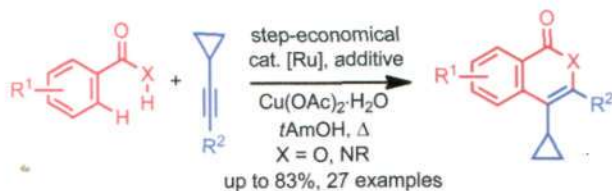


## N-Heterocyclic carbene catalyzed synthesis of oxime esters

Dieter Enders,\* André Grossmann and David Van Craen

A selective, redox- and atom economical synthesis of oxime esters via N-heterocyclic carbene catalyzed redox esterification has been developed. The new protocol opens a convenient and direct entry to the title compounds in very high yields without any significant limitations in the substrate scope.

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## Ruthenium-catalyzed C-H/O-H and C-H/N-H bond functionalizations: oxidative annulations of cyclopropyl-substituted alkyne

Monica Deponti, Sergei I. Kozhushkov, Dmitry S. Yufit and Lutz Ackermann\*

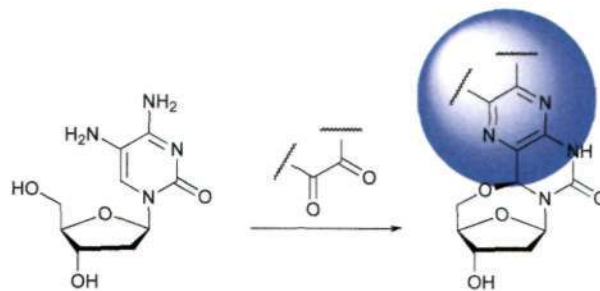
Ruthenium-catalyzed syntheses of cyclopropyl-substituted isocoumarins and isoquinolones were accomplished through highly chemo-, site- and regioselective C-H/O-H and C-H/N-H bond functionalizations with cyclopropylacetylenes.

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### Syntheses and photophysical properties of 5'-6-locked fluorescent nucleosides

Kristmann Gislason, Dnyaneshwar B. Gophane and Snorri Th. Sigurdsson\*

The condensation reaction between 5-amino-2'-deoxycytidine and 1,2-diketones was utilized to obtain nine 5'-6-locked fluorescent nucleosides with diverse photophysical properties. One nucleoside was also studied in model DNA structures, including a nicked duplex.

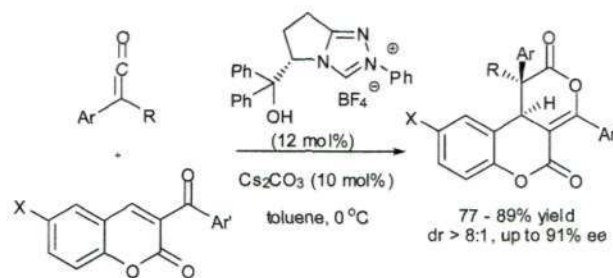


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### N-heterocyclic carbene-catalyzed [4 + 2] cycloaddition of ketenes and 3-aryl coumarins: highly enantioselective synthesis of dihydrocoumarin-fused dihydropyranones

Teng-Yue Jian, Xiang-Yu Chen, Li-Hui Sun and Song Ye\*

Dihydrocoumarin-fused dihydropyranones were synthesized in high yield with good diastereo- and enantioselectivity via NHC-catalyzed [4 + 2] cyclization of ketenes and 3-aryl coumarins.



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### Photochemically induced radical alkylation of C(sp<sup>3</sup>)-H bonds

Tamaki Hoshikawa, Shin Kamijo and Masayuki Inoue\*

The present direct alkylation of C(sp<sup>3</sup>)-H bonds enables construction of various tri- and tetra-substituted carbons from heteroatom-substituted methylenes, methines and alkanes in a highly chemoselective fashion.



170

### Mechanistic investigation of the oxidation of hydrazides: implications for the activation of the TB drug isoniazid

Ruth I. J. Amos, Brendon S. Gourlay, Brian F. Yates, Carl H. Schiesser, Trevor W. Lewis and Jason A. Smith\*

In the oxidation of arylhydrazides the diimide intermediate is the source for nucleophilic acyl substitution products and the precursor for acyl radical formation, there is no evidence for the formation of acyl cations.

