

# Organic & Biomolecular Chemistry

An international journal of synthetic, physical and biomolecular organic chemistry

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## IN THIS ISSUE

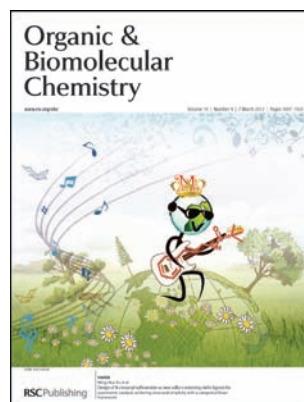
ISSN 1477-0520 CODEN OBCRAK 10(9) 1697–1932 (2012)



### Cover

See Andrew C. Benniston *et al.*, pp. 1775–1784.

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### Inside cover

See Ming-Hua Xu *et al.*, pp. 1764–1768.

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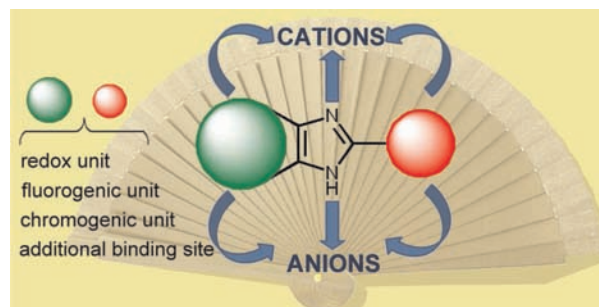
## PERSPECTIVE

1711

### Imidazole derivatives: A comprehensive survey of their recognition properties

Pedro Molina,\* Alberto Tárraga\* and Francisco Otón\*

Imidazole derivatives: a range of possibilities in the field of molecular recognition.



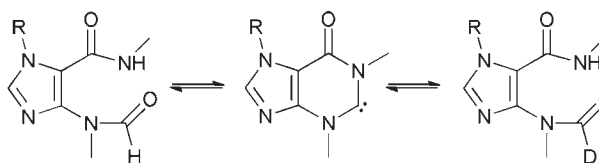
## COMMUNICATIONS

1725

### Studies of the H–D exchange mechanism of malonganone B

Peter G. K. Clark, Matthias Lein and Robert A. Keyzers\*

Studies of malonganone B analogues indicated that its H–D exchange in deuterium NMR solvents goes through a novel mechanism involving a rare mixed amine–amide NHC.



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# Organic & Biomolecular Chemistry

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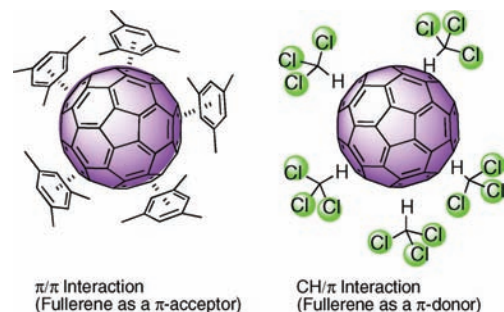
## COMMUNICATIONS

1730

**First kinetic evidence for the CH/ $\pi$  and  $\pi/\pi$  solute–solvent interaction of C<sub>60</sub> in the Diels–Alder reaction with cyclohexadiene**

Takumi Oshima,\* Tsubasa Mikie, Naohiko Ikuma and Hajime Yakuma

The first CH/ $\pi$  solute–solvent interaction of C<sub>60</sub> was evidenced by the kinetic solvent effects in the Diels–Alder reaction with 1,3-cyclohexadiene.

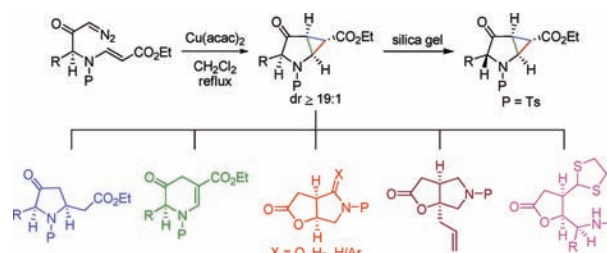


1735

**Stereoselective synthesis and applications of nitrogen substituted donor–acceptor cyclopropanes (N-DACs) in the divergent synthesis of azacycles**

Santosh J. Gharpure,\* U. Vijayasree and S. Raja Bhushan Reddy

Stereoselective intramolecular cyclopropanation of vinylogous carbamates with carbenes using Cu(acac)<sub>2</sub> as the catalyst has been developed for the construction of N-DACs. Regioselective cleavage of each of the cyclopropane bonds of the N-DACs led to a diverse array of azacycles.

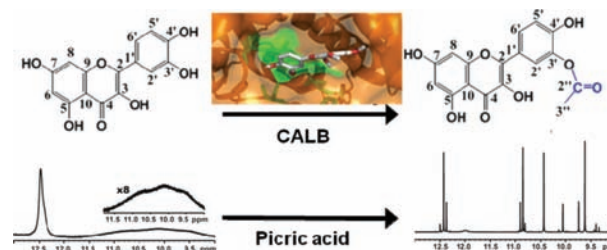


1739

**Unexpected enzyme-catalyzed regioselective acylation of flavonoid aglycones and rapid product screening**

Eleni Kyriakou, Alexandra Primikyri, Pantelis Charisiadis, Maria Katsoura, Ioannis P. Gerothanassis, Haralambos Stamatis and Andreas G. Tzakos\*

CALB-mediated regioselective polyphenol aglycon transformations are reported and rapid product formation screening is achieved through an internal sensor.

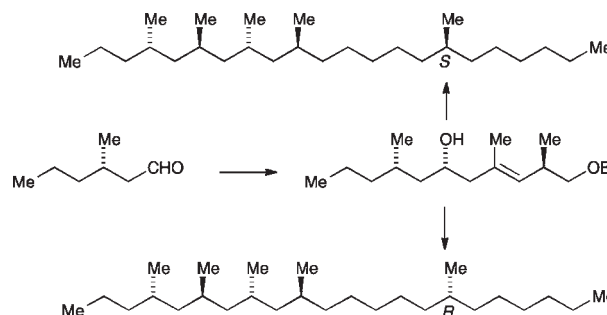


1743

**Total synthesis of a cuticular hydrocarbon from the cane beetle *Antitrogus parvulus*: confirmation of the relative stereochemistry**

Norazah B. Basar, Hao Liu, Devendra Negi, Hasnah M. Sirat, Gareth A. Morris and Eric J. Thomas\*

Synthesis and comparison of (4*S*,6*R*,8*R*,10*S*,16*S*)- and (4*S*,6*R*,8*R*,10*S*,16*R*)-4,6,8,10,16-pentamethyltricosanes confirmed that the (16*S*)-epimer was the hydrocarbon isolated from *Antitrogus parvulus*.



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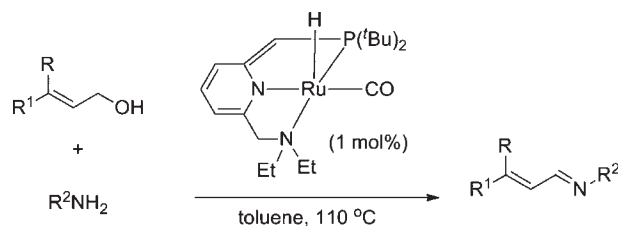
## COMMUNICATIONS

1746

 **$\alpha,\beta$ -Unsaturated imines via Ru-catalyzed coupling of allylic alcohols and amines**

Jared W. Rigoli, Sara A. Moyer, Simon D. Pearce and Jennifer M. Schomaker\*

The conversion of allylic alcohols and amines to  $\alpha,\beta$ -unsaturated imines is reported using 1 mol% of a Ru-pincer complex.

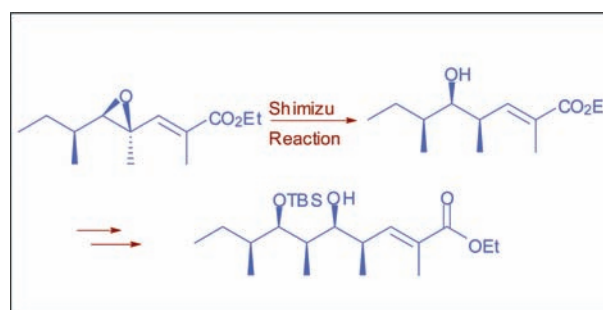


1750

**An iterative Shimizu non-aldol approach for the stereoselective synthesis of C13-C22 fragment of callistatin A**

Sandip A. Pujari and Krishna P. Kaliappan\*

A stereoselective synthesis of C13-C22 fragment of callistatin A by using an iterative Shimizu non-aldol approach is reported.



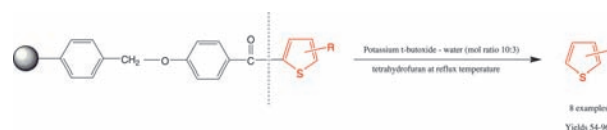
## PAPERS

1754

**Polymer-supported syntheses of thiophene-containing compounds using a new type of traceless linker**

Abderrazak Ben-Haida and Philip Hodge\*

A traceless linker that is based on the Haller–Bauer cleavage of polymer-supported aryl 2-thienyl ketones is described.

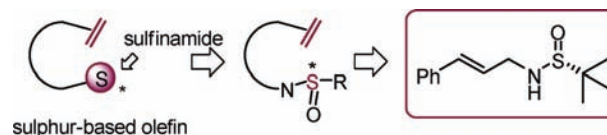


1764

**Design of *N*-cinnamyl sulfinamides as new sulfur-containing olefin ligands for asymmetric catalysis: achieving structural simplicity with a categorical linear framework**

Shen-Shuang Jin, Hui Wang, Ting-Shun Zhu and Ming-Hua Xu\*

The design and development of an extraordinarily simple new class of chiral sulfur–olefin hybrid ligands with remarkable structural simplicity were described. These unique sulfinamide–olefin ligands have been proved to be highly effective ligands in Rh-catalyzed asymmetric 1,4-additions.



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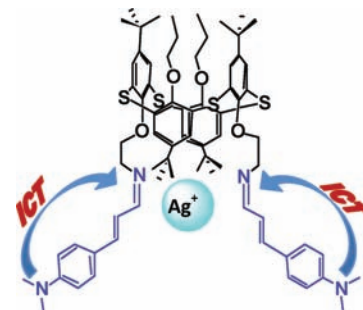
## PAPERS

1769

**Thiacalix[4]arene-cinnamaldehyde derivative: ICT-induced preferential nanomolar detection of Ag<sup>+</sup> among different transition metal ions**

Manoj Kumar,\* Naresh Kumar and Vandana Bhalla

A new thiacalix[4]arene-cinnamaldehyde derivative **3**, which undergoes red shift in the fluorescence spectrum in the presence of Ag<sup>+</sup> ions, has been synthesized.



1775

**Colour-responsive fluorescent oxy radical sensors**

Baris Yucel,\* Bahar Sanli, Huseyin Akbulut, Suheyra Ozbey and Andrew C. Benniston\*

Chameleon molecules change fluorescence colour by altering their oxidation state.

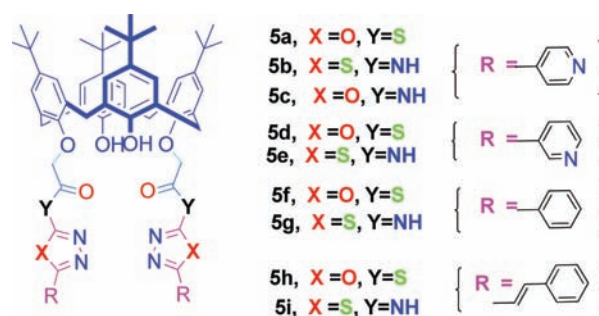


1785

**Calix[4]arene based 1,3,4-oxadiazole and thiadiazole derivatives: Design, synthesis, and biological evaluation**

Manishkumar B. Patel, Nishith R. Modi, Jignesh P. Raval and Shobhana K. Menon\*

Some novel calix[4]arene based 1,3,4-oxadiazole and 1,3,4-thiadiazole were synthesized as antioxidant, antimycobacterial, antimicrobial and antifungal agents and evaluated for their *in vitro* activities against different strains.

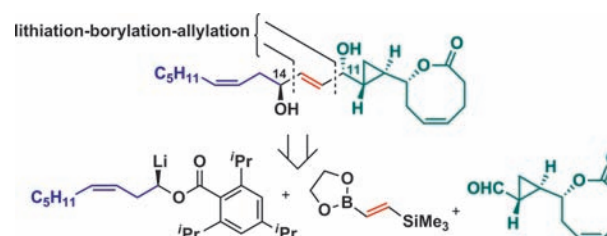


1795

**Stereocontrolled asymmetric synthesis of syn-E-1,4-diol-2-enes using allyl boronates and its application in the total synthesis of solandelactone F**

Anna Robinson and Varinder K. Aggarwal\*

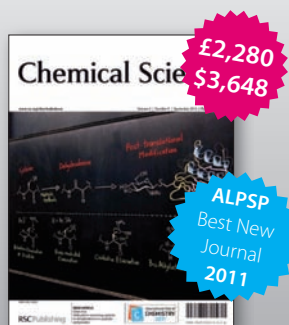
A highly stereocontrolled total synthesis of solandelactone F is reported using lithiation–borylation–allylation as the key step.



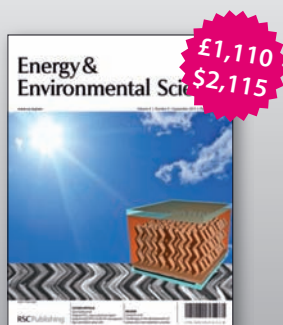
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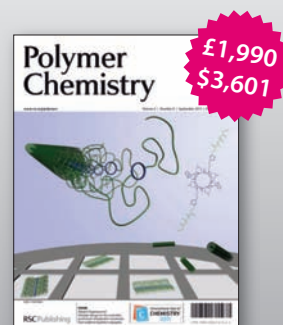
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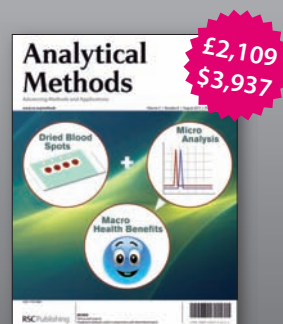
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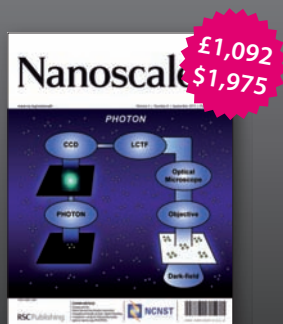
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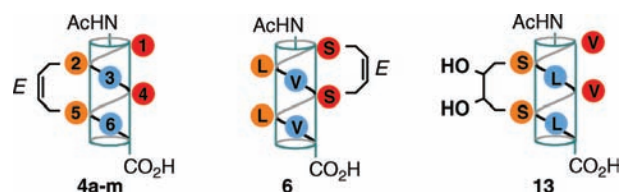
## PAPERS

1802

**Conformational stability studies of a stapled hexa- $\beta^3$ -peptide library**

Romila D. Gopalan, Mark P. Del Borgo, Ylva E. Bergman, Sharon Unabia, Roger J. Mulder, Matthew C. J. Wilce, Jacqueline A. Wilce, Marie-Isabel Aguilar\* and Patrick Perlmutter\*

An extensive library of new, 14-helical, hexa  $\beta^3$ -peptides demonstrates that appropriately stapled hexa- $\beta^3$ -peptides can allow for a number of variations, including staple size, location and functionalization, without significant perturbation of the 14-helix.

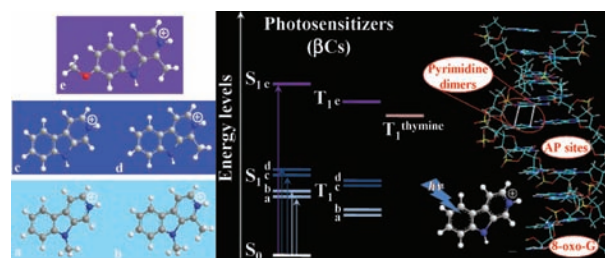


1807

**Photosensitization of DNA by  $\beta$ -carbolines: Kinetic analysis and photoproduct characterization**

M. Micaela Gonzalez, Mariana Vignoni, Magali Pellon-Maison, Matias A. Ales-Gandolfo, Maria R. Gonzalez-Baro, Rosa Erra-Balsells, Bernd Epe\* and Franco M. Cabrerizo\*

The type and extent of DNA modifications photosensitized by  $\beta$ -carboline alkaloids strongly depend on both the pH and the chemical structure of the photosensitizer.

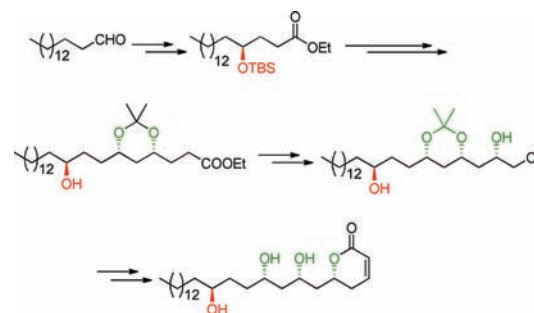


1820

**Organocatalytic stereoselective synthesis of passifloricin A**

Pradeep Kumar,\* Menaka Pandey, Priti Gupta and Dilip D. Dhavale

An efficient enantioselective synthesis of passifloricin A has been achieved in high diastereomeric excess by a combination of iterative proline-catalyzed sequential  $\alpha$ -aminoxylation, Horner–Wadsworth–Emmons olefination and ring-closing metathesis.

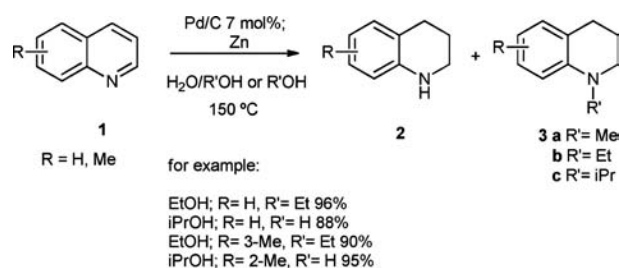


1826

**An efficient one pot transfer hydrogenation and N-alkylation of quinolines with alcohols mediated by Pd/C/Zn**

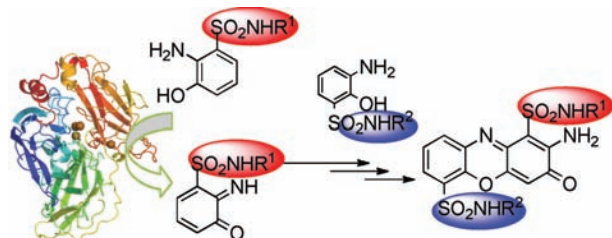
Belén Abarca,\* Rosa Adam and Rafael Ballesteros\*

1,2,3,4-Tetrahydroquinolines and N-alkylated tetrahydroquinolines from quinolines have been obtained with excellent yields in one step.



## PAPERS

1834

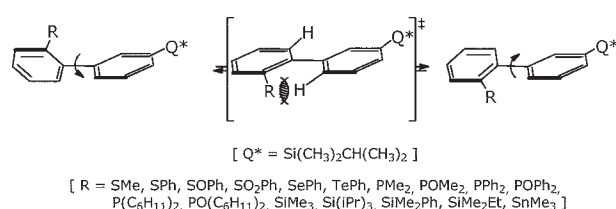


### Non-symmetrically substituted phenoxazinones from laccase-mediated oxidative cross-coupling of aminophenols: an experimental and theoretical insight

Frédéric Bruyneel, Georges Dive and Jacqueline Marchand-Brynaert\*

The study of laccase-mediated cross-coupling reactions of pairs of aminophenol substrates brought evidence that the first Michael addition step proceeds outside the catalytic pocket.

1847

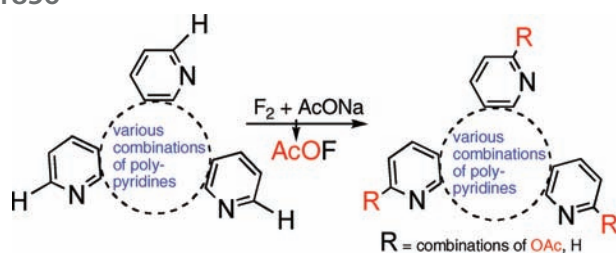


### Rotational barriers of biphenyls having heavy heteroatoms as *ortho*-substituents: experimental and theoretical determination of steric effects

Lodovico Lunazzi, Michele Mancinelli, Andrea Mazzanti,\* Susan Lepri, Renzo Ruzziconi\* and Manfred Schlosser\*

The free energies of activation for the aryl–aryl rotation of 17 biphenyl derivatives, bearing a heavy heteroatom (S, Se, Te, P, Si, Sn) *ortho* substituent, have been measured by variable temperature NMR. The present values extend the available list of steric *B* values.

1856

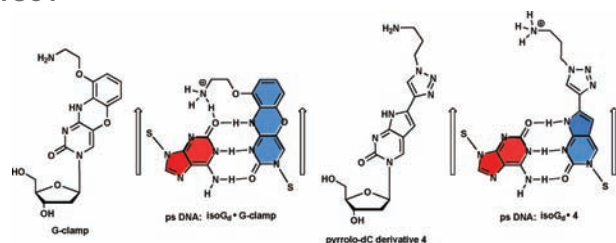


### Activation of a CH bond in polypyridine systems by acetyl hypofluorite made from F<sub>2</sub>

Julia Gateno, Youlia Hagooly, Inna Vints and Shlomo Rozen\*

General method for oxygenation of polypyridine systems at the  $\alpha$ -position using AcOF.

1861



### Parallel-stranded DNA: Enhancing duplex stability by the 'G-clamp' and a pyrrolo-dC derivative

Xin Ming, Ping Ding, Peter Leonard, Simone Budow and Frank Seela\*

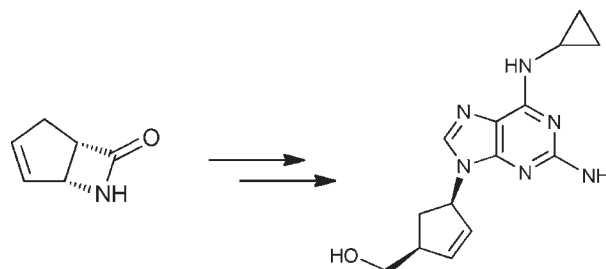
G-clamp **3** and derivative **4** were incorporated into ps and aps DNA, and **3** forms extraordinary stable base pairs in ps DNA.

## PAPERS

1870

**Enantioselective synthesis of the carbocyclic nucleoside (–)-abacavir**

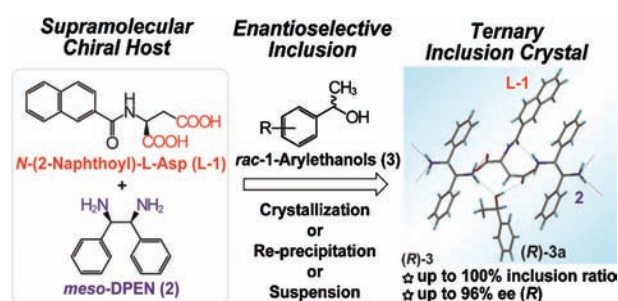
Grant A. Boyle, Christopher D. Edlin, Yongfeng Li, Dennis C. Liotta, Gareth L. Morgans\* and Chitalu C. Musonda

A route to the carbocyclic nucleoside (–)-Abacavir starting from a readily available  $\beta$ -lactam is described.

1877

**Enantioseparation of 1-arylethanols via a supramolecular chiral host consisting of *N*-(2-naphthoyl)-L-aspartic acid and an achiral diamine**

Koichi Kodama,\* Ayaka Kanno, Eriko Sekine and Takuji Hirose\*

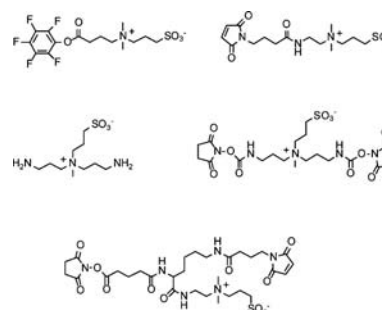
A new supramolecular chiral host was constructed from *N*-acylated L-aspartic acid and an achiral diamine for enantioseparation of 1-arylethanols.

1883

**Zwitterionic reagents for labeling, cross-linking and improving the performance of chemiluminescent immunoassays**

Anand Natrajan,\* David Sharpe and David Wen

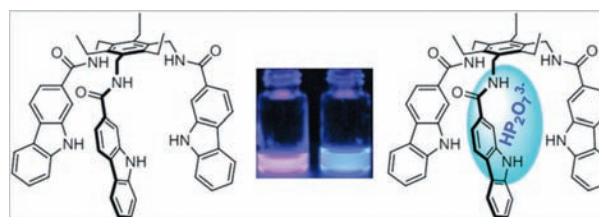
Reactive zwitterionic reagents improve the solubility of hydrophobic antigens, reduce the non-specific binding of proteins and improve chemiluminescent immunoassay performance.



1896

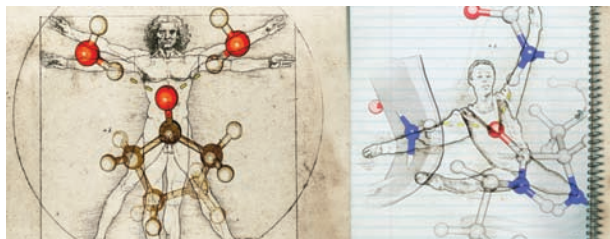
**Combined study of anion recognition by a carbazole-based neutral tripodal receptor in a competitive environment**

David Curiel,\* Guzmán Sánchez, Carmen Ramírez de Arellano, Alberto Tárraga and Pedro Molina

Receptors based on the novel carbazole-2-carboxamide unit have been evaluated in anion complexation studies to reveal a good affinity towards  $\text{HP}_2\text{O}_7^{3-}$  anion in highly polar media.

## PAPERS

1905

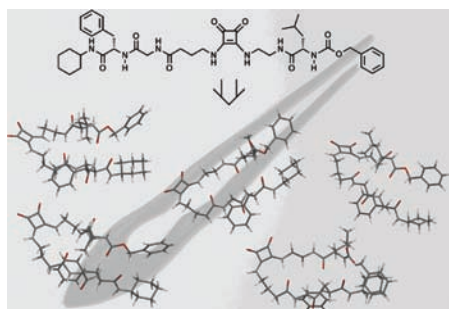


### Hydrogen-bond stabilization in oxanion holes: *grand jeté* to three dimensions

Luis Simón\* and Jonathan M. Goodman\*

Molecular dynamics and ONIOM calculations reaffirm the conclusion from crystallographic studies that oxanion holes are not usually stabilized by planar arrangements of H-bonds, but by 3D arrangements. This sub-optimal transition state stabilization leads to better overall catalysis.

1914

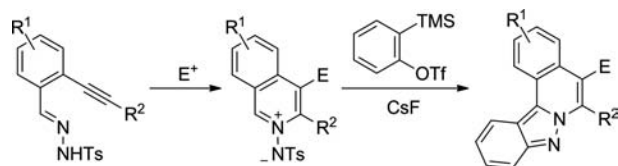


### Synthesis and conformational studies of peptido-squaramide foldable modules: a new class of turn-mimetic compounds

Luis Martínez, Angel Sampedro, Elena Sanna, Antoni Costa\* and Carmen Rotger\*

The combination of a dissecondary squaramide with 4-aminobutyric acid affords a new turn module that can be functionalized with dipeptide chains to induce the formation of a hairpin-like structure.

1922



### Aryne [3 + 2] cycloaddition with *N*-sulfonylpyridinium imides and *in situ* generated *N*-sulfonylisoquinolinium imides: a potential route to pyrido[1,2-*b*]indazoles and indazolo[3,2-*a*]isoquinolines

Jingjing Zhao, Pan Li, Chunrui Wu,\* Hongli Chen, Wenying Ai, Renhong Sun, Hailong Ren, Richard C. Larock and Feng Shi\*

*N*-Tosylpyridinium imides and *in situ* generated *N*-tosylisoquinolinium imides react with arynes in a [3 + 2] cycloaddition fashion and readily afford pyrido[1,2-*b*]indazoles and indazolo[3,2-*a*]isoquinolines after elimination of a sulfinate anion.