

# Organic & Biomolecular Chemistry

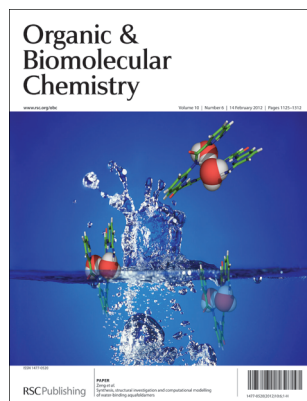
An international journal of synthetic, physical and biomolecular organic chemistry

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## IN THIS ISSUE

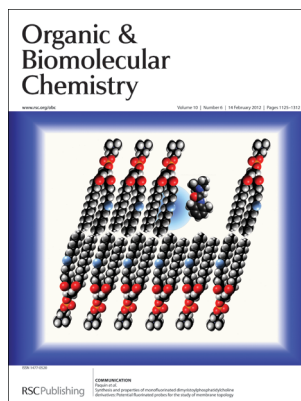
ISSN 1477-0520 CODEN OBCRAK 10(6) 1125–1312 (2012)



### Cover

See Zeng *et al.*, pp. 1172–1180.

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### Inside cover

See Paquin *et al.*, pp. 1145–1148.

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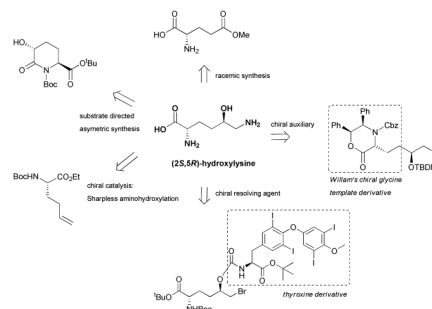
## PERSPECTIVE

1137

### Synthesis of glycosylated 5-hydroxylysine, an important amino acid present in collagen-like proteins such as adiponectin

Katherine R. Herbert, Geoffrey M. Williams, Garth J. S. Cooper and Margaret A. Brimble\*

This perspective highlights the importance of glycosylated (2*S*,5*R*)-hydroxylysine in the observed bioactivity of collagen and related collagen-like proteins such as adiponectin, an important target for the treatment of type II diabetes.



## COMMUNICATIONS

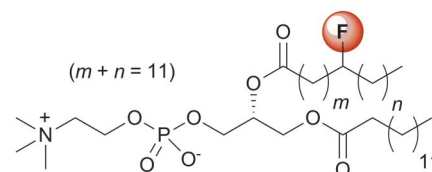
1145

### Synthesis and properties of monofluorinated dimyristoylphosphatidylcholine derivatives: Potential fluorinated probes for the study of membrane topology

Jonathan Guimond-Tremblay, Marie-Claude Gagnon, Jozy-Ann Pineault-Maltais, Vanessa Turcotte, Michèle Auger\* and Jean-François Paquin\*

The synthesis of three monofluorinated dimyristoylphosphatidylcholine derivatives (F-DMPC) is described. FTIR studies suggest that the presence of the fluorine atom does not significantly perturb the lipid conformational order and phase transition temperature.

### Potential NMR probes for the study of membrane topology



monofluorinated dimyristoylphosphatidylcholine derivatives (F-DMPC)

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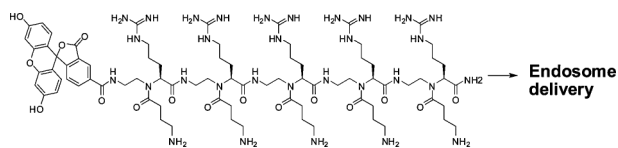
## COMMUNICATIONS

1149

**Cellular uptake of an  $\alpha$ -AApeptide**

Ge Bai, Shruti Padhee, Youhong Niu, Rongsheng E. Wang, Qiao Qiao, Robert Buzzeo, Chuanhai Cao\* and Jianfeng Cai\*

Here we show that an  $\alpha$ -AApeptide is able to penetrate the membranes of living cells from an extracellular environment and enter the endosome and cytoplasm of cells. The efficiency of the cellular uptake is unexpectedly comparable to a Tat peptide (48–57) of the same length and superior to an  $\alpha$ -peptide with identical functional groups.

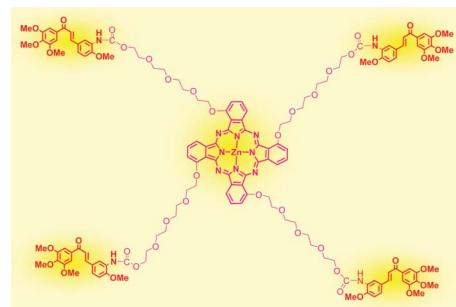


1154

**Towards dual photodynamic and antiangiogenic agents: design and synthesis of a phthalocyanine-chalcone conjugate**

Sinem Tuncel, Jérémie Fournier-dit-Chabert, Florian Albrieux, Vefa Ahsen,\* Sylvie Ducki\* and Fabienne Dumoulin\*

A phthalocyanine-chalcone conjugate has been designed and prepared to combine the vascular disrupting effect of chalcones with the photodynamic effect of phthalocyanines.

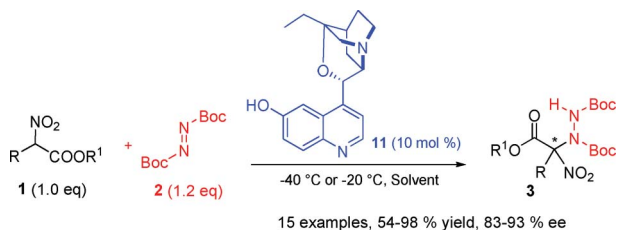


1158

**Direct amination of  $\alpha$ -substituted nitroacetates using di-*tert*-butyl azodicarboxylate catalyzed by Hatakeyama's catalyst  $\beta$ -ICD**

Cong-Bin Ji, Yun-Lin Liu, Xiao-Li Zhao, Yin-Long Guo,\* Hao-Yang Wang and Jian Zhou\*

We report the first example of catalytic asymmetric direct amination of  $\alpha$ -monosubstituted nitroacetates using di-*tert*-butyl azodicarboxylate, catalyzed by Hatakeyama's catalyst  $\beta$ -ICD.



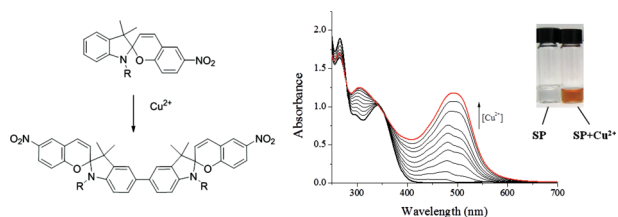
## PAPERS

1162

**Interaction studies between photochromic spiropyrans and transition metal cations: the curious case of copper**

Manuel Natali and Silvia Giordani\*

We report the first example of cross-coupling mediated by copper(II) in mild conditions causing the symmetric dimerization of spiropyran dyes.



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(Merck Award Winner)  
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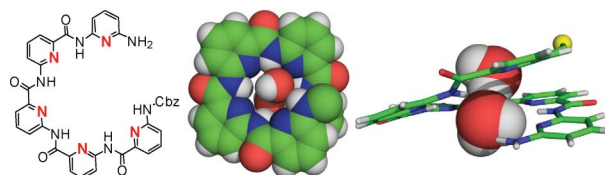
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1172

### Synthesis, structural investigation and computational modelling of water-binding aquafoldamers

Huaiqing Zhao, Wei Qiang Ong, Xiao Fang, Feng Zhou, Meng Ni Hii, Sam Fong Yau Li, Haibin Su and Huaqiang Zeng\*

A series of water-binding aquafoldamers have been studied in detail, illustrating their potential use for recognizing larger water clusters of diverse topologies and as synthetic water channels.

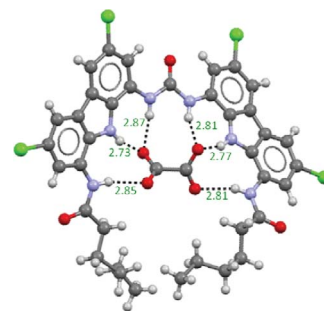


1181

### Bis-amidocarbazolyl urea receptor for short-chain dicarboxylate anions

M. Belén Jiménez, Victoria Alcázar, Rafael Peláez, Francisca Sanz, Ángel L. Fuentes de Arriba and M. Cruz Caballero\*

High values of association constants in DMSO-*d*<sub>6</sub> were observed for malonate and succinate diTBA salts with bis-amidocarbazolylurea receptor **1**. X-ray structures of the 1 : 1 complexes with oxalate and malonate show good geometrical complementarity between host and guests. Lower association is obtained with larger succinate dianion.

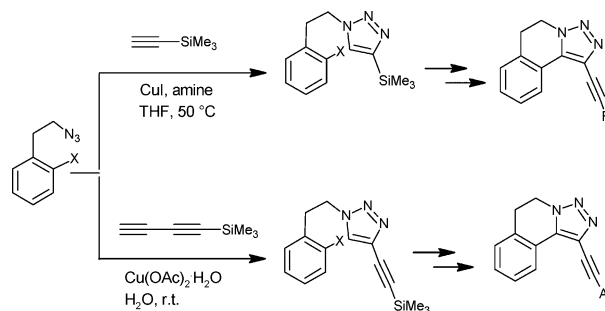


1186

### A general procedure for the synthesis of alkyl- and arylethynyl-1,2,3-triazole-fused dihydroisoquinolines

Vito Fiandanese,\* Silvia Maurantonio, Angela Punzi and Giacomo G. Rafaschieri

A general procedure for the synthesis of the title compounds has been devised by means of click reactions followed by a transition metal-catalyzed functionalization of C–H bond.

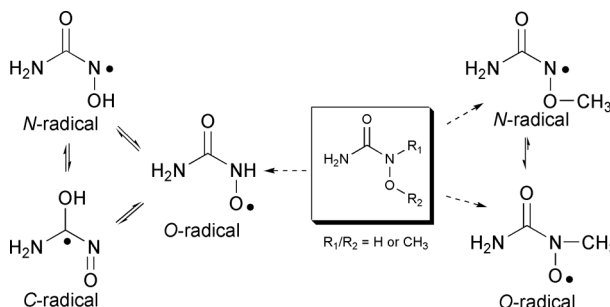


1196

### Computational study of radicals derived from hydroxyurea and its methylated analogues

Ivana Vinković Vrček, Davor Šakić, Valerije Vrček,\* Hendrik Zipse and Mladen Birus

Stabilities and reactivities of *N*- and *O*-centered free radicals derived from hydroxyurea derivatives have been investigated by using quantum-chemical methods.



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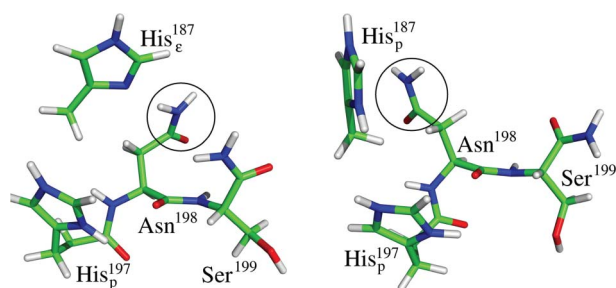
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1207

**Mechanism of C-terminal intein cleavage in protein splicing from QM/MM molecular dynamics simulations**

Jon I. Mujika,\* Xabier Lopez and Adrian J. Mulholland

We investigate the mechanism and determinants of reaction in the GyrA intein focusing on the requirements for asparagine activation for its cyclization.

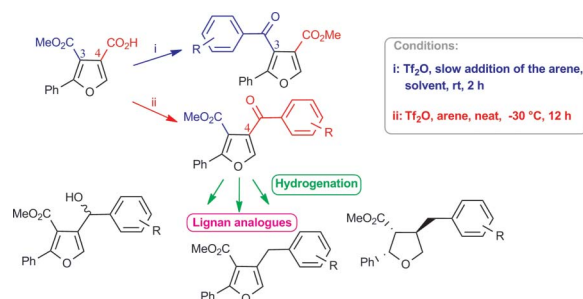


1219

**Regiodivergent synthesis of trisubstituted furans through Tf<sub>2</sub>O-catalyzed Friedel–Crafts acylation: a tool for access to tetrahydrofuran lignan analogues**

Daniela Comegna,\* Marina DellaGreca,\* M. Rosaria Iesce, Lucio Previtera, Armando Zarrelli and Simona Zuppolini

3- or 4-Aroylfurans have been prepared selectively and in high yields from a common precursor by simple tuning of reaction conditions in Friedel–Crafts acylation promoted by triflic anhydride.

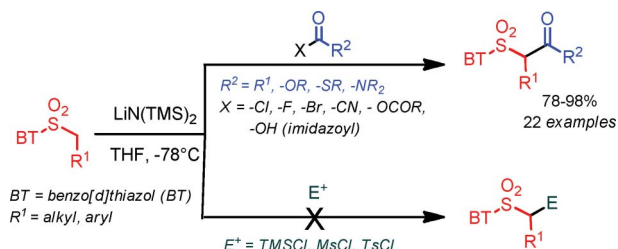


1225

**Practical synthesis of β-oxo benzo[d]thiazolyl sulfones: Scope and limitations**

Jiří Pospíšil,\* Raphaël Robiette, Hitoshi Sato and Kevin Debrus

A systematic study of the reactivity of generated benzo[d]thiazolyl (BT) sulfone α-anions towards electrophiles (X–CO–Y, TMSCl and RSO<sub>2</sub>X) is reported.

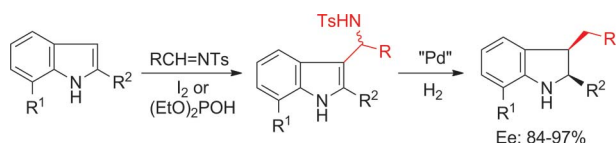


1235

**Pd-Catalyzed asymmetric hydrogenation of 3-(toluenesulfonamidoalkyl)-indoles**

Ying Duan, Mu-Wang Chen, Qing-An Chen, Chang-Bin Yu and Yong-Gui Zhou\*

A series of 2-substituted 3-(toluenesulfonamidoalkyl)indoles was synthesized and hydrogenated using chiral Pd catalyst, giving the 2,3-disubstituted indolines with up to 97% ee.



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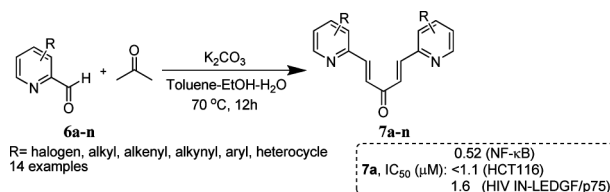


1239

### Synthesis of the pyridinyl analogues of dibenzylideneacetone (pyr-dba) *via* an improved Claisen–Schmidt condensation, displaying diverse biological activities as curcumin analogues

Bin Cao, Yong Wang, Kan Ding, Nouri Neamati and Ya-Qiu Long\*

An efficient synthesis of pyr-dba was achieved by Claisen–Schmidt condensation using  $K_2CO_3$  as base and Toluene–EtOH– $H_2O$  as solvents. These curcumin analogues display diverse biological activities.

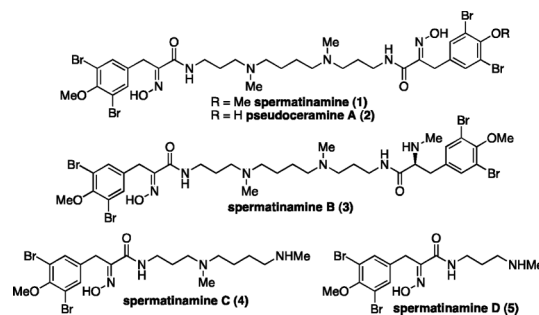


1246

### Syntheses of pseudoceramines A–D and a new synthesis of spermatinamine, bromotyrosine natural products from marine sponges

J. Mikael Hillgren, Christopher T. Öberg and Mikael Elofsson\*

The total syntheses of five bromotyrosine alkaloid natural products have been achieved, utilising a direct acyl substitution of  $\alpha$ -hydroxyiminoesters with amine nucleophiles as the key transformation.

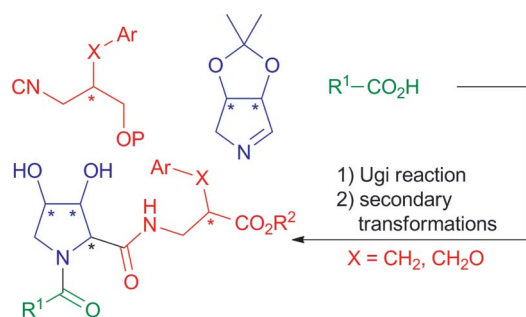


1255

### Diversity oriented and chemoenzymatic synthesis of densely functionalized pyrrolidines through a highly diastereoselective Ugi multicomponent reaction

Valentina Cerulli, Luca Banfi, Andrea Basso, Valeria Rocca and Renata Riva\*

A highly diastereoselective Ugi reaction allows the convergent preparation of enantiomerically pure pyrrolidine-based peptidomimetics.

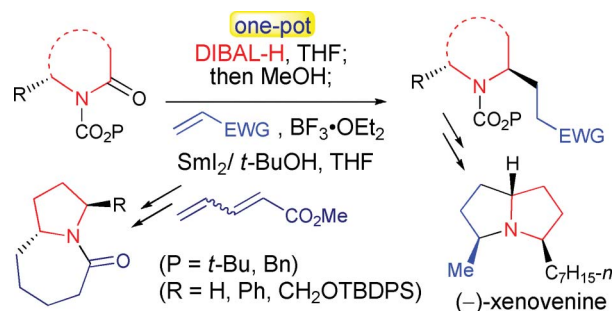


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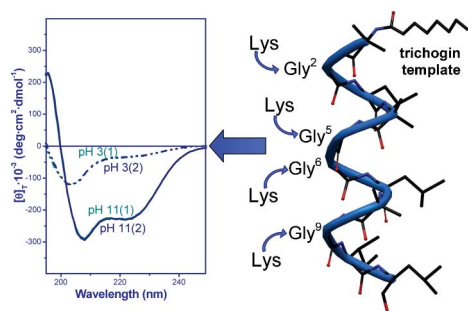
### One-pot reductive coupling of *N*-acylcarbamates with activated alkenes: application to the asymmetric synthesis of pyrrolo[1,2-*a*]azepin-5-one ring system and (–)-xenovenine

Xue-Kui Liu, Xiao Zheng, Yuan-Ping Ruan, Jie Ma and Pei-Qiang Huang\*

The one-pot reductive coupling of *N*-acylcarbamates with activated alkenes was developed, which opened a ready entry to pyrrolo[1,2-*a*]azepin-5-one and (–)-xenovenine.



1285

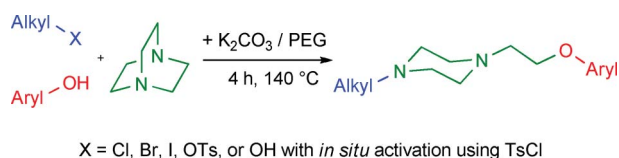


### Trichogin GA IV: A versatile template for the synthesis of novel peptaibiotics

Marta De Zotti, Barbara Biondi, Cristina Peggion, Fernando Formaggio, Yoonkyung Park, Kyung-Soo Hahm and Claudio Toniolo\*

One or more Gly-to-Lys replacements on the hydrophilic face of the peptide-template trichogin GA IV modulate its biological properties and promote a pH-mediated, reversible,  $3_{10}$ - to  $\alpha$ -helix transition.

1300



### Ring-opening reactions of 1,4-diazabicyclo[2.2.2]octane (DABCO) derived quaternary ammonium salts with phenols and related nucleophiles

Nenad Maraš, Slovenko Polanc and Marijan Kočevar\*

1,4-Dialkylpiperazines were synthesized from DABCO, alkyl halides or sulfonates, or *in situ* activated alcohols, using also phenolates and other nucleophiles.

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