

JOURNAL OF THE CHEMICAL SOCIETY

## Perkin Transactions 1

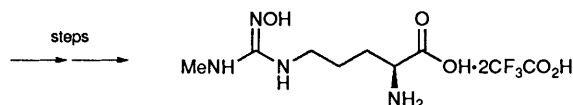
Organic and Bio-organic Chemistry

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## Perkin Communications

769 Synthesis and biological activity of the novel nitric oxide synthase inhibitor *N*<sup>ω</sup>-hydroxy-*N*<sup>ω</sup>-methyl-L-arginine

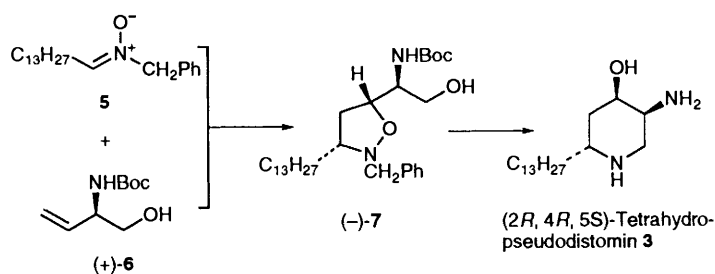
Humphrey A. Moynihan, Stanley M. Roberts, Hazel Weldon, Graham H. Allcock, Erik E. Ånggård and Timothy D. Warner



*N*<sup>ω</sup>-Hydroxy-*N*<sup>ω</sup>-methyl-L-arginine has been synthesised in eight steps from *N*<sup>δ</sup>-Z-L-ornithine and has been found to inhibit the biosynthesis of nitric oxide

773 Cycloaddition of a nitron to 2-aminobut-3-en-1-ol for large-scale preparation of 3-aminopiperidin-4-ols: a new asymmetric synthesis of (2*R*,4*R*,5*S*)-tetrahydropseudodistomin

Takeaki Naito, Miho Ikai, Mitsuko Shirakawa, Kuniko Fujimoto, Ichiya Ninomiya and Toshiko Kiguchi

777 New stereoselective synthesis of *trans*-2,5-disubstituted pyrrolidines by cyclization of aminyl radicals generated from 2- and/or 5-substituted *N*-chloro-*N*-alkylalk-4-enylamines with Bu<sub>3</sub>SnH-azoisobutyronitrile

Masao Tokuda, Hirotake Fujita and Hiroshi Suginome

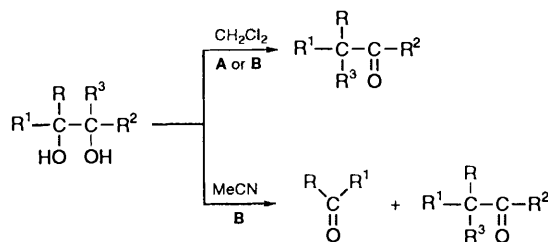


$R^1, R^2$  or  $R^3 = H, \text{alkyl or phenyl}$

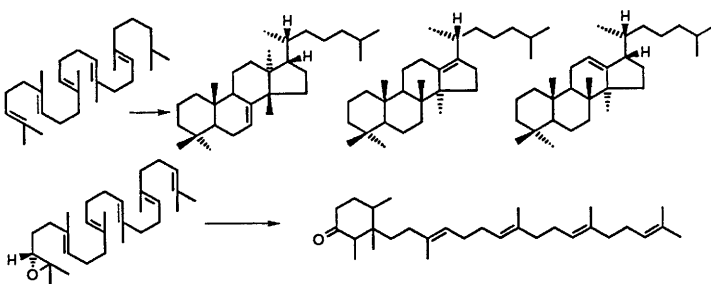
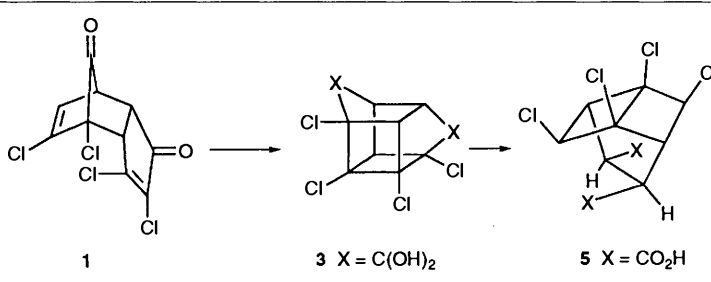
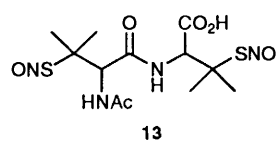
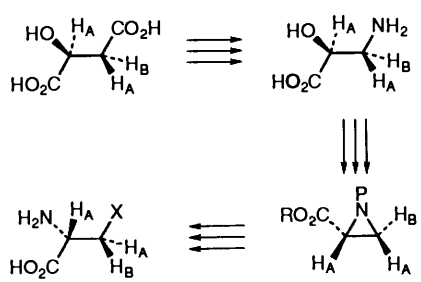
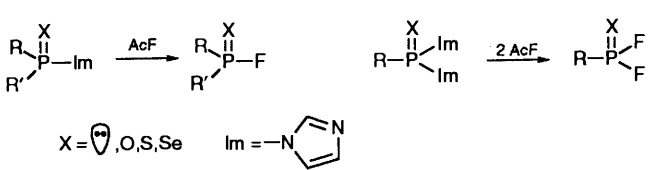
Reagents and conditions: i, NCS, benzene; ii, Bu<sub>3</sub>SnH-AIBN, benzene, reflux

## 779 Pinacol-pinacolone rearrangement induced by aminium salts

Luigi Lopez, Giuseppe Mele and Costanza Mazzeo

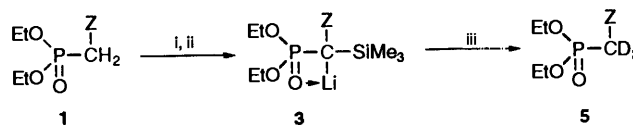


## Articles

- 783 **Enzymic cyclization of 2,3-dihydrosqualene and squalene 2,3-epoxide by squalene cyclases: from pentacyclic to tetracyclic triterpenes**
- Ikuro Abe and Michel Rohmer
- 
- 793 **Synthesis of (1*S*\*,2*S*\*,3*S*\*,4*S*\*,5*S*\*,6*S*\*,7*S*\*,8*S*\*)-1,2,7,8-tetrachlorotricyclo[4.2.0.0<sup>3,8</sup>]-octane-4,5-dicarboxylic acid. Novel entry into the C<sub>2</sub>-bisecocubane system**
- Colin J. Baker, Gordon I. Fray, Graham R. Geen and Keith Ryan
- 
- 797 **Preparation of some novel *S*-nitroso compounds as potential slow-release agents of nitric oxide *in vivo***
- Humphrey A. Moynihan and Stanley M. Roberts
- 
- Some novel bis-nitrosothiols have been prepared, for example 13. This compound shows hypotensive and platelet aggregation-inhibitory activities
- 807 **Versatile synthesis of stereospecifically labelled D-amino acids *via* labelled aziridines—preparation of (2*R*,3*S*)-[3-<sup>2</sup>H<sub>1</sub>]- and (2*R*,3*R*)-[2,3-<sup>2</sup>H<sub>2</sub>]-serine; (2*S*,2'*S*,3*S*,3'*S*)-[3,3'-<sup>2</sup>H<sub>2</sub>]- and (2*S*,2'*S*,3*R*,3'*R*)-[2,2',3,3'-<sup>2</sup>H<sub>4</sub>]-cystine; and (2*S*,3*S*)-[3-<sup>2</sup>H<sub>1</sub>]- and (2*S*,3*R*)-[2,3-<sup>2</sup>H<sub>2</sub>]-β-chloroalanine**
- B. Svante Axelsson, Kevin J. O'Toole, Philip A. Spencer and Douglas W. Young
- 
- 817 **New general synthesis of organophosphorus P-F compounds *via* reaction of azolides of phosphorus acids with acyl fluorides: Novel route to 2-deoxynucleosidyl phosphorofluoridates and phosphorodifluoridates**
- Wojciech Dąbkowski, Jan Michalski, Jacek Wasiak and Friedrich Cramer
- 
- R, R' = alkyl, alkoxy, aryl, aryloxy, 3' or 5'-nucleosidyloxy

821 General synthesis of  $\alpha,\alpha$ -dideuteriated phosphonic esters

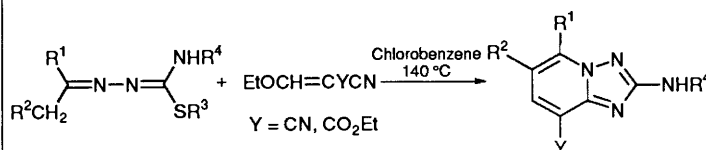
Sylvie Berté-Verrando, François Nief, Carl Patois and Philippe Savignac



Reagents and conditions: i, LDA (2 equiv.)–THF,  $-78\text{ }^{\circ}\text{C}$ ; ii,  $\text{ClSiMe}_3$ ,  $0\text{ }^{\circ}\text{C}$ ; iii,  $\text{LiOD-D}_2\text{O-THF}$ ,  $20\text{ }^{\circ}\text{C}$

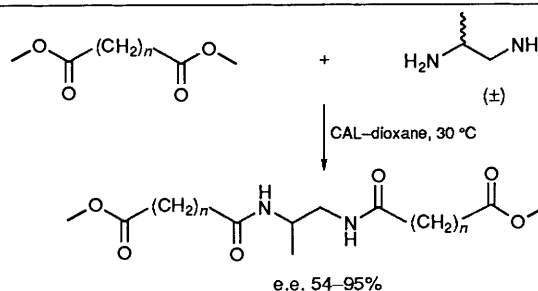
825 Cyclization of isothiosemicarbazones. Part 10. A novel route to 2-amino[1,2,4]triazolo-[1,5-*a*]pyridine derivatives

Chiji Yamazaki, Yoshiko Miyamoto and Hiromi Sakima

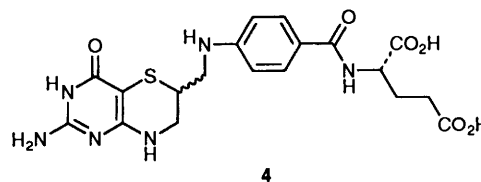


## 829 Enzymatic aminolysis of non-activated diesters with diamines

Covadonga Astorga, Francisca Robolledo and Vicente Gotor

833 Synthesis of a novel 5-deaza-5-thia analogue of tetrahydrofolic acid, *N*-(*p*-{[(2-amino-6,7-dihydro-4-oxo-3*H*,8*H*-pyrimido[5,4-*b*][1,4]-thiazin-6-yl)methyl]amino}benzoyl)glutamic acid

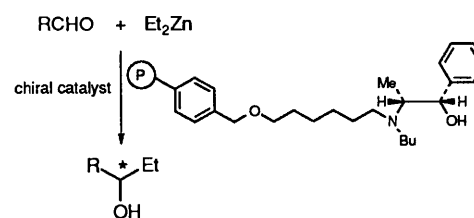
Reiko Totani, Magoichi Sako, Kosaku Hirota and Yoshifumi Maki



A 5-deaza-5-thia analogue of tetrahydrofolic acid, **4**, is synthesised as a diastereoisomeric mixture

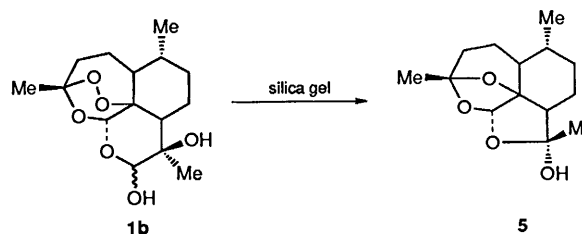
## 837 Enantioselective addition of diethylzinc to aldehydes using chiral polymer catalysts possessing a methylene spacer

Masami Watanabe and Kenso Soai



## 843 Tandem silica gel-catalysed rearrangements and subsequent Baeyer–Villiger reactions of artemisinin derivatives

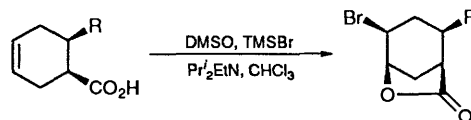
Boris Yagen, Yu Ming Pu, Herman J. C. Yeh and Herman Ziffer



The silica gel catalysed conversion of **1b** into **5** is described. A mechanism of the reaction and its implication are discussed

- 847 **Dimethyl sulfoxide–trimethylsilyl bromide–amine system as a bromonium ion source containing a potential internal nucleophile; unusual bromolactonisation of cyclohex-3-enecarboxylic acid derivatives**

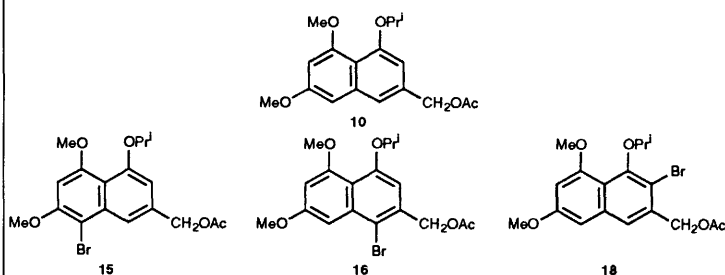
Kazuyuki Miyashita, Akira Tanaka, Hiroaki Mizuno, Masahiro Tanaka and Chuzo Iwata



Unusual *cis*-bromolactonisation of the cyclohex-3-enecarboxylic acid derivatives (R = H, Ph) occurred by employing a DMSO–TMSBr–amine system

- 853 **Regioselective bromination, debromination and bromine migration in a 2-acetoxymethyl-4,5,7-trialkoxynaphthalene**

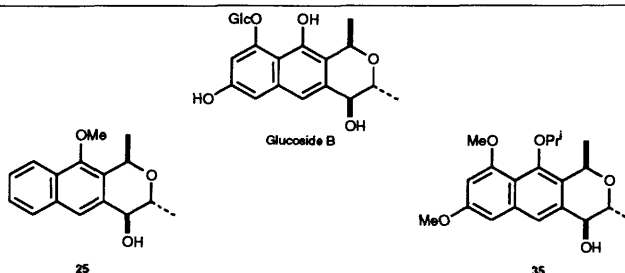
Robin G. F. Giles, Ivan R. Green, Lorraine S. Knight, Vanessa R. Lee Son, Peter R. K. Mitchell and Selwyn C. Yorke



Compound **10** may be selectively monobrominated to afford products **15**, **16** or **18**

- 859 **Stereoselective base-induced conversion of naphthalenic precursors into naphthopyrans related to the aphin-derived glucoside B**

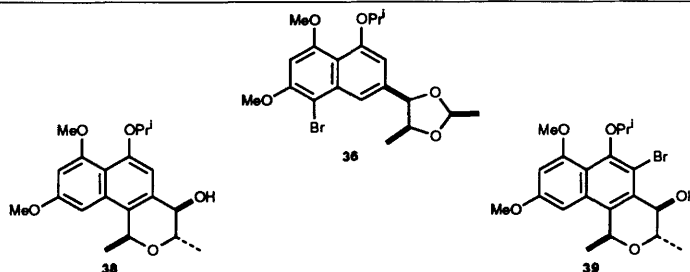
Robin G. F. Giles, Ivan R. Green, Lorraine S. Knight, Vanessa R. Lee Son and Selwyn C. Yorke



Racemic analogues **25** and **35** of Glucoside B are assembled from appropriate naphthalenes

- 865 **The stereoselective formation of naphtho[1,2-*c*]-pyrans, angular analogues of the aphin-derived glucoside B, by an intramolecular version of the Mukaiyama reaction of 4-naphthyldioxolanes**

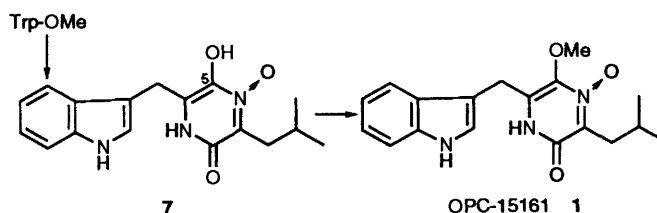
Robin G. F. Giles, Ivan R. Green, Lorraine S. Knight, Vanessa R. Lee Son and Selwyn C. Yorke



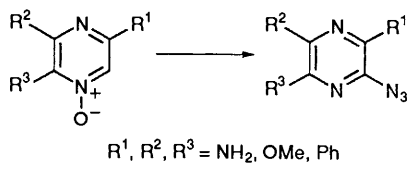
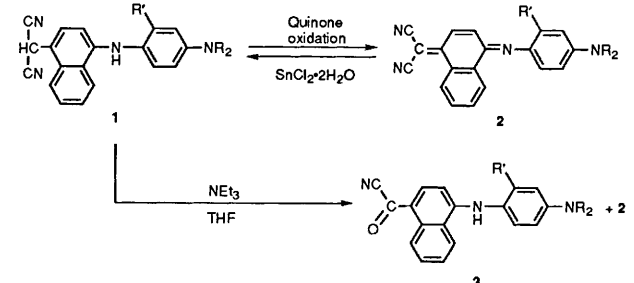
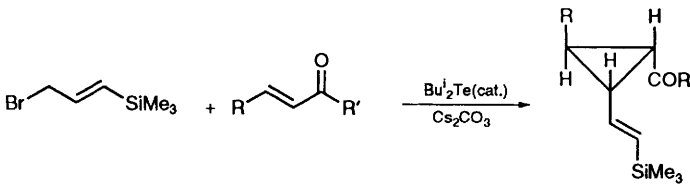
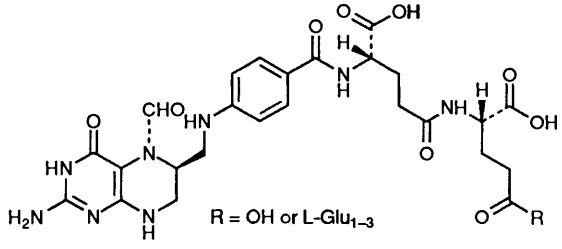
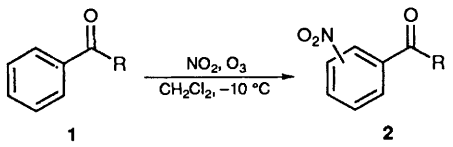
Products **38** and **39** are formed from the dioxolane **36**, using titanium tetrachloride

- 875 **Synthesis of 2,5-dioxygenated pyrazine 4-oxides: total synthesis of a new inhibitor of superoxide anion generation, OPC-15161**

Yasuyuki Kita, Shuji Akai, Hiromichi Fujioka, Yasumitsu Tamura, Hitoshi Tone and Youichi Taniguchi



Total synthesis of OPC-15161 **1** has been achieved in 4 or 6 steps in 9.9–10.6% overall yields through methylation of the 5-hydroxy group of the key precursor **7** prepared from tryptophan methyl ester

<p>885 <b>Studies on pyrazines. Part 27. A new deoxidative nucleophilic substitution of pyrazine <i>N</i>-oxides; synthesis of azidopyrazines with trimethylsilyl azide</b></p> <p>Nobuhiro Sato, Naoko Miwa and Noriko Hirokawa</p>	 <p style="text-align: center;"><math>R^1, R^2, R^3 = \text{NH}_2, \text{OMe}, \text{Ph}</math></p>
<p>889 <b>Naphthoquinone methide-type near-IR colour formers: their synthesis and oxidation properties</b></p> <p>Yuji Kubo</p>	
<p>893 <b>An efficient and highly stereoselective synthesis of trimethylsilylvinylcyclopropane derivatives via an organotelluronium salt: first example of catalytic Wittig-type cyclopropanation</b></p> <p>Yao-Zeng Huang, Yong Tang, Zhang-Lin Zhou, Wei Xia and Li-Ping Shi</p>	
<p>897 <b>First completely chemical synthesis of [(6<i>S</i>)-<i>N</i><sup>5</sup>-formyltetrahydropteroyl]poly-<math>\gamma</math>-L-glutamic acid derivatives</b></p> <p>Anthony L. Fitzhugh, Rhone K. Akee, Faith C. Rueti, Jerry Wu, John R. Klose and Bruce A. Chabner</p>	 <p style="text-align: center;"><math>R = \text{OH or L-Glu}_{1-3}</math></p>
<p>903 <b>Ozone-mediated nitration of aromatic ketones and related compounds with nitrogen dioxide</b></p> <p>Hitomi Suzuki and Takashi Murashima</p>	 <p style="text-align: center;"><i>ortho</i> : <i>meta</i> = 1.1–3.8/1.0</p>

## Corrigenda

- 909 **New synthetic applications of sialic acid aldolase, a useful catalyst for KDO synthesis. Relation between substrate conformation and enzyme stereoselectivity** Udo Kragl, Astrid Gödde, Christian Wandrey, Nadège Lubin and Claudine Augé
- 909 **Tumour-targetted boranes. Part 1. Coupling of *closo*-carboranes to substituted 2-nitroimidazoles via 1,3-dipole cycloaddition** Martin Scobie, Mary F. Mahon and Michael D. Threadgill
- 910 **Fluorination of 5'-deoxy-5'-(methylthio)adenosine with xenon difluoride provides an expedient synthesis of (fluoromethylthio)adenosine** George Guillerm and Marie Gâtel

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NOTE: An asterisk in the heading of each paper indicates the author who is to receive any correspondence.

## Forthcoming Articles in *Perkin Transactions 1*

Double Enantioselective Transesterification of Racemic Carboxylic Esters and Cyclic *meso*-Diols by Lipase Catalysis  
**F. Theil, A. Kunath, M. Ramm, T. Reiher and H. Schick**

Reaction of 1,1-Dichloro-2-(chloromethyl)cyclopropane with Some Carbanions: A Simple Synthesis of 1,2-Disubstituted Methylene-cyclopropanes  
**A. Jonczyk, I. Kmiotek-Skarzynska and T. Zdrojewski**

Novel Conversion of *E* Stereoselectivity to *Z* Stereoselectivity in Trifluoromethylated  $\alpha,\beta$ -Unsaturated Esters and Nitriles by way of *O*-Methylation of an Ylide Anion  
**Y. Shen and S. Gao**

The Chemistry of the Herbicidins. Reactivity of Silyl Enol Ethers Derived from Simple and Carbohydrate-based Tetrahydropyrans  
**T.C. Gallagher, P.J. Cox, A.M. Griffin, N.J. Newcombe, S. Lister, M.V.J. Ramsay and D. Alker**

The High-pressure Diels-Alder Reaction of Buckminsterfullerene with Several Tropones. Characterisation of the 1:1-Cycloadducts  
**H. Takeshita, J.-F. Liu, N. Kato, A. Mori and R. Isobe**

Synthesis of the Indole Nucleoside Antibiotics Neosidomycin and SF-2140  
**R.H. Wightman, J. Stoddart and J.G. Buchanan**

On the Stereochemistry of Epoxidation of Allylic and Homoallylic Cyclohexene Alcohols  
**P. Kocovsky**

Approaches to Pseudopeptidic Ergopeptines. Part 2. Consequences of the Incorporation of an  $\alpha$ -Azaproline Residue into the Oxacyclic System  
**G. Lucente, F. Pinnen, G. Luisi, A. Calcagni, E. Gavuzzo and S. Cerrini**

Quinaphthin, a Binaphthyl Quinonoid Secondary Metabolite produced by *Heliconia richonis*  
**J.S. Whitehurst, P. Adriaenssens, A.E. Anson, P.J. Fisher, K.G. Orrell, J. Webster and M.J. Begley**

Absolute Stereochemistry of Spiciferones and Spiciferin, Bioactive Metabolites of the Fungus *Cochliobolus spicifer*: Evidence for their Unique Biosynthesis  
**H. Nakajima, K. Fukuyama, H. Fujimoto, T. Baba and T. Hamasaki**

New Phenol-containing Bis(azacrown ether)s: Synthesis and Complexing Properties  
**A.V. Bordunov, N.G. Lukyanenko, V.N. Pastushok, V.I. Vetrogon, N.I. Vetrogon and J.S. Bradshaw**

Preparation of Dithiadiazafulvalene Precursors: 2-Piperidino-1,3-thiazolines or 2-Unsubstituted-1,3-thiazolines from the Reduction of the Corresponding 2-Piperidino Mesoionic Thiazoles  
**A. Robert, M. Bssaibis and A. Souzi**

Magnesium Mediated *ortho*-specific Formylation and Formaldoximation of Phenols  
**D. Levin, R. Aldred, R. Johnston and J. Neilan**

Stereoselective Synthesis of 7-Substituted Jasmonic Acid Derivatives and Investigation of their Biological Activity  
**S. Blechert, T. Taapken, E.W. Weller and M.H. Zenk**

Chemoenzymatic Large-scale Synthesis of L-Leuovorin  
**Y. Kuge, K. Inoue, K. Ando, T. Eguchi, T. Oshiro, K. Mochida, T. Uwajima, T. Sugaya, J. Kanazawa, M. Okabe and S. Tomioka**

Synthesis of Optically Active Tetracyclic Quassinoid Skeleton  
**T.K.M. Shing and Y. Tang**

A Reinvestigation of the Synthesis of *trans*-(+)-1,2,3,4,4a,10a-Hexahydrobenzo[1,4]dioxino[2,3-*c*]pyridine  
**P.A. Procopiou, P.C. Cherry, M.J. Deal and B. Lamont**

Asymmetric Synthesis of (–)-(1*R*,2*S*)-Cispentacin and Related *cis*- and *trans*-2-Amino Cyclopentane- and Cyclohexane-1-carboxylic Acids  
**S.G. Davies, O. Ichihara, I. Lenoir and I.A.S. Walters**

# 5<sup>TH</sup>

# BELGIAN ORGANIC SYNTHESIS SYMPOSIUM

organized under the auspices of  
the Royal Belgian Academy of Sciences, Letters and Fine Arts.  
Namur, July 11-15, 1994

## The symposium will include:

- The Merck-Schuchardt Chair in Organic Synthesis: a one-day course on a specific topic;
- A series of plenary lectures with ample time for discussion;
- A special lecture delivered by the recipient of Dr. Paul Janssen Prize for Creativity in Organic Synthesis;
- Poster sessions;
- An exhibition of scientific instruments, books and chemicals.

The conference will take place in the auditorium "Pedro Arrupe" of the "Facultés Universitaires Notre Dame de la Paix, Namur"

## The Merck-Schuchardt Chair

will be held on July 11 by Prof. D. Seebach, Zürich, Switzerland.  
Chairman: J.M. Lehn (Strasbourg) France

## The following distinguished scientists

have accepted the invitation to present a plenary lecture:

- Adam W. (Würzburg) Germany
- Fraser-Reid B. (Durham) U.S.A.
- Ghosez L. (Louvain-la-Neuve) Belgium
- Greene A. (St. Martin D'Herès) France
- Lehn J.-M. (Strasbourg) France
- Masamune (Cambridge) U.K.
- Murahashi S.-I. (Osaka) Japan
- Nicolaou K.C. (La Jolla) U.S.A.
- Smith A. (Philadelphia) Pennsylvania - U.S.A.
- Stoddart J.F. (Birmingham) U.K.
- Zard S.Z. (Palaiseau) France

## The organizing committee:

Prof. A. Krief, Chairman (Namur), Dr. W. Dumont (Namur), Prof. L. Hevesi (Namur)

## The scientific committee:

P. De Clercq (Gent), L. Ghosez (Louvain-la-Neuve), A. Krief (Namur), G. L'abbé (Leuven), J. Nasielski (Bruxelles), M. Vandewalle (Gent), H.G. Viehe (Louvain-la-Neuve)

## General Information:

Those who intend to contribute a poster should submit an abstract on a special form (mailed with the second circular letter).

For further information contact the chairman of the symposium:

Prof. A. Krief, Facultés Universitaires Notre Dame de la Paix, Department of Chemistry  
61, rue de Bruxelles, B-5000 Namur, Belgium  
Tel. 32.81.72.45.39 - Fax 32.81.72.45.36

Financial support such as sponsoring, publicity in the programme or participation in the exhibition is gratefully welcomed.



## The Merck-Schuchardt Chair in Organic Synthesis:

The Merck-Schuchardt Chair is a one-day course on a specific topic in organic synthesis. A scientist, who has performed excellent work in this field, gives a seminar reviewing both his own research and results of other scientists. The Merck-Schuchardt Chair will be awarded every other year on the occasion of the "Belgian Organic Synthesis Symposium" (BOSS) and carries DM 10,000. The scientific committee of the BOSS selects the subject and nominates the lecturer. The chair will be moderated by a distinguished scientist. Merck-Schuchardt's intention is to create an institution providing the latest scientific knowledge in a special field of synthesis.

## Dr. Paul Janssen Prize for Creativity in Organic Synthesis:

The Dr. Paul Janssen Prize for Creativity in Organic Synthesis will be awarded to an organic chemist by Janssen Chimica and the Janssen Research Foundation, on the occasion of the "Belgian Organic Synthesis Symposium". Nominations must be made on a form available from Dr. Marcel Janssen, Janssen Research Foundation, Turnhoutseweg 30, B-2340 Beerse, Belgium. Deadline for nominations: November 30, 1993. Rules for nominations are available at the above address. The prize consists of a medal, a citation as well as Ecu 7,500. The laureate will be announced in a special advertisement.

Jury: Prof. L. Ghosez (Chairman), Prof. H. Kagan, Prof. C. Szantay, Prof. M. Vandewalle, Prof. F. Arcamone, Prof. G. Pattenden, Prof. E. Winterfeldt, Dr. M. Janssen (Secretary)

