Advances in Heterocyclic Chemistry

Volume 102

Advances in

HETEROCYCLIC CHEMISTRY

VOLUME 102

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VOLUME **102**

Editor

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Amsterdam • Boston • Heidelberg • London • New York • Oxford
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Academic Press is an imprint of Elsevier

Academic Press is an imprint of Elsevier
Linacre House, Jordan Hill, Oxford OX2 8DP, UK
84 Theobald's Road, London WC1X 8RR, UK
Radarweg 29, PO Box 211, 1000 AE Amsterdam, The Netherlands
30 Corporate Drive, Suite 400, Burlington, MA 01803, USA
525 B Street, Suite 1900, San Diego, CA 92101-4495, USA

First edition 2011

ISBN: 978-0-12-385464-3
ISSN: 0065-2725

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Printed and bound in USA
11 12 13 14 10 9 8 7 6 5 4 3 2 1

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PREFACE TO VOLUME 102

Volume 102 of our Advances commences with the next of our continuing surveys of the "The Literature of Heterocyclic Chemistry, Part X, 2005–2007" this one covering reviews published in the period 2005–2007, again authored by Professor L.I. Belen'kii and his colleagues V.N. Gramenitskaya and Yu.B. Evdokimenkova all affiliated with the Zelinsky Institute of Organic Chemistry.

A comprehensive treatment of the "Friedländer Annulation in the Synthesis of Azaheterocyclic Compounds" has been provided by M. Shiri and Z. Tanbakouchian (Alzahra University, Tehran, Iran), M.A. Zolfigol (Bu-Ali Sina University, Hamedan, Iran), and H.G. Kruger (University of KwaZulu-Natal, Durban, South Africa); it details the wide variety of heterocyclic systems available with this methodology.

This volume closes with another contribution by A.P. Sadimenko (University of Fort Hare, Alice, South Africa) in his ongoing series covering "Organometallic Complexes of Heterocycles." The present chapter deals with "Organometallic Complexes of Aminopyridines" of which a very large number have been described with highly diverse structures and important applications in a variety of areas.

Alan R. Katritzky
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CHAPTER 1

The Literature of Heterocyclic Chemistry, Part X, 2005–2007

L.I. Belen'kii[a], V.N. Gramenitskaya[a] and Yu.B. Evdokimenkova[b]

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Advances in Heterocyclic Chemistry, Volume 102
ISSN 0065-2725, DOI 10.1016/B978-0-12-385464-3.00001-7

1. INTRODUCTION

This survey is a sequel to nine already published survey's in *Advances in Heterocyclic Chemistry* (66AHC(7)225, 79AHC(25)303, 88AHC(44)269, 92AHC(55)31, 98AHC(71)291, 99AHC(73)295, 01AHC(79)199, 04AHC(87)1, 06AHC(92)145). It includes monographs and reviews published during the period 2005–2007 as well as some published earlier but omitted in Part IX.

Like Parts III–IX, this survey is based partly on bibliographic papers published by the authors in *Khimiya Geterotsiklicheskikh Soedinenii* since 2006 (09KGS466, 09KGS939, 09KGS1107). Sources not only in English but also in Russian, Japanese, Chinese, Czech, and other languages are surveyed and classified. This feature of the survey should cause no problem because some of the sources are available in English translations and practically all others have informative English abstracts as well as quite understandable and useful schemes and lists of references. As before, carbohydrates are not covered. Such compounds are mentioned only in general cases (e.g., anomeric effect) as well as when carbohydrates serve as starting compounds for the synthesis of other heterocycles or they are present as fragments of a complex system including another heterocyclic moiety such as nucleosides.

2. GENERAL SOURCES AND TOPICS

2.1 General books and reviews

2.1.1 Textbooks and handbooks

Synthesis and properties of heteroaromatic compounds: 05MI1.

2.1.2 Annual reports

2.1.2.1 Comprehensive reports

07PHC1, 08PHC1, 09PHC1.

2.1.2.2 Specialized reports devoted to basic series of heterocycles

Three-membered heterocycles: 07PHC55, 07PHC81, 08PHC70, 09PHC47.
Four-membered heterocycles: 07PHC106, 08PHC92, 09PHC74.
Pyrrole and its benzo derivatives: 07PHC150, 08PHC135, 09PHC122.
Furan and its benzo derivatives: 07PHC187, 08PHC176, 09PHC152.
Thiophenes, selenophenes, and tellurophenes: 07PHC126, 08PHC112, 09PHC94.
Five-membered heterocycles with more than one N atom: 07PHC218, 08PHC208, 09PHC190.
Five-membered heterocycles with N and S (Se) atoms: 07PHC247, 08PHC242, 09PHC220.
Five-membered heterocycles with O and S (Se, Te) atoms: 07PHC276, 08PHC277, 09PHC253.
Five-membered heterocycles with O and N atoms: 07PHC288, 08PHC288, 09PHC265.
Pyridine and its benzo derivatives: 07PHC310, 08PHC314, 09PHC289.
Diazines and their benzo derivatives: 08PHC353, 08PHC383, 09PHC333.
Triazines, tetrazines, and fused polyaza-systems: 07PHC371, 08PHC414, 05PHC337.
Six-membered heterocycles with O and/or S atoms: 07PHC376, 09PHC365, 09PHC399.
Seven-membered heterocycles: 07PHC402, 08PHC437, 09PHC432.
Heterocycles with eight-membered and large rings: 07PHC430, 08PHC465, 09PHC459.

2.1.2.3 Reports devoted to individual problems

Annual survey of organometallic metal cluster chemistry for the year 2003: 05CCR(249)2763.
Transition metals in organic synthesis, highlights for the year 2003: 06CCR(250)300.
Transition metals in organic synthesis, highlights for the year 2004: 06CCR(250)2411.
Bioinspired organic chemistry: 06AR(B)377.
Catalysis (particularly, the use of ionic liquids in catalysis): 06AR(B)325.
Chemical genetics: 06AR(B)138.
Computational organic chemistry: 06AR(B)219.

Heterocyclic chemistry: 06AR(B)81.
Highlights of natural product synthesis (2004, 2005): 06AR(B)98.
Marine natural products: 06AR(B)123.
Mechanisms of radical and radical ion reactions: 06AR(B)247.
N-Heterocyclic carbenes in transition metal and organic catalysis: 06AR(B)168.
Oxidation and reduction methods: 06AR(B)34.
Recent developments in palladium-catalyzed heterocycle synthesis and functionalization: 05COC625.
Supramolecular chemistry (molecular recognition, structure and assembly, and functional systems): 06AR(B)148.
TEMPO (2,2,6,6-tetramethylpiperidine-*N*-oxyl) as an important reagent in alcohol oxidation and its application in synthesis of natural products between 2000 and 2004: 06MRO155.

2.1.3 Nomenclature

IUPAC nomenclature (general monograph): 04MI1.

2.1.4 History of heterocyclic chemistry, biographies

Heterocyclic chemistry in Moscow State University: 05KGS31.
History of aromaticity (heteroaromaticity) concept: 05CRV3436.
The history of Woodward–Doering/Rabe–Kindler total synthesis of quinine: 07AG(E)1378.
Input of Prof. K. C. Nicolaou in chemical biology and medicinal chemistry: 05JMC5613.

2.1.5 Bibliography of monographs and reviews

The literature of heterocyclic chemistry, 2002–2004: 06AHC(92)145.
Specialized surveys: 09KGS466, 09KGS939, 09KGS1107.

2.2 General topics by reaction type

We have classified the many reviews dealing with these materials under the following headings:

1. *General Sources and Topics.*
2. *Structure and Stereochemistry (it is self-subdivided into Theoretical Aspects, Stereochemical Aspects, Betaines and Other Unusual Structures, Miscellaneous Substituted Heterocycles).*
3. *Reactivity (General Topics: Reactions with Electrophiles and Oxidants, Reactions with Nucleophiles and Reducing Agents, Reactions Toward Free Radicals, Carbenes, etc., Reactions with Cyclic Transition State, Reactivity of Substituents, Heterocycles as Intermediates in Organic Synthesis).*
4. *Syntheses (General Topics and Nonconventional Synthetic Methodologies, Synthetic Strategies and Individual Methods, Versatile Synthons and Specific*

Reagents, Ring Synthesis from Nonheterocyclic Compounds, Syntheses by Transformation of Heterocycles).

5. *Properties and Applications (Dyes and Intermediates, Substances with Luminescent and Related Properties, Organic Conductors, Coordination Compounds, Polymers, Ionic Liquids, Miscellaneous).*

2.2.1 General sources and topics

All-metal aromaticity and antiaromaticity of inorganic heterocycles: 05CRV3716.

Anion receptors: 05H(66)689.

Conjugated polymers (particularly, polythiophene, polypyrrole, and polyfuran) and aromaticity: 05CRV3448.

Estimating aromatic stabilization energies, particularly, in heterocycles: 05CRV3773.

From macrocyclic oligo-acetylenes to aromatic ring carbomers, pericyclynes and hetero-pericyclynes: 06CRV5317.

The influence of different main group elements on relative stability of valence isomers: 07BCJ1241.

Interactions C−F...H, F...F, C−F...π in crystalline compounds including fluorine heterocycles: 05CSR22.

Intervalence charge transfer in trinuclear and tetranuclear Fe, Ru, and Os complexes with heterocyclic ligands: 06CRV2270.

Liquid chromatography – mass spectrometry: 03MI1.

Locking self-assembly of supramolecular structures possessing heterocyclic fragments: 07CSR856.

Memory of chirality and asymmetric synthesis: 05S1.

Modern HPLC: 06MI8.

Molecular recognition of oxoanions based on guanidinium receptors: 07CSR198.

Nucleus-independent chemical shifts (NICS) as an aromaticity (heteroaromaticity) criterion: 05CRV3842.

Palladium-catalyzed dynamic kinetic asymmetric allylic alkylation with the DPPBA ligands: 07AA59.

Principles of environmental chemistry (general monograph): 05MI9.

Recent advances in heterolytic nucleofugal, particularly, heterocyclic leaving groups: 07T5103.

Recent progress in stable radical chemistry (including heterocyclic radicals and radicals with heterocyclic substituents): 07OBS1321.

Tautomeric equilibria in relation to π-electron delocalization (particularly, tautomerism of heteroaromatic compounds): 05CRV3561.

Spherical aromaticity of fullerenes, polyhedral boranes, and related structures including heterofullerenes: 05CRV3613.

2.2.2 Structure and stereochemistry

2.2.2.1 Theoretical aspects

H-Bond-assembled supramolecular architectures of fullerenes functionalized by various heterocyclic fragments: 07CJO153.

Planar tetracoordinate carbon and fenestranes, in particular, azafenestranes: 06CRV4787.

Supramolecular chemistry in water (noncovalent interactions with participation of heterocycles): 07AG(E)2366.

Supramolecular species bearing quaternary azaaromatic moieties: 06H(68)1467.

2.2.2.2 Molecular dimensions

Atomic resolution crystallography of proteins and X-ray absorption fine structure studies of metalloproteins: 05CCR197.

Bioactive conformations, and structure–activity relationship of taxol and its analogs: 05ZOR329. Configuration, conformation, reactivity, and applications of hexahydropyrrolo[2,3-b]indoles in synthesis: 07ACR151.

Conformationally constrained peptide nucleic acid (PNA) analogs and DNA/RNA binding selectivity: 05ACR404.

Conformationally locked nucleotides and their analogs: 06Y681.

Coordination modes of 5-pyrazolones (X-ray diffraction data): 07CCR1561.

Effect of preferential conformations on base properties and thermodynamics of conformation conversion along with type of ring fusion on *cis–trans* conversion of bicycle in pyrrolizidines: 06KGS1443.

Metalloproteins three-dimensional structure determination using multiple-scattering analysis of X-ray absorption fine structure: 05CCR(249)141.

Rules to predict the conformational behavior of saturated seven-membered heterocycles: 05ARK(6)88.

Structural (X-ray) properties of homoleptic, mononuclear transition metal complexes of 1,2-dioxolenes: 06CCR(250)2000.

X-ray structural chemistry of cobalamins: 06CCR(250)1332.

2.2.2.3 Stereochemical aspects

Asymmetric domino reactions based on the use of chiral auxiliaries: 06T1619.

Asymmetric domino reactions based on the use of chiral catalysts and biocatalysts: 06T2143.

Asymmetric organocatalysis (general monograph): 05MI13.

Asymmetric organocatalysis of Diels–Alder, [3 + 2] and [4 + 3] cycloaddition reactions: 05Y464.

Asymmetric catalysis by chiral hydrogen-bond donors, particularly, by proline and alkaloids: 06AG(E)1520.

Asymmetric heterogeneous catalysis (heterocycles as catalysts, starting compounds and products): 06AG(E)4732.

Catalytic enantioselective construction of all-carbon quaternary stereocenters: 06S369.

Crystallization-induced diastereomer transformations: 06CRV2711.

Dynamic stereochemical rearrangements in chiral organometallic complexes with heterocyclic ligands: 07CSR551.

Efficiency in nonenzymatic kinetic resolution of chiral heterocycles: 05AG(E)3974.

Enantioselective organocatalysis (general monograph): 07MI3.

Exciting supramolecular architectures: Light-induced processes and synthetic transformations in noncovalent assemblies: 05EJO4041.

Lewis acid–base bifunctional asymmetric catalysis, particularly, in the Reissert reaction: 05SL1491.

One-pot synthesis of fused aromatics, particularly, those with diazapentaphene skeleton possessing helical chirality and assignment of absolute configuration assisted by theoretical circular dichroism: 05Y798.

Principles and recent applications of chiral auxiliaries: 06S1899.

Structure of fullerenes and fullereno-annulated heterocycles; problem of fullerene chirality: 06CRV5049.

Synthesis and the potential of thiaheterohelicenes: 06OBC2518.

Use of chiral sulfoxides as chiral auxiliaries in asymmetric synthesis of bioactive products: 06T5559.

2.2.2.4 Betaines and other unusual structures

Azafulvenium methides chemistry: 06ARK(7)89.

The chemistry of functionalized *N*-heterocyclic carbenes: 07CSR592.

Development of organic photochromic radical compounds: 07CJO696.

Formation of [5,6]- and [6,6]-open fulleroide structures, particularly, fullerenoheterocycles: 07UK768.

Synthesis and properties of cationic π-conjugated systems stabilized by bicyclo[2.2.2]octene units including annulated silatropilium ion, thiophene, 1,2-dithiine, 1,4-dithiine, and oligothiophenes: 05SL187.

Synthesis and properties of molecular rods containing heterocyclic fragments: 05CRV1197.

2.2.2.5 Miscellaneous substituted heterocycles

Chemistry of arene- and hetarene-based hydrazonoalkanenitriles: 07H(71)2545.

Fluorinated ethers, thioethers, and amines, derivatives of heterocycles: 05CRV827.

Heterofullerenes: 06CRV5191.

Persulfurated aromatic compounds including hetarenes: 06AG(E)1686.

Products of [2 + 3] cycloaddition to [60] fullerene: 05CJO159.

2.2.3 Reactivity

2.2.3.1 General topics

Activation of heteroaromatics by metal sulfonate catalysts in C–C bond forming reactions: 06Y752.

Alkynylation of chiral alkoxy-, amino-, and thio-substituted hetarencarbaldehydes: 06CRV2355.

Anion binding involving π-acidic heteroaromatic rings: 07ACR435.

Baylis–Hillman reactions in nontraditional media (water, ionic liquids, supercritical CO_2): 07CJO322.

Baylis–Hillman reaction in synthetic chemistry (particularly, cyclic amines as catalysts): 07CSR1581.

Carbonylation of heterocycles by homogeneous catalysts: 07CC657.

Chemistry of *N*-(1-haloalkyl)azinium halides: 07ARK(3)96.

Cycloacylation of thioamides and their derivatives by compounds with activated multiple bond: 07KGS1283.

Homologation of heterocycles by a sequential reductive opening lithiation–electrophilic substitution–cyclization: 06AHC(91)135.

Electrochemical fluorination of heterocyclic compounds: 06AHC(90)239.

Microwave irradiation for accelerating reactions of three- to five-membered heterocycles: 05AHC(88)1.

Microwave acceleration of reactions of six-, seven-membered, spiro and fused heterocycles: 06AHC(90)1.

Modern Pummerer-type reactions including those concerning heterocycles: 06T5003.

Proton-coupled electron transfer: 07CRV5004.

Recent advances in intermolecular direct arylation reactions of heterocycles and arenes: 07AA35.

Some aspects of the Willgerodt–Kindler reaction and connected reactions: 05H(65)411.

Templated photoreactions (including reactions of heterocycles) in homogeneous solutions: 06CRV5413.

Water as a reagent in regio- and stereoselective reactions including syntheses of heterocycles and ring opening of epoxides: 05Y18.

2.2.3.2 Reactions with electrophiles and oxidants

Advances in singlet oxygen chemistry including photooxidation of five-membered heteroaromatics with one and two heteroatoms: 05T6665.

Electrophilic reactions of aromatic and heteroaromatic compounds in ionic liquids: 06ZOR1761.

Photooxygenation of heterocycles: 05COC109.

Use of solid catalysts in Friedel–Crafts acylation reactions, particularly, in acylation of heterocycles: 06CRV1077.

Uses of sodium chlorite and sodium bromate in oxidation of heterocycles, particularly, to give lactones: 06OPP177.

2.2.3.3 Reactions with nucleophiles and reducing agents

Asymmetric hydrogenation of heteroaromatic compounds: 07ACR1357.

Carbanion generation through tin–lithium exchange of vinyl-, aryl-, and hetarylstannanes: 06ARK(9)265.

Halogen dance reaction and its application in synthesis of heteroaromatics: 07COC637, 07CSR1046.

Nucleophilic aromatic substitution of hydrogen as a tool for heterocyclic ring annulation: 07AHC(93)57.

Position-flexible elaboration of halogenated heterocycles into metalated species as key intermediates for synthesis: 07SL3096.

Recent advances in asymmetric hydrogenation of heteroaromatic compounds (quinoxaline, pyridine, and furan derivatives): 05CJO634.

Stereoselective conjugate addition reactions to heterocyclic acceptors using *in situ* metallated terminal alkynes and the development of novel chiral P,N-ligands: 07BCJ1635.

Vicarious nucleophilic substitution of hydrogen in nitro aromatics and nitro heteroaromatics: 07CJO17.

2.2.3.4 Reactions toward free radicals, carbenes, etc.

Catalysis of stannane-mediated radical chain reactions by benzeneselenol, particularly, reactions with formation and/or participation of heterocycles: 07ACR453.

Intermolecular reactions of electron-rich heterocycles with copper and rhodium carbenoids: 07CSR1109.

2.2.3.5 Reactions with cyclic transition state

The Diels–Alder cycloadditions of 3,5-dibromo-2-pyrone and its derivatives: 07PHC1.

Nucleophilic addition, 1,3-dipolar cycloaddition, and Diels–Alder reactions of indoles substituted at the 2- or 3-position with electron-withdrawing groups (NO_2, $PhSO_2$): 05COC1493.

Porphyrins in Diels–Alder and 1,3-dipolar cycloaddition reactions: 08PHC44.

2.2.3.6 Reactivity of substituents

Advances in catalytic, enantioselective aminations and oxygenations of carbonyl compounds (heterocycles and their complexes as catalysts): 05AG(E)4292.

Asymmetric benzoin reactions with *N*-heterocyclic carbene catalysts: 07Y370.

Progress in protection of carbonyl compounds as acetals, thioacetals, and oxothioacetals: 07CJO576.

2-Pyridylsilyl group as useful multifunctional group in organic synthesis: 06SL157.

2.2.3.7 Heterocycles as intermediates in organic synthesis

Advances in liquid-phase organic synthesis using functional ionic liquid as supports: 07CJO1188.

Advances in homogeneous oxidation with metalloporphyrins as catalysts: 07CJO1039.

Application of N-oxides in asymmetric catalytic reactions: 05CJO272.

Applications of *N*-chlorosuccinimide in organic synthesis: 07S3599.

Asymmetric enamine catalysis, in particular, reactions with participation of heterocycles as catalysts, starting compounds, and/or products: 07CRV5471.

Asymmetric organocatalysis, particularly, using heterocyclic, mainly, proline-derived organocatalysts: 07T9267.

The atroposelective synthesis of axially chiral biaryls *via* configurationally unstable, lactone-bridged biaryls: 02JOM(661)31.

Bifunctional organocatalysts, particularly, (S)-3-(*N*-isopropyl-*N*-3-pyridinylaminomethyl) BINOL for enantioselective aza-Morita–Baylis–Hillman (aza-MBH) reactions: 07Y1089.

Bisoxazoline ligands with chiral backbones and their applications in asymmetric catalytic reactions: 07CJO1087.

Catalytic enantioselective α-fluorination of carbonyl compounds using chiral transition metal complexes with heterocyclic ligands: 07Y1099.

Chiral glycolate equivalents for the asymmetric synthesis of α-hydroxycarbonyl compounds (heterocycles as chiral auxiliaries): 07BCJ1451.

Chiral heterocyclic amines as organocatalysts for asymmetric conjugate addition to nitroolefins and vinyl sulfones *via* enamine activation: 07CC3123.

Chiral N-oxides in asymmetric catalysis: 07EJO29.

Chiral sulfur-containing heterocyclic ligands for asymmetric catalysis: 07T1297.

1-Chloromethyl-4-fluoro-1,4-diazoniabicyclo[2.2.0]octane bis(tetrafluoroborate), Selectfluor, as fluorinating agent: 05AG(E)192.

Complete reversal of enantioselection using oxazoline-containing Schiff base ligand: 07Y969.

Creation of chiral heterocyclic ligands having acetal structures and their applications to Pd-catalyzed asymmetric reactions: 07Y1191.

Cyclic derivatives of dihydroxyacetone, particularly, 2,2-dimethyl-1,3-dioxan-4-one in organic synthesis: 05AG(E)1304.

Development of the optically active chiral ligands ferrocenyloxazolinylphosphines (FOXAP) and their application to catalytic asymmetric reactions: 07Y761.

The directed synthesis of axially chiral ligands, reagents, catalysts, and natural products through the "lactone methodology": 02JOM(661)49.

Enantiopure cyclic nitrones as a useful class of building blocks for asymmetric syntheses: 07S485.

Heterocyclic hydroperoxides in selective oxidations: 07CRV3338.

Iminium catalysis, in particular, reactions with participation of heterocycles as catalysts, starting compounds, and/or products: 07CRV5416.

Intelligent catalysts, especially, *N*,*N*'-{[(2S,5S)-1-(4-pyridinyl)-2,5-pyrrolidinediyl]dicarbonyl}bis[L-tryptophan] 3,3'-dioctyl ester in regioselective acylation of carbohydrates: 07Y1081.

Ionic-liquid-supported synthesis as a novel liquid-phase strategy for organic synthesis: 06ACR897.

Metalated heterocycles in organic synthesis: 07ARK(10)152.

New advances in N-oxidation of nitrogen-containing heterocyclic compounds: 07CJO1050.

New advances in palladium-catalyzed aerobic oxidations (*N*-heterocycles as ligands): 06CJO397.

New advances of asymmetric cyclopropanation reactions using chiral metal catalysts including metal complexes with oxazoline, porphyrin, bipyridine, and terpyridine derivatives: 07CJO438.

N-Heterocyclic carbenes catalyzed umpolung reactions *via* homoenolates: 07Y1009.

Nucleophilic organocatalysis through umpolung (pyridine, triazole, thiazole, benzimidazole, imidazole derivatives, and cyclic amines as catalysts): 07CJO545.

O/C rearrangements: a powerful strategy for the synthesis of functionalized carbocycles, mainly, from saturated *O*-heterocycles: 07T3081.

Organocatalysis by *N*-heterocyclic carbenes: 07CRV5606.

Organocatalytic asymmetric Mannich reactions (heterocycles as chiral ligands and products): 07EJO5797.

Peptide-based asymmetric catalysts: 07CJO1195.

Recent advances in kinetic resolution using nonenzymatic catalysts, mainly heterocycles: 07CJO1345.

Recent advances in the catalytic asymmetric nitroaldol (Henry) reaction including application of heterocyclic organocatalysts and ligands: 07EJO2561.

Recent development and application of chiral phase-transfer catalysts including heterocycles as catalysts: 07CRV5656.

Silica-supported Pd catalysts with heterocyclic ligands for Heck coupling reactions: 07T6949.

Small molecule activation at uranium coordination complexes, particularly, with *N*-heterocyclic ligands: 06CC1353.

Small-molecule H-bond donors in asymmetric catalysis including reactions with participation of heterocycles as catalysts, starting compounds, and/or products: 07CRV5713.

Stereoselective anhydride openings (heterocycles as catalysts): 07CRV5683.

Synthesis of chiral heterocyclic phosphines for application in asymmetric catalysis: 07COC61.

Synthetic applications and oxidation kinetics of *N*-hetarenium halochromates and dichromates: 07T4367.

Tandem oxidation processes using manganese dioxide: 05ACR851.

Transition metal-catalyzed carbochalcogenation of alkynes: 06CL1320.

Zwitterions in carbon–carbon and carbon–nitrogen bond-forming reactions as a promising synthetic strategy leading, in particular, to various heterocycles: 06ACR520.

2.2.4 Synthesis

2.2.4.1 General topics, nonconventional synthetic methodologies

Advances in microwave-assisted combinatorial synthesis: 06CJO1500.

Amine-catalyzed Baylis–Hillman reaction (proline, 1-azabicyclo[2.2.2] octane, imidazole, etc. as catalysts): 05CJO763.

Application to the preparation of useful compounds based on enzyme- and microorganism-mediated reactions: 06Y664.

Application of polymer-supported oxazo-type compounds (mainly, oxazolidinone, oxazoline, and oxazaborolidine derivatives) to asymmetric reactions: 05CJO1039.

Aryne-mediated synthesis of heterocycles: 07H(74)89.

Borylation of saturated heterocycles with several heteroatoms: 06KGS643.

Carbene complexes of nonmetals: 05EJO237.

Catalyzed reactions of alkynes in water (particularly, syntheses of *O*-heterocycles by hydroalkoxylation, *N*-heterocycles by hydroamination, syntheses of triazoles, imidazoles, furans, and hetero [2 + 2 + 2] trimerizations): 06ASC1459.

Cationic organometallic complexes of scandium, yttrium, and the lanthanoids in synthesis of heterocycles using Diels–Alder reactions and intramolecular hydroamination: 06CRV2404.

Cerium(III) chloride promoted reactions and their applications to organic synthesis: 05CJO1029.

Cp*Ir complex-catalyzed hydrogen transfer reactions directed toward environmentally benign organic synthesis including *N*-heterocyclization and hydrogenation of quinolines: 05SL560.

Convenient and general microwave-assisted protocols for the synthesis of heterocycles: 06H(70)655.

Cupreines and cupreidines as bifunctional cinchona organocatalysts: 06AG(E)7496.

N-Cyanoimines in synthesis of heterocyclic compounds: 05KGS963.

The development of enantioselective rhodium-catalyzed hydroboration of olefins (chiral bidentate P,P and P,N ligands): 05ASC609.

1,3-Dipolar cycloadditions in aqueous media: 06H(68)2177.

Domino reactions in organic synthesis: 06MI5.

Dynamic combinatorial chemistry of various heterocycles: 06CRV3652.
Environmentally benign solvents (supercritical fluids, ionic liquids, low melting polymers (especially PEG), perfluorinated solvents and water) in organic synthesis, particularly, that of heterocycles: 05COC195.
From multicomponent-reactions (MCRs) toward multifunction-component-reactions (MFCRs): 07H(73)149.
Heterocycles from transition metal catalysis. Formation and functionalization: 05MI11.
Highly efficient synthesis of heterocyclic compounds based on reactivity of trifluoroacetyl group: 06Y1282.
β-Hydride elimination in Pd(II)-catalyzed reactions proceeding in the presence of nitrogen ligands (pyridine, bipyridine, phenanthroline, etc.) for the regeneration of the Pd(II) species: 05CJO1182.
High-pressure organic synthesis (including hetero Diels–Alder and 1,3-dipolar [3 + 2] cycloaddition reactions and epoxide ring opening): 05Y770.
High-throughput and parallel screening methods in asymmetric hydrogenation: 06CRV2912.
Indium reagents in heterocyclic chemistry: 05COC1205.
The intramolecular coiodination of alkenes with internal oxygenated nucleophiles as a methodology for the preparation of heterocyclic compounds: 05COS393.
Methods for the synthesis of hetaryl-substituted 1,4-benzo- and 1,4-naphthoquinones: 05KGS803.
MCRs (general monograph): 05MI5.
The multisubstrate screening of asymmetric catalysts: 05ASC737.
New advances of Fe- and Co-catalyzed C–C coupling reactions: 07CJO703.
Nucleophilic radicals with low-lying unoccupied orbitals, such as acyl, oxyacyl, silyl, stannyl, and germyl radicals providing high regiocontrol and efficient methods for the synthesis of heterocycles: 07ACR303.
Organic chemistry in water (including syntheses of heterocycles): 06CSR68.
Organocatalytic asymmetric Mannich reactions (heterocycles as chiral ligands and products): 07EJO5797.
Photoinduced electron-transfer processes in the synthesis of heterocycles: 07ACR128.
Platinum-catalyzed hydrofunctionalization of unactivated alkenes with carbon, nitrogen, and oxygen nucleophiles in synthesis of heterocycles: 07CC3607.
Progress in application of Merrifield resin in solid phase organic synthesis including reactions with participation and formation of heterocycles: 07CJO1069.
Progress in isocyanide-based MCRs in synthesis of heterocycles: 06CRV17.

Progress of Rh-catalyzed [2 + 2], [2 + 2 + 1], [2 + 2 + 2], [3 + 2], [3 + 4], [4 + 2] cycloadditions: 07CJO958.
Recent advances in the development and applications of post-Ugi transformations: 07H(73)125.
Recent advances in microwave-assisted synthesis of heterocyclic compounds: 05COS333.
Recent approaches to the construction of 1-azaspiro[4.5]decanes and related 1-azaspirocycles: 06T3467.
Recent developments in the electrochemistry of some nitro compounds of biological significance, in particular, 1,4-dihydropyridine, imidazole, and furan derivatives: 05COC565.
Recent progress in [3 + 2] cycloaddition using azomethine ylides: 06CJO9.
Recent progress of Mitsunobu reaction, particularly, for heterocycles as reactants and products: 06CJO454.
Ru-catalyzed reconstructive synthesis of functional organic molecules *via* C−C bond cleavage: 05CL1462.
S-Alkyl-*N*-cyanoisothioureas-based synthesis of azoles, azines, and their fused derivatives: 07KGS1443.
Selected patented cross-coupling reaction technologies with participation of heterocycles as ligands in catalyst molecules, reactants, and products: 06CRV2651.
Synthesis and transformations of heterocycles under the action of microwave irradiation: 05KGS1123.
Synthesis of heterocycles using mercuric triflate as a catalyst: 06Y744.
Synthesis of heterocyclic dimethyl *o*-dicarboxylates using cycloaddition reactions of dimethyl acetylenedicarboxylate: 05JHC337.
Synthetic use of molecular iodine for organic synthesis, particularly, to form heterocycles: 06SL2159.
A synthon approach to spiro and hetero spiro compounds: 06T779.
Stereoselective construction of quaternary stereocenters: 05ASC1473.
Theoretical studies on domino cycloaddition reactions: 05MRO47.
Utilization of heteropalladation (halopalladation; oxypalladation; aminopalladation; acetyloxypalladation) of unsaturated hydrocarbons in organic synthesis, particularly, in synthesis of heterocycles: 07CJO819.
Versatile reactivity and catalytic effects of copper(II) halides in organic syntheses including formation of lactones and lactams, pyrrolidinones, pyridinones, pyrroles, and dihydrofurans; dehydrogenation of heterocycles to yield uracils, dihydroisoquinolines, and pyridazines: 05COC1737.

2.2.4.2 Synthetic strategies and individual methods

Aldolase-mediated synthesis of iminocyclitols and novel heterocycles: 06AA63.

Amination with oximes, particularly, intramolecular amination to give five- and six-membered *N*-heterocycles: 05EJO4505.

1,3-Aminoalcohols and their derivatives in asymmetric organic synthesis of *N*- and *N*,*O*-heterocycles: 07CRV767.

C−C, C−O, C−N bond formation on sp^2 carbon by Pd(II)-catalyzed reactions involving oxidant agents, particularly, alkene–hetarene coupling and syntheses of various *O*- and *N*-heterocycles: 07CRV5318.

Cascade reactions in the synthesis of heterocycles: 06ARK(7)416, 07AHC (94)1.

Chemical and biochemical transformations in ionic liquids: 05T1015.

Chemical routes for the transformation of biomass into chemicals, particularly, into furfural, *N*-heterocycles, and epoxides: 07CRV2411.

Chemistry of the anionically activated aromatic CF_3 group and synthesis of heterocycles: 07MRO183.

The chemistry of mercapto- and thione-substituted 1,2,4-triazoles and their utility in heterocyclic synthesis: 06ARK(9)59.

Design of highly functional small-molecule catalysts and related reactions based on acid–base combination chemistry (particularly, synthesis of oxazolines and thiazolines and L-histidine-derived sulfonamide as a minimal artificial acylase for the kinetic resolution of racemic alcohols): 07SL686.

Development and applications of electron-transfer-initiated cyclization reactions, particularly, applications to heterocycle synthesis: 07SL191.

Domino synthesis of carbo- and heterocycles involving a 1,3 or 1,4 C→O silyl migration: 07SL177.

Enantioselective synthesis of heterocycles mediated by organoselenium reagents: 06ARK(7)186.

Environmentally benign solvent systems and a greener [4 + 2] cycloaddition process for synthesis of heterocycles: 07MRO89.

Gold-catalyzed organic reactions, particularly, olefin epoxydation and hydroamination or hydroalkoxylation of alkynes to give *N*- or *O*-heterocycles: 07CRV3180.

Heterocycles as versatile building blocks in different synthetic strategies: 06ARK(7)395.

Hypervalent iodine(V) reagents, particularly, benzoiodoxole oxide in organic synthesis: 06ARK(9)26.

Microwave-assisted synthesis in water as solvent: 07CRV2563.

Microwave irradiation for accelerating reactions with participation and/or formation of heterocycles: 05ZOR1757, 06ARK(9)1.

Modern strategies in organic catalysis based on iminium activation using imidazolidinones as catalysts: 06AA79.

New heterocyclization reactions with Fischer carbene complexes: 06ARK (7)129.

New routes to organofluorine compounds (including fluorinated heteroaromatics) based on ketenes and the radical transfer of xanthates: 07OBC205.

Nitrobutadienes from β-nitrothiophenes as valuable building-blocks in the overall ring-opening/ring-closure protocol to homo- or heterocycles: 06ARK(7)169.

Nonconventional methodologies for Heck reaction, particularly, with participation of heteroaromatics: 05T11771.

Nucleophilic dearomatizing (D_NAr) reactions of aromatic C,H-systems, particularly, of heterocycles: 07CRV1580.

Poly(ethylene glycol) supported synthesis of heterocyclic libraries: 05CJO1157.

Preparation of heterocyclic compounds *via* Ti- or Zr-metallacycles: 05Y102.

Progress in the synthesis of heterocycle-fused C_{60}-fullerene derivatives: 06CJO168.

Radical conjugate additions with participation and formation of heterocycles including stereoselective reactions, cascade reactions, and application of conjugate additions in total synthesis of natural products: 05T10377.

Rapid, "green," predictable microwave-assisted synthesis with participation and formation of heterocycles: 05ACR653.

Recent advances in synthesis of condensed heterocycles on polymeric supports: 05CJO152.

Recent developments in the synthesis of heterocyclic derivatives by PdI_2-catalyzed oxidative carbonylation reactions of suitably functionalized alkynes: 03JOM(687)219.

Recent applications of the dealkoxycarbonylation reactions of malonate esters, β-keto esters, α-cyanoesters, and related analogs including preparation and participation of heterocycles: 07ARK(2)1, 07ARK(2)54.

1,4-Sila- and stannatropic strategy for generation of 1,3-dipoles and its application to heterocyclic synthesis: 06ARK(7)370.

Solvent-free and highly concentrated reactions, particularly, those with formation and/or participation of heterocycles as a green chemistry approach to asymmetric catalysis: 07CRV2503.

The Sonogashira reaction in syntheses with formation and/or participation of heterocycles: 07CRV874.

C_2-Symmetric chiral bis(oxazoline) ligands in asymmetric catalysis: 06CRV3561.

Synthesis of azaheterocycles by one-electron reduction of oximes: 06ARK (7)245.

Synthesis of cyclic nitrones by bromocyclization of unsaturated oximes: 06ARK(7)76.

Synthesis of heterocycles *via* palladium-catalyzed oxidative addition: 06CRV4644.

Synthesis of heterocyclic compounds by carbenoid transfer reactions: 07COC177.

Synthesis of β-tropolone and fused heterocycle derivatives by acid-catalyzed and photoreactions of *o*-quinones with quinolines and benzimidazoles: 06ARK(7)439.

Synthetic uses of ynolate-initiated heterocyclizations: 07T10.

Transformation of carbon dioxide, particularly, in synthesis of oxazolidinones and cyclic carbonates or copolymerization with oxiranes: 07CRV2365.

Transition metal catalyzed [2 + 2 + 2] cycloaddition and application in organic synthesis, particularly, that of heterocycles: 05EJO4741.

2.2.4.3 Versatile synthons and specific reagents

Aldehyde *N*,*N*-dialkylhydrazones as neutral acyl anion equivalents, particularly, in synthesis of heterocycles: 07EJO5629.

Application of allenic compounds tandem cyclizations, particularly, in ring opening of ethynylaziridines with participation of organocopper compounds, Pd-catalyzed stereoselective cyclization of allenes leading to aziridines, pyrrolidines, and benzoindoles: 05YZ899.

Application of 2-iodoxybenzoic acid as oxidizing agent in organic synthesis: 06CJO1623.

Asymmetric hetero-Diels–Alder reactions of Danishefsky's and Brassard's dienes with aldehydes: 07SL2147.

Asymmetric synthesis of *N*-heterocycles (including alkaloids) using *N*-sulfinyl imines: 06ARK(7)120.

Boron-substituted building blocks in Diels–Alder and other cycloaddition reactions, particularly, those with participation and formation of heterocycles: 05S2091.

Cerium(IV) ammonium nitrate – A versatile single-electron oxidant, particularly, in heterocyclic chemistry: 07CRV1862.

CF_3-bearing aromatic and heterocyclic building blocks: 06AG(E)5432.

The chemistry of hydrazonates including their use for the synthesis of heterocycles: 07COC773.

Cyclic nitriles, particularly, those of heterocyclic series in synthesis: 05T747.

Derivatives of polyfluoroalkanethioic acids in synthesis of fluorine-containing heterocycles: 05RKZ(6)107.

Development of boron-based reactions and reagents for organic synthesis, particularly, of heterocycles: 07Y1048.

4,4-Dialkoxybutan-2-ols as synthons, particularly, for preparation of heterocycles: 05ZOR9.

Domino reactions of 1,3-bis(silyl enol ethers) with 4-(trialkylsilyloxy)benzopyrylium triflates to give various carbo- and heterocycles: 07SL1016.

Enantiopure sulfinamides in asymmetric synthesis of aziridines, pyrrolidine phosphonates, and natural bioactive heterocycles: 05AA93.
Formation of halogen and chalcogen derivatives via hetaryllithium intermediates: 05S3477.
Functionalized magnesium organometallics as versatile intermediates for the synthesis of polyfunctional heterocycles: 06CC583.
Generation, properties, and synthetic applications of nitrile ylides: 07COC741.
Guanidines in organic synthesis, particularly, that of heterocycles: 06S737.
N-Heterocyclic carbenes as versatile organocatalysts and reagents: 06CJO745.
Lithium aminoborohydrides in reduction of *N*-alkyllactams and synthesis of 2-dialkylaminopyridines: 05AA61.
New synthetic applications and biological activity of diazenes: 05JHC401.
Nitrenium ions and direct electrophilic aromatic amination (cyclic nitrenium ions, formation of *N*-heterocycles by intramolecular amination, *N*-aminopyridinium salts): 05ZOR487.
Organotrifluoroborates in organic synthesis (reactions with oxiranes, epoxidation of, hetaryltrifluoroborates in Suzuki reaction): 05AA49.
Pd-catalyzed direct arylation of simple arenes (hetarenes) in synthesis of biaryl molecules: 06CC1253.
Pd-catalyzed domino oligocyclizations of acyclic 2-bromoenynes and 2-bromoenediynes: 05ACR413.
Preparation and reactions of heteroaryl organomagnesium compounds: 06CL2.
Privileged building blocks in chemo-differentiating ABB MCRs (reactions introducing into the final product one molecule of component A and two molecules of component B): 07CSR484.
Recent advances in stereoselective syntheses using *N*-acylimines (particularly, with formation of heterocycles): 07S159.
Recent developments of biaryl synthesis including that of arylhetarenes *via* cross-coupling reactions of areneboronic acid derivatives: 05Y312.
Recycle application of catalyst supported by ionic liquids to asymmetric reaction: 06CJO1362.
Solid-supported reagents, catalysts, and their use to solution-phase combinatorial synthesis: 06CJO885.
Synthesis and application of functionalized chiral ionic liquids: 06CJO1031.
Synthesis and stereoselective functionalization of silylated heterocycles as a new class of formyl anion equivalents: 06CC4881.
Synthesis of enaminones and their utility in organic (mainly, heterocyclic) synthesis: 06CJO1192.
Synthesis of heterocycles by using thiones and selones: 05Y791.

Synthetic applications of Buchwald's phosphines in palladium-catalyzed aromatic-bond-forming reactions, mainly those leading to *N*- and *O*-heterocycles: 06AA17.
The use of phosphorus-centered radicals in organic chemistry, particularly, syntheses and transformations of heterocycles: 05CSR858.
Titanium complexes in enantioselective Friedel–Crafts, [4 + 2] and [3 + 2] cycloaddition reactions, epoxidation and other oxidation processes: 06CRV2126.
2-(Trimethylsilyl)ethanesulfonyl (or SES) group in amine protection and activation: 06CRV2249.
Utilities of some carbon nucleophiles in heterocyclic synthesis: 07COC853.
Utility of cyanoacetic acid hydrazide in heterocyclic synthesis: 06ARK(9)113.

2.2.4.4 Ring synthesis from nonheterocyclic compounds

Arsonium ylides in synthesis of heterocycles: 05T1385.
Asymmetric MCRs: 05AG(E)1602.
Asymmetric organocatalytic domino reactions: 07AG(E)1570.
CH/π hydrogen bonds in organic reactions, particularly, in hetero Diels–Alder reaction: 05T6923.
Cascade reactions of carbonyl ylides for heterocyclic synthesis: 09PHC20.
Citronellal as key compound in organic synthesis, particularly, of heterocycles: 07T6671.
Condensation of thioamides with acetylencarboxylic acids derivatives: 06ZOR807.
The construction of C$-$N double bonds by aza-Wittig reaction in the preparation of heterocycles: 07T523.
Criss-cross cycloaddition reactions with hexafluoroacetone azine to give partially fluorinated heterocycles and polymers: 06H(67)443.
gem-Disubstituent effect in formation of heterocycles: 05CRV1735.
The domino way to heterocycles: 07T5341.
Fluorine-containing 2-functionalized 1,3-dicarbonyl compounds for heterocyclic synthesis: 06H(69)593.
Formation of furoxans by cyclization of 1,2-dinitroso compounds: 05CSR797.
Formation of heterocycles by cyclizations of allylic substrates *via* palladium catalysis: 05T3457.
Formation of heterocycles by transition metal-catalyzed hydroarylation of alkynes: 05S167.
Generation of iminium ylides from carbenes and carbenoids and their synthetic application: 05UK183.
Gold catalysis, particularly that of reactions leading to formation of heterocycles: 06AG(E)7896.

Heterocycle synthesis by copper-facilitated addition of heteroatoms to alkenes, alkynes, and arenes: 07CSR1153.

Heterocyclization reactions with participation of thiols: 06ZOR647.

Hexamethyldisilathiane-based thionation of carbonyl compounds as a versatile approach to sulfur-containing heterocycles, particularly, to isothiazoles: 05SL1965.

Intramolecular dipolar cycloaddition reactions of azomethine ylides: 05CRV2765.

1-Lithio-1,3-dienes as useful building blocks for heterocyclic compounds: 07BCJ1021.

Metal vinylidenes and allenylidenes in catalysis of anti-Markovnikov additions to terminal alkynes and alkene metathesis, particularly with participation and/or formation of heterocycles: 06AG(E)2176.

Multicomponent syntheses of heterocycles by transition-metal catalysis: 07CSR1095.

N-Vinylic phosphazenes as a tool for the synthesis of acyclic and heterocyclic compounds: 06COC2371.

One- or two-step sequences for the synthesis of heterocycles from versatile nitroalkanes: 07T12099.

Pd-catalyzed bicyclization in the one-step formation of heteropolycyclic ring systems: 06COC1325.

Quinoneimides in synthesis of heterocycles: 05SL2407.

Reactions of functionalized alkoxyethylenes with nucleophilic reagents in synthesis of five-, seven-membered heterocycles and macroheterocycles: 06ZOR167.

Recent advances in the chemistry of α,β-unsaturated trifluoromethylketones including their use for synthesis of heterocycles: 07T7753.

Ring closing enyne metathesis as a powerful tool for the synthesis of heterocycles: 07CSR55.

Ring-closing metathesis as a basis for the construction of aromatic and heteroaromatic compounds: 06AG(E)2664.

Ring-closing metathesis of heteroatom-substituted dienes: 06H(70)705.

Ring-closing metathesis of substrates containing more than two C–C double bonds as a rapid access to functionalized heterocycles: 06COC1363.

Ring-closure reactions of *ortho*-vinyl-*tert*-anilines via the *tert*-amino effect: 06S2625.

Secondary amides as *ortho*-directed metallation groups for arenes; a useful construction way for polysubstituted aromatic and heteroaromatic systems: 06COC1817.

Stereoselective allylboration using (*B*)-γ-alkoxyallyldiisopinocampheylboranes for the synthesis of heterocycles: 07ARK(2)121.

Synthesis of enamines, enol ethers, and related compounds by cross-coupling reactions, particularly, synthesis of heterocycles including natural products: 05CC973.

Synthesis of heterocycles by transition metal-catalyzed cyclocarbonylation reactions: 06COC1397.

Synthesis of heterocycles through hydrosilylation, silylformylation, silylcarbocyclization, and cyclohydrocarbonylation reactions: 06COC1341.

Synthesis of partially fluorinated heterocycles from 4,4-bis(trifluoromethyl)-substituted hetero-1,3-dienes *via* C−F bond activation and their application as trifluoromethyl-substituted building blocks: 06H(69)569.

Synthesis of perfluoroalkyl heterocycles using perfluoroolefins containing a reactive group at the double bond: 05AHC(88)231.

Synthesis of polyfluoroalkyl-substituted heterocycles based on unsaturated compounds possessing polyfluoroalkyl groups: 06KGS323.

Synthesis of polyfunctional heterocycles based on aromatic nitriles: 06IVUZ(4)3.

Synthesis of pyrrolidine derivatives and lactones based on conjugate addition of nitroalkanes to electron-poor alkenes: 05CRV933.

Thiourea and selenourea and their applications, particularly, in the preparation of heterocycles: 06COS439.

The use of anthranilic acid as a starting material for synthesis of heterocycles: 06COS379.

2.2.4.5 Syntheses by transformation of heterocycles

Advances in siloxane-based aryl–aryl coupling reactions for synthesis of bihetaryls: 05T12201.

Aryl-hetaryl bond formation by transition metal-catalyzed direct arylation: 07CRV174.

Carbon-carbon coupling reactions of heterocycles catalyzed by heterogeneous Pd catalysts: 07CRV133.

Catalytic direct arylation of heteroaromatic compounds: 07CL200.

Direct transition metal-catalyzed functionalization of heteroaromatic compounds: 07CSR1173.

Hypervalent iodine chemistry (I-heterocycles and synthesis of heterocycles): 05AG(E)3656.

Innovative catalytic protocols for the ring-closing Friedel–Crafts-type alkylation and alkenylation of arenes and hetarenes: 06EJO3527.

Modern synthetic methods for fluorine-substituted target molecules, particularly, with participation or preparation of heterocycles: 05AG(E)214.

Regioselective (site-selective) functionalization of unsaturated halogenated nitrogen, oxygen, and sulfur heterocycles by Pd-catalyzed cross-couplings and direct arylation processes: 07CSR1036.

Ring-closure reactions of heterocyclic analogs of *o*-vinyl-*tert*-anilines *via* the *tert*-amino effect: 06S2625.

Synthesis of biaryls and arylhetarenes using Pd/C-catalyzed Suzuki–Miyaura reaction: 06EJO2679.

Thioisomünchnones in synthesis of heterocycles: 05ACR460.

2.2.5 Properties and applications (except drugs and pesticides)

2.2.5.1 Dyes and intermediates

Advances in study of the indocyanine dyes: 06CJO442.

Current developments in optical data storage with organic dyes including *N*-heterocycles and their metal complexes: 06AG(E)2016.

Improving the properties of organic dyes by molecular encapsulation (particularly, cyclodextrin-encapsulated dyes and rotaxane dyes): 05EJO4051.

2.2.5.2 Substances with luminescent and related properties

Classification and mechanism of the action of sun-protecting natural and synthetic organic UV filters: 06IVUZ(11)3.

Design, synthesis, and biological activity of unnatural enediynes (including those with heterocyclic fragments) and related analogs equipped with pH-dependent or phototriggering devices: 07CRV2861.

Designing tridentate ligands, 2,2′-bipyridine and 2,2′:6′2″-terpyridine derivatives, for ruthenium(II) complexes with prolonged room temperature luminescence lifetimes: 05CSR133.

Electron and energy transfer modulation with photochromic switches (dithienylethenes and spiro[chromene-2,2′-indolines]): 05CSR327.

Fluorescent oligopyridines and their photo-functionality as tunable fluorophores: 07COC195.

Functional bioluminescent imidazopyrazinone dyes: 06Y1062.

Heterocycles as materials for fluorescence resonance energy transfer analysis: 06AG(E)4562.

Heterocycle-based dendrimers for use in organic light-emitting diodes and solar cells: 07CRV1097.

Heterocyclic photochromes as molecular switches controlled by light: 06CC1169.

Luminescent sensors and switches in the early 21st century: 05T8551.

Molecular engineering of octupolar NLO molecules and materials based on bipyridyl metal complexes: 05ACR691.

Photoactive metallocyclodextrins: 05CSR120.

Recent development of 6π-electrocyclic photochromic systems: 06CL1204.

Research of electrophosphorescent organometallic complexes (*N*-heterocycles as ligands): 05CJO873.

Research progress of 8-hydroxyquinoline metal complexes in electroluminescent devices: 06CJO783.

Spiro compounds for organic optoelectronics: 07CRV1011.

A survey of the synthesis of photochromic material diarylethenes, mainly, dithienylethenes: 07CJO175.

2.2.5.3 Organic conductors (except polymers)

Charge carrier transporting heterocycle-derived molecular materials and their applications in devices: 07CRV953.

Functionalized acenes and heteroacenes for organic electronics: 06CRV5028.

Organic semiconducting thiophene-, selenophene-, and tetrathiafulvalene-derived oligomers for use in thin film transistors: 07CRV1066.

Organic synthesis and device testing for molecular electronics, in particular, synthesis of oligo(2,5-thiophene ethynylenes): 06AA47.

Research progress of tetrathiafulvalene and its derivatives in organic photoelectric functional materials: 05CJO1167.

2.2.5.4 Coordination compounds

Amide-based macrocyclic ligands for anion coordination: 06AG(E)7882.

Applications of transition-metal catalysts, particularly complexes with *N*-heterocyclic ligands, to textile and wood-pulp bleaching by using hydrogen peroxide: 06AG(E)206.

Coordination chemistry and catalytic applications of functionalized *N*-heterocyclic carbene ligands: 07COC1491.

Coordination modes of 5-pyrazolones (X-ray diffraction data): 07CCR1561.

Coordination properties of bidentate alcohols and aldehydes derived from imidazole, pyrazole, or pyridine toward Cu(II), Co(II), Zn(II), and Cd (II) ions in the solid state and aqueous solution: 05CCR(249)2259.

Crystal engineering of coordination polymers using 4,4′-bipyridine as a bond between transition metal atoms: 06CC4169.

Cyclophosphazene-based multisite coordination ligands: 07CCR1045.

Evolution and applications of second-generation ruthenium olefin metathesis catalysts, particularly, using heterocyclic ligands: 07AA45.

Macroheterocyclic ligands in coordination of anions: 05ACR671.

Mössbauer studies of coordination compounds (including those with *N*-heterocyclic ligands) using synchrotron radiation: 05CCR(249)255.

Molecular assemblies of functional (mostly *N*-heterocyclic) molecules on gold electrode surfaces studied by electrochemical scanning tunneling microscopy: 06BCJ1167.

N-Heterocycle-based multicoordinate ligands for actinide/lanthanide separations: 07CSR367.

Salicylidenealdimine-based metal chelates for gas (O_2 and NO) binding and catalysis: 05ACR765.

Selective extraction of naturally occurring radioactive Ra^{2+} with crown ethers and related compounds: 05CSR753.

C_2-Symmetric chiral bis(oxazoline) ligands in asymmetric catalysis: 06CRV3561.

The synthesis and coordination chemistry of 2,6-bis(pyrazolyl)pyridines and their terpyridine analogs: 05CCR(249)2880.

Synthesis and application of new *N*-heterocyclic carbene ruthenium complexes in catalysis: 06COC203.

Synthesis, structures, metal coordination chemistry and applications of 4-acyl-5-pyrazolone ligands: 05CCR(249)2909.

Transition metal complexes as molecular machine prototypes: 07CSR358.

2.2.5.5 Polymers

The chemistry of organic nanomaterials (oligothiophenes and perilenediamides): 05AG(E)5592.

Chemical sensors based on amplifying fluorescent conjugated polymers functionalized, particularly, by crowns, thymine, or cyclophanes: 07CRV1339.

Conducting-polymer-based chemical sensors: 07BCJ2074.

The C_{60}/-conjugated oligothiophenes as materials for organic solar cells: 05CSR31.

Effect of different factors on curing process of tetrazole-containing polymers: 06IVUZ(6)3.

High-content photochromic polymers based on dithienylethenes: 05EJO1233.

Linear-conjugated systems (mainly, poly- and oligothiophenes) derivatized with C_{60}-fullerene as molecular heterojunctions for organic photovoltaics: 05CSR483.

Main-chain organometallic polymers: synthetic strategies, applications, and perspectives: 07CSR729.

Metal containing polymers with heterocyclic rigid main chains: 07CCR2104.

Modular and dynamic functionalization of polymeric scaffolds using *N*-heterocycles: 07ACR63.

New approaches to the analysis of high connectivity materials, coordination framework polymers derived from lanthanide metal ions with *N*,*N*′-dioxide ligands, such as 4,4′-bipyridine, pyrazine, 1,2-bis(pyridin-4-yl)ethane, and *trans*-1,2-bis(pyridin-4-yl)ethane *N*,*N*′-dioxides: 05ACR335.

Organocatalytic ring-opening polymerization involving lactones, morpholine-2,6-diones, cyclosiloxanes, and oxadisilacyclohexanes as monomers and/or heterocyclic carbenes as catalysts: 07CRV5813.

Relationships between molecular structure, electrochemical properties, and solid-state organization of salts of extended tetrathiafulvalene analogs: 05CSR69.

The roles of molecular architecture and redox matching in conducting metallopolymers possessing heterocyclic fragments: 05CC23.

Study of structuration processes of tetrazole-containing polymers under the action of various curing systems: 06IVUZ(8)3.

Synthesis and properties, particularly, as composite and electroluminescent materials, of polyquinolines and polyantrazolines: 05UK739.

Synthesis of oligo(*p*-aniline) analogs, indolocarbazole and diindolocarbazole derivatives: 05SL1223.

Synthesis of uniform, nonnatural oligomers, particularly, oligopyridines, oligo(pyridine–pyrimidine)s, and oligothiophenes: 06SL1793.

N-Vinylpyrrolidone in radical copolymerization reactions: 06IVUZ(2)3.

2.2.5.6 Ionic liquids

Advances in liquid-phase organic synthesis using functional ionic liquid as supports: 07CJO1188.

Application of ionic liquid-anchored catalyst or reagent to organic synthesis: 07CJO1236.

Approaches to crystallization from ionic liquids: 06CC4767.

Asymmetric catalysis in ionic liquids: 07COS370.

Catalysis in ionic liquids: 07CRV2615.

Chemical and biochemical transformations in ionic liquids: 05T1015.

The chemistry of the C(2) position of imidazolium room temperature ionic liquids (mainly, deprotonation to give *N*-heterocyclic carbenes): 07COS381.

Computer simulation of clusters, liquids, and crystals of dialkylimidazolium salts: 07ACR1156.

Electrochemical aspects of ionic liquids: 05MI12.

Electrophilic reactions of aromatic and heteroaromatic compounds in ionic liquids: 06ZOR1761.

Evolution of ionic liquids from solvents and separations to advanced high energy materials: 07ACR1182.

Functional design of ionic liquids: 06BCJ1665.

Functionalized imidazolium salts for task-specific ionic liquids and their applications: 06CC1049.

Intermolecular dynamics, interactions, and solvation in ionic liquids: 07ACR1217.

Ionic green solvents from renewable resources: 07EJO1049.

Ionic liquids and catalyst immobilization: 05Y503.

Ionic liquids as environmentally benign solvents in organic synthesis, particularly, that of heterocycles: 05COC195.

Ionic liquids as solvents for catalyzed oxidations (in particular, epoxidation) of organic compounds: 06ASC275.

Ionic liquids in separations including extractions, gas chromatography, and supported liquid membrane processes: 07ACR1079.

Ionic liquids in the range of solvents (general monograph): 05MI7.

Lanthanides and actinides in ionic liquids: 07CRV2592.

Local structure formation in alkyl-imidazolium-based ionic liquids as revealed by linear and nonlinear Raman spectroscopy: 07ACR1174.

Metal catalyzed reactions in ionic liquids: 05MI10.

Progress in the research of guanidinium ionic liquids including cyclic ionic liquids: 06CJO1335.

Reactivity of ionic liquids: 07T2363.

Recycle application of catalyst supported by ionic liquids to asymmetric reaction: 06CJO1362.

Solvation, solute rotation and vibration relaxation, and electron-transfer reactions in room temperature ionic liquids: 07ACR1130.

Solubility of CO_2, CH_4, C_2H_6, C_2H_4, O_2, and N_2 in 1-hexyl-3-methylpyridinium bis(trifluoromethylsulfonyl)imide (comparison to other ionic liquids): 07ACR1208.

Synthesis and application of functionalized chiral ionic liquids: 06CJO1031.

The use of quaternary phosphonium salts and 1,3-dialkylimidazolium hexafluorophosphates in "green" organic synthesis: 07COC107.

2.2.5.7 Miscellaneous

Chemistry of important flavor compounds, *viz.*, 2-acetyl-1-pyrroline, 6-acetyl-1,2,3,4-tetrahydropyridine, 2-acetyl-2-thiazoline, and 5-acetyl-2,3-dihydro-4*H*-thiazine: 06CRV2299.

Discotic liquid crystals (particularly, heterocyclic compounds) and their applications from tailor-made synthesis to plastic electronics: 07AG(E) 4832.

2,4,6,8,10,12-Hexanitro-2,4,6,8,10,12-hexaazatetracyclo[5.5.0.0^{3,11}.0^{5,9}] dodecane (high-energy material hexanitrohexaazaisowürtzitane): 05UK830.

Hydrogen-bonding of fullerenes with heterocycles: 05AG(E)5374.

Microporous porphyrin solids: 05ACR283.

Multidimensional crystal structures and unique solid-state properties of heterocyclic thiazyl radicals and related materials: 06BCJ25.

Multisubstituted olefins (including those with heterocyclic substituents), their synthesis and applications to materials science and pharmaceutical chemistry: 06BCJ811.

Self-organized functional liquid-crystalline assemblies (including those with *N*-heterocyclic fragments) as soft materials: 06AG(E)38.

Synthesis of aryl- and heteroaryl-substituted cyclopentadienes and indenes and their use in transition metal chemistry: 06COC937.

Synthesis of self-organizing thiophene-, thiazole-, or 1,3,4-thiadiazole-based mesogenic materials: 07CSR2046.

Synthesis, structure, and molecular encapsulation of open-cage fullerenes, particularly, possessing heterocyclic fragments: 05SL2117.

Synthesis, structure, and properties of thiols, particularly, those of heterocyclic series: 05ZOR647.

2.3 Specialized heterocycles

α-Haloacetylenic and acetylenic alcohols (including their use for synthesis of *O*- and *N*-heterocycles): 07ZOR169.

Methods for the synthesis of polycyclic nitramines: 07UK724.

New catalytic reactions of oxa- and azabicycloalkenes, including oxa- and azabenzonorbornadienes: 07ACR971.

Polar acetalization and transacetalization in the gas phase to form cyclic acetals (the Eberlin reaction): 06CRV188.

2.3.1 Nitrogen heterocycles (except alkaloids)

2.3.1.1 General sources and topics

Abnormal *N*-heterocyclic carbenes: 07CCR596.

Anionic tethered *N*-heterocyclic carbene chemistry: 07CSR1732.

Aryl and 1,3-dioxoindanyl derivatives of azoles and azines: 07KGS1603.

f-Block *N*-heterocyclic carbene complexes: 06CC3959.

Coordination chemistry and catalytic applications of functionalized *N*-heterocyclic carbene ligands: 07COC1491.

Formation and reactions of osmium−carbon double bonds, particularly, in carbene analogs derived from osmacyclopropene, osmafurane, and osmapyrrole: 07CCR795.

From the reactivity of *N*-heterocyclic carbenes to new chemistry in ionic liquids: 06CC1809.

Inclusion complexes containing quaternary azaaromatic moieties: 07H(71)1685.

Metal complexes with "pincer"-type ligands incorporating *N*-heterocyclic carbene functionalities: 07CCR610.

Metal-mediated asymmetric alkylation using chiral *N*-heterocyclic carbenes derived from chiral amines: 07CCR702.

Mixed oxazoline-carbenes as stereodirecting ligands for asymmetric catalysis: 07CCR718.

NHC–Ru complexes-friendly catalytic tools for manifold chemical transformations: 07CCR765.

N-Heterocyclic carbenes as organocatalysts: 07AG(E)2988.

N-Heterocyclic carbene–silver complexes as a new class of antibiotics: 07CCR884.

Organosilicon derivatives of nitrogen heterocycles with hypervalent silicon atom: 07UK885.

Palladium complexes of *N*-heterocyclic carbenes as catalysts for cross-coupling reactions: 07AG(E)2768.

Preparation and application of *N*-heterocyclic carbene complexes of Ag(I): 07CCR642.

Preparation of NHC–ruthenium complexes (NHC is *N*-heterocyclic carbene) and their catalytic activity in metathesis reaction: 07CCR726.

Progress in five-membered heterocyclic metallocene *N*-analogs: 07CJO329.

Stereoelectronic parameters associated with *N*-heterocyclic carbene ligands: 07CCR874.

Structural and catalytic properties of chelating bis- and tris-*N*-heterocyclic carbenes: 07CCR841.

Supported *N*-heterocyclic carbene complexes in catalysis: 07CCR860.

Synthesis, structure, and application of Ag(I) *N*-heterocyclic carbene complexes: 05CRV3978.

Transition metal-catalyzed reactions using alkynes as precursors of carbene and vinylidene complexes used in cyclizations leading to *N*-heterocycles: 05CL1068.

2.3.1.2 Structure and stereochemistry

Mössbauer studies of coordination compounds (including those with *N*-heterocyclic ligands) using synchrotron radiation: 05CCR(249)255.

Self-organization of disc-like molecules (in particular, formation of discotic crystals by *N*-heterocycles): 06CSR83.

2.3.1.3 Reactivity

Direct sp^3 C−H bond activation adjacent to nitrogen in heterocycles: 07CSR1069.

Oxidation of *N*-heterocycles (a green approach): 07JHC1223.

4-RSO_2-substituted azetidin-2-ones, pyrrolidin-2-ones, and piperidin-2-ones as stable precursors of reactive N-acylimino derivatives: 05CRV3949.

2.3.1.4 Synthesis

N-Acylhydrazones as versatile electrophiles for the synthesis of nitrogen heterocycles: 05AG(E)5176.

Azide rearrangements in electron-deficient systems (particularly, Schmidt lactam synthesis): 06CSR146.

A catalytic enantioselective aza-Michael reaction with participation of *N*-heterocycles in asymmetric synthesis of β-amino carbonyl compounds: 05EJO633.

1,5- or 1,7-electrocyclization of conjugated azomethine ylides: 06EJO2873.

Five-, six-, and seven-membered lactim ethers. Synthesis and chemical properties: 05KGS645.

Gold-catalyzed hydroamination of C−C multiple bonds, particularly, in synthesis of *N*-heterocycles: 06EJO4555.

Imidoyl radicals in organic synthesis, particularly, in construction of indoles, phenanthridines, pyrrolidines, quinolines, quinoxalines, and fused poly-cyclic N-containing derivatives: 07COC1366.

Olefin metathesis of amine-containing systems to give nitrogen heterocycles: 07ASC1829.

Oximes as reagents, particularly, for syntheses of *N*-heterocycles: 06UK884.

Preparation of cyclic S,S-dioxides and N-oxides using elemental fluorine and HOF·CH_3CN: 05EJO2433.

Synthesis of azabicyclic systems using nitrogen-directed radical rearrangements: 07OBC3071.

Synthesis of bridged and nonbridged *N*-heterocycles by cyclocondensation of bis(silyl enol ethers) with iminium salts: 07EJO2233.

Synthesis of *O*- and *N*-heterocycles by cyclization of unsaturated organolithiums: 02JOM(646)59.

Synthesis of *N*-heterocycles (indole, quinoline, pyrrole, pyrrolizine, indolizine derivatives, lactams, pumiliotoxin C, and lycopodine) using Ti-activated molecular nitrogen: 04JOM(689)4210.

Synthesis of unusual azides, 1,2,3-triazoles, and azirines using hexadecyltributylphosphonium azide as a highly potent reagent: 07S3431.

Synthesis, properties, and applications of *N*-phosphino lactams: 06CCR (250)2867.

2.3.2 Oxygen heterocycles

2.3.2.1 Chemistry of individual classes of O-heterocycles

Calorimetric and computational study of sulfur-containing six-membered rings including saturated *S*- and *O*-heterocycles: 05CSR347.

Cyclic organic peroxides: 07OBS3895.

Recent developments in main group metal complexes catalyzed/initiated polymerization of lactides and related cyclic esters: 06CCR(250)602.

2.3.2.2 Reactivity

Cyclopropanation of unsaturated *O*-heterocycles, particularly, sugar derivatives: 05T321.

2.3.2.3 Synthesis

Benzoic anhydride and its derivatives in the synthesis of lactones: 05Y2.

Gold-catalyzed reactions of oxo-alkynes in preparation of *O*-heterocycles: 07ARK(5)6.

Oxidative cyclization of dienes and polyenes mediated by transition-metal-oxo species (formation of *O*-heterocycles): 07S2585.

Preparation of optically active fluorine-containing γ-lactones with four chiral centers: 05Y26.

Reactions of 2,3-allenols and their derivatives, particularly those leading to *O*-heterocycles: 06CJO1468.

Synthesis of *O*- and *N*-heterocycles by cyclization of unsaturated organolithiums: 02JOM(646)59.

Transition metal-catalyzed reactions using alkynes as precursors of carbene and vinylidene complexes used in cyclizations leading to *O*-heterocycles: 05CL1068.

2.3.3 Sulfur heterocycles

2.3.3.1 Chemistry of individual classes of S-heterocycles

Benzo-annulated cyclic polysulfides: 07MRO15.

Cyclic 1,2-disulfonium dications and their Se and Te analogs: 05RKZ(6)77.

Persistent *S*-heterocyclic π-radical cations, their self-association, and its steric control in the condensed phase: 05OBC561.

2.3.3.2 Structure and stereochemistry

Calorimetric and computational study of sulfur-containing six-membered rings including saturated *S*- and *O*-heterocycles: 05CSR347.

2.3.3.3 Reactivity

Chiral sulfur ligands (*S*-, *S,N*-, *S,O*-heterocycles and heterocycles bearing S-containing substituents) for asymmetric catalysis: 07CRV5133.

Oxidation of sulfides to sulfoxides: 05T1933, 05T8315.

2.3.3.4 Synthesis

Catalytic syntheses and reactions of *S*-heterocycles (thiacycloalkanes, thiophenes, thiolane 1,1-dioxides): 05MI4.

Preparation of cyclic S,S-dioxides and N-oxides using elemental fluorine and $HOF{\cdot}CH_3CN$: 05EJO2433.

Heterocyclic chemistry of sulfur chlorides – Fast ways to complex heterocycles: 06EJO849.

2.4 Natural and synthetic biologically active heterocycles

We have classified the many reviews dealing with these materials under the following headings:

1. *General Sources and Topics (it is self-subdivided into Biological Functions, Syntheses).*
2. *Alkaloids (General, Syntheses, Individual Groups).*
3. *Antibiotics (General, Antitumor, β-Lactam, Macrocyclic, Miscellaneous).*
4. *Vitamins.*
5. *Drugs (General, Activity Types, Individuals and Groups).*
6. *Pesticides.*
7. *Miscellaneous (Enzymes, Amino Acids and Peptides, Plant Metabolites, Marine, Cyclodextrins, Other).*

2.4.1 General sources and topics

Advances in natural product synthesis by using intramolecular Diels–Alder reactions: 05CRV4779.

Bioinspired electron-transfer systems and their applications: 06BCJ177.

Bioinspired hydrogen bond motifs in ligand design: 05ACR54.

Biologically active molecules with a "light switch": 06AG(E)4900.

Bioluminescence mechanism in the limpet-like snail, *Latia neritoides*: 05BCJ1197.

Biomedical applications of carbon nanotubes functionalized by heteryl groups: 05CC571.
Biosynthetic and biomimetic electrocyclizations: 05CRV4757.
The chemistry of nitroxyl (HNO) and implications in biology: 05CCR(249)433.
Chiral trisoxazolines in asymmetric catalysis and molecular recognition: 05CSR664.
Current and emerging approaches for natural product biosynthesis in microbial cells: 05ASC927.
Effects of small heterocyclic molecules on morphogenesis and cell anatomy; on interaction of protein molecules; and on DNA and RNA: 05CSR472.
Exogenous carcinogens in foodstuffs and carcinogens produced in technological processes: 05CLY3.
Functionalized organolithium compounds in total synthesis: 05T3139.
The impact of bacterial genomics on natural product research: 05AG(E)6828.
Mechanistic studies of relevance to the biological activities of chromium: 05CCR(249)281.
Metathesis reactions in total synthesis: 05AG(E)4490.
Microscale approach for configurational analysis using combined cross metathesis/CD·FDCD protocols in natural products research: 06Y382.
Misassigned natural products and the role of synthesis in modern structure elucidation: 05AG(E)1012.
N-Containing compounds of macromycetes: 05CRV2723.
New synthetic methods useful for total synthesis of nitrogen-containing bioactive natural products, particularly, dendrobatid alkaloids, marine alkaloids, monoterpene alkaloid incarvillateine: 05CPB1375.
A novel method for construction of heterocycles using organometallic complexes and its application to the total syntheses of natural products: 05CPB457, 05YZ51.
Novel strategies for labeling biomolecules: 05OBC20.
Photoreduction of polyazaaromatic Ru(II) complexes by biomolecules: 06CCR(250)1627.
Polyketide stereotetrads in natural products (lactones, particularly, erythromycin): 05CSR677.
Progress in the synthesis of heterocyclic natural products by the Staudinger/intramolecular aza-Wittig reaction: 05ARK(2)98.
Revision of the stereochemistries of natural products through synthetic study: synthesis of fudecalone and kaitocephalin: 07Y511.
Rigid-rod molecules in biomembrane models: 05ACR79.
Ru(II) and Ru(III) complexes with cyclam and related species: 05CCR(249)405.

Small organic molecules (preferably, heterocycles) as initiators of chemical biology: 06Y529.

Strategy and design of total synthesis of selected bioactive natural products: 05CRV4707.

Synthetic nonpeptide mimetics of α-helices (terpyridines, 1,4-benzodiazepin-2,5-dione, polycyclic ethers, trisubstituted imidazoles as mimetics): 07CSR326.

Tandem cyclizations based on the novel reactions of allenic compounds useful for preparation of bioactive heterocycles: 05CPB1211.

2.4.1.1 Biological functions of natural and synthetic bioactive heterocycles

Advances in the biological activities and synthesis of 2-arylbenzo[*b*]furans: 05CJO25.

Biologically active pyrazines of natural and synthetic origin: 06CLY959.

Chemical modification of biomolecules to change their functionalities: 06Y1306.

Enzymatic reactions, mechanisms, structures, and biological functions of nicotinamide adenine dinucleotide: 07OBC2541.

Pilicides – 5-oxo-2,3-dihydrothiazolo[3,2-*b*]pyridine-3-carboxylic acid derivatives targeting bacterial virulence: 07OBC1827.

Reactions and biological activity of oximes of five-membered heterocyclic compounds with two heteroatoms: 07KGS1123.

Recent advance in quinoline derivatives with biological activities: 07CJO1318.

Recent progress on squaric acid research in bioorganic fields: 07Y32.

Recent progress on the lipase-catalyzed asymmetric syntheses: 07Y772.

Representative natural products used in chemical genetics studies: 06CRV2476.

Structural analogs of tetrapyrrole macrocycles and their biological properties: 07IZV663.

Synthesis and role of glycosylthio heterocycles in carbohydrate chemistry: 06T2943.

Synthetic studies on cyclic guanidine natural products for the elucidation of their controlling mechanism of protein–protein interaction: 06Y539.

Synthetic studies on oligosaccharide mimics carrying sulfur atoms in the pyranose rings: 06Y766.

Transition metal cage complexes (including complexes with porphyrin, phthalocyanine, cyclic polyamines as ligands) in biochemistry and medicine: 07IZV555.

2.4.1.2 General approaches to syntheses of biologically active heterocycles

Advances in iron catalyzed cross coupling reactions, selected applications to the total synthesis of bioactive natural products and pharmaceutically relevant compounds: 05CL624.

Advances on application of low valent titanium to organic synthesis (McMurry reaction, particularly, in synthesis of natural products and their analogs): 05CJO1342.
Applications of Baeyer–Villiger monooxygenase in organic synthesis (mainly, for five- and seven-membered lactones): 05CJO1198.
Application of hydrolytic kinetic resolution (HKR) in the synthesis of bioactive compounds: 07T2745.
Application of Ramberg–Bäcklund reaction to the synthesis of functional organic molecules, particularly, of natural compounds: 06CJO158.
Asymmetric intramolecular conjugate addition of chiral enolates to form pyrrolidine, piperidine, indoline, and tetrahydroisoquinoline derivatives with contiguous quaternary and tertiary stereocenters: 06YZ617.
Asymmetric syntheses of natural products using acetals: 05YZ699.
Carbon–carbon and carbon–hydrogen bond transformations mediated by highly reactive radicals and their application to the synthesis of bioactive compounds: 07Y665.
Cascade reactions in total synthesis: 06AG(E)7134.
Catalytic asymmetric synthesis of biologically active molecules: 07Y439.
Chemo- and stereoselective reduction of enaminones for the preparation of biologically active compounds: 06ARK(6)104.
Cyclooctyne and 4-cyclooctyn-1-ol as versatile building blocks, especially, in synthesis of natural products: 05EJO4231.
α,β-Dehydroamino acids as versatile intermediates in organic synthesis and structural motives in natural products and biologically active compounds: 06S1.
Development of amino thiourea catalysts as an artificial enzymes and their application to catalytic enantioselective synthesis of natural products and medical supplies: 06Y1139.
Development of enantioselective fluorination reaction and its application to the synthesis of biologically active compounds: 06Y14.
Development of Stetter reaction, particularly, in the synthesis of natural compounds: 06CJO906.
Enantio- and diastereoselective construction of α,α-disubstituted α-amino acids for the synthesis of biologically active compounds: 05EJO5127.
A formal [3 + 3] cycloaddition approach to natural-product synthesis: 05EJO23.
The hetero Diels–Alder approach to carbohydrate-containing molecular scaffolding: 07COS47.
High-throughput synthesis of combinatorial libraries based on natural products: 07Y795.
The intramolecular Stille reaction in some target natural product syntheses (mainly, authors results): 02JOM(653)261.

Long-range stereocontrol and asymmetric desymmetrization in total synthesis of merrilactone A: 07Y419.
Macrocyclization by ring-closing metathesis in the total synthesis of natural products: 06AG(E)6086.
Macrolactonizations in the total synthesis of natural products: 06CRV911.
Methods for constructing the peroxy bonds of organic peroxides: 05CJO1372.
Natural products syntheses based on biotransformations using hydrolases: 07Y783.
New development of photoinduced electron transfer reaction and total synthesis of natural products, cyclic amines and *O*-macroheterocycles: 05YZ16.
New developments in the asymmetric synthesis of heterocyclic natural products: 06ARK(7)57.
New metal-catalyzed carbocyclization reactions for the construction of complex natural products: 06ARK(7)338.
The Nicholas reaction as a powerful tool for the stereoselective synthesis of bioactive compounds, particularly, cyclic polyethers: 07SL343.
Organocatalytic synthesis of drugs and bioactive natural products: 07EJO2575.
Oxidative C–C bond formation in synthesis of natural products: 06ARK(7)310.
Palladium-catalyzed α-arylation of carbonyl derivatives and its applications in the synthesis of natural products: 05CJO282.
Palladium-catalyzed cross-coupling reactions in total synthesis: 05AG(E)4442.
Rapid syntheses of biologically active quinazolinone natural products using microwave technology: 07COS223.
Recent advances in asymmetric nitroso Diels–Alder reactions leading, particularly, to natural products: 06EJO2031.
Regio- and stereoselective synthesis of tri- and tetrasubstituted alkenes by introduction of CO_2 and alkylzinc reagents into alkynes in preparation of natural products: 07EJO4981.
Regioselective ortho metalation-cross coupling for biaryls and heterobiaryls in aromatic and heteroaromatic natural product synthesis: 02JOM(653)150.
Separations and syntheses of chiral compounds and application to the synthesis of natural products using fluorous chemistry: 06Y617.
Strategies for the formation of tetrahydropyran rings in the synthesis of natural products: 06EJO2045.
Strategies for the synthesis of C_2 symmetric natural products: 07T1487.
Syntheses of natural products having an epoxyquinone structure: 05CRV4515.

Titanocene-mediated radical cyclization in the synthesis of natural products: 06EJO1627.
Total syntheses of useful bioactive compounds and significance of their developments: 06Y458.
Total synthesis of epoxyquinonoid natural products: 07BCJ1672.
Total synthesis of α-pyrone meroterpenoids, novel bioactive microbial metabolites: 05CRV4559.
Total synthesis of some natural *O*-heterocycles as possible agents in combating neurodegenerative disorders: 06ACR539.
Total synthesis of structurally unique γ-lactam natural products: 07Y460.
The use of catalytic asymmetric sulfide oxidations to the syntheses of bioactive sulfoxides: 05ASC19.

2.4.2 Alkaloids

2.4.2.1 General The quest for quinine: 05AG(E)854.

2.4.2.2 Structure Design, synthesis, and binding affinity of new nicotinic ligands: 06ARK(8)50.

2.4.2.3 Synthesis

Advances in the synthesis of analogs of the *Delphinium* alkaloid methyllycaconitine: 05SL1809.
Applications of Stevens rearrangement of ammonium ylides to the synthesis of alkaloids: 06T1043.
Comparison of synthetic strategies and routes of total synthesis of the lupin alkaloid cytisine: 07T1885.
[3 + 3] Cycloadditions and related strategies in alkaloid natural product synthesis: 05OBC1349.
Intermolecular Pictet–Spengler condensation with chiral carbonyl derivatives in the stereoselective syntheses of optically active isoquinoline and indole alkaloids: 05ARK(12)98.
New strategies for the synthesis of monoterpene indole alkaloids: 05COC1465.
Novel routes to pyrroles, indoles, and carbazoles – Applications in total synthesis of alkaloids including fused indolizidines, yohimbanes, Amaryllidaceae alkaloids, and carbazoles: 05COC1601.
Oxidative amidation of phenols *via* the use of hypervalent iodine reagents in alkaloid synthesis: 07S3759.
Progress in the synthesis of morphine alkaloids (2000–2004): 05SL388.
Pyrroles and gem-dihalocyclopropanes as building blocks for alkaloid synthesis: 05COC1589.
Reserpine: a challenge for total synthesis of natural products: 05CRV4671.
Stereo-controlled construction of quaternary carbon stereocenters in total synthesis of complex cyclotryptamine alkaloids: 07AG(E)5488.
Syntheses of morphine and codeine (1992–2002): 06COS99.

Synthesis of antitumor bis-tetrahydroisoquinoline alkaloids, saframycins and ecteinascidins: 05CJO42.

Synthesis of benzo[*c*]phenanthridine alkaloids using a Pd-catalyzed aryl–aryl coupling reaction: 05H(65)697.

Synthesis of hydroxylated phenanthridines, Amaryllidaceae constituents (1996–2004): 05SL365.

Synthesis of poison frog alkaloids as novel, potent, and subtype-selective blockers of neuronal nicotinic acetylcholine receptors: 06Y49.

Synthesis of cinchona alkaloids: 07Y598.

Synthetic studies of hetisan-type aconite alkaloids (total synthesis of (±)-nominine): 06Y237.

Total synthesis of calycotomine, 1-hydroxymethyl-substituted tetrahydroisoquinoline: 05S339.

Trends in total synthesis of alkaloids: 05CLY298.

A unified synthetic approach to the pyrazinone-containing dragmacidin alkaloids: 06CC3769.

2.4.2.4 Individual groups of alkaloids

Advances in development of dopaminergic aporphinoids: 07JMC171.

Advances in the chemistry of macroline, sarpagine, and ajmaline-related indole alkaloids: 06T8655.

Benzo[*c*]phenanthridine alkaloids sanguinarine and chelerythrine: 06H(68)2403.

Chemistry of indole alkaloids related to the corynanthe-type from *Uncaria*, *Nauclea*, and *Mitragyna* plants: 05COC1445.

The chemistry of recently isolated naturally occurring quinazolinone alkaloids: 06T9787.

Chemoenzymatic synthesis of morphine, pancratistatin, and their analogs: 06ARK(7)276.

Diazine analogs of the pyridocarbazole alkaloids: 06COC363.

β-Isocupreidine-catalyzed asymmetric Baylis–Hillman reactions (β-isocupreidine is (3*R*,8*R*,9*S*)-10,11-dihydro-3,9-epoxy-6-hydroxycinchonane): 06Y1132.

Luotonin A (a novel pyrroloquinazolinoquinoline alkaloid isolated from the aerial parts of *Peganum nigellastrum* Bunge) as a lead toward anticancer agent development: 05H(65)2203.

Progress in the study of *Daphniphyllum* alkaloids: 07CJO565.

Quaternary isoquinoline alkaloids sanguinarine and chelerythrine, *in vitro* and *in vivo* effects: 06CLY30.

Research progress on sugar-mimic polyhydroxy alkaloids derived from plants: 07CJO1337.

Stereochemistry, syntheses, and biological activity of lupine alkaloids: 07YZ1557.

The structure, biological activities, and synthesis of 3-hydroxypyrrolizidine alkaloids and related compounds: 05COC1393.
Survey of chemical syntheses of the pyrrolizidine alkaloids turneforcidine and platynecine: 07H(74)125.
Synthesis and absolute configuration of (+)-lentiginosine, indolizidine alkaloid, a potent and selective inhibitor of amyloglucosidase: 07EJO1551.
Total synthesis of antitumor pyrroloiminoquinone alkaloids discorhabdins and makaluvamines: 05COC1567.

2.4.3 Antibiotics

2.4.3.1 Antitumor antibiotics

Antitumor antibiotics: bleomycin, enediynes, and mitomycin: 05CRV739.
Hybrid molecules based on distamycin A as potential antitumor agents: 06ARK(7)20.
Recent developments in the syntheses of macrocyclic antitumor compounds epothilones and analogs: 06EJO4071, 06MRO49.
Recent developments in the chemistry and biology of epothilones: 06ARK (8)131.
Rifamycin – mode of action, resistance, and biosynthesis: 05CRV621.
Studies on natural anticancer macrolactones cryptophycins and their mimetics: 06CJO27.
Synthetic studies on fostriecin and related natural products with antitumor properties: 07Y874.
Total syntheses of anticancer natural product FR901464: 07Y119.
Total syntheses of antitumor antibiotic FR900482, its derivatives and analogs through the eight-membered *N*-hydroxybenzazocinone key intermediate: 07Y470.
Total syntheses of macrolactones with excellent antitumor activity, epothilones: 07CJO298.

2.4.3.2 β-Lactam antibiotics

Bacterial resistance to β-lactam antibiotics: 05CRV395.
New progress in enzymatic *semi*-synthesis of β-lactam antibiotics: 06CJO292.

2.4.3.3 Macrocyclic antibiotics

Chemistry and biology of the 23-membered macrocyclic streptogramin A antibiotics: 07MRO159.
A comparative analysis of the total syntheses of the amphidinolide T natural products: 05OBC2675.
Enantioselective disposition of lansoprazole and rabeprazole in human plasma: 06YZ395.
The role of macrolides in translation and protein synthesis: 05CRV499.
Synthesis and antibacterial activity of macrolides and ketolides related to erythromycin: 06T3171.

Synthesis of nine-membered dilactone antibiotics antimycins: 06CJO1370.

2.4.3.4 Miscellaneous antibiotics

Biomimetic synthesis of heteroaromatic thiopeptide antibiotics from amino acids: 07AG(E)7930.

Biosynthesis and mode of action of lantibiotics: 05CRV633.

Chemistry and biology of ramoplanin, a lipoglycodepsipeptide with potent antibiotic activity: 05CRV449.

2-Deoxystreptamine as central scaffold of aminoglycoside antibiotics: 05CRV775.

Derivatization and isotope labeling of amphotericin B aiming at elucidation of the ion-channel structure: 06Y502.

Genetic approaches to polyketide antibiotics: 05CRV543.

Glycopeptide and lipoglycopeptide antibiotics: 05CRV425.

Molecular insights into aminoglycoside action and resistance: 05CRV477.

N-Heterocyclic carbene–silver complexes as a new class of antibiotics: 07CCR884.

An overview of synthetic and biological studies of ambruticin and analogs: 05COC405.

Streptogramins, oxazolidinones, and other inhibitors of bacterial protein synthesis: 05CRV529.

Synthetic routes toward the pyranonaphthoquinone antibiotic pentalongin and syntheses of corresponding nitrogen derivatives: 07SL829.

Synthetic study of nucleoside antibiotics: 06YZ579.

Thiopeptide antibiotics: 05CRV685.

Total synthesis of spiroketal containing natural products, reveromycin A and spirofungin B: 06ARK(7)105.

2.4.4 Vitamins

Asymmetric synthesis of vitamin E: 06CJO1353.

Chemistry and enzymology of vitamin B_{12}: 05CRV2075.

Design and synthesis of highly potent vitamin D receptor antagonists based on the structural development of vitamin D_3-26,23-lactone: 07Y947.

Green molecular transformations mediated by vitamin B_{12} related metal complexes as homogeneous and heterogeneous catalysts: 05Y780.

Luminescent transition metal complex biotin conjugates: 06CCR(250)1724.

Properties and chemical modification of L-ascorbic acid: 05KFZ(5)26.

Search for and development of active vitamin D_3 analogs: 05Y728.

Thiamine models and perspectives on the mechanism of action of thiamine-dependent enzymes: 06CSR684.

2.4.5 Drugs

2.4.5.1 General

Analysis of the reactions used for the preparation of drug candidate molecules: 06OBC2337.

Applications of total synthesis toward the discovery of clinically useful anticancer agents: 07CSR1207.
Asymmetric synthesis of active pharmaceutical ingredients: 06CRV2734.
Chemistry and biology of a series of antitumor sulfonamides: exploiting transcriptomic and quantitative proteomic analyses for exploring drugable chemical space: 06Y1171.
Design and structure–activity relationships of MT_2 selective melatonin receptor antagonists: 06ARK(8)8.
Design and synthesis of heterocycle-fused enediyne prodrugs activable at will: 06ARK(7)261.
Designing heterocyclic selective kinase inhibitors: from concept to new drug candidates: 06ARK(7)496.
Development of environmentally benign organometallic catalysis for drug discovery and its application to heterocyclic chemistry: 07CPB1099, 07YZ1383.
Development of novel catalytic asymmetric reactions mediated by chiral Pd complexes as acid–base catalysts and their application to synthesis of drug candidates: 06CPB1351.
Human pharmaceuticals in the aquatic environment: 07CRV2319.
Intramolecular cyclization utilized for release of active substances from prodrugs: 05CLY21.
Large-scale oxidations in the pharmaceutical industry: 06CRV2943.
Metabolism of endogenic compounds: 06MI1.
Metabolism of exogenous compounds. Drugs and other xenobiotics: 06MI2.
Neuronal nicotinic acetylcholine receptors: 05JMC4705.
New methodologies for entry to targets of therapeutic interest: 05SL2571.
New targets and screening approaches in antimicrobial drug discovery: 05CRV759.
Noncovalent interactions in adducts of platinum drugs with nucleobases in nucleotides and DNA as revealed by using chiral substrates: 06CCR(250)1315.
Organic chemistry of food, feed, and biologically active additives: 06MI3.
Organic synthesis of small organic molecules and biomolecules, especially of pharmaceutically relevant heterocycles with light-fluorous reagents, reactants, catalysts, and scavengers: 06AA3.
Organolithium reagents in pharmaceutical asymmetric processes: 06CRV2596.
Ozonolysis applications in drug synthesis: 06CRV2990.
Potent agents based on ligands for cell-surface receptors (particularly, indoloazepines) and drug discovery: 06ACR831.
Practical synthesis of multifunctional compounds, particularly, intermediates for drug synthesis through Pd/C-catalyzed coupling reactions: 06Y853.

Process design and development of drug candidates using cross-coupling reactions as key steps in Merck: 02JOM(653)279.

Renin inhibitors as antihypertensive agents and factor Xa inhibitors as anticoagulants: 07H(73)47.

S-Adenosyl-L-homocysteine hydrolase as an attractive target for antimicrobial drugs: 07YZ977.

Search and structural elucidation of medicinal products from the vegetable kingdom, including flavonoids: 07YZ1975.

Selective reductions with boron reagents in synthesis of pharmaceutical and other products: 06CRV2617.

Silatranes in medicine and agriculture: 05MI3.

Structure, orientation, and conformational changes in transmembrane domains of multidrug transporters: 05ACR117.

Synthesis and pharmaceutical evaluation of positron emission tomography [^{11}C] radiopharmaceuticals for the clinical application: 06YZ737.

Use of the furoxan system in the design of new NO-donor antioxidant hybrids: 06ARK(7)301.

Synthesis and biological evaluation of potential positron emission tomography ligands for brain visualization of dopamine D_3 receptors: 06ARK (8)102.

Tyrosine kinase drug discovery: 06ARK(8)38.

2.4.5.2 Definite types of activity

Activation of p53 protein as tumor suppressor by small heterocyclic molecules: 05JMC4491.

Adverse cardiovascular effects of the coxibs: 05JMC2251.

Antibacterial natural products in medicinal chemistry: 06AG(E)5072.

Antitumor properties of 16-membered ketolactone fludelone: 05AG(E)2838.

Applications and synthesis of the antiepileptic drug oxcarbazepine and related structures: 07COC1385.

CB1 cannabinoid receptor antagonists for treatment of obesity and prevention of comorbid metabolic disorders: 06JMC4008.

A comparative QSAR study of nonsteroidal anti-inflammatory drugs: 05CRV3235.

Development of and industrial process for synthesizing 9-(2,3-deoxy-2-fluoro-β-D-*threo*-pentofuranosyl)adenine – anti-HIV agent Lodenosine: 05Y864.

Discovery and development of small-molecule chemokine coreceptor CCR5 antagonists (oximino-substituted piperidinopiperidines, chiral piperazines, spirodiketopiperazines): 06JMC2851.

Dithiol proteins as guardians of the intracellular redox milieu in parasites, old and new drug targets in trypanosomes and malaria-causing plasmodia: 05AG(E)690.

Dopamine D3 receptor partial agonists and antagonists as potential drug abuse therapeutic agents: 05JMC3663.

Drug discovery research for microbial metabolites: 06Y548.

5-Hydroxytryptamine 2C (5-HT_{2C}) receptor agonists as potential antiobesity agents: 06JMC4023.

Industrial syntheses of the central core molecules of HIV protease inhibitors: 06CRV2811.

Macrolide antibiotics and other macroheterocycles as heat shock protein 90 inhibitors: 05JMC7503.

Medicinal chemistry of human ether-a-go-go related gene (hERG) optimizations: 06JMC5029.

A medicinal chemistry perspective of control of hepatitis C: 05JMC1.

Melanin-concentrating hormone-1 receptor antagonists for the treatment of obesity: 06JMC4017.

Melanocortin-4 receptor (MC4R) agonists for the treatment of obesity: 06JMC4035.

Molecular recognition of protein kinase binding pockets for design of potent and selective kinase inhibitors: 07JMC409.

Neurotoxins and drugs for treatment of Parkinson disease: 05KFZ(9)3, 05KFZ(10)14, 05KFZ(11)3.

New approaches toward anti-HIV chemotherapy: 05JMC1297.

The next generation of phosphodiesterase inhibitors: 05JMC3449.

Norepinephrine reuptake inhibitors for neuropsychiatric disorders: 06H(69)539.

Oral cholesteryl ester transfer protein (CETP) inhibitors for treating coronary artery disease: 06JMC1.

Peptidomimetics, glycomimetics, and scaffolds from carbohydrate building blocks: 07EJO4177.

Progress in total synthesis of orlistat (antiobesity drug): 05CJO902.

Prospects for metabotropic glutamate 1 receptor antagonists in the treatment of neuropathic pain: 07JMC2563.

Pyridazine derivatives as novel acyl-CoA:cholesterol acyltransferase (ACAT) inhibitors: 05JHC395.

Rational drug design of δ opioid receptor agonist TAN-67: 06Y371.

Research on nonnucleoside inhibitors of HIV-1 reverse transcriptase from 4,5,6,7-tetrahydro-5-methylimidazo[4,5,1-*jk*](1,4)benzodiazepin-2(1*H*)-one (TIBO) to etravirine (TMC125): 05JMC1689.

Structural and functional basis of cyclooxygenase inhibition: 07JMC1425.

The structure–activity relationship of allyl-substituted oxopyrimidines: 05YZ73.

Thrombin receptor (protease activated receptor-1) antagonists as potent antithrombotic agents with strong antiplatelet effects: 06JMC5389.

Transient receptors as potential vanilloid type 1 channel modulators for the treatment of pain: 07JMC2589.

2.4.5.3 Individual substances and groups of compounds

Antitumor metal-containing carboranes: 07IZV620.

Azetidine-2-carboxylic acid as one of key pharmaceuticals: 06OPP427.

Carboranyl thymidine analogs for neutron capture therapy: 07CC4978.

CO and NO in medicine: 07CC4197.

Drug targets, lead compounds, and potential therapeutic applications of endocannabinoids: 05JMC5059.

Induction of apoptosis in several cancer cell lines by a natural product, Apoptolidin (20-membered macrolide): 06AG(E)872.

Medicinal chemistry approach to the synthesis of pyrazolo[4,3-*e*][1,2,4]triazolo[1,5-*c*]pyrimidine template: 06COC259.

Medicinal chemistry of combretastatin A4: 06JMC3033.

Oxazolidine derivatives of ephedrine with retarded activity: 05CLY318.

Porphyrin-based, chlorophyll-based, or phthalocyanine-based compounds as synergic photosensitizers for photodynamic therapy: 05COC813.

3,3'-Pyrrolidinyl-spirooxindole natural products as inspirations for the development of potential therapeutic agents: 07AG(E)8748.

Quinolone and pyridone antibacterial agents, bacterial topoisomerase inhibitors: 05CRV559.

Recent progress during 2000–2005 in development of dopamine receptor subtype-selective agents, potential therapeutics for neurological and psychiatric disorders: 07CRV274.

Recent progress in the synthesis of artemisinin and its derivatives: 06OPP1.

The structure-phototoxicity relationship and photostability of fluoroquinolones: 05YZ255.

Synthesis and pharmacology of (4a*S*,6*R*,8a*S*)-5,6,9,10,11,12-hexahydro-3-methoxy-11-methyl-4a*H*-[1]benzofuro[3a,3,2-*ef*] [2]benzazepin-6-ol (galantamine): 06CRV116.

Synthetic approaches to 1,2,4-triazoles and their pharmacological importance: 06KGS1605.

Synthetic strategies toward O(6)-substituted guanine derivatives and their use in medicine: 05COS215.

Tetrazole derivatives as drugs: 07KGS3.

2.4.6 Pesticides

Cross-coupling and metallation reactions of 3(2*H*)-pyridazinones in syntheses of fungicides and herbicides: 05JHC427.

Pyrazole chemistry in crop protection: 07H(71)1467.

Pyrimidine chemistry in crop protection: 06H(68)561.

Structural determination of the ComX pheromone: synthesis studies on ComXRO-E-2 pheromone and ComXRO-E-2 peptides containing modified tryptophan residue with a geranyl group: 07Y608.

Synthetic studies of fluorine-containing compounds for household insecticides, particularly, 3-[1R-trans-(2-trifluoromethyl)cyclopropanecarbonyl]-4-hydroxy-6-methyl-2-pyrone with outstanding insecticidal activity against *Blattella germanica*: 07Y620.

2.4.7 Miscellaneous

2.4.7.1 Enzymes, coenzymes, and their models

Artificial metalloenzymes: 05CSR337.

Artificial ribonucleases: 06OBC15.

Aryloxo and thiolato vanadium complexes as chemical models of the active site of vanadium nitrogenase: 05CCR(249)2144.

Chemical structure and biological activity of α- and β-glucosidase inhibitors: 06T10277.

Chemistry and biochemistry of insulin-mimetic vanadium and zinc complexes (including pyridine, pyrimidine, thiazole, and porphyrin derivatives): 06BCJ1645.

Chemistry relating to the nickel enzymes CODH and ACS: 05CCR(249)1582.

Computational and synthetic approaches for the discovery of HIV-1 integrase inhibitors: 06ARK(7)224.

Cytochromes *c*: 06CRV2550.

Development of novel C1 domain ligands of protein kinase C to clarify the precise structure and activation mechanism: 06Y515.

Development of ruthenium catalyzed oxidation reactions inspired by cytochrome P-450: 07Y2.

DFT investigations of models related to the active site of [NiFe] and [Fe] hydrogenases: 05CCR(249)1620.

cis-Dihydroarenediols and -hetarenediols formation by arene dihydroxylating dioxygenase enzymes and application of the *cis*-dihydrodiol bioproducts: 06OBC181.

Discovery of nonpeptide bradykinin B_2 receptor agonists and antagonists: 05Y852.

Diversity of epoxide hydrolase biocatalysts: 06COC1145.

Domain relationships in thiamine diphosphate-dependent enzymes: 06ACR550.

Enantioselective C−C bond synthesis catalyzed by enzymes: 05CSR530.

Friedel–Crafts-type mechanism for the enzymatic elimination of ammonia from histidine and phenylalanine: 05AG(E)3668.

Fifteen years of Raman spectroscopy of engineered heme containing peroxidases: 05ACR433.

Functional roles of the heme architecture and its environment in tetraheme cytochrome c: 07ACR171.

Spectroelectrochemistry of hydrogenase enzymes and related compounds: 05CCR(249)1536.
Structural and functional models related to the nickel hydrogenases: 05CCR(249)1555.
Structural and spectroscopic models of the A-cluster of acetyl coenzyme A synthase/carbon monoxide dehydrogenase: 05CCR(249)3007.
Structure-based inhibitor design of human hematopoietic prostaglandin D synthase: 05Y739.
Structure–function relationships of nickel–iron sites in hydrogenase and a comparison with the active sites of other nickel–iron enzymes: 05CCR(249)1609.
Synthesis of gem-diamine 1-*N*-iminosugars, a new family of glycosidase inhibitors: 06H(67)461.
Synthesis of inhibitors against glycosyltransferases: 06Y894.
Synthetic models of the active site of catechol oxidase: 06CSR814.
Synthetic, structural, and reactivity studies of iron-only hydrogenase model compounds: 05CCR(249)1641.
The substrate spectrum of mandelate racemase: 05ASC951.
Thiamine models and perspectives on the mechanism of action of thiamine-dependent enzymes: 06CSR684.
Total synthesis of lactacystin and salinosporamide, natural proteasome inhibitors: 07CAJ20.
Total synthesis of (+)-scyphostatin, a neutral sphingomyelinase inhibitor: 07Y358.
Transient-state kinetic approach to mechanisms of enzymatic catalysis: 05ACR157.
X-ray magnetic circular dichroism – a high energy probe of magnetic properties, particularly of metalloproteins and metalloenzymes: 05CCR(249)3.

2.4.7.2 Amino acids and peptides

Assembly-line enzymology for polyketide and nonribosomal peptide antibiotics: 06CRV3468.
Asymmetric catalysis mediated by synthetic peptides including cyclic dipeptides and peptides formed by proline or histidine: 07CRV5759.
Atomic resolution crystallography of proteins and X-ray absorption fine structure studies of metalloproteins: 05CCR197.
Biological activity and synthesis of diketopiperazines: 07T9923.
Biomimetic synthesis of heteroaromatic thiopeptide antibiotics from amino acids: 07AG(E)7930.
Characterization of "spectroscopically quiet" metals in metalloproteins: 05CCR161.
Chemical and biological aspects of Cu^{2+} interactions with peptides and aminoglycosides: 05CCR(249)2323.

Conjugation of 1,4,7,10-tetraazacyclododecane-1,4,7,10-tetraacetic acid (DOTA) and its derivatives to peptides, applications and future prospects of the conjugates: 07MRO281.

A crosslinking amino acid histidinoalanine: 07T9033.

Expanding the genetic code (possible use of 30 new amino acids for creating new proteins and organisms): 05AG(E)34.

Heterocycles as ligands in catalytic asymmetric synthesis of α-amino acids: 07CRV4584.

Heme-iron in lipid oxidation: 05CCR(249)485.

Hexafluoroacetone as protecting and activating reagent; new routes to amino, hydroxy, and mercapto acids and their application for peptide and glyco- and depsipeptide modification: 06CRV4728.

Mechanism of electron transfer in heme proteins and models: 05CRV2627.

Metallacyclopeptides as artificial analogs of naturally occurring peptides: 05CSR496.

Metalloproteins three-dimensional structure determination using multiple-scattering analysis of X-ray absorption fine structure: 05CCR(249) 141.

Natural occurrence, syntheses, and the use of 1-aminocyclopropanecarboxylic acid (ACA) and other 2,3-methanoamino acids for preparation of cyclic peptides and heterocyclic derivatives of ACA: 07CRV4493.

Neurohormonal preparations of ergo peptide series related to ergo alkaloids: 05RKZ(1)125.

Peptide-activated G protein-coupled receptors recognizing ligands with turn structure: 05CRV793.

Peptide hormones, particularly, cyclopeptides: 05RKZ(1)11.

Plant cyclopeptides: 06CRV840.

Polypeptides and 100 years of chemistry of α-amino acid *N*-carboxyanhydrides: 06AG(E)5752.

Process development of micafungin, a novel cyclopeptide antifungal agent: 06Y1294.

Protein-reactive natural products (mainly, heterocycles): 05AG(E)5788.

Recent advances in the synthesis of unnatural α-amino acids (heterocycles are of considerable importance both during the synthetic build-up and as targets): 07COC801.

Recent progress of the synthetic studies of biologically active marine cyclic peptides and depsipeptides: 05CRV4441.

Recognition of proline-rich motifs by protein–protein interaction domains: 05AG(E)2852.

Stereoselective synthesis of aza- and diazabicyclo[X.Y.0]alkane dipeptide mimetics: 05S1031.

Strategies for targeting protein–protein interactions with synthetic agents: 05AG(E)4130.

Structure and thermodynamic stability of lanthanide complexes with amino acids and peptides: 05CCR(249)567.

The synthesis of 16-membered macrocyclic depsipeptides, cryptophycins: 06S3747.

Syntheses of peptidomimetics based on pyranose and polyhydroxylated piperidine scaffolds: 06COS403.

Thiopeptide antibiotics: 05CRV685.

X-ray magnetic circular dichroism – a high energy probe of magnetic properties, particularly of metalloproteins and metalloenzymes: 05CCR(249)3.

2.4.7.3 Plant metabolites

Advances in the chemistry of furano-diterpenoids from *Teucrium* genus (2000–2005): 05H(65)1221.

Asymmetric synthesis of styryl-lactones isolated from several species of the genus *Goniothalamus* (Annonaceae): 06COS41.

Biosynthetic pathway leading to the fungal secondary metabolite phomoidride B: 06SL354.

A combined resonance Raman and quantum chemical study of the chromophore structural changes during the photocycle of phytochrome: 07ACR258.

Bioactive compounds from mushrooms: 07H(72)45.

Core-modified ginkgolides, terpene trilactones from *Ginkgo biloba* extract: 05H(66)743.

Enantiotopic synthesis of merrilactone and guanacastepene: 05H(66)711.

Naturally occurring iridoids (cyclopentano[*c*]pyran monoterpenoids), a review compiling data reported between 1994 and 2005: 07CPB159.

Naturally occurring secoiridoids and bioactivity of naturally occurring iridoids and secoiridoids: 07CPB689.

New biologically active metabolites from Chinese higher fungi: 06COC849.

Progress in the research on naturally occurring flavones and flavonols: 06COC873.

Progress in total synthesis of anti-HIV pyranocoumarin calanolide A and its analogs: 07CJO685.

Recent advancement in synthesis of green tea catechins: 07CJO1502.

Structure and bioactivity of the furan-diterpenoids from the genera *Leonotis* and *Leonurus*: 07H(74)31.

Survey of briarane-related diterpenoids obtained from Gorgonacea (genus *Briareum*, *Erythropodium*, *Ellisella*, and *Junceella*) and Pennatulacea (genus *Cavernularia* and *Pteroeides*): 05H(65)195.

Synthesis, bioactive conformations, and structure–activity relationship of taxol and its analogs: 05ZOR329.

Synthesis of phytochrome, a protein-bound linear tetrapyrrole: 05SL2861.

Synthetic strategies of fostriecin: 05H(66)727.

2.4.7.4 Heterocycles produced by marine organisms

Absolute configuration of ciguatoxin and development of immunoassay systems: 07BCJ1870.

Advances in the total synthesis of biologically important marine macrolides: 05CRV4237.

Anti-inflammatory metabolites from marine sponges: 05CSR355.

The asymmetric total synthesis of nakadomarin A, a marine manzamine alkaloid: 05Y200.

Bioactive nitrogenous metabolites from ascidians: 07H(74)53.

Biomimetic synthesis of *trans*, *syn*, *trans*-fused polycyclic ethers (polyoxepanes and polypyrans): 06SL1816.

Chemistry and biology of anti-inflammatory marine natural ts, phospholipase A_2 inhibitors: 05COC1419.

Construction of fused polycyclic ethers (particularly, ciguatoxins and gambieric acids) by strategies involving ring-closing metathesis: 06CC3571.

Convergent strategies for syntheses of trans-fused polycyclic ethers: 05CRV4379.

Development and application of a convergent strategy for the total synthesis of polycyclic ether natural products: 07BCJ856.

The first asymmetric total synthesis of tetrodotoxin, a puffer fish toxin: 07Y492.

Iminium alkaloids from marine invertebrates, structure, biological activity, and biogenesis: 05CL454.

New methods of imidazole functionalization – from imidazole to marine alkaloids: 06SL965.

The phomactins. A novel group of terpenoid platelet activating factor antagonists related biogenetically to the taxanes: 06ACR354.

Progress in the study of marine bromopyrrole alkaloids: 05CJO788.

Pursuit of novel bioactive marine metabolites: 06Y471.

Recent research progresses on lamellarins and analogous hexacyclic polyaromatic pyrrole alkaloids of bioactive marine mollusk: 05CJO641.

The structural revision of marine natural product palau'amine and total synthesis of the latter and related pyrrole–imidazole alkaloids: 07AG(E)6586.

Studies on total synthesis of the cylindricine/fasicularin/lepadiformine family of tricyclic marine alkaloids: 06CRV2531.

Synthesis and antitumor activity of actin-depolymerizing macrolides from the sea hare *Aplysia kurodai*: 06Y1273.

The synthesis of aplysinopsins, meridianines, and related compounds of marine origin: 05MRO211.
Synthesis of prelaureatin, laurencin, and the medium-ring ether parts of ciguatoxins: 07Y502.
Synthetic strategies of marine polycyclic ethers *via* intramolecular allylations. The iterative total synthesis of hemibrevetoxin B and the convergent total synthesis of gambierol: 05ACR423.
Synthetic studies of marine polycyclic ether ciguatoxins: 06Y418.
Systematic synthesis of diastereomeric poly-THF ring cores and total synthesis of antitumor Annonaceous acetogenins: 06SL993.
Total synthesis and structure–activity relationship of a cytotoxic polycyclic ether Gymnocin-A: 06Y808.
Total synthesis of brevetoxin B: 07Y430.
Total synthesis of marine polycyclic ethers: 05CRV4314.
Total synthesis of oxacyclic macrodiolide natural products: 05CRV4348.
Total synthesis of tricyclic marine alkaloids (−)-lepadiformine, (+)-cylindricine C, and (−)-fasicularin: 07Y805.

2.4.7.5 Cyclodextrins

Noncovalently bound cyclodextrin dimers and related compounds: 05KGS1603.
Photoactive metallocyclodextrins: 05CSR120.

2.4.7.6 Other topics

C-Alkoxycarbonyl nitrones as building blocks for the synthesis of butenolides, lactams, and modified nucleosides: 05MRO59.
The asymmetric total synthesis of epoxyquinols A, B, and C and epoxytwinol A: 07EJO3783.
Bioactive *O*-heterocycles from insect pathogenic fungi: 05ACR813.
Cellulose as fascinating biopolymer and sustainable raw material: 05AG(E) 3358.
Chemistry and biology of wortmannin (steroidal furan isolated from *Penicillium wortmanni* Klocker): 05OBC2053.
Chemistry of some pyrrolidine derivatives (domoic acid, isodomoic acids) and their analogs: 05T5713.
Creation of highly potent vitamin D receptor antagonists: 07YZ1215.
Design and synthesis of new classes of heterocyclic C-glycoconjugates and carbon-linked sugar and heterocyclic amino acids by asymmetric MCRs: 06ACR451.
Development of novel nuclear receptor ligands based on receptor-folding inhibition hypothesis: 07YZ341.
Directly linked polyazoles (polythiazoles, polyoxazoles, and thiazolyloxazoles), important moieties in natural products: 05S1907.

Discovery of progesterone receptor agonists and antagonists inspired by the fungal metabolite PF1092C: 06Y559.
Heteroditopic receptors: 07H(72)53.
Natural occurrence, synthesis, and biological activity of piperidine homoazasugars: 05ARK(3)110.
Natural products with maleic anhydride structure: 07CRV1777.
Noncovalent binding of luminescent transition metal polypyridine complexes to avidin, indole-binding proteins and estrogen receptors: 07CCR2292.
Novel bioactive compounds from insect pathogenic fungi: 07Y700.
Novel bioactive *O*-heterocycles isolated from unexploited organisms, cellular slime molds: 07YZ1431.
Progress in the total synthesis of antitumor styryl lactones: 05MRO333.
Recent advances in the total synthesis of piperidine azasugars: 05EJO2159.
Recent methodologies for the synthesis of furan-2(5*H*)-ones and their application in total synthesis of natural products with this subunit: 05MRO139.
Rh(I)-catalyzed cyclizations *via* rhodacycle intermediates and its application to the synthesis of (+)-epiglobulol: 07Y183.
Search for the functions of glyco-linkages in natural glycosides by using trans-glycosylation: 06Y34.
Stereoselective syntheses of naturally occurring 5,6-dihydropyran-2-ones: 07T2929.
Stereoselective synthesis of procyanidin oligomers and their bioactivity: 05Y982.
Structures, sources, and synthetic strategies of natural nonanomeric spiroketals: 05CRV4406.
Study on myxomycetes as a new source of bioactive natural products: 07YZ1369.
Thioglycosides in sequential glycosylation strategies: 05CSR769.
The use of titanocene(III)-mediated radical epoxide opening in syntheses of natural products: 06MRO23.

3. THREE-MEMBERED RINGS

3.1 General topics

Asymmetric synthesis of epoxides and aziridines from aldehydes and imines: 06MI10.
Aziridines and epoxides in organic synthesis (general monograph): 06MI9.
Epoxidation and aziridination reactions using chalcogenides as organocatalysts: 07CRV5841.
Epoxides and aziridines: 06ARK(3)6.

Epoxides and aziridines in click chemistry: 06MI21.
Expanding the utility of lithiated epoxides and aziridines in synthesis: 06SL1.
Metalated epoxides and aziridines in synthesis: 06MI14.
Recent advances in silver-catalyzed nitrene, carbene, and silylene-transfer reactions leading, particularly, to aziridines and silacyclopropanes: 06EJO4313.

3.2 One heteroatom

3.2.1 One nitrogen atom

Advances in nitrogen transfer reactions involving aziridines: 06ACR194.
Asymmetric ring-opening of aziridines with carbon nucleophiles: 06EJO4979.
Asymmetric syntheses with aziridinecarboxylate and aziridinephosphonate building blocks: 06MI12.
Azide compounds as nitrogen sources for atom-efficient and ecologically benign nitrogen-atom-transfer reactions such as aziridination: 05CL1304.
Aziridination by metal-mediated carbon−nitrogen bond formation reactions: 05COC657.
Aziridination of telluronium and sulfonium ylides: 05SL2720.
Aziridine natural products – discovery, biological activity, and biosynthesis: 06MI20.
Aziridines in parallel- and solid-phase synthesis: 07EJO1717.
Chiral nonracemic sulfinimines as versatile reagents for asymmetric synthesis, particularly, of aziridines: 06T8869.
Investigations of the [2,3]-sigmatropic rearrangements of vinylaziridines and allylic amines: 07SL1190.
Methyleneaziridines in organic synthesis: 06SL3205.
N-Phosphinoylimines in synthesis of aziridines: 05S1205.
Recent development of regio- and stereoselective aminohalogenation reaction of alkenes, particularly, aziridinium mechanism of the reaction: 07EJO2745.
Recent progress in the study of aziridination reaction: 06CJO1173.
Synthesis and reactivity of C-heteroatom-substituted aziridines: 07CRV2080.
Synthesis of aziridines: 06MI13.
Toward reaction selectivities of imines and aziridines: 06SL181.
The utility of lithiated aziridines in synthesis: 06SL1.
Vinylaziridines in organic synthesis: 06MI11.

3.2.2 One oxygen atom

3.2.2.1 Reactivity of oxiranes

Asymmetric ring opening of epoxides: 05COC1, 06CJO1208, 06EJO4979.
Catalytic asymmetric epoxide ring-opening chemistry: 06MI16.

Chiral cyclohexene oxide-derived 1,2-amino alcohols and 1,2-diamines in asymmetric synthesis: 06SL2699.

Development of new synthetic reactions featuring tandem carbon−carbon bond formation, particularly, tandem reactions involving α,β-epoxysilanes: 07YZ1399.

Epoxides in complex molecule synthesis: 06MI17.

Epoxysilane rearrangement, its mechanistic studies, and synthetic applications: 06Y1148.

Expanding the utility of lithiated epoxides in synthesis: 05SL1359, 06SL1.

Opening of epoxide ring with intramolecular participation of oxygen-containing nucleophilic group: 05ZOR167.

Reactions of epoxides with oxygen-containing nucleophiles: 06ZOR327.

Recent advances in the semi-pinacol rearrangement of α-hydroxy epoxides and related compounds: 07CSR1823.

Ring opening of epoxides in enantioselective additions of organolithiums: 05S2271.

Synthesis of 1,2-difunctionalized fine chemicals through catalytic, enantioselective ring-opening reactions of epoxides: 06S3919.

Use of titanocene monochloride in organic synthesis, for example, for ring opening of epoxides: 06CJO145.

Vinylepoxides in organic synthesis: 06MI18.

3.2.2.2 Synthesis of oxiranes

Advances in homogeneous and heterogeneous catalytic asymmetric epoxidation: 05CRV1603.

The biosynthesis of epoxides: 06MI19.

Chiral-auxiliary-controlled diastereoselective epoxidations: 05SL1047.

Chromium- and manganese-salen promoted epoxidation of alkenes: 05CRV1563.

Enantioselective epoxidation of olefins with chiral metalloporphyrin catalysts: 05CSR573.

Epoxidation of olefins with dioxiranes: 05CJO745.

Epoxidation of telluronium and sulfonium ylides: 05SL2720.

Frontier of asymmetric epoxidation: utilization of aqueous hydrogen peroxide: 06Y869.

Metal-catalyzed synthesis of epoxides: 06MI15.

Progress in the epoxidation of olefins using hydrogen peroxide as oxidant: 05CCR(249)1944, 07CJO358.

Recent developments in epoxide preparation: 06COS457.

Salen–metal complex catalyzed asymmetric epoxidation of the olefins: 05CJO347.

3.2.3 One sulfur atom

Synthesis and reactions of 2-alkylidene thiiranes and thietanes: 07S2755.

3.3 Two heteroatoms

3.3.1 Two nitrogen atoms

Diazirines as carbene precursors: 06ACR267.

Endeavors to make the photophore, phenyldiazirine easy to use: 07YZ1693.

3.3.2 Two oxygen atoms

Epoxidation of olefins with dioxiranes: 05CJO745.

Recent advances in the study and application of dioxiranes: 05CJO386.

3.3.3 One nitrogen and one oxygen atom

N-Phosphinoylimines in synthesis of oxaziridines: 05S1205.

4. FOUR-MEMBERED RINGS

4.1 General topics

Recent developments in the use of catalytic asymmetric ammonium denolates in chemical synthesis including preparation of β-lactones and β-lactams: 07CRV5596.

4.2 One heteroatom

4.2.1 One nitrogen atom

α-Amino acid derivatives with a Cα–P bond in synthesis of β-lactam antibiotics: 07ARK(6)193.

Benzazetines and their derivatives: 07KGS1763.

β-Lactams as building blocks for the stereoselective synthesis of nonβ-lactam products: 07CRV4437.

Synthesis and applications of sulfur-containing chiral ferrocene derivatives, particularly, ferrocene-containing β-lactams: 07SL360.

Synthesis of β-lactams using the copper(I)-catalyzed cycloaddition of a terminal alkyne and a nitrone, the Kinugasa reaction: 07SL2321.

Theoretical studies on the ring opening of β-lactams in solution and in enzymatic media: 06COC805.

4.2.2 One oxygen atom

Ring opening of oxetanes in enantioselective additions of organolithiums: 05S2271.

Synthesis of polymers with well-defined structures by novel ring-opening reactions of oxetanes: 06Y934.

4.2.3 One sulfur atom

Methods for the synthesis of 3-aminothietane derivatives: 07KGS655.

Synthesis and reactions of 2-alkylidene thiiranes and thietanes: 07S2755.

4.3 Two heteroatoms

Structural aspects of 1,2-dioxetanes active toward intramolecular charge-transfer-induced chemiluminescent decomposition: 05BCJ1899.

5. FIVE-MEMBERED RINGS

5.1 General topics

Application of organocatalysts (particularly, proline, imidazole, thiazole derivatives) to asymmetric synthesis: 06CJO618, 06CJO899.
Asymmetric 1,3-dipolar cycloadditions: 07T3235.
Asymmetric 1,3-dipolar cycloadditions of cyclic stabilized ylides derived from chiral 1,2-amino alcohols: 06SL2349.
Azolium cyclophanes: 06MRO333.
"Click chemistry" and its applications in synthesis of azoles: 06CJO271.
Five-membered *vic*-dioxo heterocycles: 04MI2.
Formation of five-membered heterocyclic rings under radical cyclization conditions: 05T10603.
Intramolecular 1,3-dipolar cycloaddition reactions in targeted syntheses: 07T12247.
Reactions of functionalized alkoxyethylenes with nucleophilic reagents in synthesis of five-membered heterocycles: 06ZOR167.
Recent progress of halogen-dance reactions in heterocycles: 05H(65)2005.
Regioselective cross-coupling reactions of multiple halogenated *N*-, *O*-, and *S*-heterocycles: 05T2245.
Theoretical investigations of [3 + 2] cycloaddition reactions: 06UK1045.

5.2 One heteroatom

We have classified the many reviews dealing with these materials under the following headings:

1. *General.*
2. *One Nitrogen Atom (it is self-subdivided into Monocyclic Pyrroles, Hydropyrroles, Porphyrins and Related Systems, Indoles, Carbazoles, Related Systems, and Hydrogenated Derivatives, Isoindoles Including Phthalocyanins and Porphyrazines, Polycyclic Systems Including Two Heterocycles).*
3. *One Oxygen Atom (Furans, Hydrofurans, Annulated Furans, Five-Membered Lactones).*
4. *One Sulfur Atom (Thiophenes, Annulated Thiophenes).*

5.2.1 General

Advances in the chemistry of trihetarylmethanes (trithienyl-, trifuryl-, and triindolylmethanes): 06T6731.

Formation of five-membered heterocycles under radical cyclization conditions: 07T793.
Functionalized acetylenes as versatile building-blocks for the multicomponent assembling of polysubstituted furans and pyrroles: 07H(73)87.
New reactions in fullerene chemistry, particularly, retro-cycloaddition processes of fullerenopyrrolidines and fullerenoisoxazolines: 07SL3077.
Palladium-catalyzed carboetherification and carboamination reactions of γ-hydroxy- and γ-aminoalkenes for the synthesis of tetrahydrofurans and pyrrolidines: 07EJO571.

5.2.2 One nitrogen atom

Noncovalent interactions, particularly, those in five-membered *N*-heterocycles: 07CSR172.
Preparation of chiral 4-substituted γ-lactams and corresponding γ-amino acids: 07KGS803.
Recent progress in the synthesis of pyrrole, dihydropyrrole, and pyrrolidine compounds: 05CJO1311.
Synthesis and biological activity of pyrrole, pyrroline, and pyrrolidine derivatives with two aryl groups on adjacent positions: 06T7213.
Synthesis and biological properties of sequence-specific DNA-alkylating pyrrole–imidazole polyamides: 06ACR935.

5.2.2.1 Monocyclic pyrroles

Advances in the chemistry of dipyrrins and their complexes (dipyrrin = dipyrrolylmethane): 07CRV1831.
Aminomethylated pyrroles (chemistry and applications): 06MRO167.
Applications of planar-chiral pyrrole derivatives as ligands in asymmetric catalysis: 06ACR853.
Dipyrinones, constituents of the pigments of life: 06OPP347.
Metal-mediated synthesis of pyrroles: 07ARK(10)121.
Palladium-catalyzed cross-coupling and related reactions involving pyrroles: 06EJO3043.
Pyrrole protection: 06T11531.
Ring contraction methodology for the synthesis of pyrroles: 05COC261.
Supramolecular chemistry of "acyclic" π-conjugated oligopyrroles: 07EJO5313.
Synthesis and reactions of pyrrolecarbodithioates: 05RKZ(6)97.
The synthesis of highly functionalized pyrroles: 07S3095.
Synthesis of new pyrroles with potential antimycobacterial, antifungal, and selective COX-2 inhibiting activities: 07COC1092.

5.2.2.2 Hydropyrroles

Asymmetric synthesis of hydroxylated pyrrolidines and related bioactive compounds: 06SL1133.

Chemistry and biology of pyrroline and pyrrolidine nitroxides: 05JHC437.

Construction of enantiopure pyrrolidine ring system *via* asymmetric [3 + 2] cycloaddition of azomethine ylides: 06CRV4484.

Development of proline-derived chiral aminophosphine ligands for palladium-catalyzed asymmetric allylic alkylation: 06Y628.

Intramolecular aminopalladation of alkenes as a key step to pyrrolidines and related heterocycles: 07CSR1142.

N-Methylpyrrolidone (general monograph): 05MI8.

Optically active proline-catalyzed enantioselective organic reactions: 05CJO1619.

Progress in the study on the reaction mechanisms of proline-catalyzed asymmetric direct aldol reactions: 06CJO1463.

Recent advances in the synthesis of nicotine and its derivatives: 07T8065.

Recent progress in the application of *N*-bromosuccinimide to organic chemical reactions: 06CJO1518.

Synthetic approaches to enantiomerically pure 8-azabicyclo[3.2.1]octane derivatives: 06CRV2434.

N-Vinylpyrrolidone in radical copolymerization reactions: 06IVUZ(2)3.

5.2.2.3 Porphyrins and related systems

Advances in the synthesis and chemistry of carbaporphyrins and related porphyrinoids: 07EJO5461.

Application of magnetic circular dichroism spectroscopy to porphyrinoids: 07CCR429.

Applications of magnetic circular dichroism spectroscopy to porphyrins and phthalocyanines: 07CC4077.

Application of metalloporphyrins in catalytic oxidation reactions: 07CJO34.

Artificial photosynthetic systems: assemblies of slipped cofacial porphyrins and phthalocyanines showing strong electronic coupling: 07OBC1679.

Asymmetric heterogeneous catalysis by metalloporphyrins: 06CCR(250)2212.

Bending, stretching, and twisting porphyrins: 06CC243.

Benziporphyrins, synthetic porphyrin analogs with one of pyrrole rings replaced by a benzenoid ring: 05ACR88.

The chemistry of *N*-confused porphyrin (NCP) and its analogs: 05ACR10, 05Y211, 06PAC(78)29.

Chiral self-discriminative self-assembling of *meso*–*meso* linked diporphyrins: 07CCR2743.

Complexing properties of porphyrins: 05UK839.

N-Confused calix[4]pyrroles: 06CCR(250)2929.

Corrole-based applications: 07CC1987.

Creation of porphyrin–fullerene-linked artificial photosynthetic systems: 07BCJ621.
Crown porphyrins: 06CCR(250)519.
Crystal engineering of porphyrin framework solids: 05CC1243.
Developments in heteroporphyrins and their analogs during 1999–2005: 06CCR(250)468.
Dynamic supramolecular porphyrin systems: 05T13.
Electronic structures of highly deformed iron(III) porphyrin complexes: 06CCR(250)2271.
Enantioselective epoxidation of olefins with chiral metalloporphyrin catalysts: 05CSR573.
Fullerene-porphyrin supramolecular discrete host–guest complexes: 05ACR235.
Ground- and excited-state tautomerism in porphycenes: 06ACR945.
Heme–copper/dioxygen adduct formation, properties, and reactivity: 07ACR563.
Hetero-arrays of porphyrins and phthalocyanines: 07CCR2334.
High-valent iron(IV)–oxo complexes of heme and nonheme ligands in oxygenation reactions: 07ACR522.
Metalloporphyrin-NO bonding: 05ACR943.
Metalloporphyrin receptors of bases: 07IZV636.
Modern aspects of porphyrin IX chemistry: 07ZOR3.
New ring-contracted porphyrinoids, corrolazines in high-valent metalloporphyrinoid stabilization, and activation of dioxygen: 07ACR626.
Novel aspects of corrole chemistry: 05MRO355.
Nucleophilic substitution as a tool for the synthesis of unsymmetrical porphyrins: 05ACR733.
Optically active supramolecular porphyrin-based systems: 06UK820.
Photoinduced electron transfer in supramolecular systems of fullerenes functionalized with ligands capable of binding to zinc porphyrins and zinc phthalocyanines: 05CCR(249)1410.
Photophysical properties of metal-mediated assemblies of porphyrins: 06CCR(250)1471.
Photophysics and photochemistry of kinetically labile, water-soluble porphyrin complexes: 06CCR(250)1792.
The photophysics and photochemistry of cofacial free base and metallated bisporphyrins held together by covalent architectures: 07CCR401.
Porphyrin-calix[4]arenes: 05ZOR807.
Porphyrin-containing molecular squares: 06CCR(250)1710.
Porphyrins in Diels–Alder and 1,3-dipolar cycloaddition reactions: 08PHC44.
Progress in the chemical reactions of chlorophyll-*a* derivatives and synthesis of polysubstituted chlorin or porphyrin: 05CJO1353.

Progress in the study of fluoroporphyrins: 07CJO24.
Reactivity and electronic structure of stable high-spin nickel(II) or copper (II) derivatives of core modified porphyrins (21-heteroporphyrins and 2-aza-21-carbaporphyrin): 05CCR(249)2510.
Recent advances in the synthesis of hydroporphyrins: 07COC1310.
Structure and properties of sterically twisted porphirins: 05UK268.
Supported metalloporphyrin catalysts for alkene epoxidation: 06OBC599.
Synthesis and properties of calixarene-porphyrin conjugates: 05CJO375.
Synthesis of *meso*-phenyl substituted porphyrins as starting compounds for preparation of porphyrin-containing polymers: 07IZV680.
Synthesis, structure, reactivity, and photoluminescence of lanthanide(III) monoporphyrinate complexes: 07CCR2386.
Synthetic receptors based on porphyrins and their calyx[4]arene conjugates: 06MI6.
Synthetic routes to porphyrins bearing fused rings: 06T10039.
Synthetic strategies and structural aspects of metal-mediated multiporphyrin assemblies: 06ACR841.
Tailoring porphyrins and chlorins for self-assembly in biomimetic artificial antenna systems: 05ACR612.
Transition-metal complexes of expanded porphyrins: 07ACR371.
Transition metal-mediated fuctionalization and multiplication of porphyrins toward giant functional systems: 07Y298.
Understanding binding interactions of cationic porphyrins with B-form DNA: 05CCR(249)1451.
X-ray structural chemistry of cobalamins: 06CCR(250)1332.

5.2.2.4 Indoles, carbazoles, related systems, and hydrogenated derivatives

Acid–base catalysis of asymmetric fluorination of oxindoles: 05YZ785.
Annulated indoles: 06ARK(7)67.
Bartoli indole synthesis (reaction of vinyl magnesium halides with *o*-substituted nitroarenes): 05COC163.
Catalytic and stereoselective alkylation of indoles: 05SL1199.
Catalytic arylations in indole syntheses: 07SL507.
Catalytic asymmetric hydrogenation of indoles: 07Y109.
5,6-Dihydroxyindoles and indole-5,6-diones: 05AHC(89)1.
Direct and efficient organic synthesis using indium catalysts, particularly, Friedel–Crafts alkylation of indoles with allylic or benzylic alcohols: 07Y99.
Enantioselective fluorination reactions of active methine compounds, particularly, oxindole derivatives catalyzed by chiral palladium complexes: 06SL1467.
2-Indolylacyl radicals in the synthesis of indole compounds: 09PHC1.
Methods for synthesis and chemical properties of isogramins: 05KGS483.

Naphthostyryl chemistry (general monograph): 05MI6.

Nucleophilic addition, 1,3-dipolar cycloaddition, and Diels–Alder reactions of indoles substituted at the 2- or 3-position with electron-withdrawing groups (NO_2, $PhSO_2$) to give a variety of indoles, pyrroloindoles, and carbazoles: 05COC1493.

Polymer-supported indole chemistry; chemical properties of polymer-bound indole system: 06IVUZ(10)3.

Practical methodologies for the synthesis of indoles: 06CRV2875.

Recent development of Fischer indole synthesis in technology: 06CJO1025.

Recent uses of palladium chemistry in indole synthesis: 06COS477.

Synthesis and applications of amphiphilic fullerenopyrrolidine derivatives: 06OBC1629.

Synthesis and functionalization of indoles through palladium-catalyzed reactions: 05CRV2873.

Synthesis and functionalization of indoles through rhodium catalyzed reactions: 07COS201.

Synthesis of indole derivatives *via* isocyanides: 06OBC757.

An update on catalytic enantioselective alkylations of indoles: 07MRO115.

The Witkop–Winterfeldt oxidation of indoles (formation of quinolones): 07COC159.

5.2.2.5 Isoindoles (including phthalocyanins and porphyrazines)

Advanced methods for the synthesis of 3-substituted 1*H*-isoindol-1-ones: 05COC1277.

Advances in the studies of photochemical reactions and synthetic applications of arenedicarboximides, in particular, phthalimide and *N*-methylnaphthalimide: 06CJO278.

Applications of magnetic circular dichroism spectroscopy to porphyrins and phthalocyanines: 07CC4077.

Approaches to the formation of condensed isoindolones: 05COC1261.

Axially modified gallium phthalocyanines and naphthalocyanines for optical limiting: 05CSR517.

Effects of substituents on the photochemical and photophysical properties of main group metal (Zn, Al, Ge, Si, Sn, Ga, and In) phthalocyanines: 07CCR1707.

Hetero-arrays of porphyrins and phthalocyanines: 07CCR2334.

Metalloporphyrin hosts for supramolecular chemistry of fullerenes: 07CSR189.

Novel families of phthalocyanine-like macrocycles – porphyrazines with annulated electron-withdrawing 1,2,5-thia/selenodiazole rings: 06CCR(250)1530.

Photochemical addition reactions involving phthalimides: 05H(65)2221.

Photoinduced electron transfer in supramolecular systems of fullerenes functionalized with ligands capable of binding to zinc porphyrins and zinc phthalocyanines: 05CCR(249)1410.

Phthalocyanines in nanotechnology: 07CC2000.

Synthesis and excitation energy transfer of cyclic porphyrin arrays as artificial photosynthetic antenna: 07CSR831.

Synthesis and structure modification of unsymmetrically substituted phthalocyanines: 07UK732.

Synthesis, properties, and applications of ruthenium phthalocyanine and naphthalocyanine complexes: 07CCR1128.

Vibrational spectroscopy of phthalocyanine and naphthalocyanine in sandwich-type (na)phthalocyaninato and porphyrinato rare earth complexes: 06CCR(250)424.

5.2.2.6 Polycyclic systems including two heterocycles

Chemistry of indoloindoles: 07UK348.

Configuration, conformation, reactivity, and applications of hexahydropyrrolo[2,3-*b*]indoles in synthesis: 07ACR151.

Effect of preferential conformations on base properties and thermodynamics of conformation conversion along with type of ring fusion on *cis*--*trans*-conversion of bicycle in pyrrolizidines: 06KGS1443.

Excited-state double-proton transfer in the 7-azaindole dimer in the gas phase: 06BCJ373.

11*H*-Isoindolo[2,1-*a*]benzimidazoles: 07KGS323.

Methods for construction of isoindolo[1,2]-fused benzazepines and benzazocines: 06KGS963.

Methods for construction of isoindolo[1,2]-fused quinolines and isoquinolines: 06KGS1123.

Organometallic methods for the synthesis and functionalization of azaindoles: 07CSR1120.

Pyrrolo[2,1-*b*]thiazoles: 07H(71)761.

Recent developments in the synthesis of indolizines: 07CJO1060.

Recent developments on the synthesis of indolizidine derivatives (−)-swainsonine and analogs: 05COS39.

Synthesis and chemical transformations of 2,3-dihydropyrrole-2,3-diones [*a*] annulated with azaheterocycles: 06KGS3.

Synthesis and reactivity of 5- and 6-azaindoles: 07T8689.

Synthesis and reactivity of 7-azaindole (1*H*-pyrrolo[2,3-*b*]pyridine): 07T1031.

Synthesis of 1*H*-pyrrolo[1,2-*b*][1,2,4]triazole as a novel heterocyclic cyan dye-forming coupler for color photographic use: 06Y222.

5.2.3 One oxygen atom

5.2.3.1 Furans

2-Aminofurans and 3-aminofurans: 06AHC(92)1.

Furan oxidations (using furan ring as a C-1 or C-4 synthon) in organic synthesis: 07COC1076.

Intramolecular thermal and catalytic [4 + 2] cycloaddition in 2-alkenylfurans: 05UK707.

Metal-mediated synthesis of furans: 07ARK(10)121.

Recent methodologies for the synthesis of furan-2(5*H*)-ones and their application in total synthesis of natural products with this subunit: 05MRO139.

Syntheses of polysubstituted furans: 06OBC2076.

5.2.3.2 Hydrofurans

Recent advances in the stereoselective synthesis of tetrahydrofurans: 07T261.

Synthesis of dihydrofurans substituted in position 2: 05EJO4929.

Synthesis, properties, and application of 5-alkoxy-2,5-dihydrofuran-2-ones: 05CJO239.

Synthetic strategies toward naturally occurring tetronic acids: 06S3157.

Transition metal-catalyzed asymmetric ring opening reactions of oxabenzonorbornadienes: 06CJO1613.

5.2.3.3 Annulated furans

Advances in the biological activities and synthesis of 2-arylbenzo[*b*]furans: 05CJO25.

Naphtho[2,3-*c*]furan-4,9-diones and related compounds: 05T9929.

The palladium-catalyzed assembly and functionalization of benzo[*b*]furans: 06COC1423.

Progress in syntheses of 3-*n*-butylphthalide and its analogs: 07COC833.

Recent advances in the Hauser annulation of phthalides and their analogs: 07CRV1892.

Use of stabilized phthalide anion annulation reactions in synthesis: 07S643.

5.2.3.4 Five-membered lactones

Applications of Baeyer–Villiger monooxygenase for the synthesis of five-membered lactones: 05CJO1198.

Chemistry and biology of resorcylic acid lactones: 07CC22.

Synthesis of butenolides by one-pot cyclization reactions of silyl enol ethers with oxalyl chloride: 06SL3369.

5.2.4 One sulfur atom

5.2.4.1 Thiophenes

Cascade heterocyclization in synthesis of thiophene derivatives and their fused analogs: 05RKZ(6)11.

C–C Cross-coupling reactions for the combinatorial synthesis of oligothiophenes: 02JOM(653)200.

Electronic energy transfer in a dinuclear Ru/Os complex containing a photoresponsive dithienylethene derivative as bridging ligand: 05CCR(249)1327.

Photochromic bisthienylethenes as multifunction switches: 07CC781.

Nitrobutadienes from β-nitrothiophenes as valuable building-blocks in the overall ring-opening/ring-closure protocol to homo- or heterocycles: 06ARK(7)169.

Stability of thiophenium ions and features of reactions of thiophenes with electrophiles: 05RKZ(6)59.

Synthesis and reactivity of thioaurones (2-benzylidenebenzo[*b*]thiophen-3 (2*H*)-ones): 05H(65)451.

Synthesis of uniform, nonnatural oligomers, particularly, oligothiophenes: 06SL1793.

Tetraphenylenes and helical β-oligothiophenes as annulated, chiral π-conjugated systems: 07SL1799.

Thiophene 1,1-dioxides as building blocks in organic synthesis and materials chemistry: 06UK1139.

5.2.4.2 Annulated thiophenes

Chemistry of benzo[*c*]thiophene: 07MI1.

Chemistry of thienothiophenes: 05UK235, 06AHC(90)125.

Chemistry of thienopyridines and related systems: 06MI4.

Dithienothiophenes, their syntheses and electronic and optical properties: 05T11055.

Chemistry of thienopyridines: 07AHC(93)117.

Chemistry of thienopyrimidines: 06AHC(92)83.

Progress in the study of thieno[3,4-*c*]thiophenes: 05CJO475.

Synthesis of polynuclear aromatic compounds incorporating a fused thiophene ring: 05H(65)1491.

5.2.4.3 Hydrothiophenes

Cascade reactions of unsaturated xanthates and related reactions in the synthesis of hydrogenated isobenzothiophenes: 05YZ469.

Single-site metal catalysts in the hydrogenation of thiophenes: 04JOM(689)4277.

5.3 Two heteroatoms

We have classified the many reviews dealing with these materials under the following headings:

1. *General.*
2. *Two Nitrogen Atoms (it is self-subdivided into Pyrazoles, Imidazoles, and Annulated Imidazoles).*

3. *One Nitrogen and One Oxygen Atom (1,2-Heterocycles, 1,3-Heterocycles).*
4. *One Nitrogen and One Sulfur Atom.*
5. *Two Oxygen Atoms.*
6. *Two Sulfur Atoms.*

5.3.1 General

Azoles with two pyridine substituents at carbon atoms; their synthesis and application in coordination chemistry: 05KGS1290.

Cross-coupling reactions on azoles with two and more heteroatoms: 06EJO3283.

Reactions and bioactivity of oximes of five-membered heterocycles with two heteroatoms: 07KGS1123.

Synthesis and structure of oximes of five-membered heterocycles with two heteroatoms: 07KGS483.

Synthetic utility of five-membered heterocycles – chiral functionalization and applications (2-oxazolone, 1,3-dihydro-2-imidazolone, 2-thiazolone, chiral synthons, chiral auxiliaries, chiral ligands): 05T8073.

5.3.2 Two nitrogen atoms

5.3.2.1 Pyrazoles

Classification of hydrogen-bond motives in crystals of NH-pyrazoles: 06ARK(2)15.

Coordination modes of 5-pyrazolones (X-ray diffraction data): 07CCR1561.

Coordination properties of didentate alcohols and aldehydes derived from imidazole, pyrazole, or pyridine toward Cu(II), Co(II), Zn(II), and Cd(II) ions in the solid state and aqueous solution: 05CCR(249)2259.

Metal derivatives of bis(pyrazolyl)alkanes: 05CCR(249)663.

Metal derivatives of tris(pyrazolyl)alkanes: 05CCR(249)525.

Pyrazole chemistry in crop protection: 07H(71)1467.

Silver(I) salts as useful reagents in pyrazole synthesis: 07ARK(2)224.

Stereoselective cycloadditions of nitrilimines as a source of enantiopure 4,5-dihydropyrazoles: 05H(65)2513.

The synthesis and coordination chemistry of 2,6-bis(pyrazolyl)pyridines and their terpyridine analogs: 05CCR(249)2880.

Synthesis, structures, metal coordination chemistry, and applications of 4-acyl-5-pyrazolone ligands: 05CCR(249)2909.

Utilization of chiral enaminones and azomethine imines in the synthesis of functionalized pyrazoles: 06ARK(7)35.

5.3.2.2 Imidazoles

Approaches to crystallization from ionic liquids: 06CC4767.

Bio-inspired membranes for advanced polymer electrolyte fuel cells (anhydrous proton-conducting membrane *via* molecular self-assembled acid–base complex such as one consisting of an acidic surfactant, mono-dodecylphosphate, and a basic surfactant, 2-undecylimidazole): 07BCJ2110.

The chemistry of the C(2) position of imidazolium room temperature ionic liquids (mainly, deprotonation to give *N*-heterocyclic carbenes): 07COS381.

Chemistry of ureidocarboxylic and ureylenedicarboxylic acids: 06UK217.

Computer simulation of clusters, liquids, and crystals of dialkylimidazolium salts: 07ACR1156.

Coordination properties of didentate alcohols and aldehydes derived from imidazole, pyrazole, or pyridine toward Cu(II), Co (II), Zn(II), and Cd(II) ions in the solid state and aqueous solution: 05CCR(249)2259.

Crystal engineering of binary metal imidazolate and triazolate frameworks: 06CC1689.

2,3-Dihydroimidazol-2-ylidenes and their main group element chemistry: 05CCR(249)829.

From the reactivity of *N*-heterocyclic carbenes to new chemistry in ionic liquids: 06CC1809.

Functional design of ionic liquids: 06BCJ1665.

Functionalized imidazolium salts for task-specific ionic liquids and their applications: 06CC1049.

Imidazolium receptors for the recognition of anions: 06CSR355.

New guanidines from 2-imidazolidinones through 2-chloroamidinium derivatives: 06ARK(7)148.

New methods of imidazole functionalization – from imidazole to marine alkaloids: 06SL965.

Recent progress in the catalytic synthesis of imidazoles: 07CAJ568.

Synthesis and biological activity of vicinal diaryl-substituted 1*H*-imidazoles: 07T4571.

Synthesis and biological properties of sequence-specific DNA-alkylating pyrrole–imidazole polyamides: 06ACR935.

5.3.2.3 Annulated imidazoles

1- and 3-Deazapurines (imidazopyridines): 05AHC(89)159.

11*H*-Isoindolo[2,1-*a*]benzimidazoles: 07KGS323.

Magnetism of metal-nitroxide compounds involving bis-chelating imidazole and benzimidazole substituted nitronyl nitroxide free radicals: 05CCR(249)2591.

Stable heteroaromatic carbenes of the benzimidazole series: 05ARK(8)10.

Synthesis and reactivity of 4-azaindoles: 07T8689.

5.3.3 One nitrogen and one oxygen atom

5.3.3.1 1,2-Heterocycles

Preparations and utility of aminoisoxazoles in the synthesis of fused systems: 07AHC(94)173.

Recent advances in the synthesis and reactivity of isoxazoles: 05COC925.

Syntheses of polyfunctionalized compounds using nitroisoxazolones: 05Y1232.

Synthesis of isoxazolidines and isoxazolines from nitrones by 1,3-dipolar cycloaddition on solid supports: 05CSR507.

5.3.3.2 1,3-Heterocycles

Applications and modifications of oxazolidin-2-ones in synthetic organic chemistry: 07COS238.

The construction of the oxazolidin-2-one ring: 07COS81.

The diverse chemistry of oxazol-5-(4*H*)-ones: 07CSR1432.

C_2-Symmetric chiral bis(oxazoline) ligands in asymmetric catalysis: 06CRV3561.

Synthesis and use of bisoxazolinyl-phenyl pincers: 07CSR1133.

Synthetic applications of chiral 3-acyloxazolidin-5-ones: 06ARK(7)292.

5.3.4 One nitrogen and one sulfur atom

Isothiazolium salts and their use as components for the synthesis of *S,N*-heterocycles: 07AHC(94)215.

Pyrrolo[2,1-*b*]thiazoles: 07H(71)761.

Recent progress in the synthesis of thiazolopyrimidine analogs: 07CJO166.

Synthesis and applications of chalcogenoamides, particularly, for preparation of thiazoles: 07COS15.

Synthesis and chemical properties of 2-substituted thiazoline-4,5-diones: 06KGS1283.

Synthesis of fused thiazoles: 06KGS167.

Synthetic approaches toward 2-iminothiazolidines: 06T513.

5.3.5 Two oxygen atoms

Electrochemical and structural (X-ray) properties of homoleptic, mononuclear transition metal complexes of 1,2-dioxolenes: 06CCR(250)2000.

Synthetic applications of chiral 1,3-dioxolan-4-ones: 06ARK(7)292.

5.3.6 Two sulfur atoms

Electronic communication in tetrathiafulvalene (TTF)/C_{60} systems: 07ACR1015.

Synthesis and applications of tetrathiafulvalenes and ferrocene-tetrathiafulvalenes and related compounds as electroactive organic materials: 05T3889.

Synthesis of 1,3-dithiole-2-thiones and tetrathiafulvalenes using oligo(1,3-dithiole-2,4,5-trithione): 06KGS483.

5.4 Three heteroatoms

Cross-coupling reactions on azoles with two and more heteroatoms: 06EJO3283.

5.4.1 Three nitrogen atoms

5.4.1.1 Monocyclic systems

Click chemistry (triazole synthesis and beyond): 07S1589.

Copper-catalyzed azide–alkyne cycloaddition producing only 1,4-disubstituted-1,2,3-triazoles at room temperature in excellent yields: 07AA7.

Cu^I-catalyzed alkyne-azide "click" cycloadditions, a mechanistic and synthetic perspective: 06EJO51.

The growing applications of click chemistry: 07CSR1249.

Mechanistic studies in triazolinedione ene reactions: 05SL713.

N-Heterocyclic carbene-copper(I) complexes in homogeneous catalysis, particularly, of [3 + 2] cycloaddition of azides and alkynes: 07SL2158.

Organic azides in reactions with participation and preparation of 1,2,3-triazoles: 05AG(E)5188.

Recent applications of the Cu^I-catalyzed Huisgen azide–alkyne 1,3-dipolar cycloaddition reaction in carbohydrate chemistry, particularly, synthesis of *N*-glycosyl-1,2,3-triazoles: 07OBC1006.

Stable heteroaromatic carbenes of the 1,2,4-triazole series: 05ARK(8)10.

Synthesis of triazoles and tetrazoles by 1,3-dipolar cycloaddition ("click chemistry") in bioconjugate chemistry: 06CJO1640.

Synthetic approaches to 1,2,4-triazoles and their pharmacological importance: 06KGS1605.

1,2,3-Triazole and its derivatives, general review: 05UK369.

Triazole as the keystone in glycosylated molecular architectures constructed by a click reaction: 07CAJ700.

5.4.1.2 Annulated triazoles

Acylbenzotriazoles as advantageous *N*-, *C*-, *S*-, and *O*-acylating agents (2000–2005): 05SL1656.

Benzotriazole-mediated amino-, amido-, alkoxy-, and alkylthio-alkylation: 05T2555.

Benzotriazole-stabilized acyl anion synthons: 06ARK(4)119.

Synthesis and biological activity of 1,2,4-triazolo[1,5-*a*][1,3,5]triazines (5-azapurines): 06H(68)1723.

Synthesis of 1*H*-pyrrolo[1,2-*b*][1,2,4]triazole as a novel heterocyclic cyan dye-forming coupler for color photographic use: 06Y222.

5.4.2 Two nitrogen atoms and one oxygen atom

1,3-Dipolar cycloaddition of nitrones to free and coordinated nitriles as routes to govern the process of the synthesis of 2,3-dihydro-1,2,4-oxadiazoles: 06IZV1803.

Synthesis and synthetic applications of 1,2,4-oxadiazole-4 oxides: 07COC959.

Synthesis of 2,5-disubstituted 1,3,4-oxadiazoles and their activity as insect growth regulators: 06CJO1647.

5.4.3 Two nitrogen atoms and one sulfur atom

Porphyrazines with annulated strongly electron-withdrawing 1,2,5-thiadiazole rings: 06CCR(250)1530.

5.4.4 One nitrogen atom and two sulfur atoms

1,2,3-Dithiazole chemistry in heterocyclic synthesis: 06ARK(7)207.

5.5 Four heteroatoms

Development of conductive organic molecular assemblies (organic metals, superconductors, and exotic functional materials, particularly, tetrathia(selena)fulvalenes and their derivatives): 07BCJ1.

Magnetic exchange interactions in perfluorophenyl 1,2,3,5-dithiadiazolyl radicals: 05CCR(249)2631.

Metal derivatives of tetrazoles: 06UK569.

Methods of functionalization of tetrazoles – strategy and prospects: 06ZOR487.

Organic azides in reactions with participation and preparation of tetrazoles: 05AG(E)5188.

Organometallic tetrazole derivatives, their preparation, and synthetic application: 05ZOR1599.

Properties of tetrazole-containing polymers: 06IVUZ(6)3, 06IVUZ(8)3.

Protolytic equilibria of tetrazoles: 06ZOR1599.

Synthesis of triazoles and tetrazoles by 1,3-dipolar cycloaddition ("click chemistry") in bioconjugate chemistry: 06CJO1640.

Tetrazole derivatives as drugs: 07KGS3.

Tetrazolium salts applications in inorganic analysis: 07UK187.

6. SIX-MEMBERED RINGS

6.1 General

Design of heterocyclic systems based on vinyl sulfones of polyfluorinated benzene and halopyridine series: 05RKZ(6)69.

Dihydroazines based on α,β-unsaturated ketones reactions: 06COC297.

Environmentally benign solvent systems and a greener [4 + 2] cycloaddition process for the synthesis of heterocycles: 07MRO89.

Formation of six-membered heterocyclic rings under radical cyclization conditions: 05T10603, 07T793.

The hetero Diels–Alder approach to carbohydrate-containing molecular scaffolding: 07COS47.

Nitroso and azo compounds in modern organic synthesis, particularly, nitroso and azo hetero-Diels–Alder reactions: 07BCJ595.

Recent progress in the synthesis of heterocycles (pyridines, pyridones, pyrans, pyrimidinediones, etc.) using [2 + 2 + 2] cycloaddition reactions catalyzed by transition metal complexes: 06ASC2307.

Stereoselective aza-Diels–Alder reactions: 06COC981.

Substituent tautomerism of six-membered ring heterocycles: 06AHC(91)1.

6.2 One heteroatom

We have classified the many reviews dealing with these materials under the following headings:

1. *One Nitrogen Atom (it is self-subdivided into Pyridines, Pyridinium Compounds, Ylides, Pyridine N-Oxides, Applications of Pyridines, Bipyridines and Related Systems, Hydropyridines, Biologically Active Pyridines and Hydropyridines, Pyridines Annulated with Carbocycles, Pyridines Annulated with Heterocycles).*
2. *One Oxygen Atom (Pyrans and Hydropyrans, Annulated Pyrans and Pyrylium Salts).*

6.2.1 One nitrogen atom

6.2.1.1 Pyridines

The Bohlmann–Rahtz pyridine synthesis: 07SL2459.

Chemistry of 3-cyanopyridine-2(1*H*)-chalcogenones: 06UK645.

Convergence of spectroscopic and kinetic electron transfer parameters for mixed-valence binuclear dipyridylamide ruthenium amine complexes: 05CCR(249)507.

Coordination properties of didentate alcohols and aldehydes derived from imidazole, pyrazole, or pyridine toward Cu(II), Co(II), Zn(II), and Cd(II) ions in the solid state and aqueous solution: 05CCR(249) 2259.

Metallocomplexes of bis(2-pyridylamides) and their applications in catalytic reactions: 05CCR(249)727.

Methodology for the synthesis of pyridines and pyridones: 07H(74)101.

New synthesis of pyridones using conjugate addition reactions of active methine compounds to alkynyl imines and ketones: 06Y251.

New synthetic methods to 2-pyridone rings: 05COC1757.

Nitropyridines, their synthesis and reactions: 05JHC463.

Organometallic complexes of B-, Si- (Ge-), and P- (As-, Sb-) analogs of pyridine: 05AHC(89)149.

Organometallic complexes of the η^2 (*n*,*c*)-coordinated derivatives of pyridine: 05AHC(88)111.

Organometallic chemistry of polypyridine ligands: 07AHC(93)179, 07AHC(94)107.

Pyridine-containing macrocycles *via* comediated [2 + 2 + 2] cycloadditions of α,ω-bis-alkynes: 07ARK(12)7.

Regiochemical control and completeness in elaboration of pyridines through organometallic intermediates: 07CSR1161.

Regioselective functionalization of unreactive carbon–hydrogen bonds, in particular, arylation of pyridines by using aryl iodide, silver acetate, and catalytic palladium acetate: 06SL3382.

Synthesis of optically active 1-(2-pyridinyl)ethyl derivatives: 07Y127.

Synthesis of uniform, nonnatural oligomers, particularly, oligopyridines and oligo(pyridine–pyrimidine)s: 06SL1793.

Transition metal-catalyzed [2 + 2 + 2] cycloaddition reactions to form pyridine ring systems: 07CSR1085.

6.2.1.2 Pyridinium compounds, ylides, pyridine N-oxides

Chemistry of *N*-fluoropyridinium salts: 05CSR1031.

Mechanistic studies of photochemical transformations of pyridinium salts and applications in synthesis: 07OBC2735.

Preparation of new nitrogen-bridged heterocycles using pyridinium salts and *N*-ylides: 05Y222.

Solubility of CO_2, CH_4, C_2H_6, C_2H_4, O_2, and N_2 in 1-hexyl-3-methylpyridinium bis(trifluoromethylsulfonyl)imide (comparison to other ionic liquids): 07ACR1208.

Switchable nonlinear optical metallochromophores with pyridinium electron acceptor groups: 06ACR383.

6.2.1.3 Applications of pyridines

Bis(imino)pyridines, surprisingly reactive ligands and a gateway to new families of catalysts: 07CRV1745.

Applications of planar-chiral pyridine derivatives as ligands in asymmetric catalysis: 06ACR853.

Chiral dialkylaminopyridine catalysts in asymmetric synthesis: 07CRV5570.

Chiral pyridine-containing ligands in asymmetric catalysis: 07CCR2188.

Crystal engineering of coordination polymers using 4,4′-bipyridine as a bond between transition metal atoms: 06CC4169.

Ethylene oligomerization, homopolymerization, and copolymerization by iron and cobalt catalysts with 2,6-(bis-organylimino)pyridyl ligands: 06CCR(250)1391.

Metal complexes of 4,4′-dipyridyldisulfide – structural diversity derived from a twisted ligand with axial chirality: 06CCR(250)2595.

P-Phos and its variants as versatile and effective atropisomeric dipyridylphosphine ligands in asymmetric catalysis: 06ACR711.

2-Pyridylsilyl group as useful multifunctional group in organic synthesis: 06SL157.

Pyridylsilyl group-driven cross-coupling reactions: 02JOM(653)105.

Synthesis of camphor-based pyridine ligands and their application in asymmetric catalysis: 06CSR1230.

6.2.1.4 Bipyridines and related systems

Emerging host–guest chemistry of synthetic nanotubes (nanotubes containing polypyridines and calixcrown tubes): 07CC3891.

Functional bi- and tripyridyl-based ruthenium(II)- and iridium(III)-containing polymers for potential electro-optical applications: 07CSR618.

The higher oligopyridines and their metal complexes: 06COS19.

Metal-organic molecular architectures with 2,2′-bipyridyl-like and carboxylate ligands: 05CCR(249)545.

Photophysical properties and use in photoactive supramolecular assemblies of $[Ru(bipy)(CN)_4]^{2-}$ and its derivatives: 06CCR(250)3128.

Photophysics in bipyridyl and terpyridyl platinum(II) acetylides: 06CCR (250)1819.

Ru(II)-bipyridine complexes in supramolecular systems, devices, and machines: 06CCR(250)1254.

The synthesis and coordination chemistry of 2,6-bis(pyrazolyl)pyridines and their terpyridine analogs: 05CCR(249)2880.

Synthesis of azolylpyridines, promising ligands for coordination chemistry: 05ARK(4)208.

2,2′:6′,2″-Terpyridines (from chemical obscurity to common supramolecular motifs): 07CSR246.

6.2.1.5 Hydropyridines

Asymmetric organocatalytic reductions mediated by dihydropyridines: 07OBC3407.

Asymmetric synthesis of hydroxylated piperidines and related bioactive compounds: 06SL1133.

Chemistry and biology of piperidine and tetrahydropyridine nitroxides: 05JHC437.

Chemistry of quinuclidines as nitrogen bicyclic bridged-ring structures: 06JHC1397.

Intramolecular cation–π interaction in organic synthesis (particularly, synthesis of chiral dihydropyridines): 07OBC2903.

Progress in investigation of oxidation of dihydropyridines and their analogs: 07UK27.

Stereoelectronic substituent effects, particularly, in glycosides and hydroxylated piperidines: 06ACR259.

Synthetic approaches to enantiomerically pure 8-azabicyclo[3.2.1]octane derivatives: 06CRV2434.

TEMPO (2,2,6,6-tetramethylpiperidine-*N*-oxyl) as reagent in alcohol oxidation and in synthesis of natural products: 06MRO155.

6.2.1.6 Biologically active pyridines and hydropyridines

Biological labeling reagents and probes derived from luminescent transition metal polypyridine complexes: 05CCR(249)1434.

Chemical and pharmacological significance of 1,4 dihydropyridines: 07COC847.

Comparison of the ability of pyridinium aldoximes to reactivate human red blood cell cholinesterases inhibited by ethyl- and methyl-paraoxon: 07COC1624.

Fine tuning of structure and reactivity of copper complexes using pyridylalkylamine ligands – active site models for copper proteins: 05Y1240.

Progress in coenzyme NADH model compounds and asymmetric reduction of benzoylformate: 07SL2785.

Recent advances in the synthesis of nicotine and its derivatives: 07T8065.

6.2.1.7 Pyridines annulated with carbocycles

Recent advance in quinoline derivatives with biological activities: 07CJO1318.

Recent progress in the synthesis of quinolines: 05COC141.

Ruthenium(II)-catalyzed alkyne cyclotrimerizations, in particular, synthesis of bicyclic pyridines *via* [2 + 2 + 2] cycloaddition of nitriles and dinitriles: 05Y112.

Selected recent advances in the stereoselective synthesis of isoquinoline and β-carboline derivatives with the use of chiral auxiliaries of natural origin: 05COS301.

Synthesis and properties of quinolines spiro annulated at heterocyclic fragment: 05JHC39.

Synthesis, reactions, and biological activity of quinoline oximes: 05KGS163.

Traditional and modern approaches to Skraup and Dőbner-Miller syntheses of quinolines: 06KGS803.

The Witkop–Winterfeldt oxidation of indoles (formation of quinolones): 07COC159.

6.2.1.8 Pyridines annulated with heterocycles

Advances in the area of pyridodiazinium systems containing bridgehead-nitrogen atom: 06COC319.

Advances in the chemistry of naphthyridines: 06AHC(91)189.

Biologically active benzo[*b*]naphthyridines: 05UK1001.

Chemistry of thienopyridines: 07AHC(93)117.

Chemistry of thienopyridines and related systems: 06MI4.

1- and 3-Deazapurines (imidazopyridines): 05AHC(89)159.

Excited-state double-proton transfer in the 7-azaindole dimer in the gas phase: 06BCJ373.

Methods for construction of isoindolo[1,2]-fused quinolines and isoquinolines: 06KGS1123.

Organometallic methods for the synthesis and functionalization of azaindoles: 07CSR1120.

Progress in synthesis of pyridopyrimidine analogs: 05CJO1530.

Raman scattering and photophysics in spin-state-labile d^6 Ru(II)- and Fe (II)dipyridophenazine complexes: 06CCR(250)1696.

Recent development in the synthesis of indolizines: 07CJO1060.

Synthesis and reactivity of 4-, 5-, and 6-azaindoles: 07T8689.

Synthesis and reactivity of 7-azaindole (1*H*-pyrrolo[2,3-*b*]pyridine): 07T1031.

Synthetic approaches to pyrrolo-, furo-, thienoquinolines, and their benzo or heterocyclic annulated derivatives: 05H(65)901.

6.2.2 One oxygen atom

6.2.2.1 Pyrans and hydropyrans

The Diels–Alder cycloadditions of 3,5-dibromo-2-pyrone and its derivatives: 07PHC1.

2-Methyl-4-oxo-4*H*-1-benzopyran-4-one as a synthon in heterocyclic chemistry: 06JHC813.

Photoinduced reactions of chromones and bischromones: 06ARK(9)239.

Six-membered ring spiranes: carbocycles and heterocycles with oxygen: 05COC1287.

Stereoselective syntheses of naturally occurring 5,6-dihydropyran-2-ones: 07T2929.

Strategies for the formation of tetrahydropyran rings in the synthesis of natural products: 06EJO2045.

Synthetic approaches to α,β-unsaturated δ-lactones and lactols: 07EJO225.

6.2.2.2 Annulated pyrans and pyrilium salts

Biomimetic synthesis of *trans*,*syn*,*trans*-fused polycyclic ethers (polyoxepanes and polypyrans): 06SL1816.

Chemistry of 4-oxo-4*H*-1-benzopyran-3-carbonitrile: 05JHC1035.

C–C Cross-coupling reactions for the combinatorial synthesis of coumarin dyes: 02JOM(653)200.

Cycloaddition reactions of transition metal-containing benzopyrylium and related zwitterionic intermediates: 06CL1082.

Flavonoids, a general monograph: 06MI7.

Methods for isolation and analysis of natural coumarins: 06KFZ(6)47.

3-Phenoxychromones, their occurrence in nature, methods for synthesis and modification, and biological properties: 06KPS203.

Spiropyrans. Synthesis, properties, and applications: 05KGS323.
Strategies and approaches for constructing 1-oxadecalins: 06T10785.
Synthesis and modification of 4-arylcoumarins (neoflavones): 05KPS199.
The synthesis of chromenes, chromanes, coumarins, and related heterocycles *via* tandem reactions of salicylic aldehydes or salicylic imines with α,β-unsaturated compounds: 07OBC1499.
Synthesis of isochromanes and related pyran-type heterocycles using oxa-Pictet–Spengler cyclization: 06S187.

6.3 Two heteroatoms

We have classified the many reviews dealing with these materials under the following headings:

1. *Two Nitrogen Atoms (it is self-subdivided into 1,2-Heterocycles, 1,3-Heterocycles, Monocyclic Pyrimidines and Hydropyrimidines Except Pyrimidine Nucleoside Bases and Nucleosides, Annulated Pyrimidines Except Purines, Pteridines, and Flavins, Pyrimidine Nucleoside Bases and Purines, Nucleotides and Nucleosides, Nucleic Acids, Pyrazines and Hydropyrazines).*
2. *One Nitrogen and One Oxygen Atom.*
3. *One Nitrogen and One Sulfur Atom.*
4. *Two Oxygen Atoms.*

6.3.1 Two nitrogen atoms

Advances in the area of pyridodiazinium systems containing a bridgehead-nitrogen atom: 06COC319.

6.3.1.1 1,2-Heterocycles

Cross-coupling and metallation reactions of 3(2*H*)-pyridazinones in syntheses of fungicides and herbicides: 05JHC427.
Cyclizations to new azolopyridazines and related ring systems: 05JHC421.
Functionalization and synthetic application of pyridazin-3(2*H*)-ones: 05JHC353.
Palladium-catalyzed cross-coupling reactions on pyridazine moieties: 06SL3185.
Palladium-catalyzed reactions on 1,2-diazines: 06COC377.
Pyridazine derivatives as novel acyl-CoA:cholesterol acyltransferase (ACAT) inhibitors: 05JHC395.
Nucleophilic substitution of hydrogen in pyrimido[4,5-*c*]pyridazine-5,7(6*H*,8*H*)-diones: 05JHC375.
Synthesis of biologically active pyridazinoquinoxalines: 05JHC387.
Synthesis of functionalized compounds containing pyridazine and related moieties: 05JHC361.
Synthesis and heterocyclization of 3-alkynyl-6,8-dimethylpyrimido[4,5-*c*]pyridazine-5,7(6*H*,8*H*)-diones and their lumazine analogs: 05JHC413.

Use of polyfunctionalized pyridazines as reactive species for building chemical diversity: 06COC277.

6.3.1.2 1,3-Heterocycles: monocyclic pyrimidines and hydropyrimidines (except pyrimidine nucleoside bases and nucleosides)

Kost–Sagitullin rearrangement and other isomerization recyclization of pyriymidines: 05KGS1445.

Novel ring closure and ring opening reactions of 2-aminopyrimidines into imidazo[1,2-*c*]pyrimidines and guanidines, respectively: 06ARK(7)5.

Pyrimidine chemistry in crop protection: 06H(68)561.

Synthesis of biologically interesting 6-substituted pyrimidines and 4(3*H*)-pyrimidinones: 06ARK(7)452.

Synthesis of uniform, nonnatural oligomers, particularly, oligo(pyridine–pyrimidine)s: 06SL1793.

6.3.1.3 Annulated pyrimidines (except purines, pteridines, and flavins)

Chemistry of thienopyrimidines: 06AHC(92)83.

Coenzyme 5,10-methylene and methenyltetrahydrofolate models in organic synthesis: 06AHC(91)159.

Photophysics and photochemistry of pterins in aqueous solution: 06ACR395.

Progress in synthesis of pyridopyrimidine analogs: 05CJO1530.

Recent progress in the synthesis of thiazolopyrimidine analogs: 07CJO166.

6.3.1.4 Pyrimidine nucleoside bases and purines

Artificial nucleobases for hole transport: 07Y204.

2,4-Difluorotoluene as a thymine analog in shape and DNA replication without hydrogen bonds: 06CC3665.

Fused six- and more-membered heterocyclo-purinediones, -purinones, and -purineimines: 05AHC(88)175.

Isolation, characterization, and independent synthesis of guanine oxidation products: 06EJO1351.

Synthetic strategies toward O(6)-substituted guanine derivatives and their use in medicine: 05COS215.

6.3.1.5 Nucleotides and nucleosides

Application of microwaves in the synthesis of nucleoside analogs: 07CJO449.

Chemistry and properties of nucleoside and oligonucleoside boranophosphates: 07CRV4746.

Chiral synthesis of heterosubstituted nucleoside analogs from noncarbohydrate precursors: 07COC1017.

Conformationally locked nucleotides and their analogs: 06Y681.

Coordination chemistry of polyamines and their interactions in ternary systems including metal ions, nucleosides, and nucleotides: 05CCR(249)2335.

Creation of low toxic reverse-transcriptase inhibitory nucleosides that prevent the emergence of drug-resistant HIV variants: 06Y716.

Cycloadditions as a method for oligonucleotide conjugation: 06COS9.

cycloSal Phosphates as chemical trojan horses for intracellular nucleotide and glycosylmonophosphate: 06EJO1081.

Development and applications of fluorescent oligonucleotides: 06COC491.

Four hydrogen-bonding motifs in oligonucleotides: 06ARK(7)326.

Metathesis strategy in nucleoside chemistry: 05T7067.

Molecular design of polyamines and related compounds for controlled DNA folding transition: 06Y1122.

Molecular mechanisms of mammalian global genome nucleotide excision repair: 06CRV253.

Noncovalent interactions in adducts of Pt drugs with nucleobases in nucleotides: 06CCR(250)1315.

Novel synthetic methods for manufacturing 2′-deoxynucleosides: 05Y594.

Nucleobases as supramolecular motifs: 05CSR9.

Nucleoside chemistry in crop protection (synthetic issues, mode of action, and biological efficacy of nucleoside classes active as herbicides, fungicides, and insecticides are presented): 05H(65)667.

Nucleoside monomers with the DNA base replaced by fluorescent hydrocarbons and heterocycles, and their assembly into DNA-like molecules in which the all bases are fluorescent: 06OBC4265.

Nucleoside 5-triphosphates: self-association, acid–base, and metal ion-binding properties: 05CSR875.

An overview of diazine nucleoside analogs: 06COC333.

Palladium-catalyzed C–N and C–C cross-couplings as versatile avenues for modifications of purine 2′-deoxynucleosides: 02JOM(653)234.

Phosphate mimic of nucleotides, influences on the ribofuranose conformations: 06H(67)823.

Progress in synthesis, fluorescent, and biochemical properties of etheno-substituted purine nucleoside derivatives: 06CJO1457.

Prokaryotic nucleotide excision repair: 06CRV233.

Recent developments in mass spectrometry for the characterization of nucleosides, nucleotides, oligonucleotides, and nucleic acids: 05CRV1869.

Recent developments in the chemistry of nucleosides: 07PHC27.

Recent highlights in modified oligonucleotide chemistry: 07OBC3260.

Supramolecular architectures generated by self-assembly of guanosine derivatives: 07CSR296.

Synthesis and properties of oligonucleotides with acylamido substituents: 07SL1.

Synthesis and properties of *psico*-nucleosides: 06OPP101.

Synthesis of 7-deazapurine (pyrrolo[2,3-*d*]pyrimidine) 2′-deoxyribonucleosides: 07COC427.

Synthesis of biologically important nucleoside analogs by Pd-catalyzed C–N bond-formation: 05COS83.

Synthesis of 2′,3′-didehydro-2′,3′-dideoxynucleosides having variations at either or both of the 2′- and 3′-positions: 06T9085.

Synthesis of 2′,3′-didehydro-2′,3′-dideoxynucleosides *via* nucleoside route: 06COS261.

Synthesis of modified purine nucleosides and related compounds mediated by adenosine deaminase (ADA) and adenylate deaminase (AMPDA): 05S509.

50 Years of chemical synthesis of oligo- and poly-nucleotides: 05OBC3851.

6.3.1.6 Nucleic acids

The basic concept of “click chemistry” as a new synthetic method and its applications in DNA research: 06CJO271.

Bio-inspired programmable self-assembly on DNA templates: 06CL694.

The chemical biology that controls DNA function and structure (mainly, the sequence and conformation specific chemical reactions that occur in DNA): 05Y1016, 07BCJ823.

Conformationally constrained PNA analogs and DNA/RNA binding selectivity: 05ACR404.

Covalent bonding of modified nucleic acids with proteins in study of specific protein–nucleic interactions: 05UK84.

Direct reversal of DNA alkylation damage: 06CRV215.

DNA-based molecular design of nanobiomaterials: 05CL1206.

DNA-binding properties of anticancer active dinuclear Rh, Re, and Ru compounds: 05ACR146.

DNA-metal base pairs: 07AG(E)6226.

DNA nanomachines and nanostructures involving quadruplexes: 06OBC3383.

DNA-programmed assembly of nanostructures: 05OBC4023.

DNA–protein interactions, bionanotechnology, and molecular recognition: 05AG(E)1166.

DNA and RNA synthesis: 05CRV593.

Elucidating DNA damage and repair processes by independently generating reactive and metastable intermediates: 07OBC18.

Functionalization of PNAs and novel strategy for active control of DNA recognition by external factors using peptide ribonucleic acids: 05Y63.

Lone pair–aromatic interactions in nucleic acids: 07ACR197.

Metal complexes of phenanthroline, bipyridine, terpyridine, dipyridophenazine, or porphyrins as agents for the control of DNA structure: 07CSR471.

The metal-conjugated PNA challenge: 05SL1984.

Metal-containing nucleic acids as new biomaterials: 07COC355.

Molecular design, chemical synthesis, and biological carbohydrate-intercalator hybrids activity of artificial and DNA photocleaving: 05Y325.

NMR structures of damaged DNA: 06CRV607.

Noncovalent interactions in adducts of platinum drugs with nucleobases in DNA: 06CCR(250)1315.

Perspectives in chemistry and therapeutic applications of locked nucleic acid: 07CRV4672.

Photochemical DNA manipulation and DNA analysis by photoresponsive artificial DNA: 07Y709.

Preparation and properties of mRNA 5′-cap structure: 05COC999.

Role of modified nucleosides in the translation function of tRNAs from extreme thermophilic bacteria and animal mitochondria: 07BCJ1253.

The structure of homo-DNA ((4,6)-linked oligo-2,3-dideoxy-D-glucopyranose nucleic acid): 07CSR31.

Supramolecular DNA recognition: 07CSR280.

Synthesis and properties of size-expanded DNAs: 07ACR141.

Synthesis, modifications, and applications of PNAs: 05CJO254.

Synthesis of ferrocenyl deoxyribonucleic acids (DNAs) and their application to electrochemical gene detection: 06Y208.

Synthesis of highly functional nucleic acids and their application to DNA technology: 05BCJ2083.

6.3.1.7 1,4-Heterocycles: pyrazines and hydropyrazines

Biological activity and synthesis of diketopiperazines: 07T9923.

Biologically active pyrazines of natural and synthetic origin: 06CLY959.

3,5-Dihalo-2(1*H*)-pyrazinones as versatile scaffolds in organic synthesis: 06S2799.

Industrial-scale palladium-catalyzed coupling of aryl halides and amines, particularly, in synthesis of arylpiperazines: 06ASC23.

6.3.1.8 Annulated pyrazines

Coenzyme 5,10-methylene and methenyltetrahydrofolate models in organic synthesis: 06AHC(91)159.

Raman scattering and photophysics in spin-state-labile d^6 Ru(II)- and Fe(II)dipyridophenazine complexes: 06CCR(250)1696.

Synthesis of quinazolinones and quinazolines: 05T10153.

6.3.2 One nitrogen and one oxygen atom

Advances in the synthesis of 2*H*-1,4-benzoxazin-3(4*H*)-ones and 3,4-dihydro-2*H*-1,4-benzoxazines: 05T7325.

Benzophenoxazine-based fluorescent dyes for labeling biomolecules: 06T11021.
Morpholine and its derivatives (general monograph): 07MI2.

6.3.3 One nitrogen and one sulfur atom

Recent progress in the chemistry of 2,1-benzothiazines: 08PHC1.
Retrosynthetic approaches to the synthesis of phenothiazines: 06AHC(90)205.

6.3.4 Two oxygen atoms

Synthesis of 2,3-dihydro-2-ylidene-1,4-benzodioxins: 05COC377.

6.4 Three heteroatoms

6.4.1 Three nitrogen atoms

Recent applications of 2,4,6-trichloro-1,3,5-triazine and its derivatives in organic synthesis: 06T9507.
Synthesis and biological activity of 1,2,4-triazolo[1,5-*a*][1,3,5]triazines (5-azapurines): 06H(68)1723.

6.5 Four heteroatoms

The magnetochemistry of verdazyl radical-based materials: 05CCR(249) 2612.
Recent advances and applications in 1,2,4,5-tetrazine chemistry: 07T4199.

7. RINGS WITH MORE THAN SIX MEMBERS

7.1 General

Formation of 7- to 12-membered heterocycles by diene and enyne metathesis: 07T3919.

7.2 Seven-membered rings

Applications and synthesis of the antiepileptic drug oxcarbazepine and related structures: 07COC1385.
Reactions of functionalized alkoxyethylenes with nucleophilic reagents in synthesis of seven-membered heterocycles: 06ZOR167.
Rules for predicting the conformational behavior of saturated seven-membered heterocycles: 05ARK(6)88.

7.2.1 One heteroatom

Application of Baeyer–Villiger monooxygenase in synthesis of seven-membered lactones: 05CJO1198.
Biomimetic synthesis of *trans*,*syn*,*trans*-fused polyoxepanes and polypyrans: 06SL1816.

Construction of fused polycyclic ethers (particularly, ciguatoxins and gambieric acids) by strategies involving ring-closing metathesis: 06CC3571.

Development and industrialization of the process of vapor-phase Beckmann rearrangement of cyclohexanone oxime: 07BCJ1280.

Methods for construction of isoindolo[1,2]-fused benzazepines: 06KGS963.

Recent developments in the synthesis of oxepines: 06T9301.

7.2.2 Two heteroatoms

1,4-Benzodiazepin-2,5-dione as synthetic nonpeptide mimetic of α-helices: 07CSR326.

5-Methyl-4,5,6,7-tetrahydroimidazo[4,5,1-*jk*](1,4)benzodiazepin-2(1*H*)-one as nonnucleoside inhibitors of HIV-1 reverse transcriptase: 05JMC1689.

7.2.3 Three and more heteroatoms

Design, synthesis, and applications of 3-aza-6,8-dioxabicyclo[3.2.1] octane-based scaffolds for peptidomimetic chemistry: 06SL331.

1,2,3,4,5-Pentathiepines and 1,2,3,4,5-pentathiepanes: 07UK219.

7.3 Medium rings

Synthesis and biological perspectives of benzannulated medium ring heterocycles: 07H(71)1011.

Chemistry of 2,8-dimethyl-6*H*,12*H*-5,11-methanodibenzo[*b*,*f*]diazocin (Tröger's base): 07AHC(93)1.

The eight-membered *N*-hydroxybenzazocinone key intermediate in total syntheses of antitumor antibiotic FR900482, its derivatives and analogs: 07Y470.

Methods for construction of isoindolo[1,2]-fused benzazocines: 06KGS963.

7.4 Large rings

7.4.1 General problems

7.4.1.1 Structure, stereochemistry, reactivity, design

Anion templated assembly of mechanically interlocked structures: 07CSR211.

Chemistry and biology of the 23-membered macrocyclic streptogramin A antibiotics: 07MRO159.

Compartmental macrocyclic Schiff bases and related polyamine derivatives: 07CCR1311.

Mass spectrometric studies of noncovalent compounds, particularly, catenanes: 06OBC2825.

Molecular recognition *via* base-pairing in self-assembled macrocyclic and high-order ensemble synthesis, supramolecular polymer preparation, molecular cage construction, and energy and electron transfer modeling: 07CSR314.

A survey of transannular interactions in some macroheterocyclic compounds: 05COC521.

7.4.1.2 Synthesis

Advances in synthesis and research of macrocyclic compounds containing tetrathiafulvalene: 07CJO1220.

Functionalized alkoxyethylenes in synthesis of macroheterocycles: 06ZOR167.

Macrocyclization by ring-closing metathesis in the total synthesis of natural products: 06AG(E)6086.

Metal-free methods in the synthesis of macrocyclic Schiff bases: 07CRV46.

Progress in study of novel supramolecular host – thiacalixarene: 07CJO907.

Pyridine-containing macrocycles *via* comediated [2 + 2 + 2] cycloadditions of α,ω-bis-alkynes: 07ARK(12)7.

Ring-closing metathesis as a basis for the construction of macroheterocycles: 06AG(E)2664.

Syntheses, structures, and interactions of heterocalixarenes: 05AHC(89) 120, 06ARK(9)17.

Synthesis and antibacterial activity of macrolides and ketolides related to erythromycin: 06T3171.

Synthesis and properties of calixarene–porphyrin conjugates: 05CJO375.

Synthesis and properties of dibenzotetraaza macroheterocycles: 05KGS1763.

Synthesis, isomerization, and functions of cyclophanes with azobenzene units in the main frame: 05Y370.

Synthesis of arylenephosphamacrocycles using three- and pentavalent phoshorus compounds: 07UK362.

Synthesis of azomethyne macrocycles by condensation of dicarbonyl compounds with diamines without the use of metal ions as template agents: 07UK843.

Synthesis of macrocyclic compounds including natural products by ring closing metathesis: 07COC1339.

Synthesis of macrosphelides, natural potent cell-cell adhesion inhibitors having novel three ester linkages in their 16-membered macrocyclic skeleton: 05H(65)1741, 05Y140.

The synthesis of 16-membered macrocyclic depsipeptides, cryptophycins: 06S3747.

Synthesis of rotaxanes and catenanes using the Cu(I)-catalyzed 1,3-dipolar cycloaddition: 07BCJ1856.
Synthesis of water-soluble azacyclophane hosts as a guest-delivering carrier: 06Y1041.
Synthetic study on globomycins (19-membered depsipeptides): 05Y51.
Total synthesis of natural 8- and 9-membered lactones: 07CRV239.

7.4.1.3 Applications

Amide-based macrocyclic ligands for anion coordination: 06AG(E)7882.
Application of crown compounds bearing photochromic moiety for metal ion separation and determination: 06Y61.
Chemical sensors based on amplifying fluorescent conjugated polymers functionalized, particularly, by crowns or cyclophanes: 07CRV1339.
Conjugation of 1,4,7,10-tetraazacyclododecane-1,4,7,10-tetraacetic acid (DOTA) and its derivatives to peptides, applications, and future prospects of the conjugates: 07MRO281.
Emerging host–guest chemistry of synthetic nanotubes (nanotubes containing polypyridines and calixcrown tubes): 07CC3891.
Macro(hetero)cycles as precursors for organic nanotubes: 07COS59.
Photo-driven molecular devices (light-powered bistable rotaxanes and catenanes): 07CSR77.
Synthesis of (*E*,*E*,*E*)-1,6,11-tris(arenesulfonyl)-1,6,11-triazacyclopentadeca-3,8,13-trienes and use of their Pd(0) complexes as catalysts in carbon–carbon bond-forming reactions: 04JOM(689)3669.
Synthetic molecular motors and mechanical machines (in particular, supramolecular complexes of porphyrins, crown ethers, cavitands, rotaxanes, catenanes, and other macroheterocycles): 07AG(E)72.
Transition metal complexes of some azamacrocycles and their use in molecular recognition: 07COS390.

7.4.2 Crown ethers and related compounds

Advances in the synthesis of *N*-substituted aza crown ethers and in their applications for extraction and selective complexation of metal ions: 05CJO619.
Phenyl-substituted and benzannulated aza crown compounds with nitrogen atom conjugated with benzene ring: 05UK503.
Recent advances in asymmetric phase-transfer catalysis using chiral nonracemic onium salts and crown ethers: 07AG(E)4222.
Stepwise two-electron-transfer reduction of cyclic ethers and lactones with alkalide K^-, $K^+(15\text{-crown-}5)_2$: 07COC1126.

7.4.3 Miscellaneous macroheterocycles

Amino acid derived macrocycles: 06AG(E)1364.
Anion-templated assembly of interpenetrated and interlocked structures: 06CC2105.

Azolium cyclophanes: 06MRO333.
Calixarene- and cavitand-based capsules: 06ARK(5)137.
Calixarenes as platforms for the construction of multimetallic complexes: 04JOM(689)4125.
Calixarenes enhanced as dendrimers: 06MRO219.
Chemistry and chirality of molecular knots and their assemblies: 05AG(E)1456.
The chemistry of calixpyrroles: 07H(71)1261.
Chemistry of cavitands: 07MRO125.
Conformation-directed macrocyclization reactions: 05EJO1949.
N-Confused calix[4]pyrroles: 06CCR(250)2929.
The cucurbit[n]uril family: 05AG(E)4844.
Elastic tubes built by cyclic chalcogenaalkynes as flexible hosts: 05CL126.
Functionalized cucurbiturils and their applications: 07CSR267.
Heteroatom-containing, carbon-bridged calix[4]arene, thiacalix[4]arene, and sulfonamide bridged calix[4]arene: 07MRO143.
Interlocked molecules containing quaternary azaaromatic moieties: 05H(65)1713.
Macroheterocyclic ligands in coordination of anions: 05ACR671.
Metal complexes of polyaza and polyoxaaza Schiff base macrocycles: 05CCR(249)2156.
Phosphorus-containing chiral cyclodextrins, calixarenes, and cyclophanes: 06COC2307.
Progress of calixarenes used as anion receptor: 06CJO419.
Progress on supramolecular building blocks of resorcinarenes: 06CJO431.
Recent progress on switchable rotaxanes: 06CSR361.
Reversible guest exchange mechanisms in supramolecular host–guest assemblies (hemicarcerands, cucurbiturils, hydrogen-bonded assemblies, and metal-ligand assemblies): 07CSR161.
Rotaxanes as ligands (from molecules to materials): 07CSR226.
Shape-persistent macroheterocycles: 06AG(E)4416.
Structure and binding properties of water-soluble cavitands and capsules: 07CSR93.
Sulfur-containing macrocyclic compounds as complexing reagents and extragents for transition and heavy metals: 05RKZ(6)47.
Thiacalixarenes: 06CRV5291.
Topologically novel multiple rotaxanes and catenanes based on tetraurea calix[4]arenas: 06CC2941.
Transition metal-complexed catenanes and rotaxanes as light-driven molecular machines prototypes: 05CL742.

8. HETEROCYCLES CONTAINING UNUSUAL HETEROATOMS

8.1 General

Element–element additions to unsaturated carbon–carbon bonds catalyzed by transition metal complexes in synthesis and reactions of Si-, Ge-, and B-heterocycles: 06CRV2320.

Homolytic substitution at higher main group heteroatoms to give Se-, Te-, and P-heterocycles: 06CC4055.

Rather exotic types of cyclic peroxides, heteradioxiranes (Het = N, Si, Ge, Sn, Sb, P, S, Se): 07CRV3247.

8.2 Phosphorus heterocycles

8.2.1 Chemistry of individual classes of P-heterocycles

1,2-Dihydro-, 1,2,3,6-tetrahydro-, and 1,2,3,4,5,6-hexahydrophosphinine 1-oxides: 06COC93.

The chemistry of phosphinines: syntheses, coordination chemistry, and catalysis: 06COC3.

The continuing development of the chemistry of phospholes: 06COC43.

Design and study of phosphocavitands: 05ACR108.

Exciting fields in P-heterocyclic chemistry: 05JHC451.

2-Phospha- and 2,3-oxaphosphabicyclo[2.2.2]octenes – synthesis, fragmentation with release of the bridging P-containing unit, and their use as phosphorylating agents: 06COC79.

Phosphametallocenes (mainly, works of the Prof. Mathey group): 02JOM(646)15.

Phosphorus-containing chiral cyclodextrins, calixarenes, and cyclophanes: 06COC2307.

Progress in five-membered heterocyclic metallocene N-, P-, and As-analogs: 07CJO329.

Recent advances in the chemistry of annulated azaphospholes: 07COC33.

Ring-fused 1,3,2-dioxa-, 1,3,2-oxaza-, and 1,3,2-diazaphosphorinanes: 07COC1610.

Synthesis and properties of 1-phospha-4-silabicyclo[2.2.2]octane derivatives: 06ARK(7)359.

Synthesis, reactivity, and stereochemistry of new phosphorus heterocycles with five- or six-membered rings, particularly, 2,3-dihydro-1,3-oxaphospholes and 1,4,2-oxazaphosphinanes: 05JOM(690)2472.

8.2.2 Structure and stereochemistry

Applications of planar-chiral phosphole derivatives as ligands in asymmetric catalysis: 06ACR853.

Cyclophosphazene-based multisite coordination ligands: 07CCR1045.

8.2.3 Reactivity

2,4-Bis(4-methoxyphenyl)-1,3-dithia-2,4-diphosphetane-2,4-disulfide (Lawesson's reagent) in organic synthesis: 07CRV5210.

8.2.4 Synthesis

Synthesis of arylenephosphamacrocycles using three- and pentavalent phosphorus compounds: 07UK362.

8.3 Boron heterocycles

8.3.1 Chemistry of individual classes of B-heterocycles

Advances in the chemistry of carboranes and metallacarboranes with more than 12 vertices: 07CCR2452.

The chemistry of [1]borametallocenophanes and related compounds: 03JOM(680)31.

Chemistry of boron clusters, synthesis, structure, and application for molecular construction: 07Y320.

Chemistry of the carba-*closo*-dodecaborate(−) anion, $CB_{11}H_{12}^{-}$: 06CRV5208.

The chemistry of the undecaborates: 03JOM(680)301.

Imide- and amide-supported group 5 and 6 metallacarboranes: 02JOM(657)9.

Developments in the chemistry of the nine-vertex monocarbaboranes: 02JOM(657)3.

Metal complexes of monocarbon carboranes bearing C-amino substituents: 04JOM(689)3891.

Poly(cyclodiborazane)s: 03JOM(680)27.

Synthesis, reactivity, and applications of cyclic and heteryl-substituted borinium, borenium, and boronium ions: 05AG(E)5016.

8.3.2 Synthesis

Syntheses and spectroscopic properties of BODIPY dyes and their derivatives (BODIPY is 4,4-difluoro-4-bora-3a,4a-diaza-*s*-indacene): 07CRV4891.

8.3.3 Applications

Advances in the synthetic applications of the oxazaborolidine-mediated asymmetric reduction: 06T7621.

A key role of orbital interaction in the main group element-containing π-electron systems (the systems were synthesized using dibenzoborole as building unit): 05CL2.

Antitumor metal-containing carboranes: 07IZV620.

Development of highly enantioselective oxazaborolidinone catalysts for the reactions of acyclic α,β-unsaturated ketones: 07SL1823.

8.4 Silicon, germanium, tin, and lead heterocycles

8.4.1 Chemistry of individual classes of heterocycles

Chemistry of stable silyl, germyl, and stannyl cations, radicals, and anions, particularly, Si-, Ge-, and Sn-heterocyclic moieties: 07ACR410.

Cross-coupling reactions in the chemistry of silole-containing p-conjugated oligomers and polymers: 02JOM(653)223.

Experimental aspects of aromaticity of group 14 organometallics (Si-, Ge-, and Sn-heterocycles): 07AG(E)6596.

A helmeted dialkylsilylene, 2,2,5,5-tetrakis(trimethylsilyl)silacyclopentane-1,1-diyl: 07BCJ258.

Heterocyclic compounds with a silicon atom and another nonadjacent different heteroatom: 06T7951.

Metallocanes of group 14 elements – silicon and germanium derivatives: 06KGS1777.

Metallocanes of group 14 elements – tin derivatives: 07KGS963.

Nanodimensional organostannoxane molecular assemblies: 07ACR420.

Organosilicon peroxides (including cyclic peroxides): radicals and rearrangements: 07T10385.

Photooxygenation of nonaromatic Si- and Ge-heterocycles: 07COC1053.

Polyhedral oligomeric silsesquioxane nanocomposites for biomedical applications: 05ACR879.

Progress in synthesis and application of silazanes, particularly, cyclosilazanes: 07CJO1358.

Synthesis and properties of 1-phospha-4-silabicyclo[2.2.2]octane derivatives: 06ARK(7)359.

8.4.2 Structure and stereochemistry

Silicon-stereogenic silanes (including cyclic silanes) in asymmetric catalysis: 07SL1629.

Transannular secondary bonding in metallocanes of type $[X(CH_2CH_2Y)_2MRR']$ and $[X(CH_2CH_2Y)_2M'R]$ (M = Ge(IV), Sn (IV), Pb(IV), M′ = As(III), Sb(III), and Bi(III); X = NR′, O, S; Y = O, S): 05CCR(249)859.

8.4.3 Reactivity

Electrochemical oxidation of cyclic polysilanes: 03JOM(685)145.

Organocatalytic ring-opening polymerization of oxadisilacyclohexanes and cyclosiloxanes: 07CRV5813.

8.4.4 Synthesis

A key role of orbital interaction in the main group element-containing π-electron systems (the systems were synthesized using silole or bis-silicon-bridged stilbene as building units): 05CL2.

A bulky silylene Tbt(Mes)Si [Tbt = 2,4,6-$(Me_3Si)_3C_6H_2$, Mes = 2,4,6-$Me_3C_6H_2$] generated under mild conditions in the synthesis of various silacyclic compounds: 07SL2483.

8.5 Selenium and tellurium heterocycles

8.5.1 General sources and topics

Advances in organic tellurium chemistry including Te-heterocycles: 05T1613.

Antiferromagnetic superconductor κ-$(BETS)_2FeBr_4$ [BETS = bis(ethylenedithio)tetraselenafulvalene]: 05BCJ1181.

Selenium heterocycles in synthesis and medicinal biology: 07ARK(6)14.

8.5.2 Chemistry of individual classes of heterocycles

Five-membered heterocycles with vicinal Te and O heteroatoms: 06AHC(92)55.

Heterocyclic tellurathianitrogen compounds: 02JOM(646)80.

Photooxygenation of nonaromatic Te-heterocycles: 07COC1053.

Porphyrazines with annulated strongly electron-withdrawing 1,2,5-selenodiazole rings: 06CCR(250)1530.

Preparations, structures, and reactions of 1-benzoseleno- and 1-benzotelluropyrylium salts: 07MRO105.

Synthesis and use of chalcogenoamides for preparation of selenazoles and selenazines: 07COS15.

Synthesis of selenophenes: 05MRO375.

8.6 Other unusual heterocycles

Benziodoxole-based hypervalent iodine reagents in organic synthesis: 05COS121.

Chemistry of pnictogen(III)-nitrogen ring systems: 07CSR650.

8.6.1 Metallacycles

The anions and dianions of group 14 metalloles: 05CCR(249)765.

Aromatic iridacycles, particularly, iridabenzene, iridafuran, iridathiophene, and iridapyrrole: 07ACR1035.

Azobenzenes and heteroaromatic nitrogen cyclopalladated complexes (palladacycles): 06CCR(250)1373.

Carbon–carbon bond-forming reactions using γ,γ-dialkoxyallylic Zr species and proceeding through the formation of zirconacyclopropane followed by β-elimination of the alkoxy group: 07Y56.

The coordination chemistry and reactivity of group 13 metal(I) heterocycles: 05CCR(249)1857.

DFT study of "all-metal" aromatic compounds: 05CCR(249)2740.

Five-membered metallacycles of Ti and Zr in organometallic chemistry and catalysis: 07CSR719.

Group 13 imido metallanes and their heavier analogs $[RMYR']_n$ (M = Al, Ga, In; Y = N, P, As, Sb): 05CCR(249)2094.

Organic transformations on σ-aryl organometallic complexes, particularly formation of metallocycles and complexes with heterocyclic ligands: 07AG(E)8558.

Palladacycles in catalysis: 04JOM(689)4055.

P,Pd-heterocycles and *N*-heterocyclic carbene Pd complexes as catalysts for CC-coupling reactions: 03JOM(687)229.

The potential of palladacycles: 05CRV2527.

Preparation of heterocyclic compounds *via* Ti- or Zr-metallacycles: 05Y102.

Recent advances in metallabenzene chemistry (Ir-, Os-, Pt-, Ru-heterocycles): 06AG(E)3914.

Rh(I)-catalyzed cyclizations *via* rhodacycle intermediates and its application to the synthesis of (+)-epiglobulol: 07Y183.

Stable five-membered cyclic alkynes, 1-metallacyclopent-3-yne complexes, mainly, 1-zirconacyclopent-3-yne complexes: 07Y347.

Studies of chromium cages and wheels: 05CCR(249)2577.

8.6.2 Metal chelates and related complexes

Biaryl-type bisphosphine ligands, in particular, those with heterocyclic fragments and their application to asymmetric synthesis: 05T5405.

Cationic Rh(I)/BINAP-type bisphosphine complexes as catalysts for chemo-, regio-, and enantioselective [2 + 2 + 2] cycloadditions to afford substituted benzenes, cyclophanes, and *N*-heterocycles: 07SL1977.

Chiral, poly(rare-earth metal) complexes in asymmetric catalysis (examples of reactions with participation or formation of heterocycles): 06AA31.

Complexes of tris(pentafluorophenyl)boron with pyridines, pyrroles, and indoles: 06CCR(250)170.

Coordination chemistry and catalytic applications of functionalized *N*-heterocyclic carbene ligands: 07COC1491.

Development of chemo-, regio-, and enantioselective [2 + 2 + 2] cycloadditions (particularly, to form *N*-heterocycles) catalyzed by cationic rhodium(I)/BINAP-type bisphosphine complexes: 07Y862.

Development of enantioselective reactions, particularly, hetero-Diels–Alder reaction and epoxidation of conjugated enones using chiral lanthanum complexes: 07Y977.

1,1-Di(heteroatom)-functionalized ferrocenes as [N,N], [O,O], and [S,S] chelate ligands in transition metal chemistry: 05CSR584.

Ephedrine derivatives, extraordinary tools for the study of stereogenic centers in tetra- to heptacoordinated complexes: 07CCR1852.

Formation and reactions of osmium–carbon double bonds, particularly, in carbene analogs derived from osmacyclopropene, osmafuran, and osmapyrrole: 07CCR795.

Formazanes and their metal complexes: 06UK980.
Guest-induced assembly of Pd(II)-linked coordination nanotubes using pyridine-based ligands: 07BCJ1473.
Intervalence charge transfer in trinuclear and tetranuclear Fe, Ru, and Os complexes with heterocyclic ligands: 06CRV2270.
Ligand design in multimetallic architectures, in particular, the incorporation of heterocycles and arene cores within the ligands and the use of weak interactions to assist self-assembly processes: 05ACR243.
Luminescent platinum(II) terpyridyl complexes: 07CCR2477.
Luminescent square-planar platinum(II) complexes, particularly with 2,2′-bipyridine or its derivatives having sensing functionalities: 07BCJ287.
N-Heterocyclic carbenes as highly efficient ligands in homogeneous and immobilized metathesis ruthenium catalytic systems: 05ARK(10)206.
Nickel complexes with P,N-chelating ligands in catalytic ethylene dimerization and oligomerization: 05ACR784.
On the nature of the active species in palladium catalyzed Mizoroki–Heck and Suzuki–Miyaura couplings (particularly, Pd complexes with iminopyridine and *N*-heterocyclic carbenes as ligands): 06ASC609.
Optically active metallosalen complexes as catalysts for atom-efficient asymmetric reactions: 05Y478.
Pd–*N*-heterocyclic carbene catalysts for cross-coupling reactions: 06AA97.
Rational syntheses of multinuclear high-spin metal complexes with pyridine or 1,4,7-triazacyclononane-derived ligands: 07BCJ608.
Reversible O_2-binding and activation with dicopper and diiron complexes stabilized by hexapyridine ligands: 07BCJ662.
Ruthenium complexes bearing bidentate Schiff base ligands as efficient catalysts for organic and polymer syntheses: 05CCR(249)3055.
Selective transformation of 1,2-diols based on formation of an activated five-membered intermediate with an appropriate metal ion, particularly, tin and copper: 07Y216.
Self-assembly directed by dinuclear zinc(II) macrocyclic species: 06CCR(250)414.
Symmetry driven self-assembly of metallo-supramolecular architectures in metal complexes with 2,2′-bipyridine and 8-hydroxyquinoline ligands: 07BCJ797.
Synthesis and properties of mono- and oligo-nuclear Ru(II) complexes of tridentate ligands: 06CCR(250)1763.
Tautomerism and different types of coordination of typical chelating ligands with metals: 05UK211.
Tethered bis(8-quinolinolato)chromium(III/II) complexes as asymmetric catalytic redox systems: 07CL1082.
Transition metal and lanthanide cluster complexes constructed with thiacalix[*n*]arene and its derivatives: 07CCR1734.

Transition metal complexes of some azamacrocycles and their use in molecular recognition: 07COS390.

Unconventional mixed-valent complexes of Ru and Os with heterocyclic ligands: 07AG(E)1778.

Uranium complexes of multidentate, for example, pyridine- and pyrrole-derived N-donors: 06CCR(250)816.

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CHAPTER 2

Friedländer Annulation in the Synthesis of Azaheterocyclic Compounds

Dedicated to Professor Dr. Habib Firouzabadi on the occasion of his 67th birthday.

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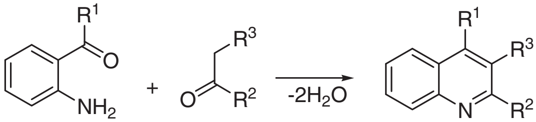
Advances in Heterocyclic Chemistry, Volume 102
ISSN 0065-2725, DOI 10.1016/B978-0-12-385464-3.00002-9

1. INTRODUCTION

Since 1882, when Friedländer reported (1882CB2572, 1892CB1752) the synthesis of quinolines from the condensation of *o*-aminoaryl carbonyl compounds and enolizable methylene groups (Scheme 1), this method has become more and more popular in azaheterocyclic synthesis.



Scheme 1.

This versatile method has opened the door for the preparation of a large number of nitrogen-containing heterocyclic compounds such as quinoline derivatives, pyridines, camptothecins, acridines, tacrine analogs, phenanthrolines, naphthyridines, anthyridines, and anthrazoline derivatives. The biological activity of these heterocyclic compounds has been widely studied. Even though some reference to biological studies will be reported, a complete review of their biological aspects falls outside the scope of this report.

A related field involves the formation of anthrazoline, polyanthrazoline, and polyquinoline derivatives using diketones and di-*o*-aminoaryl-carbonyl compounds. Vast varieties of pyridines, naphthyridines, and phenanthrolines have also been prepared similarly.

The purpose of this review is therefore to summarize the application of Friedländer type methods in a broad range of heterocyclic syntheses.

Although some short reviews have been published (77HC(32)1, 82OR37, 03MI518, 04CJOC366, 05COC141, 05UK739, 05COR141, 08COR1116, 09CJOC1), a comprehensive review article, which demonstrates the broad versatility of the Friedländer synthesis, has not yet been reported.

The basic synthesis of the simplest azaheterocyclic compounds is discussed first along with the basic mechanism of the Friedländer reaction after which a series of more and more complex azaheterocycles are covered.

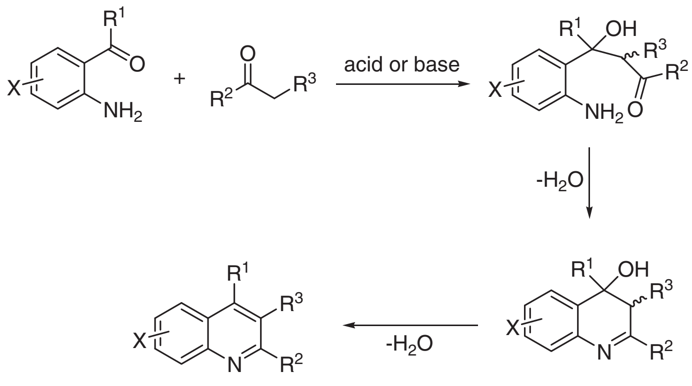
2. QUINOLINE SYNTHESIS

The simplest azaheterocycles that can be synthesized *via* the Friedländer mechanism are the quinolines.

2.1 Friedländer quinoline synthesis

Quinolines and related heterocyclic systems represent an important class of medicinal compounds (96CHEC2, 01OL1109). These ubiquitous substructures are also associated with many biologically active natural products (02NPR742). As a consequence, numerous methods to prepare quinolines have been described (84CHEC, 96CHEC2, 01OL1109, 02JOC3425, 03OL1455, 03OL1605, 03OL1765). One of the most effective and popular methods is the Friedländer synthesis (1882CB2572) that involves an acid- or base-catalyzed carbon–carbon and carbon–nitrogen bond condensation reaction followed by a cyclodehydration between an aromatic 2-aminoaldehyde or ketone with a reactive α-methylene carbonyl compound (Scheme 1).

Classical Friedländer reactions are generally carried out by refluxing either an aqueous or an alcoholic solution of reactants in the presence of a base. Alternatively, the reaction is performed by heating a mixture of the reactants at temperatures ranging from 150 to 220°C in the absence of a catalyst. Typical catalysts for the Friedlander reaction include bases (49CB41, 73JOC449, 74JOC1765, 98FA399, 99WO32100), such as piperidine (02TA227, 4TA3919), sodium ethoxide (86MI1104, 90MI532, 94CJOC626, 95CJOC99, 96JHC841, 98MI40, 01CJSC567, 02CJOC275, 02CJSC154, 07T7654, 08HAC229), sodium *tert*-butoxide (99SC4223, 00BMC2461), KOH (07JME5439), NaOH (94JME2129, 05JME2134), and MeONa (97JME2910); others involve protic acids (66JOC2899, 66JOC3852, 91JHC1301, 99H(50)479, 01BMC2727, 04MI315, 05IJB1649, 06OBC104, 06TL1059), such as *p*-toluenesulfonic acid (PTSA) (80S677, 91JME367), polyphosphoric acid (77JPR589), Tritone B (49JCS2577, 85JOC5782), silica sulfuric acid (SSA) (06M181), citric acid (08H(75)2523), NH_2SO_3H (05TL7249), HCl/microwave (MW) $H_3PMo_{12}O_{40}/SiO_2$ (08CPB1049), $H_3PW_{12}O_{40}$ (09MI1126, 06TL8811), $HClO_4$–SiO_2 (07MI114), oxalic acid (07M1249), proline (08EJO2693), CF_3CO_2H (07SC629); or Lewis acids such as $CeCl_3{\cdot}7H_2O$ (06TL813, 07T892), $FeCl_3$ and $Mg(ClO_4)_2$ (05SL2653, 07SC4071), $MgCl_2$–PTSA (08IJB1160), Bi $(OTf)_3$ (04SL963), $Yb(OTf)_3$ (05TL1647), $Ag_3PW_{12}O_{40}$ (04S2381), metal hydrogen sulfates (08H(79)2513), gold salts (03SL203, 07T2811, 08OL2159), $BiCl_3$ (06MI289), $Nd(NO_3)_3{\cdot}6H_2O$ (06S3825), $GdCl_3{\cdot}6H_2O$ (08JCM679), $SnCl_2$ (05CL314), and $ZnCl_2$ (02JOC9449, 08SL233). Other catalysts include ionic liquids (03JOC9371, 04CCL1170, 04CJC1192, 04JHC1039, 04MI1339, 08MI47, 09H(78)487), diphenyl phosphate (DPP)/MW (03TL255), I_2 (06OBC126, 07MI267), clay/MW (99SC4403), potassium dodecatangestocobaltate/MW (06H(68)549), $NaHSO_4/SiO_2$ (06ARK198, 07M659), $KAl(SO_4)_2{\cdot}12H_2O$–SiO_2 (Alum–SiO_2) (08H(75)947), $KHSO_4$ (06JHC1379), Amberlyst-15 (07MI148), clay K-10 (08CAL250), diphosgene (02JOC7884), trimethylsilyl chloride (07H(71)2397, 07S1214, 07S3891), LiBr (07SC4319), 2,4,6-trichloro-1,3,5-triazine (TCT) (07CL796), $NiCl_2{\cdot}6H_2O$ (08H(75)397), cyanuric chloride (08MI947) and *N*-bromo reagents (09TL1055).



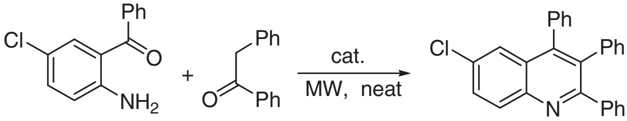
Scheme 2.

The mechanism of the Friedländer synthesis of quinolines from *o*-aminobenzaldehydes and simple aldehydes or ketones was described by Muchowski and Maddox (04CJC461). Both basic and acidic conditions have commonly been used. The first step involves a slow intermolecular aldol condensation of the aldehyde or ketone with the *o*-aminobenzaldehyde (04CJC461). The aldol adduct generated in this manner then undergoes a very rapid cyclization. Subsequent loss of water produces the quinoline derivative (Scheme 2). Both are short-lived intermediates that cannot be detected by TLC, even when deliberately generated by other routes. Overall average yields for the final product from the starting materials range between 60% and 87%. These high yields combined with versatility ensured that the Friedländer reaction became increasingly popular.

The mechanism involves a normal cross aldol condensation. The loss of water in the last step is facilitated by two factors. First, an aromatic product is formed that is thermodynamically much lower in energy. Second, the proton attached to the same carbon as R^3 is in an α-position to a C=N (C=N behaves similar to a carbonyl group) and an OH group. This proton, therefore, is quite acidic leading to facile elimination of water under both acidic (the OH group is protonated, converting it into a good leaving group) and basic conditions (the acidic α-hydrogen is easily abstracted).

Under thermal or base catalytic conditions, an *o*-aminobenzophenone fails to react with simple ketones such as cyclohexanone and deoxybenzoin (67JHC565). Zolfigol et al. have very recently discovered that *o*-aminobenzophenones react with simple ketones such as cyclohexanone and deoxybenzoin in the presence of SSA as a solid protic catalyst under MW conditions to yield new quinolines bearing hindered substituents. To show the powerful effect of SSA in the synthesis of quinolines with bulky substituents, the reaction of deoxybenzoin with 6-chloro-2,3,4-triphenylquinoline **1**

Table 1. Synthesis of 6-chloro-2,3,4-triphenylquinoline **3** from 2-amino-5-chlorobenzophenone **1** and deoxybenzoin **2**



(1) **(2)** **(3)**

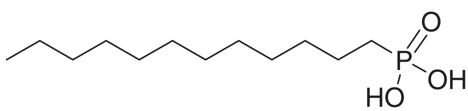
Entry	Catalyst	Equiv. cat./**1**	Time (min)	Yield (%)
1	SSA	0.1	5	85
2	$ZrOCl_2{\cdot}8H_2O$	0.1	10	0
3	$Al(HSO_4)_3$	0.25	10	80
4	$Zr(HSO_4)_4$	0.2	7	77
5	$Zr(NO_3)_4$	0.15	5	0
6	$FeCl_3{\cdot}6H_2O$	0.25	5	0
7	$Fe(NO_3)_3{\cdot}9H_2O$	0.25	5	0
8	$ZnCl_2$	1	5	0
9	$AlCl_3$	1	5	0
10	$CoCl_2{\cdot}6H_2O$	0.50	5	0
11	$Bi(NO_3)_3$	0.25	6	0
12	$NaHSO_4$	1	6	79
13	$AlPW_{12}O_{40}$	0.05	6	0
14	$ZrCl_4$	0.25	6	0

was performed with other general Lewis acids. As shown in Table 1, only hydrogen sulfate salts gave similar yields to SSA (08JICS490).

Recently, a new type of catalyst, namely "Lewis acid-surfactant-combined catalyst (LASC)," has shown high efficiencies. These reactions are promoted in water without organic cosolvents (09T587). LASCs as metal dodecyl sulfates (07MI74) are efficiently employed in the Friedländer quinoline synthesis (06MI253, 07ASC1047).

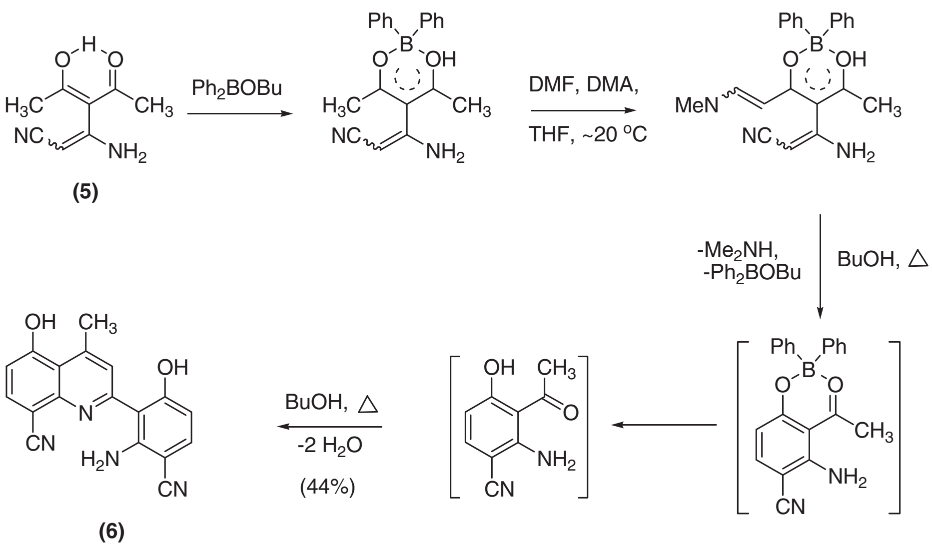
Another green route to multisubstituted quinolines using catalytic amounts of Lewis acids *via* a Friedländer annulation uses $Zr(NO_3)_4$ and $Zr(HSO_4)_4$. They are more efficient than other Lewis acids in the catalyzed condensation of *o*-aminoaryl ketones with ketones or β-diketones (07MI1214).

Ghassamipour and Sardarian have found that a catalytic amount of dodecylphosphonic acid (DPA) **4** (Figure 1) in aqueous media and solvent-free conditions prompted the synthesis in 52–89% yields (09TL514).



(4)

Figure 1.



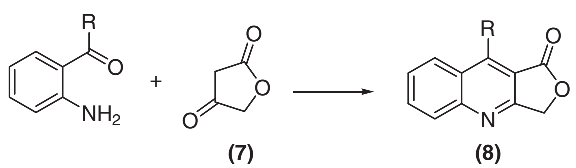
Scheme 3.

More examples where a hydrotropic aqueous medium was used under typical green conditions in Friedländer's quinoline method have been reported (07MI279). 3-Acetyl-2-amino-4-hydroxy-1,3-pentadienecarbonitrite **5** was used for the preparation of 2-(2-amino-3-cyano-6-hydroxyphenyl)-8-cyano-5-hydroxy-4-methyl-quinoline **6** based on the transformation of hydroxyvinyl ketone to the diphenylboron chelate and condensation of the latter with dimethylformamide (DMF) and dimethylacetal (DMA) (Scheme 3) (97RCB122).

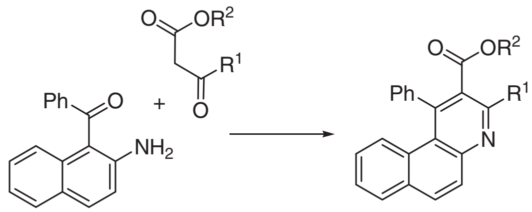
The last step involves the classical Friedländer self-condensation. Typical yields for Scheme 3 are about 44%.

A general method for the preparation of 2-hydroxymethyl-3-quinolinecarboxylic acid lactones **8** (70–100%) from an *o*-amino aromatic carbonyl compound uses tetronic acid **7** (Scheme 4) (58JOC1996).

A series of 2,3-substituted 1-phenylbenzo[*f*]quinolines **10** was synthesized from 1-benzoyl-2-naphthylamine **9** with *β*-keto esters, 50–88% yields (Scheme 5) (68JHC313).



Scheme 4.



Scheme 5.

Walser et al. have employed a combination of the Friedländer method and nucleophilic displacement of aromatic fluorines in the synthesis of quinolinoquinolines and benzochromenoquinolines. 4-(2-Fluorophenyl) quinoline **11** reacts with ethyl acetoacetate to yield **12** (83%) that is reduced to **13** followed by treatment with sodium hydride to give benzochromenoquinoline **14** in 75% yield. However, hydrolysis of **12** to **15** followed by sodium hydride leads to quinolinoquinoline **16** in 77% yield (Scheme 6) (75JHC737).

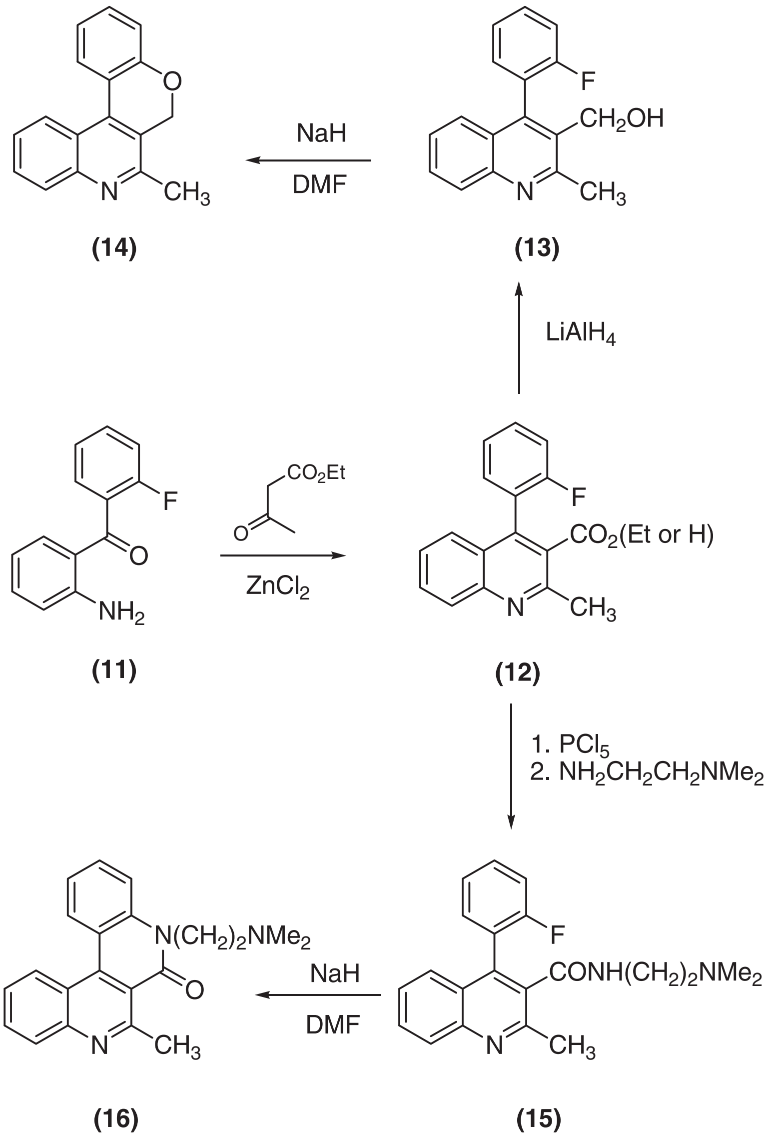
Quinolino[3,2-*d*][1]-benzazepin-6-one **19** (36%), an antitumor compound, was synthesized from [1]benzazepine-2,5-dione **18** with 2-aminobenzaldehyde **17** (55OS56, 93JCE322) in ethanol catalyzed by potassium hydroxide (Scheme 7) (98JME1299).

3-Acetyltropolone **20** reacted with 2-aminobenzaldehyde **17** in the presence of sodium ethoxide to afford 3-(2-quinolyl)tropolone **21** in good yield (Scheme 8) (90CCL101, 92CJOC85, 92MI77, 93MI85, 95JHC1373).

Condensation of **17** with 3-methyl-1-phenylpyrazolin-5-one **22** gave 3-acetylcarbostyril phenylhydrazone **23**, 3-(2-quinolyl)carbostyril **24**, and 3-methyl-1-phenylpyrazolo[4,3-*c*]-1*H*-quinolin-2-one **25** (28RZC325, 28RZC345). When **23** was heated together with nitrobenzene cyclization to 3-methyl-1-phenylpyrazolo[3,4-*b*]quinoline **26** occurred. Tomasik et al. have reported some analogs of **26** from 2-aminobenzaldehyde and 1-methyl-3-phenyl-, 1,3-dimethyl-, 1,3-diphenyl pyrazolin-5-one and pyrazolone in 20–60% yields (Scheme 9) (83JHC1539).

Compound **24** forms when **23** reacts with 2-aminobenzaldehyde **17** in a normal aldol condensation where the methyl group is converted to a nucleophile. In the second step, the amino nitrogen of **17** attacks the imine carbon of **23** with subsequent loss of N-NH-Ph. Alternatively, **23** was condensed with *o*-nitrobenzaldehyde and reduction and thermal cyclization gave pyrazoloquinoline **25** (34JIC427), a procedure similar to that used in the synthesis of an indolinquinoline (60CIL1871).

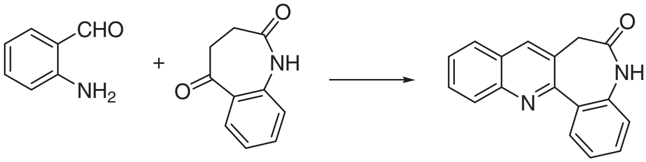
3,8-Diamino-10*H*-quindoline **29** Fluoro-Nissl Green (Scheme 10) employs a Friedländer quinoline synthesis using **27** and **28** to construct the tetracyclic quindoline ring (49% yield) (95JOC3365).



Scheme 6.

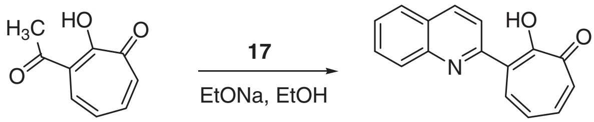
4′-Phenylquinoline **31** was readily obtained (44%) from naltrexone **30** with 2-aminobenzophenone (Scheme 11) (99JME3527).

Optically active, bicyclic ketones were subjected to a Friedländer synthesis with 2-aminobenzaldehyde (**17**) to yield *regio*-isomeric linear or



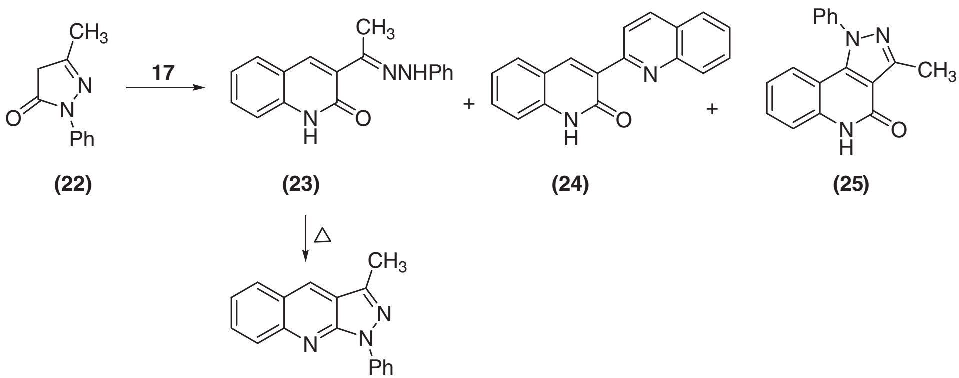
(17) (18) (19)

Scheme 7.



(20) (21)

Scheme 8.



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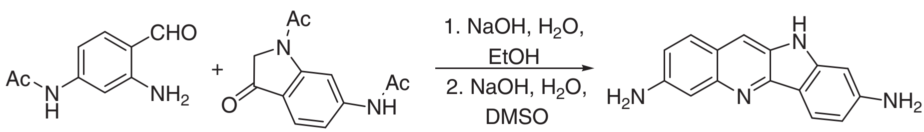
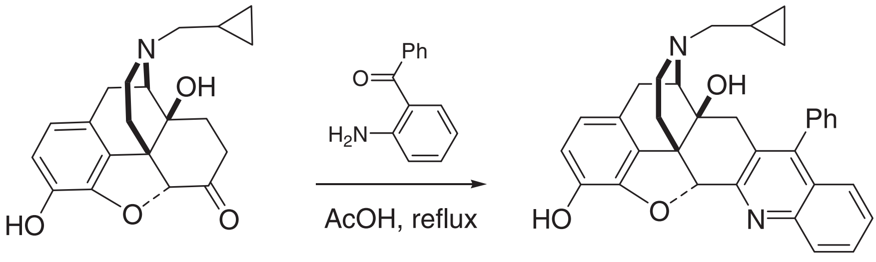
Scheme 9.



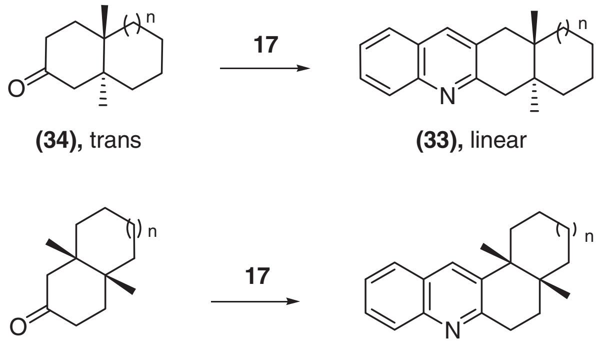
(27) (28) (29)

Scheme 10.



(30) **(31)**

Scheme 11.



(34), trans **(33),** linear

(34), cis **(35),** ,angular

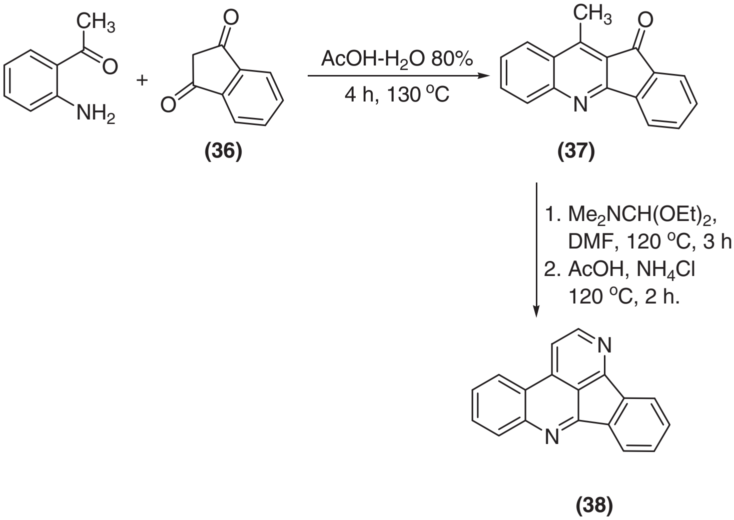
Scheme 12.

angular products. When starting from *trans*-configured ketones **32**, linear **33** were the major isomers in ratios ranging from 76:24 to >98:2. With *cis*-configured ketones **34** angular **35** were predominantly formed, although with lower regioselectivity (Scheme 12) (08EJO1811).

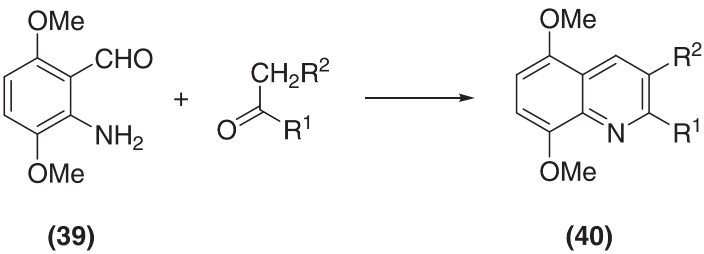
Indan-1,3-dione **36** and *o*-aminoacetophenone condense in the presence of 20% acetic acid in quantitative yields. Product **37** is an intermediate for the preparation of benzo derivatives of eupolauridine **38**, an important alkaloid (Scheme 13) (92SC1773).

Quinoline **40** was synthesized in 66–89% yields (Scheme 14) (08JMC2492). It serves as a key intermediate in the synthesis of heat shock response and angiogenesis inhibition derivatives.

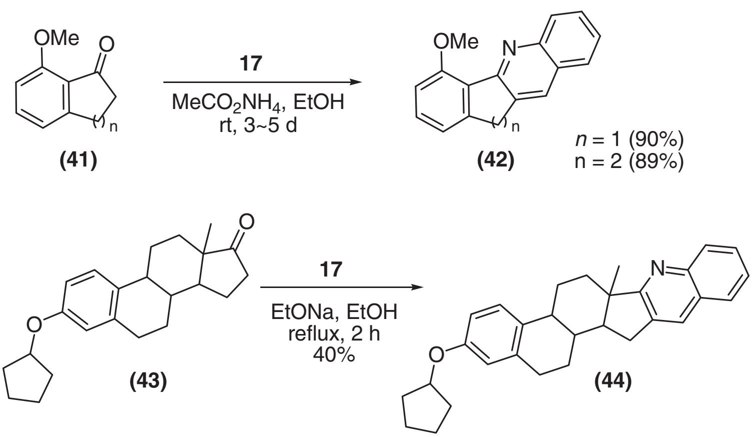
Quinolines **42** (90T2445) and **44** (02CJOC672) are prepared from *o*-aminobenzaldehyde and ketones **41** and polycyclic ketone **43**, respectively (Scheme 15). Typical yields are between 40% and 90%.



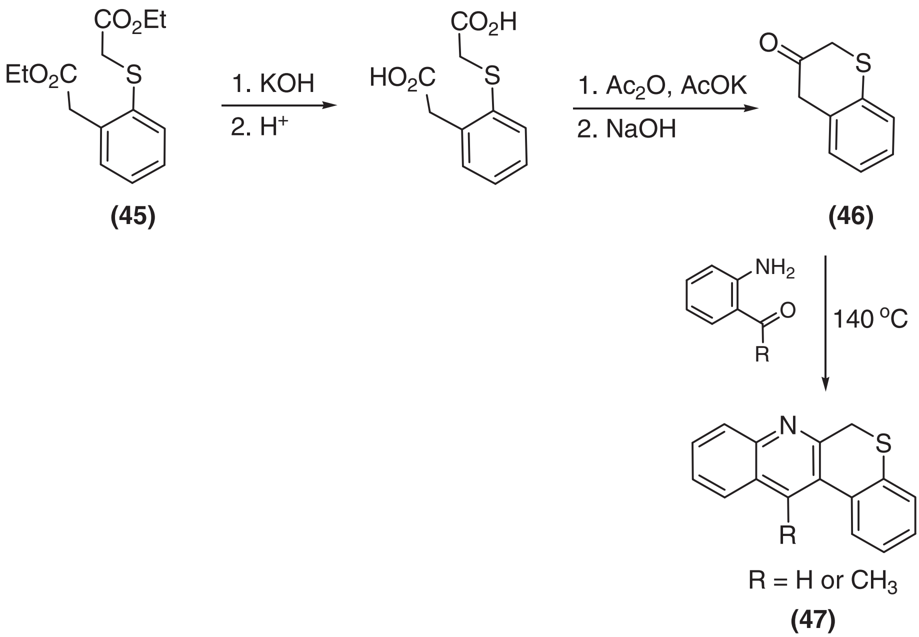
Scheme 13.



Scheme 14.



Scheme 15.



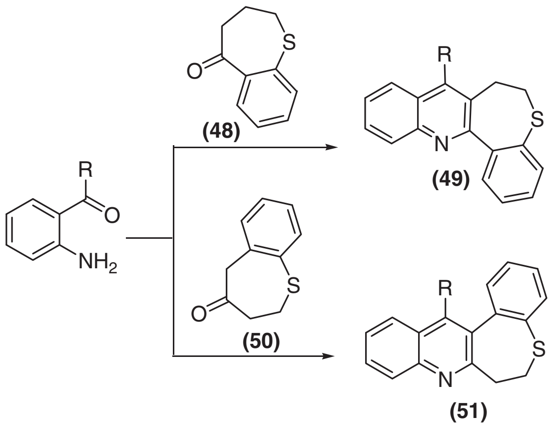
Scheme 16.

3-Thiochromanone **46** was prepared from diester **45** (Scheme 16) and used in the Friedländer quinoline synthesis in high yields to give **47**, analogs of carcinogenic nitrogen polycycles (77T2383).

Gagniant and Deluzarche (46CR808) first reported the preparation of benzothiepino[5,4-*b*]quinoline derivatives **49** and related quinolines *via* the Friedländer synthesis starting from 3,4-dihydro-1-benzothiepin-5 (2*H*)one **48**. The preparation of additional 4-substituted benzothiepino [5,4-*b*]quinolines **49** and preliminary results of their pharmacological properties were subsequently reported by Dudykina and Zagorevskii (67KGS250). Pellicciari et al. used 2,4-dihydro-1-benzothiepin-4(5*H*)-one **50** (77JC(P1)1822) similarly. Quinolines **51** were obtained in good yields R = H and R = CH_3 in 45% and 65% respective yields (Scheme 17) (78JHC927).

An efficient method (55–80% yields) gave 3-substituted carbostyrilquinone derivatives **52** from 5,8-dialkoxyquinolines followed by a cerium ammonium nitrate (CAN) oxidative demethylation (Figure 2) (93H(36) 1387). The synthesis of more complex analogs of **52** is presented in Scheme 18.

Condensation of 3,6-dimethoxy-2-aminobenzaldehyde **39** with a variety of ketones and diketones leads to 5,8-dimethoxyquinoline derivatives



Scheme 17.

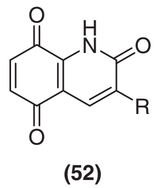
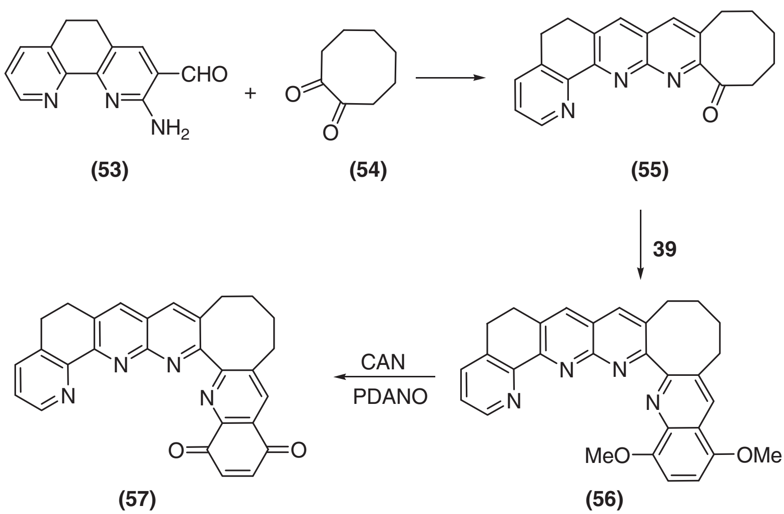


Figure 2.



Scheme 18.

56 that may be oxidized by CAN and pyridine-2,6-dicarboxylic acid *N*-oxide (PDANO) to quinones **57** (Scheme 18) in overall yields between 60% and 65%. Quinones can be incorporated into larger cavities by a selective stepwise Friedländer approach, and the CAN/PDANO oxidation works preferentially for 5,8-dimethoxyquinoline (93JOC1666).

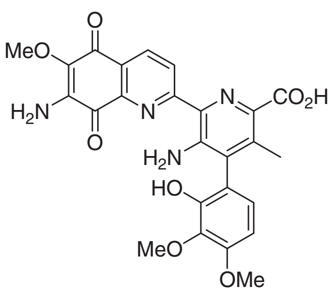
The Friedländer condensation has also been used for the synthesis of antitumor and antibiotic agents such as streptonigrin **58** (77JOC232, 78JOC121), streptonigrone **59** (07JOC8489), lavendamycin analogs **60** (07TL6014, 08T2241), and quinolinequinones **61** (04BMC1667) **62** and **63** (75JHC725, 75JHC731, 79JHC1241) (Figure 3).

Deady et al. used methyl 2-amino-3-formylbenzoate with *o*-methoxy- and *o*-nitro-phenylacetic acids, phenylpyruvic acid, and benzo[*b*]thiophen-2-one to prepare quinoline analogs (99SC4223, 00AJC143).

5-Bromo-2-aminobenzaldehyde **64** and a variety of enolizable ketones **65** afford bidentate and tridentate 6-bromoquinolines. Bromoquinoline **66** (up to 88% yield) could be coupled together to yield biquinoline **67** (Scheme 19) (03OL2251).

2,3′-Biquinoline **69** was prepared from 3-acylquinoline **68** and 2-aminobenzaldehyde **17** in 80% yields (Scheme 20) (21HCA807).

Jahng and Park prepared 2,2′-pyridylbenzo[*h*]quinoline **72** as an *N,N,C*-tridentate ligand from the condensation of 2-acetylpyridine **71** and



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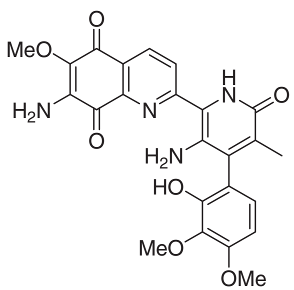

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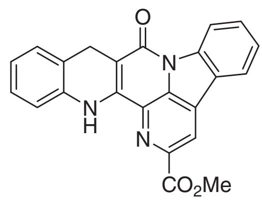

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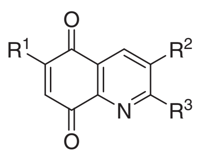
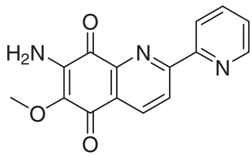


R^1 = heteroaryl, H, OMe
R^2 = H, CH_2OH, CH_2OAc
R^3 = aryl, heteroaryl, H, Cl, Me

(61)



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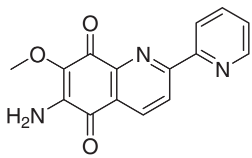
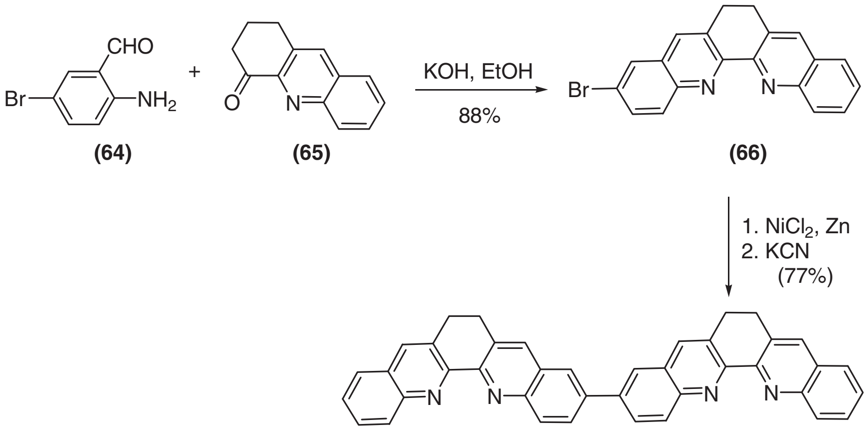


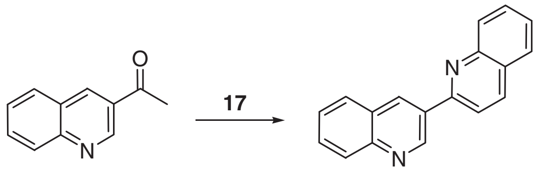
(63)

Figure 3.



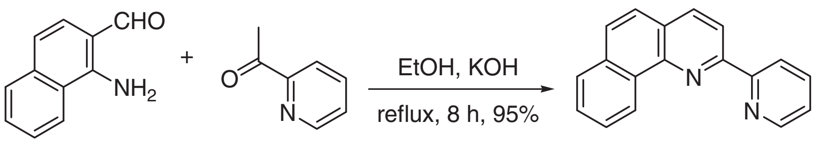
(64) (65) (66) (67)

Scheme 19.



(68) (69)

Scheme 20.



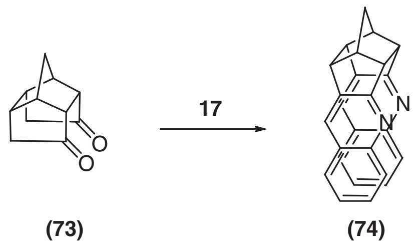
(70) (71) (72)

Scheme 21.

1-aminonaphthalene-2-carbaldehyde **70** in ethanolic KOH in 95% yields (Scheme 21) (99BKC1200).

2.2 Di- and polyquinolines synthesis

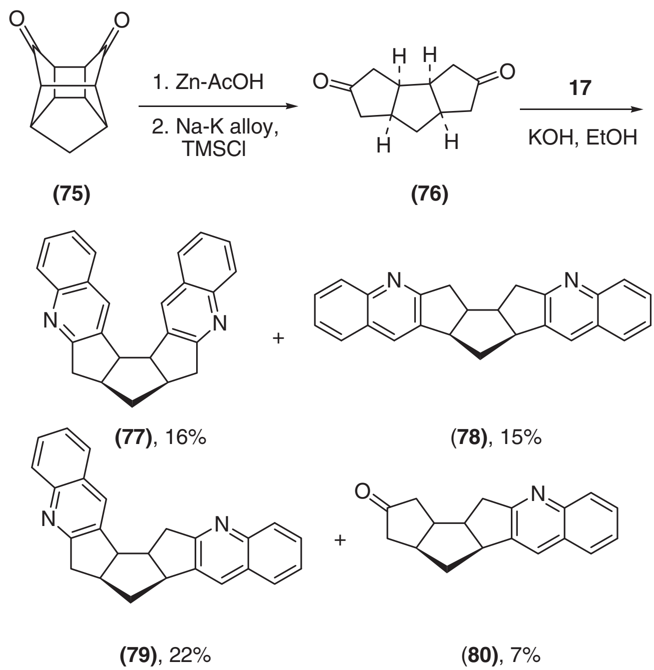
Bis-quinoline **74** was obtained from the condensation of two equivalents of tetracyclo-[6.3.0.0^{4,11}.0^{5,9}]undecane (TCU) **73** and *o*-aminobenzaldehyde **17** in 57% yields (Scheme 22) (87TL3319).



Scheme 22.

Bis-quinolines **77–79** and quinoline **80** were reported utilizing *cis*, *syn*, *cis*-triquinane dione **76**, which are readily accessible from Cookson's dione **75** (64JCS3062) and 2-aminobenzaldehyde in the presence of alcoholic KOH (Scheme 23) (92JC(P1)1747).

The condensation of other diketones **81** (85JOC666, 94JOC823, 97IC5390, 01IC3413), **82**, **83** (01IC5851), **76** (90T5077), **84** (89CC281), **73**, **85** (94JOC823, 01IC3413), **86** (97JC(P2)2099, 08MI839), **87** (08MI297), with *o*-aminobenzaldehyde, 3-amino-2-naphthaldehyde **88** or 1-amino-



Scheme 23.

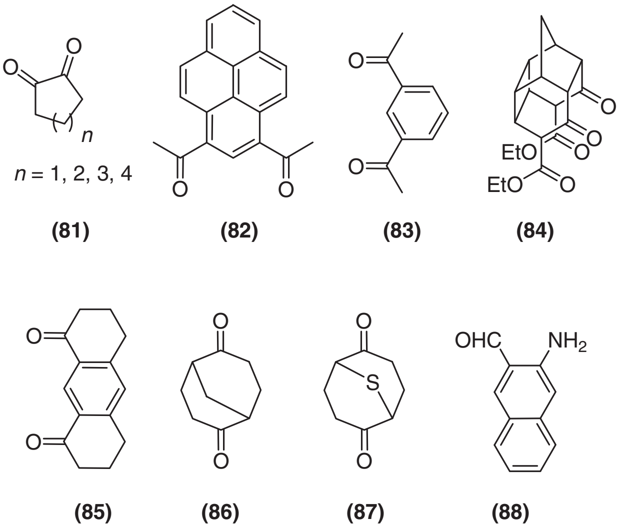
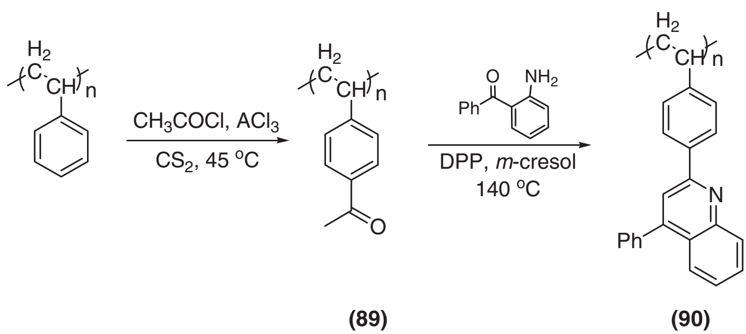


Figure 4.

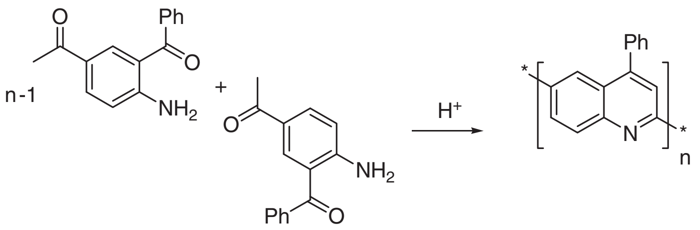
2-naphthaldehyde **70** gave the corresponding diquinolines or dibenzoquinolines (Figure 4).

Electroactive and blue light-emitting poly(vinyl diphenylquinoline) **90** was synthesized in nearly quantitative yield using a simple modification of polystyrene **89**. Polymer **90** has a glass transition temperature of 185°C, is soluble in many organic solvents, and is a weak base comparable to poly (vinylpyridine) (Scheme 24) (01MM6249).

Polyquinolines **92**, developed by Stille et al. in the 1980s, are synthesized by an acid-catalyzed condensation between monomers containing



Scheme 24.



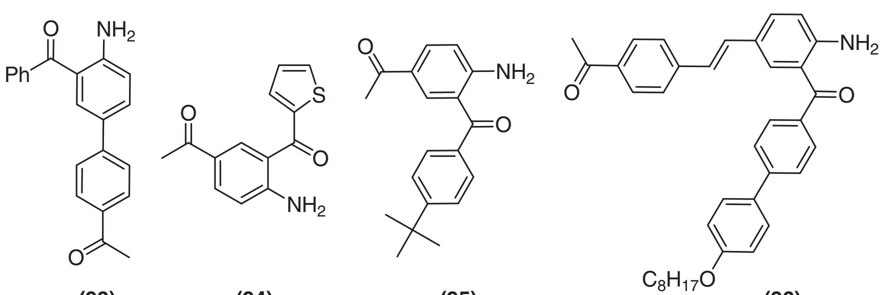
(91) (92)

Scheme 25.

both an *o*-aminoketone and also a ketomethylene **91** (Scheme 25) (81MM870, 93CM633).

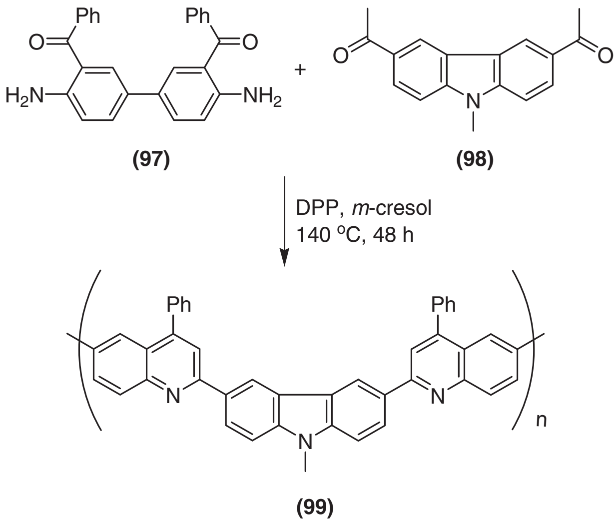
Other monomers containing both an *o*-aminoketone and a ketomethylene such as **93** (87MM258, 93CM633), **94** (87MM258), **95** (98AP52), and **96** (99PB511) were also utilized in polyquinoline synthesis (Figure 5).

The high thermal and oxidative stability, good mechanical and electrical properties, and eminent processability of such polymers make them useful as high-performance polymer materials (76MM505, 76MM512). Based on their optical and electronic properties (99MM7422), these polymers have recently been investigated as potentially useful materials in optoelectronic applications such as chemosensors (02PSA1831), photoconductive properties (92MIi1, 96MI925), electroluminescent (94MI1272, 97CM409, 99JMC2201, 99MM2065, 00MM5880), and nonlinear optical devices (91CM765, 92JPC2837, 99CM2218). Although rigid-rod polyquinolines generally exhibit high strength and possess excellent thermal properties, they have limited solubility in organic solvents, thus making fabrication difficult (92CM95). Attempts were made to enhance the



(93) (94) (95) (96)

Figure 5.



Scheme 26.

solubility of polyquinolines by incorporating flexible ether linkages or cardo units into the polymer backbone (81MM486).

Alternating carbazole–quinoline **99** and phenothiazine–quinoline donor–acceptor conjugated copolymers and a corresponding oligomer were synthesized by Jenekhe et al. and their solution and solid-state photophysics were investigated (Scheme 26) (01MM7315).

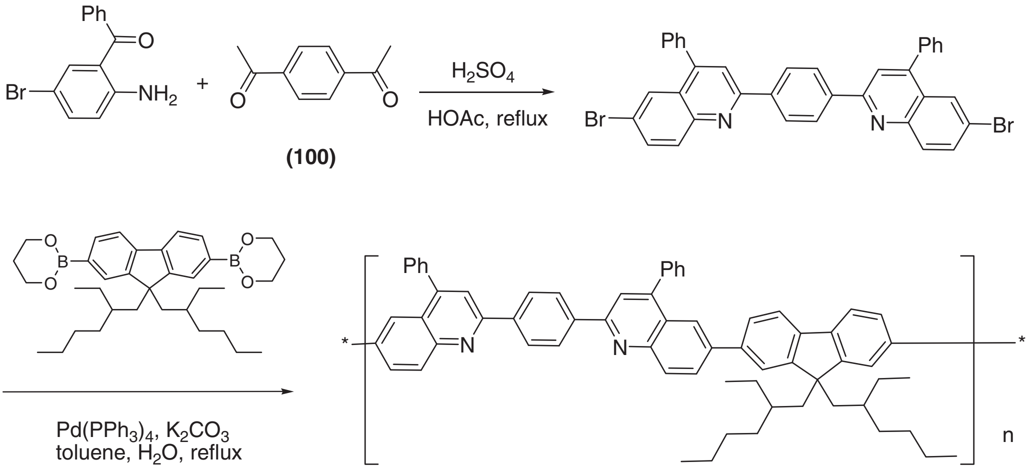
A series of conjugated copolymers **101** having the quinoline moiety in the main chain based on fluorene were synthesized in good yields by a palladium-catalyzed Suzuki coupling reaction (Scheme 27) (02MM2529, 07MCL159).

Many other di-(*o*-aminoketones) and diketones were used for the synthesis of a new class of conjugated polyquinolines (Figure 6, Table 2).

2.3 Modified Friedländer quinoline synthesis

Major modifications to the Friedländer reaction frequently involve different starting materials that deviate from the conventional examples. A one-pot combination of a modified Friedländer annulation and a Knoevenagel condensation have provided 2-styrilquinolines **152** in the presence of 1-methylimidazolium trifluoroacetate ([Hmim]TFA) (Scheme 28) (08TL5366).

3-Tetrazolylquinoline **156** was obtained in three steps involving an acid-catalyzed condensation of aminobenzophenone **153** with 3, 3-dimethoxypropionitrile **154** followed by classical tetrazole chemistry (Scheme 29) (04JME2574).



(100)

(101)

Scheme 27.

The first step involves a modified Friedländer mechanism using a nitrile known to give aldol products. The dimethoxy group is a protected aldehyde (acetal).

Several 4-perfluoroalkylquinolines **159** were prepared from 2-perfluoroalkylanilines **157** (Scheme 30) (96TL4655, 97H(45)2089, 98JFC221, 98T7947, 00JFC281). In an interesting variation where a $–CF_2$-group in an α-position to a benzene ring is converted to the ketal **158** with sodium ethoxide. While normal fluoro-alkanes are extremely resistant to both acids and bases (98JFC221) the reactivity of this $–CF_2$-group originates from the ability of the benzene ring to facilitate EtO^- displacing F^-. The second step also involves basic conditions and the enolate ion attacks the ketal. Imine formation between the amine and the carbonyl group then follows.

The same reactivity of C–F groups in an α-position to an aromatic ring is reported in Scheme 106.

Similar methodology was employed for the synthesis of methyl-1,3-dihydro-2*H*-pyrrolo[3,4-*b*]quinoline-2-carboxylate **162** in 65% yield and methyl-1,2-dihydro-3*H*-pyrrolo[3,4-*b*]quinoline-3-carboxylate **163** in 25% yield from commercial starting materials **160** and **161** (Scheme 31) (07S3319).

Synthesis of 2-aminoquinoline-3-carboxylic acids **165** from 2-tosylaminobenzaldehyde or its morpholine **164** and nitrile analogs has been reported (08RCB418) (Scheme 32).

4-Aminoquinolines **168** containing an $–CF_2CH$–heteroaryl–OH moiety are obtained in moderate yields from the electrochemical reaction of

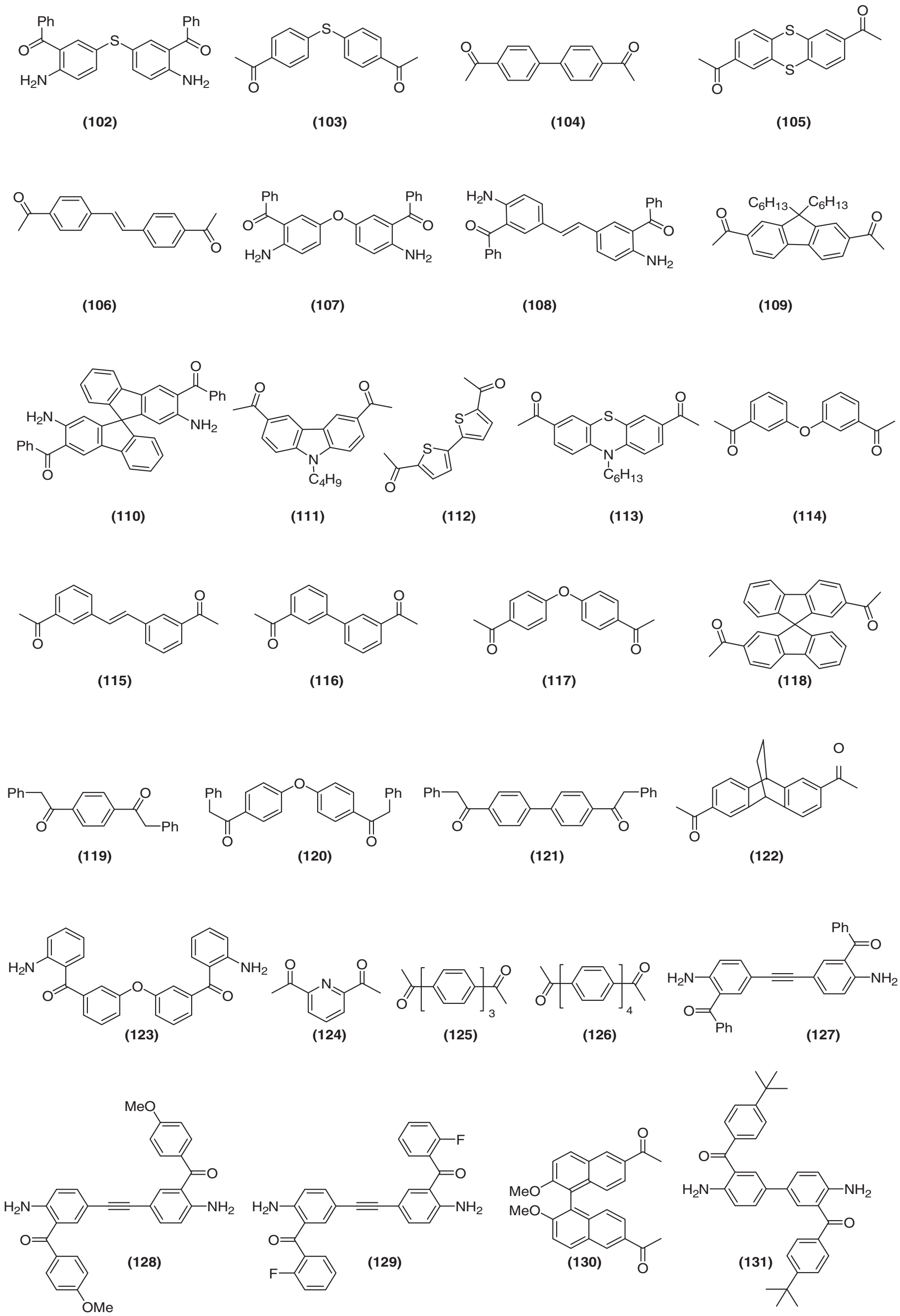

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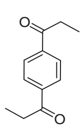
Figure 6.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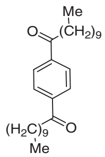
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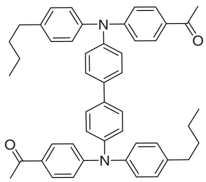
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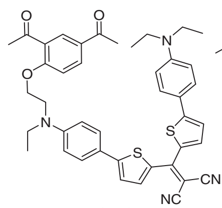
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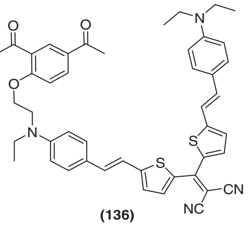
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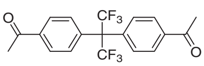
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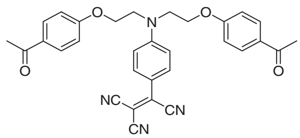
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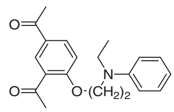
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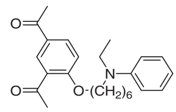
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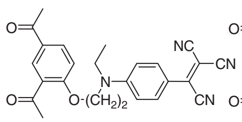
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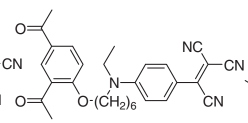
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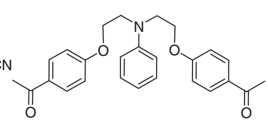
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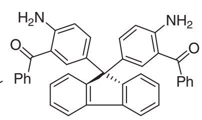
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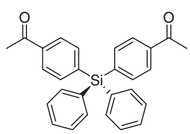
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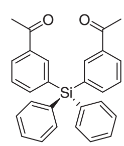
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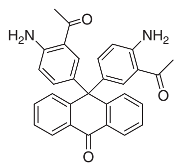
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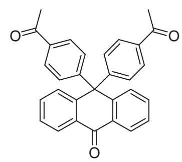
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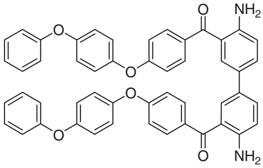
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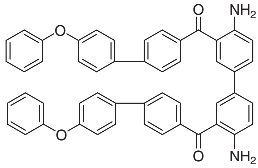

(150)

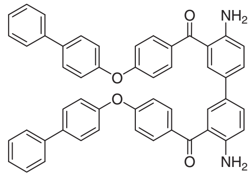
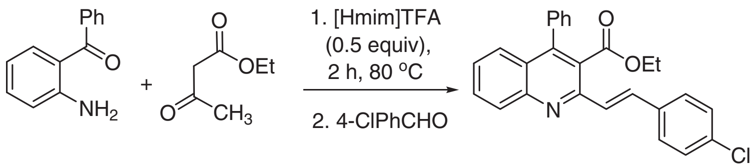

(151)

Figure 6 *(continued).*

Table 2. Monomers used in cross ketone polyquinoline synthesis

o-Aminoarylketone	Diketone	References
102	**103**	(87MM258)
97	**104**	(87MM258)
102	**105**	(87MM258)
97	**106**	(87MM258)
102, **97**, **107**, or **108**	**109**	(00MM5880)
110	**104**, **109**, **111**, **112**, or **113**	(05MM6915)
97, **107**, or **108**	**114**, **115**, or **116**	(85MM321)
110	**117**, or **118**	(02CM682)
107	**118**	(02CM682)
107	**100**, **104**, **117**, **119**, **120**, or **121**	(76MM496)
97 or **107**	**122**	(90MM2418)
123	**83**, **104**, **117**, **119**, **124**, or **132**	(76MM489)
97	**100**, **104**, **117**, **119**, **120**, **121**, **125**, or **126**	(81MM493)
107	**125** or **126**	(81MM493)
127, **128**, or **129**	**112**	(93CM633)
107	**130**	(02MI486)
131	**100**, **132**, or **133**	(98AP52)
97	**134**	(99CM27)
107	**135** or **136**, **137**	(92CM95)
97	**138**, **139**, **140**, **141**, **142**, or **143**	(96CM607)
107 or **144**	**145** or **146**	(01MM3607)
147	**148**	(81MM486)
149, **150**, or **151**	**104**	(85MM2669)



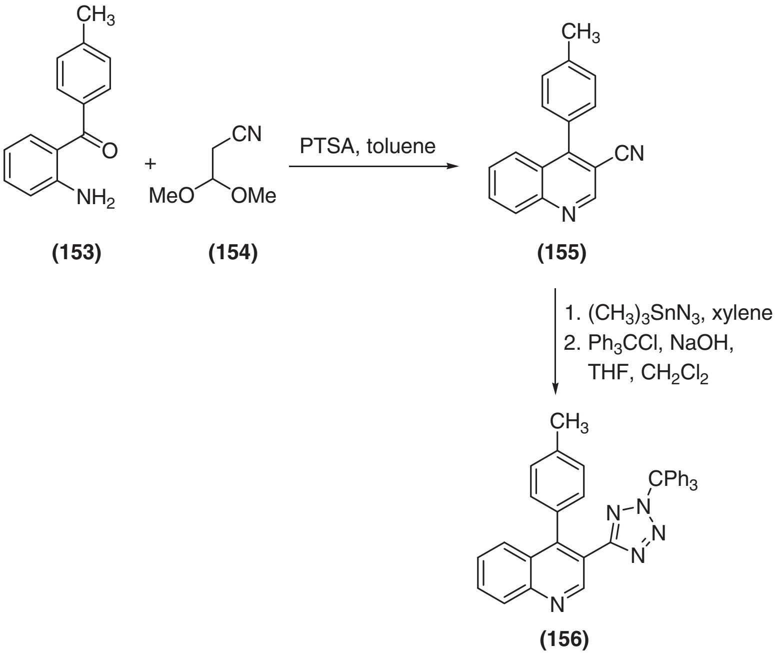
(152), 87%

Scheme 28.

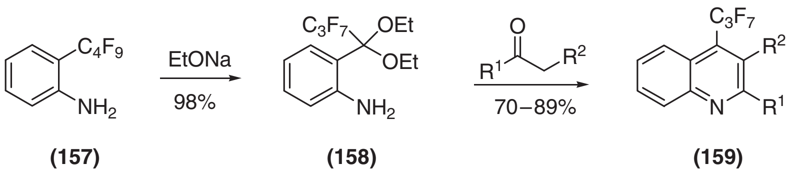
4-amino-3-chlorodifluoroacetyl-2-methoxyquinoline **167** and heteroaryl aldehydes (Scheme 33) (05TL7817).

Ru-grafted hydrotalcite is an excellent multifunctional catalyst for a one-pot synthesis of quinolines from 2-aminobenzyl alcohol **169** and various carbonyl compounds through aerobic oxidation by Ru, followed by an aldol reaction on basic sites of the hydrotalcite (Table 3) (04TL6029).

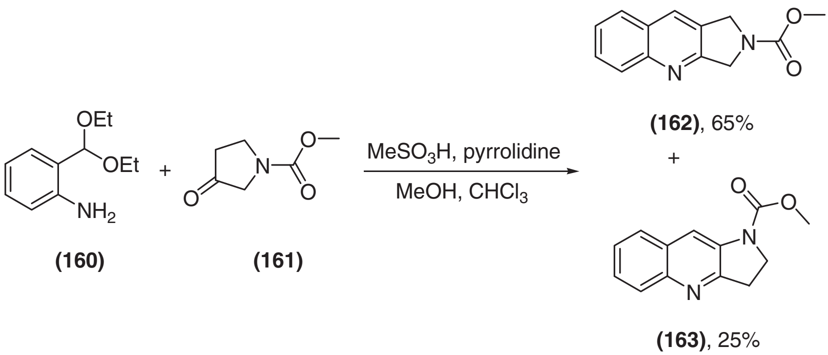
In addition, various metal complexes are proposed as catalysts for the reaction above. These include Grubbs' ruthenium complexes (01CC2576, 05BKC2038, 08EJO1625), $RuCl_2$-(dmso)$_4$ (05TL3683, 06T8988, 07EJO1599), $RhCl(PPh_3)_3$ (05JHC1219), $Pd(OAc)_2$/PEG-2000 (07JOM4182), palladium



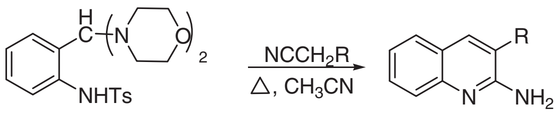
Scheme 29.



Scheme 30.



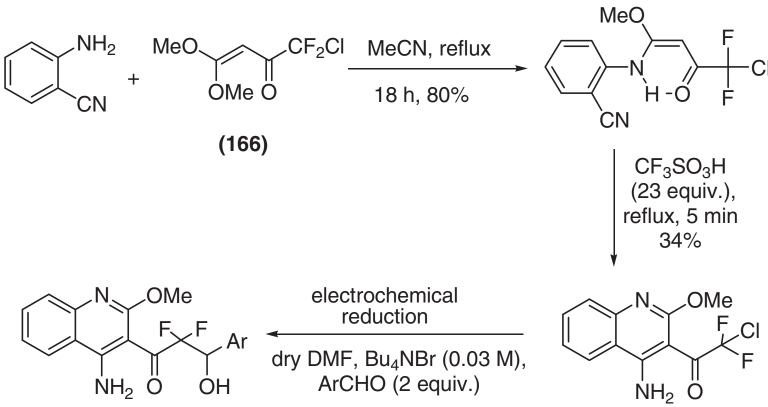
Scheme 31.



(164) **(165)**

R = COOH, COOEt, CN, $CONH_2$

Scheme 32.

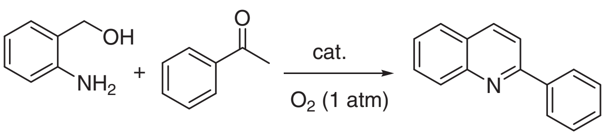


(166)

(168) **(167)**

Scheme 33.

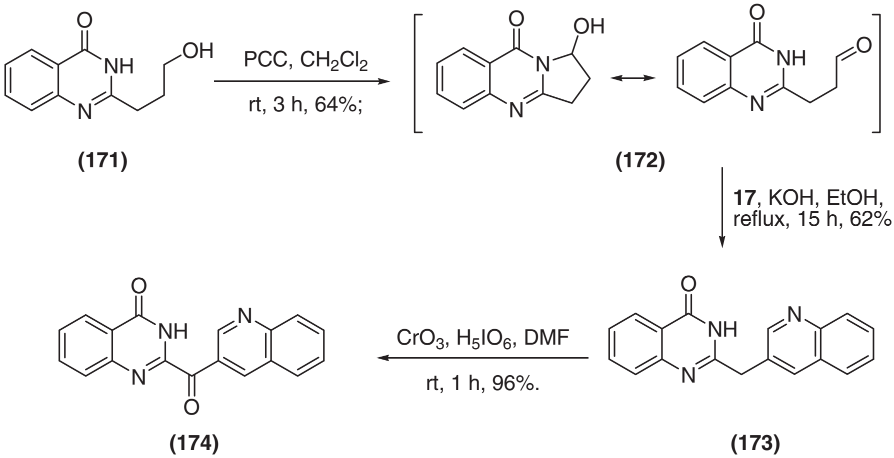
Table 3. One-pot quinoline synthesis using various catalysts[a]



(169) **(170)**

Entry	Catalyst	Conversion of **169** (%)[b]	Yield of **170** (%)[b]
1	Ru/HT-N[c]	>99	87
2	Ru/HT[d]	>99	61
3	Ru/Al_2O_3	>99[e]	3
4	Ru/MgO	37	Trace
5	Ru/$Al(OH)_3$	33	Trace
6	Ru/$Mg(OH)_2$	54	Trace
7	HT	Trace	Trace

[a] **169** (1.0 mmol), acetophenone (1.2 mmol), catalyst (0.3 g, Ru: 3.0 mol%), toluene (5 mL), 100°C, 20 h, under O2 atmosphere.
[b] Determined by GC.
[c] HT-N: hydrotalcites triethylamine.
[d] HT: hydrotalcites.
[e] Oligomeric products were formed from **169**.



Scheme 34.

on charcoal (05BKC1286), [IrCl(cod$_2$)] (05TL4539), $CuCl_2$ (06TL6781), and a reusable form of copper (09MI117).

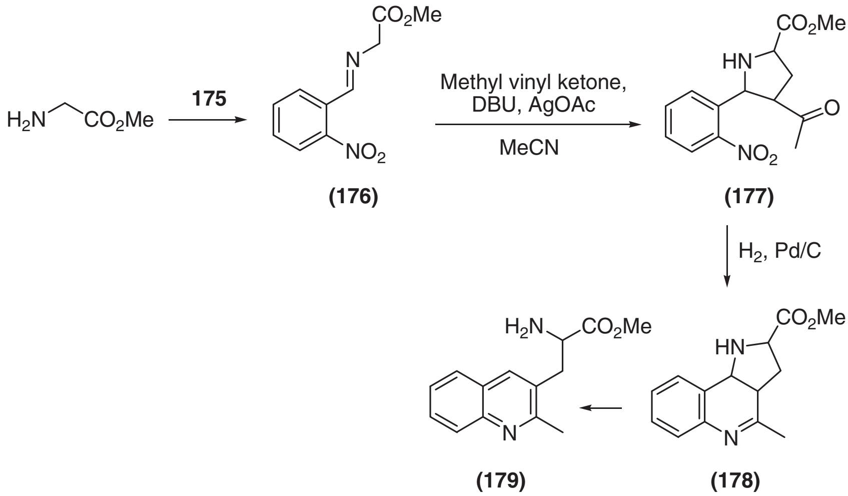
Moreover, quinolines can be prepared from an oxidative cyclization between 2-aminobenzylalcohol **169** and ketones in the presence of a Brönsted base without a transition metal catalyst (08JOC9778, 08TL6893).

A three-step biogenetic-type synthesis of bioactive natural product Luotonin F **174** (99H(51)1883) in 38% overall yield starts from the natural product pegamine **171** *via* PCC-oxidation, Friedländer condensation of **172**, and chromium trioxide–periodic acid oxidation of **173** (Scheme 34) (02S323).

Borsche (50CB78), McNaughton (03OL4257), and others (05BP911, 06ARK24, 06JCSI199) have developed a single-step method for quinoline synthesis from *o*-nitrobenzaldehyde **175** and ketones, which employs reducing agents such as $SnCl_2$ and $ZnCl_2$ (Scheme 35). The yields are between 70% and 98% (03OL4257).

In another one-pot synthesis using inexpensive reagents *o*-nitroaryl-carbaldehydes are reduced to *o*-aminoarylcarbaldehydes with iron and a catalytic amount of HCl (aq.). Subsequent *in situ* condensation with

Scheme 35.

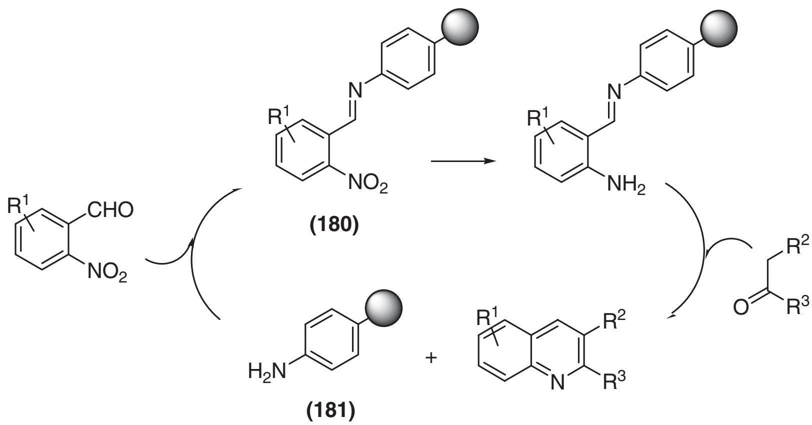


Scheme 36.

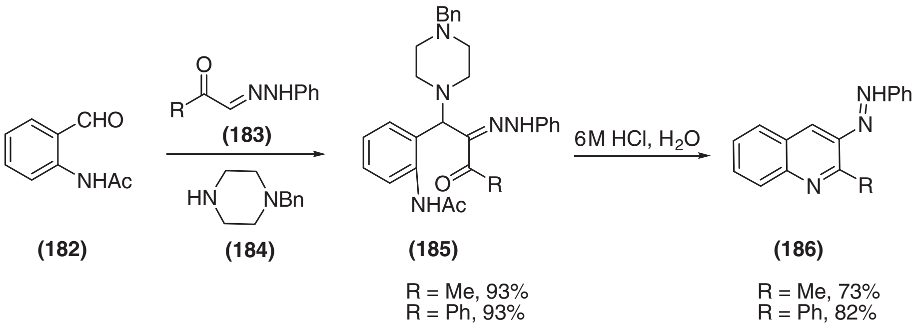
aldehydes or ketones gives mono- or disubstituted quinolines in high yields (07OBC61).

In another study, methyl 4-acetyl-5-(2-nitrophenyl)-pyrrolidine-2-carboxylate **177** was readily obtained (34%) in a one-step 1,3-dipolar cycloaddition of **176**, followed by reduction (hydrogen and Pd/C), Friedländer cyclization to **178**, and fragmentation to yield **179** (72% yield from **175**) (Scheme 36) (98RCM796).

A new solid-phase synthesis of quinolines based on a Friedländer-type reaction between a resin-bound azomethine **180** and ketones has been described. The polymer-bound aniline analog **181** was also easily recycled (Scheme 37) (03OL3061) and serves as a catalyst. Also the solution phase of this reaction has been reported (04TA3919).



Scheme 37.



(182) (184) (185) (186)

R = Me, 93%
R = Ph, 93%

R = Me, 73%
R = Ph, 82%

Scheme 38.

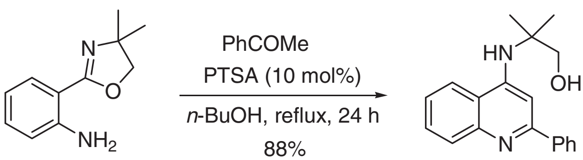
Ethyl formate in hot pressurized water is an effective green reducing system for olefins, acetylenes, and aldehydes (00GC87, 00TL3523). The nitro group can be selectively reduced in a pressure reactor at 170–200°C, yielding the Friedländer quinoline product in modest to good yields (03GC177).

In another modification, the addition of α-ketohydrazones **183** to 2-acetamido-benzaldehyde **182** proceeds efficiently in the presence of *N*-benzylpiperazine **184** to afford the expected Mannich adducts in good to high yields as **185**. These are easily converted to quinolines **186** under acidic conditions (Scheme 38) (04SC109). After hydrolysis of the Mannich product a normal Friedländer cyclization occurs.

Ketones react with *o*-oxazoline-substituted anilines **187** and a catalytic amount of PTSA in dry *n*-butanol to form 4-amino-substituted quinolines **188** or 4-quinolones in fair to good yields (Scheme 39) (06T9365).

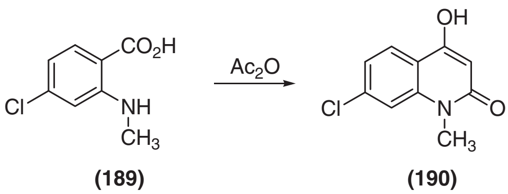
In a modified Friedländer methodology, *o*-aminobenzoic acid **189** reacts with acetic anhydride under neat conditions without base to yield 2-quinolone **190** in 43.5% yield (Scheme 40) (46JA1810).

In another modification (91JOC7288, 97SL285, 98S1176, 99S1335, 05TL767), *o*-lithiated *N*-pivaloylanilines **192** (from **191** and *sec*-BuLi and DMF) and carbonyl compounds and potassium hexamethyldisilazan

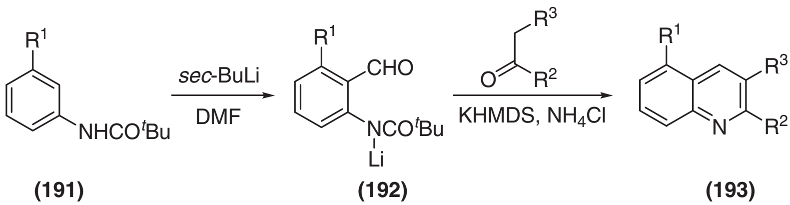


(187) (188)

Scheme 39.



Scheme 40.

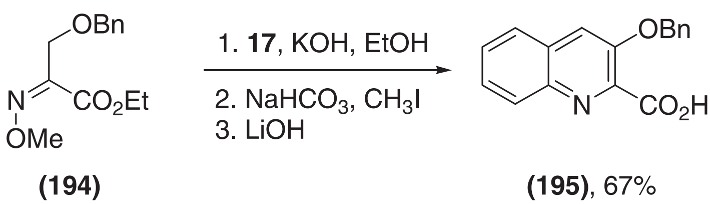


Scheme 41.

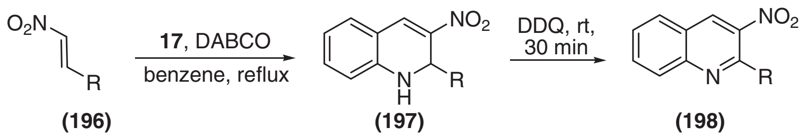
(KHMDS) afford substituted quinolines **193** in one pot (Scheme 41) (97SL285). There is a similar synthesis of diazaanthracene. The yields are between 32% and 85% (05MI374). The aldehyde originates from DMF.

Quinoline **195** is important in the total synthesis of sandramycin, quinaldopeptin, BE22179, and the luzopeptins. These compounds are symmetrical, cyclic decadepsipeptides possessing a twofold axis of symmetry that exhibit a high affinity for sequence-selective DNA binding and bis-intercalation of the pendant heterocyclic chromophores. Boger and Chen have provided a modified procedure for **195** *via o*-aminobenzaldehyde **17** and oxime **194** instead of ketones in 67% yields (Scheme 42) (95JOC7369).

A modified Friedländer synthesis involving α-nitrocarbonyl and 2-aminocarbonyl compounds was reported (50JCS395, 51JCS2992, 53JCS3914, 57JA1502). 2-Aryl-3-nitro-1,2-dihydroquinolines **197** was prepared from *β*-nitrostyrenes **196** and 2-aminobenzaldehyde **17** using DABCO. Other alkyl nitro olefins were also used. When 2,3-dichloro-5, 6-dicyanoquinone (DDQ) is added to 3-nitro-1,2-dihydroquinolines **197**,



Scheme 42.



Scheme 43.

3-nitro-2-substituted-quinolines **198** are obtained in 63–97% yields (Scheme 43) (04JOC1565).

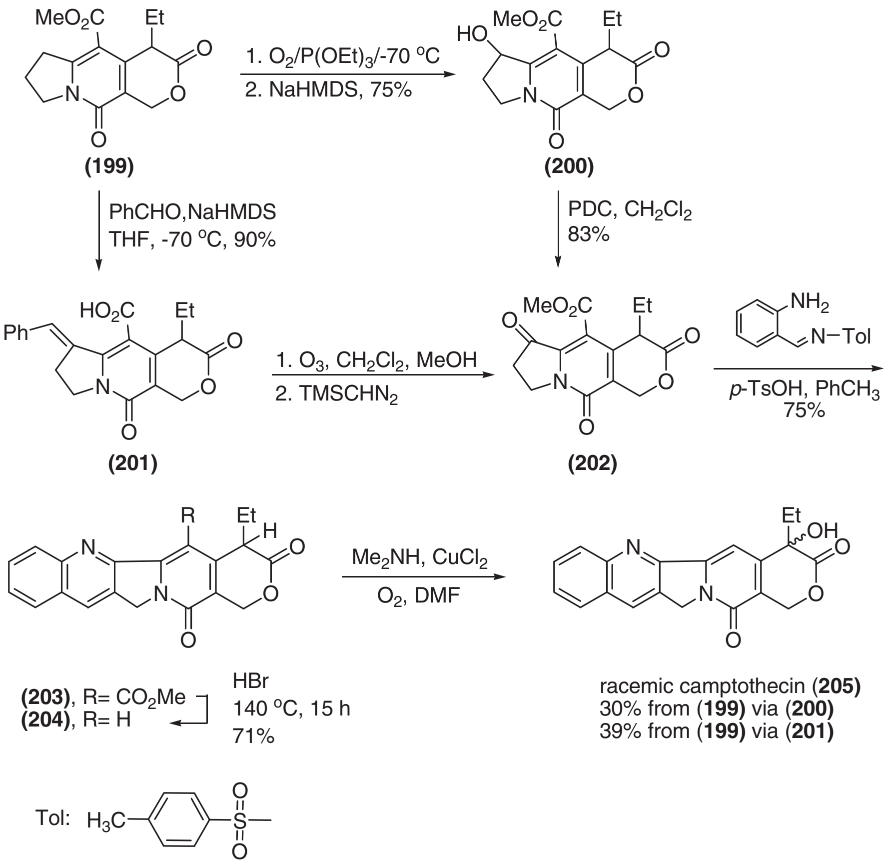
3. CAMPTOTHECIN AND ITS ANALOGS

Renewed interest in the synthesis of camptothecin and its active analogs was triggered by the discovery of its high antitumor activity in various cell lines and screening of animals models (81T1047, 83CHE753, 92MI1, 95MI1, 97BMC1481, 00BP497, 03T8649, 04BMC1585, 04JNP273). While camptothecin is a difficult alkaloid to isolate from natural sources, its analogs are synthesized in good yields from the natural precursors (73CRV385, 91CPB3183, 09CRE213).

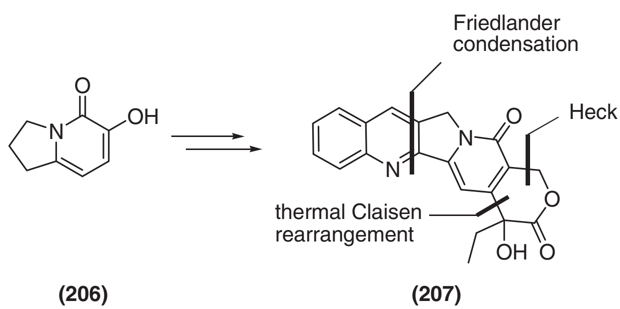
Danishefsky and coworkers reevaluated the problem and explored the chemistry to provide a two-pronged approach to an inexpensive and decent yielding route to D,L-camptothecin (71JA5576, 74JOC3430, 75JOC796, 93JOC611, 94JOC7033). In Scheme 44, the readily available tricyclic ester **199** is converted to **202** using two alternative routes. In the first sequence, **199** sodium hexamethyldisilazide (NaHMDS) and benzaldehyde afford benzylidene acid **201** in 90% yield. Ozonolysis of **201** followed by esterification provided **202** in 94% yield. The complementary route involves tricylic compound **199**, oxygen and triethylphosphite to give diastereomers **200** in 75% yield. Oxidation with pyridinium dichromate affords ketone **202** in 83% yields. Ester **202** with a Schiff base *via* a typical Friedländer condensation affords pentacyclic **203**. Heating **203** with HBr provides D,L-desoxycamptothecin **204** in 71% yield. Hydroxylation of **204** with oxygen, Me_2NH, and $CuCl_2$ gives camptothecin **205** in 91% yield. The overall yield of **205** from **200** and **201** is 30% and 39%, respectively.

A novel and efficient synthesis of racemic camptothecin **207** starting from a readily accessible hydroxy pyridine **206** was described. Key steps include a Claisen rearrangement of a functionalized allylic ether, a hindered Heck coupling, and a Friedländer condensation (Scheme 45) (05OL2989).

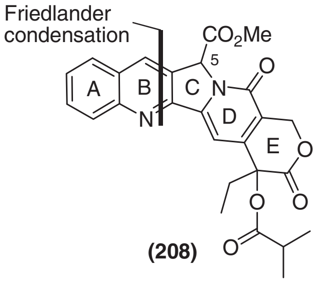
The synthesis of camptothecin analogs substituted by a carbonyl function on position 5 of ring C **208** was realized (Figure 7) (03JHC45, 05T7916, 06T3959).



Scheme 44.



Scheme 45.

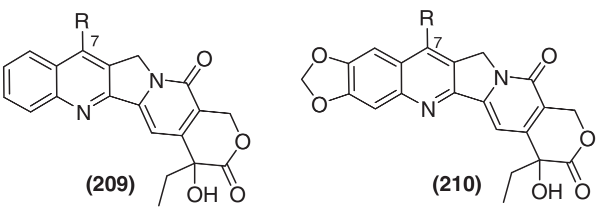


(208)

Figure 7.

By developing a new synthetic procedure for the introduction of side chains onto the camptothecin ring system, the group of Wani (93JME2689, 04BML5377) was able to prepare a number of analogs bearing bulky, hydrophobic groups directly attached to position 7, such as **209** and **210** (Figure 8). These include 7-*tert*-butylcamptothecin, 7-benzylcamptothecin, and the corresponding 10,11-methylenedioxycamptothecins. The camptothecin analogs show promising ability to inhibit the growth of selected tumor cell lines.

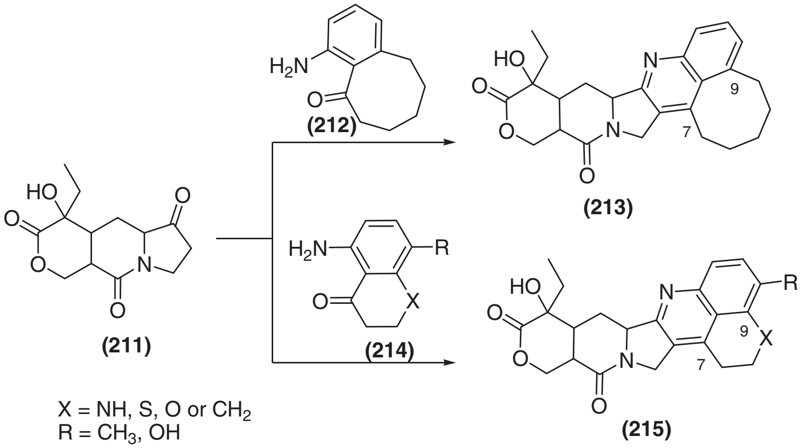
Some novel hexacyclic and 7,9-disubstituted pentacyclic derivatives of camptothecin were synthesized by Terasawa et al. They were evaluated for *in vitro* cytotoxic activity against P388, HOC-21, and QG-56 and *in vivo* antileukemic activity against P388 in mice. Hexacyclics **213** and **215** have an additional 5-, 6-, or 7-membered ring cyclized at positions 7 and 9 of the camptothecin moiety. They were prepared by intramolecular cyclization of pentacyclic camptothecin derivatives of bicyclic amino ketone **212** or **214** and a tricyclic ketone **211**, respectively (Scheme 46) (94JME3033).



(209) R = *sec*-Bu, *tert*-Bu, Bz

(210) R = Ph, Bz, *p*-tolyl, cyclopentyl, cyclohexyl, *p*-chlorophenyl, *p*-fluorophenyl, *p*-trifluoromethyl

Figure 8.



Scheme 46.

Water soluble 7-substituted quaternary ammonium salt derivatives of 10,11-(methylenedioxy)- and 10,11-(ethylenedioxy)-(20*S*)-camptothecin **216** (Figure 9) are synthesized *via* the Friedländer reaction followed by nucleophilic displacement with an aromatic amine. All are more potent than camptothecin in the *in vitro* cleavable complex assay (96JME713).

Analogs based on a novel template, 11-aza-(20*S*)-camptothecin **219**, were obtained from a total synthesis and tested as potential anticancer drugs in the topoisomerase I enzyme cleavable complex assay. The parent 11-aza-(20*S*)-camptothecin **219** is derived by cyclization of the known aminopyridine derivative 3-(3-amino-4-picoly1idene)-*p*-toluidine **217** and optically active tricyclic ketone **218** in 27% yield (Scheme 47) (95JME1106).

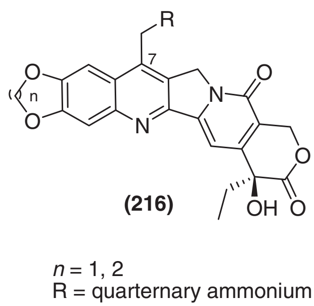
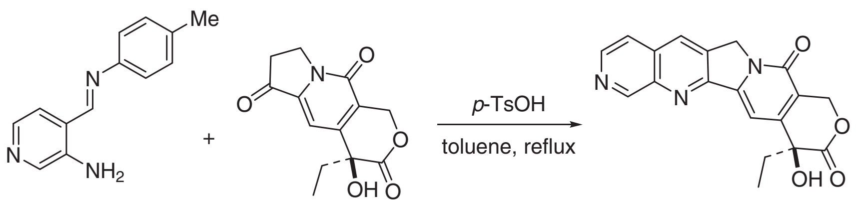


Figure 9.



(217) **(218)** **(219)**

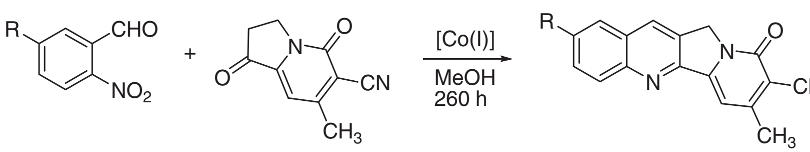
Scheme 47.

The imine in the starting material serves as the electrophile for the mixed aldol reaction in the first step. This could also rightly be classified as a modified Friedländer reaction (see Section 2.1).

An improved preparation of indolizinoquinolines using MW-assisted synthesis involves the Friedländer reaction of *o*-aminobenzaldehydes or imines with tetrahydroindolizinediones to form a quinoline (02SL2077).

Selective reduction of the aromatic nitro aldehyde **220** to the amine group by phthalocyanine cobalt (I) anion is followed by condensation to alkaloids **221** (Scheme 48) (81AGE208).

The Friedländer method is also widely used in the synthesis of camptothecin analogs for the investigation of their biological properties (69T2275, 71JA4074, 71JCS3551, 72JA3631, 86JME2358, 89TL2639, 90JC (P1)27, 98TA2285, 04TL7247, 06OBC3757). Examples are **222** (73JHC77), aromathecin (**223**) (99TL2723, 02TL1835, 08JOC1975), mappicine ketone (**224**) (95JOC2912, 99T5449), **225** (95BML2129), **226**, **227**, **228**, **229** (86JME2358), **230**, **231** (90JME972), **232** (98JME2308), **233** (87JME1774), nothapodytine B **234**, (−)-mappicine (**235**) (98JA1218, 03JHC601), 11-hydroxyacuminatines (**236**) (86JME1553, 06OBC407), aza-analog **237** (80JME554), 22-hydroxyacuminatines **238** (08BML2143), water-soluble analogs **239** (95JME395), **240** (91JME98), **241** (94BMC1397, 94CPB2518), **242** (92CPB683), E-ring modified **243** (89CPB2253), irinotecan (**244**) (91MI1164, 97JOC6588), and paclitaxel–camptothecin analogs **245** (03BMC1851) (Figure 10).



(220) **(221)**

R = H, (62%)
R = Cl, (51%)

Scheme 48.

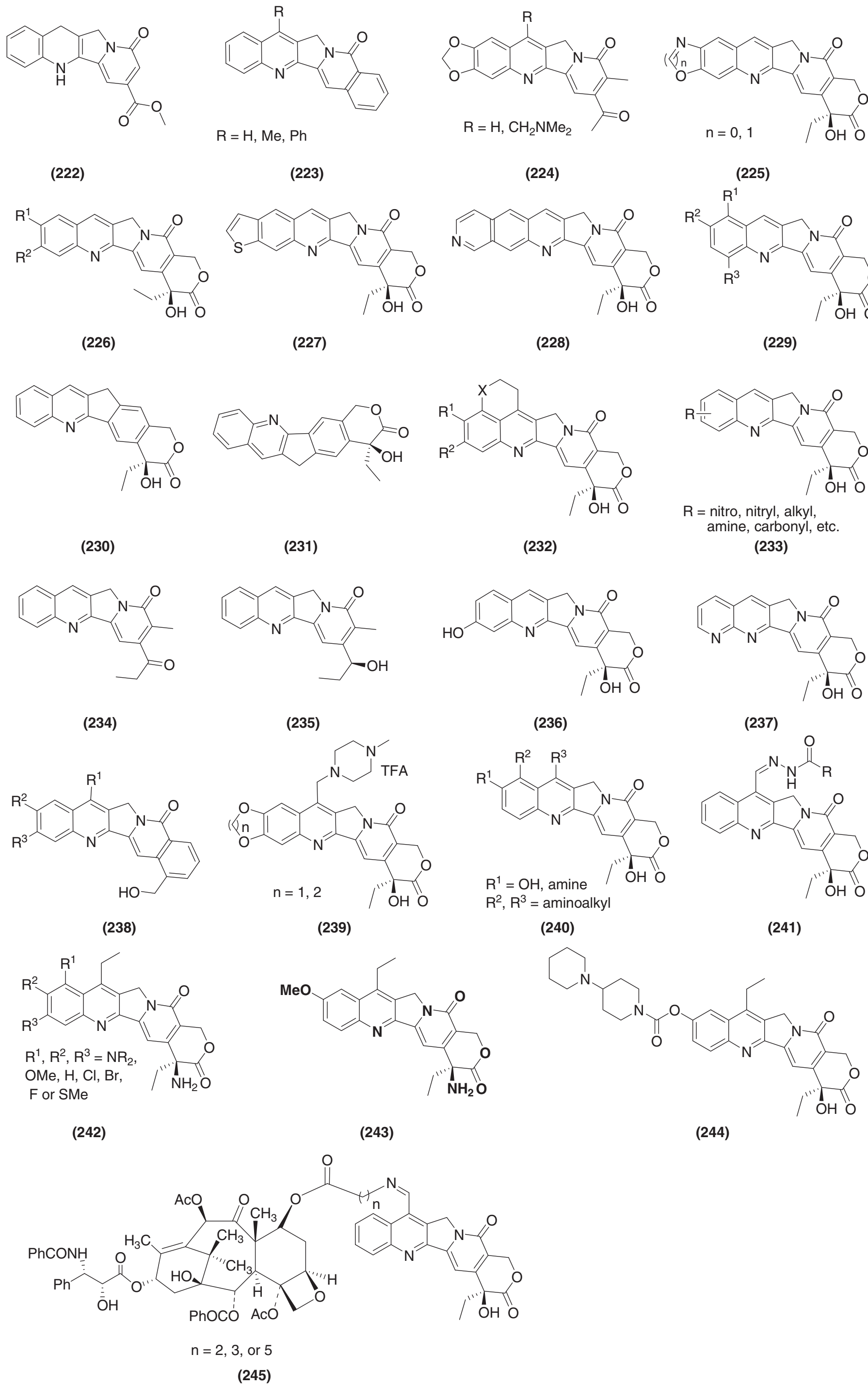

R = H, Me, Ph
R = H, CH2NMe2
n = 0, 1
(222)
(223)
(224)
(225)
(226)
(227)
(228)
(229)
R = nitro, nitryl, alkyl, amine, carbonyl, etc.
(230)
(231)
(232)
(233)
(234)
(235)
(236)
(237)
TFA
n = 1, 2
R1 = OH, amine
R2, R3 = aminoalkyl
(238)
(239)
(240)
(241)
R1, R2, R3 = NR2, OMe, H, Cl, Br, F or SMe
(242)
(243)
(244)
n = 2, 3, or 5
(245)

Figure 10.

4. PYRIDINE SYNTHESIS

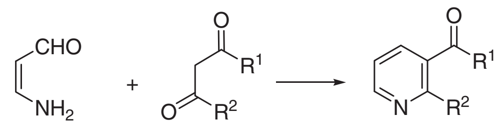
Friedländer reported the synthesis of pyridines *via* condensation of enaminoketones with *β*-ketoesters or 1,3-diketones (1883CB1833).

Breitmaier and Bayer have synthesized the 3-pyridinecarboxylates and 3-acylpyridines **247** by cyclocondensation of 3-aminoacrolein **246** with 1, 3-dicarbonyl compounds (Scheme 49) (69AGE765, 70T5907). The yields range between 30% and 80%.

β-Ketoesters and *β*-diesters also react with *β*-enaminonitriles **248** in the presence of stoichiometric amounts of tin (IV) chloride to give 4-amino-pyridines and pyridines. They also react with aromatic *ortho*-aminonitriles to give 4-amino-quinolines and quinolones (Scheme 50) (90TL3485, 95T12277). Typical yields are 45–70%.

Davies et al. have described a new synthetic strategy to obtain the Cox-2 specific inhibitor **251**. They heated a mixture of chloromalonaldehyde **249**, ammonium acetate, and ketosulfone **250** with propionic acid at 125°C. Pyridine **245** is produced in 62% yield together with furan impurity **252** in approximately 15% yield (Scheme 51) (00JOC8415).

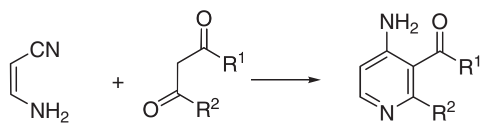
Condensation of **246** with *α*-oxo-2,3-cycloalkeno-pyridines (or thermolysis of the *o*-allyloximes **254** derived from ketones **253**) leads to the formation of 3,3′-bridged-2,2′-bipyridine **255** in 20–65% yield (Scheme 52) (85JOC3824).



(246) **(247)**

R^1, R^2 = alkyl or alkoxy

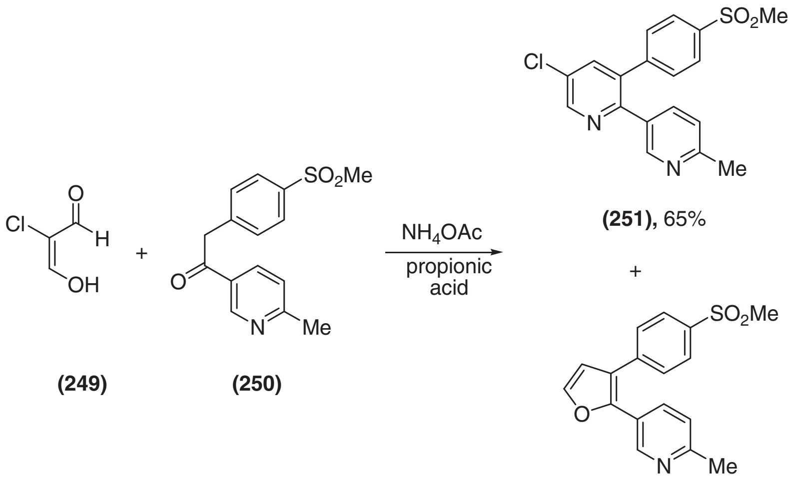
Scheme 49.



(248)

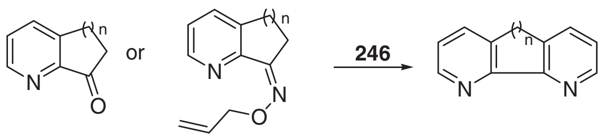
R^1, R^2 = alkyl or alkoxy

Scheme 50.



(249) **(250)** **(251),** 65% **(252),** 15%

Scheme 51.



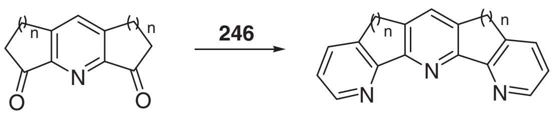
(253) **(254)** **(255)**

$n = 2, 3, 4$

Scheme 52.

Polyaza cavity terpyridines **257** were prepared from diketones **256** and **246** (Scheme 53). Similarly these diketones with *o*-aminobenzaldehyde afford bis-annulated derivatives of 2,6-di(2′-quinolyl)pyridine while condensation with 2-aminonicotinaldehyde affords bis-annulated derivatives of 2,6-di(1′,8′-naphthyrid-2′-yl)pyridine (85JOC2407).

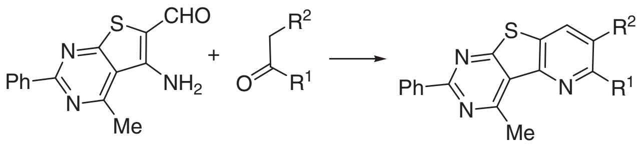
A variety of tri-, tetra- and penta-cyclic pyrido[2′,3′:4,5]thieno[2,3-*d*] pyrimidines **259** are synthesized from 5-amino-6-formyl-4-methyl-2-



(256) **(257)**

n = 2 or 4

Scheme 53.



(258) (259)

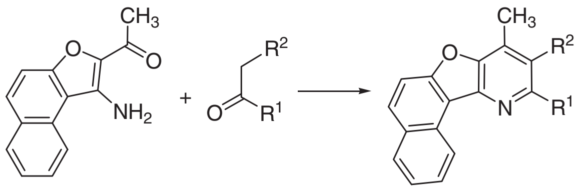
Scheme 54.

phenylthieno[2,3-*d*]pyrimidine **258***via* Friedländer condensation with aliphatic, alicyclic, and heterocyclic ketones and other active methylene compounds (Scheme 54) (07JCM689).

2-Acyl-3-aminonaphtho[2,1-*b*]furans **260** are directly synthesized from 2-hydroxy-1-naphthaldehyde oximes and then with an α-haloketone through the Thrope–Ziegler reaction to yield amino furan **260**. This amino furan undergoes cyclization with active methylenes to produce **261** in good yields (Scheme 55) (08IJB753).

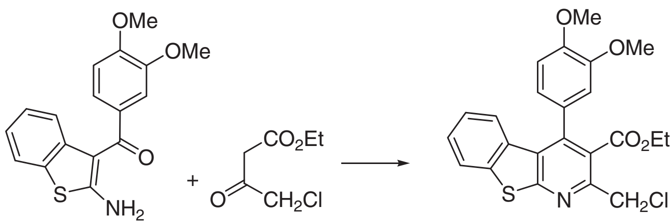
The syntheses and anti-inflammatory activities of thieno[2,3-*b*]pyridines **262** were also described. They were designed by modification of the quinoline template of a new type of disease-modifying antirheumatic drug (DMARD), TAK-603, and prepared *via* the Friedländer cyclization (75% yield) (Scheme 56) (99CPB993).

2,7-Diaryl-3-cyano-4-methylpyrano[4,3-*b*]pyridine-5-ones **266** are synthesized as shown in Scheme 57. In the first step, nitriles **264** are added to esters **263** to then form products **265** *via* a Knoevenagel condensation.



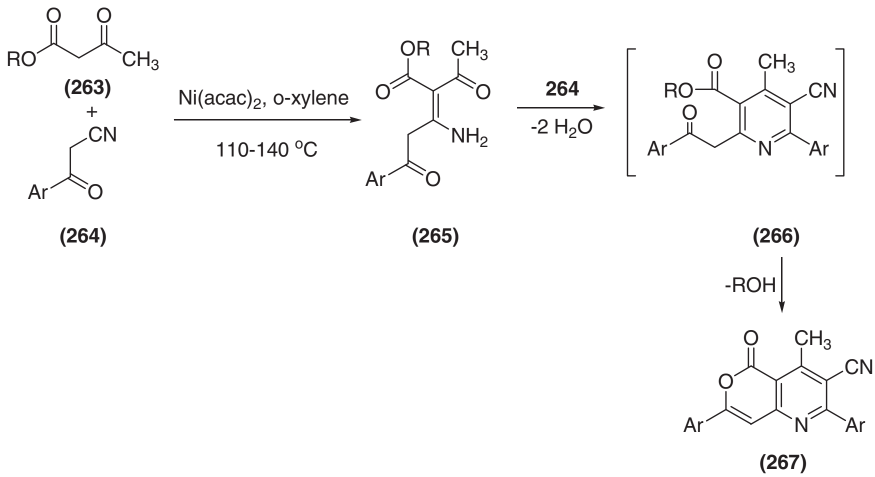
(260) (261)

Scheme 55.



(262)

Scheme 56.



Scheme 57.

265 reacts with nitriles **260** to give functionalized pyridines **266**, which undergo intramolecular cyclization to form pyranopyridines **267** in 15–19% yields (Scheme 57) (02RCB1545).

Reaction of 3-amino-2-formylimidazo[1,2-*a*]pyridine **268** with various aldehydes and ketones affords an entry to dipyridoimidazole, tri(tetra) azacyclopenta[*b*]fluorine **269**, tri(tetra)azabenzo[*b*]fluorine **270**, and triazaindeno[2,1-*b*]phenanthrene **271** derivatives (Scheme 58) (05H(65)1121).

An analogous cyclization of 2-amino-6-ethyl-4-oxo-4*H*-1-benzopyran-3-carbaldehyde **272** (86JP6110588), 5-amino-pyrazole-4-carbaldehyde **273** (07JHC343, 05JHC1311), 5-amino-1*H*-pyrazolo-4-carbaldehyde or cyano **274** (99SC4403, 04AFF510), and 5-amino-3,4-diphenylthieno[2,3-*c*]pyridazine-6-carbaldehyde **275** (96H(43)1073) with ketones was also reported. Similarly *o*-aminothiophene carbaldehydes (05H(65)2369, 06JHC101) or *o*-aminobenzaldehyde (94SC1363) react with creatinine **276** and lead to the corresponding pyridine derivatives (Figure 11). The yields of the Friedländer steps are between 30% and 85%.

2-Amino-1-methyl-6-phenylimidazo[4,5-*b*]pyridine **278** is prepared in 26% yield from 3-amino-2-phenylpropenal **277** and creatinine **276,** which are heated with *N,O*-bis(trimethylsilyl)acetamide at 120°C for

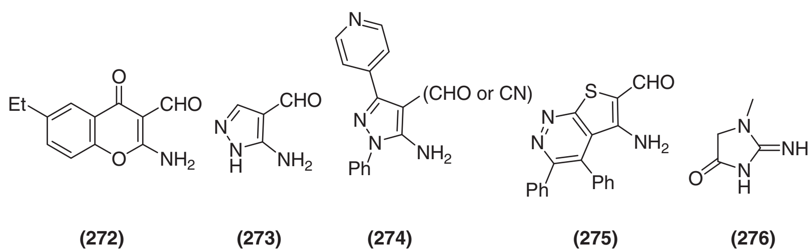
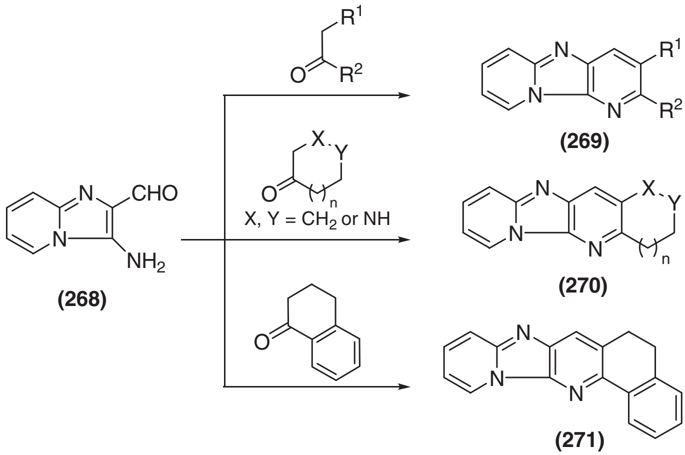
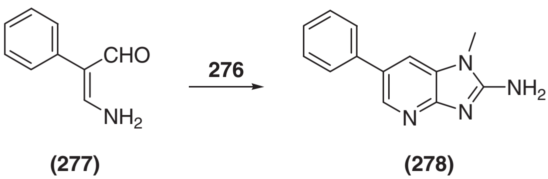


Figure 11.



Scheme 58.

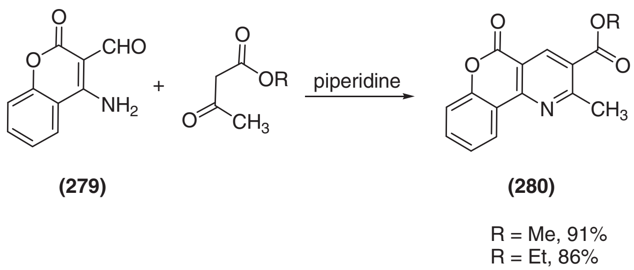


Scheme 59.

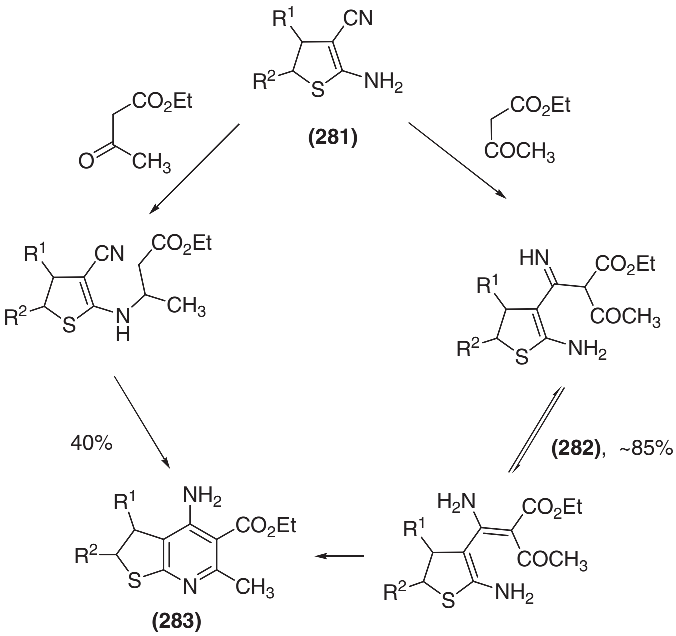
2 h (Scheme 59). By changing the conditions, a pyrimidine derivative can be the main product (95ACS361).

4-Aminocoumarin-3-formaldehyde **279** reacts with acetoacetic ester while catalyzed by piperidine to form the tricycle **280** in high yields (Scheme 60) (92LA203).

Acetoacetic esters also exhibit the characteristics of an aldol intermediate with 2-amino-4,5-dihydro-3-thiophene-carbonitriles **281**. In the first stage, the aldol adds to the nitrile group with the formation of



Scheme 60.

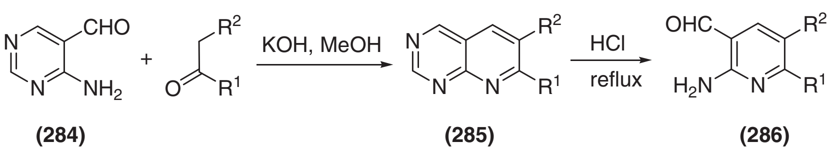


Scheme 61.

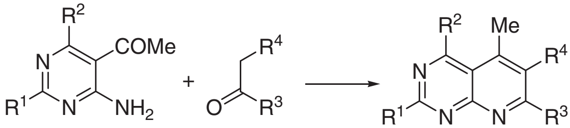
β-enaminone **282**. The latter then undergoes cyclization to ethyl-4-amino-2,3-dihydro-6-methylthieno[2,3-*b*]pyridine-5-carboxylates **283** as proposed above. Titanium tetrachloride catalyzes not only the addition of acetoacetic ester to the nitrile group (the first step) but also the last stage of cyclization (the formation of the C–N bond; Scheme 61) (93LA1269).

4-Aminopyrimidine-5-carboxaldehyde **284** with aromatic ketomethylenes provides 7- and 6,7-disubstituted pyrido[2,3-*cl*]pyrimidines **285**. Facile ring opening of the pyrimidine moiety of this heterocyclic system gives substituted 2-aminonicotinaldehydes **286** (Scheme 62) (75JOC1438).

A series of pyrido[2,3-*d*]pyrimidines **288** were obtained in good yields from pyrimidines **287** (Scheme 63) (02RCB1875, 05RCB784, 07RCB2293).



Scheme 62.



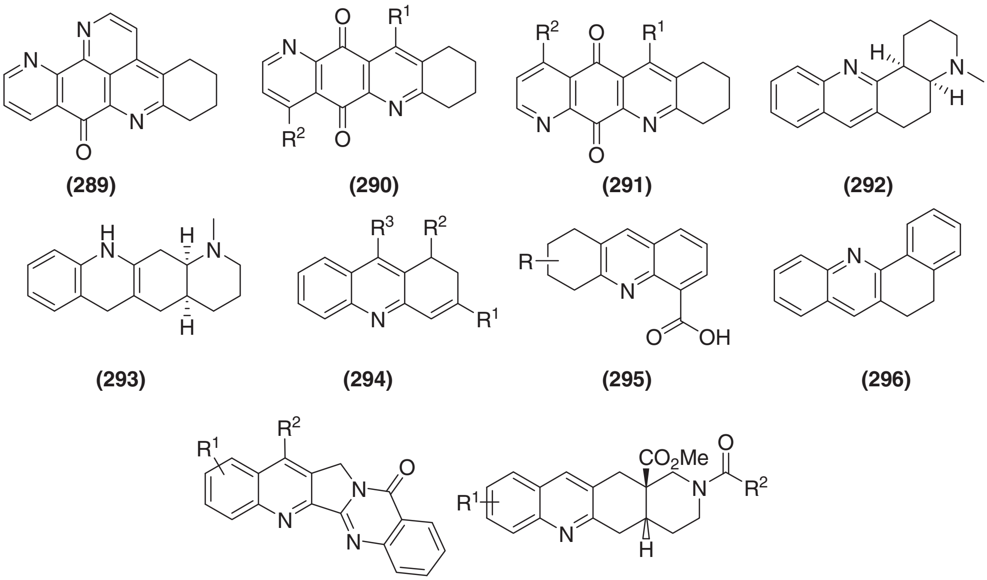
(287) (288)

Scheme 63.

5. ACRIDINE SYNTHESIS

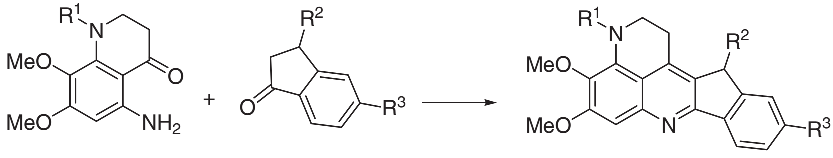
The Friedländer reaction has been used in the synthesis of acridines such as 1,2,3,4-tetrahydroascididemine **289** (99TL4097), pyrido[3,2-*b*]acridines **290**, **291** (99T12637), polyhydroacridines **292**, **293** (91JIB213), 1,2-dihydroacridines **294** (66JPR298), acridine-4-carboxylic acids **295** (99SC4223), and dihydrobenz[*c*]acridines **296** (81JHC649), luotonin A (09JOC00) and its derivatives **297** (08BKC1988) and **298** (08S2199) (Figure 12).

Molecules possessing the new skeleton of 1*H*-benzo[*c*]pyrido[2,3,4-*kl*]-acridine **301** with acyl, aminoacyl, and methoxy or aminoalkoxy substituents on the aromatic homocycles were synthesized. Most have therapeutic



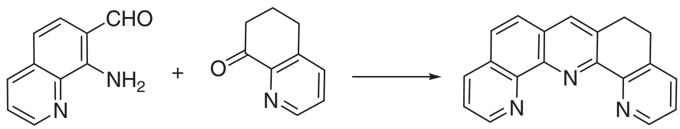
(289) (290) (291) (292) (293) (294) (295) (296) (297) (298)

Figure 12.



(299) (300) (301)

Scheme 64.



(302) (303) (304)

Scheme 65.

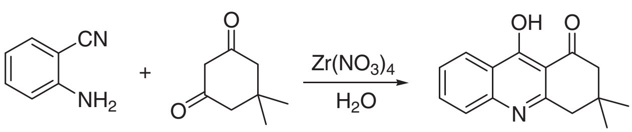
interest. The requisite 5-aminodihydroquinoline-4-ones **299** react under azeotropic conditions with substituted-tetralones **300** and acidic catalyst pyridinium *p*-toluenesulfonate (PPTS) (Scheme 64) (02JOC3502, 04JME3665).

Thummel and coworkers have synthesized dipyrido[4,3-*b*;5,6-*b*]acridines **304** from the condensation of 8-amino-7-quinolinecarbaldehyde **302** with 5,6,7,8-tetrahydro-8-quinolone (**303**) and their ruthenium(II) complexes (Scheme 65) (96IC5953).

2-Aminobenzonitril **305** and dimedone **306** in water condense to afford hydroxyl acridine **307** in 80% (Scheme 66) (07MI1214).

Chiral acridine-type ligand **308** (08TA2600) and *N*,*P*-ligands such as 5-(diphenylphosphanyl)-1,2,3,4-tetrahydroacridines **309-312** (05TL3493, 06TA1529) have been reported (Figure 13).

They were obtained utilizing the same Friedländer methodology between an amino aldehyde and a chiral methylene ketone.



(305) (306) (307)

Scheme 66.

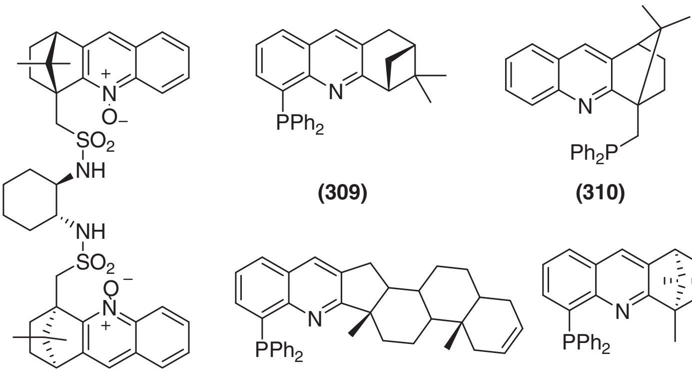


Figure 13.

6. PHENANTHROLINES

8-Amino-7-quinolinecarbaldehyde **302** is widely used in the synthesis of mono- and di-(1,10)-phenanthrolines. Examples are **313** (94T10685, 05JCD354), **314**, **315**, **316** (97IC3133), **317**, **318**, **319** (99IC5620), **320**, **321** (02OL1253), and 1,3-, 1,6-, 1,8-, and 2,7-bis(2-[1,10]phenanthrolinyl)pyrenes **322** (03EJI2774), **323** (00IC3590), and **324** (99TL7311) (Figure 14).

Chelucci and Thummel reported chiral phenanthrolines containing camphor derivatives **327**. Camphor **326**, a hindered ketone, causes the Friedländer reaction to proceed in low yields. They suggested a modification starting from **225** in which **327** forms in 51% overall yield (Scheme 67) (99SC1665).

Cyclization of (1*R*,5*S*)-(+)- and (1*S*,5*R*)-(−)-nopinone **328a**, **328b** with **302** gives enantiomerically pure (2,3-*b*)-pineno-1,10-phenanthrolines **329a**, **329b** in 40–56% yields (Scheme 68). Separate coordination with Cu(I) affords noninterconvertable chiral complexes that show equal and opposite Cotton effects in their CD spectra (98IC2145).

Di- or triacetylaromatic **330** or heteroaryl systems and **302** can be condensed to 1,10-phenanthrolines **331** (Scheme 69) (91T6851, 92SL1, 96JOC3017). Typical yields are between 50% and 86%.

Bridging ligands including a central linker appended to two 1,10-phenanthrolin-2-yl units (e.g., **333** in Scheme 70) include pyridazin-3,6-diyl, 1,8-naphthyrid-2,7-diyl, 2,2′-bipyrid-6,6′-diyl, 1,10-phenanthrolin-2,9-diyl **332**, 1,2-di(2′-pyrid-6′-yl)ethyne, and 3,6-di(2′-pyrid-6′-yl)pyridazine. Yields are ranging between 48–88% (06JOC167).

Triacetylmethane **334** with *o*-aminoquinolinecarbaldehydes on refluxing in ethanol with a catalytic amount of KOH, leads to 1,

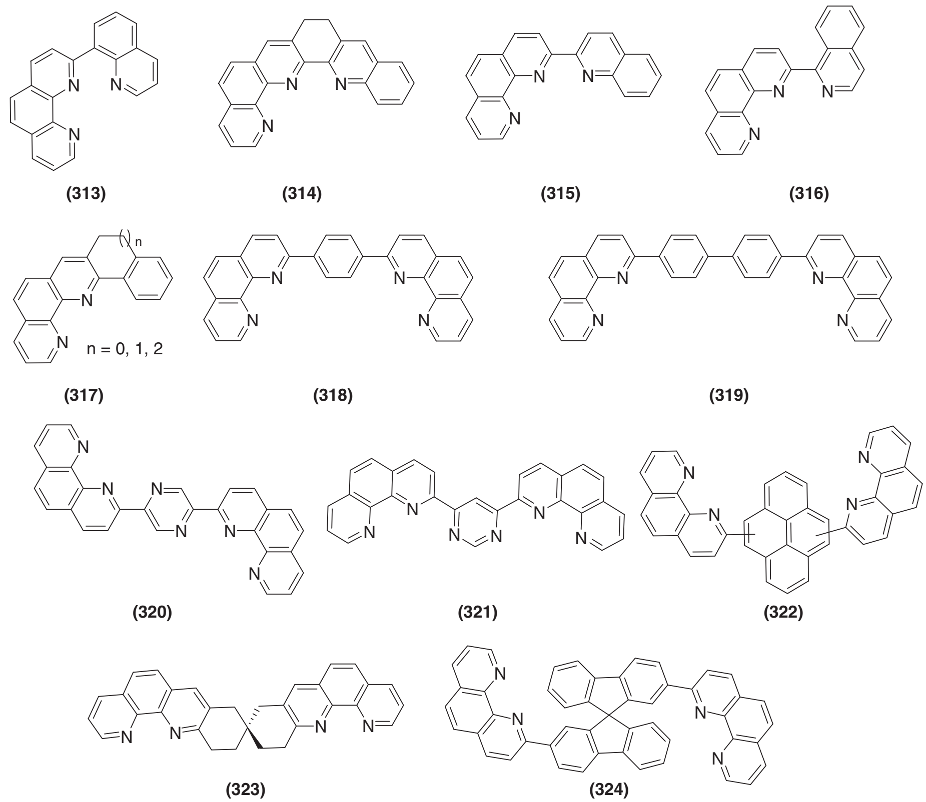
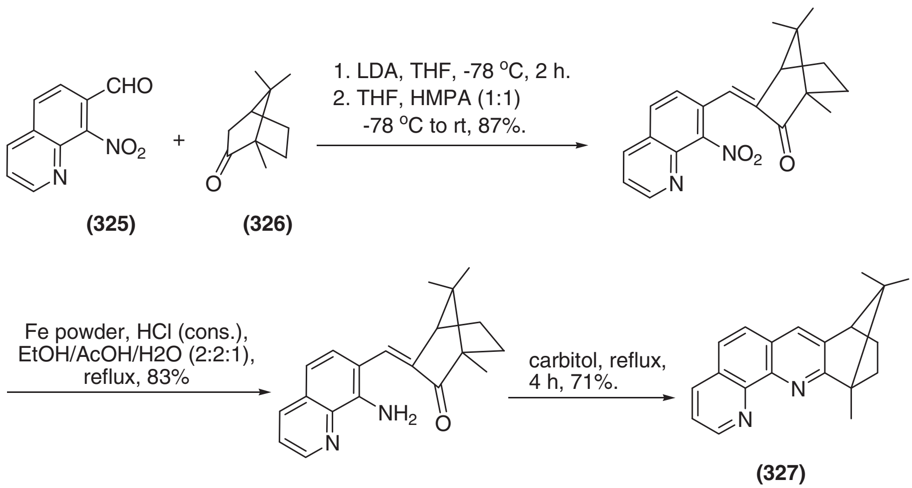
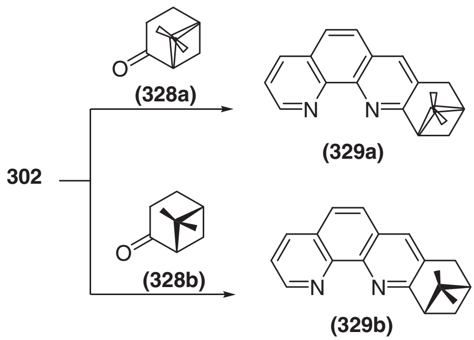


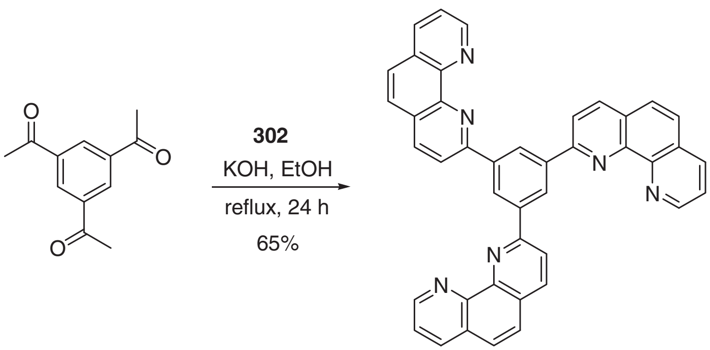
Figure 14.



Scheme 67.

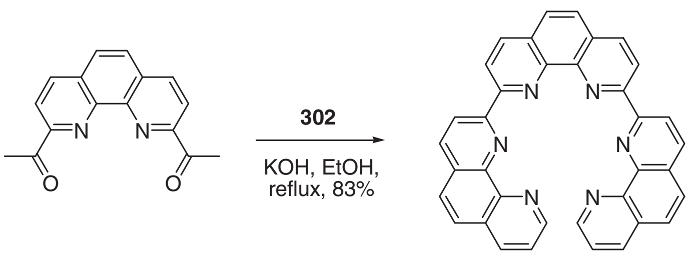


Scheme 68.



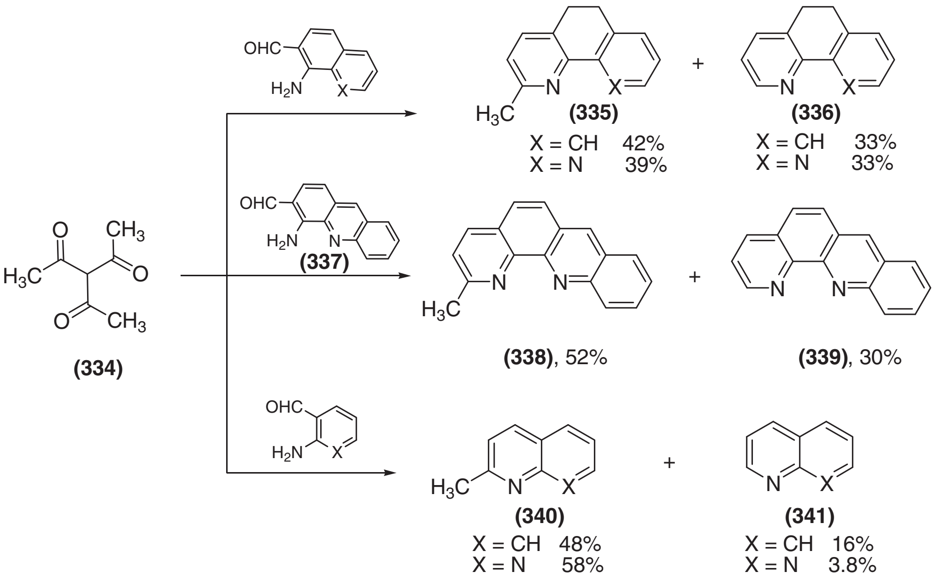
(330) (331)

Scheme 69.



(332) (333)

Scheme 70.

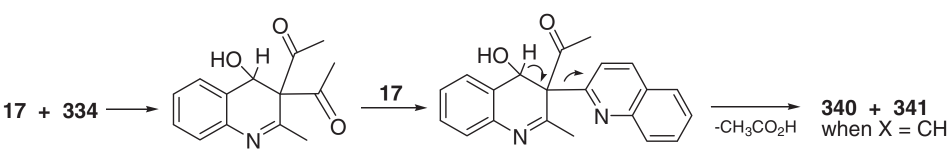


Scheme 71.

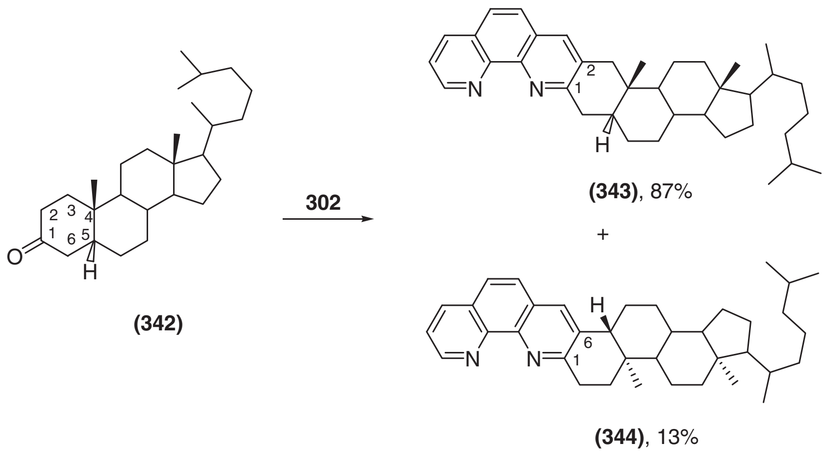
10-phenanthrolines **336** and **339** and 2-methyl-1,10-phenanthrolines **335**, **338** in moderate yields. The corresponding quinolines and 1,8-naphthyridines **340**, **341** are obtained from **334** and *o*-aminoarylaldehydes and 2-aminonicotinaldehyde under similar conditions (Scheme 71) (05H(65)2777).

The mechanisms of the above reactions were postulated (Scheme 72) (05H(65)2777).

The scope of the Friedländer condensation in the preparation of chiral alkyl-substituted 1,10-phenanthrolines was investigated. A range of chiral [*x*, *y*-*b*]-cycloalkeno-condensed phenanthrolines **343** and **344** were prepared in one step from the chiral pool of steroidal **342** or other cyclic ketones and **302** *via* base-catalyzed conditions (Scheme 73) (00MI423, 01JOC400). In the case of the major product, aldol bond formation takes place from the sterically less hindered α-carbon (C2) **343** while the sterically more hindered α-carbon (C6) **344** reacts to form the minor product (Scheme 73).



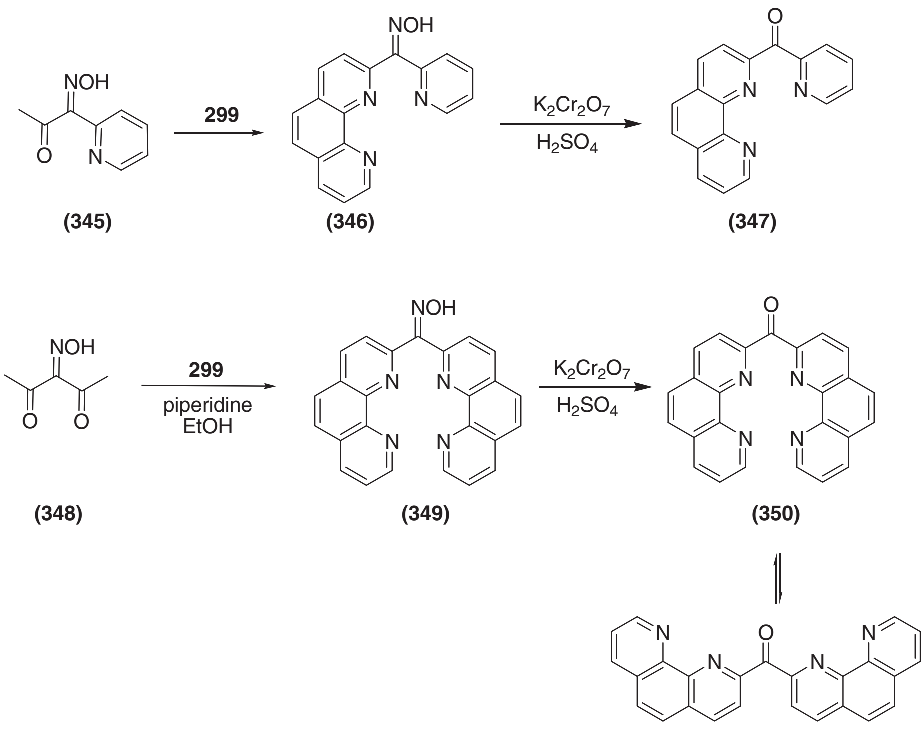
Scheme 72.



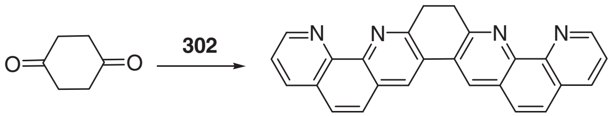
(342)

Scheme 73.

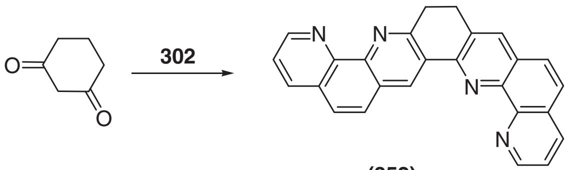
A synthetic protocol involving condensation of **302** and oximes **345** or **348** followed by potassium dichromate oxidation was applied to 1-(pyrid-2′-yl)propane-1,2-dione-1-oxime **346** and 2,3,4-pentanetrione-3-oxime **349** to provide the ligands phenanthrolin-2-yl-pyrid-2-yl-methanone **347** and di-(phenathrolin-2-yl)-methanone **350**, respectively (Scheme 74).



Scheme 74.



(351)



(352)

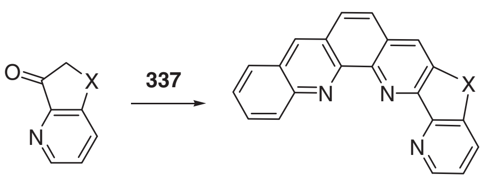
Scheme 75.

These ligands **347** and **350** were complexed with [Ru] and their properties were studied (05IC8733). The typical yield for the Friedländer condensation steps is about 70%.

The synthesis of biphen 2,2′-dimethylene-3,3′-biphen **351** and 2, 3′-dimethylene-3,2′-biphen **352** uses the same methodology (Scheme 75) (02IC3423).

4-Aminoacridine-3-carbaldehyde **337** with cycloalkane-1,2-diones (Scheme 76) (08H(75)871) and 1,3-, 1,4-di-, 1,3,5-triacetylbenzenes (07HAC650) benzo[*b*]cyclo-alkanones, 3,4-dihydro-1(2*H*)-anthracenone (07H(71)2003) ketones **353** (08H(75)2507), or simple ketones (02H(57) 1109) afford a series of mono-, di-, or tribenzo[*b*]-1,10-phenanthrolines.

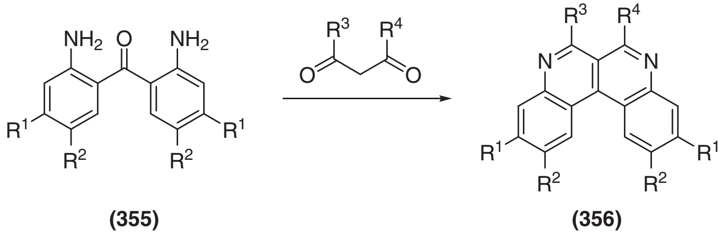
Model compounds **356** are carcinogenic materials. In the synthesis of **356**, Friedländer methodology is widely used. Typical starting materials are 2,2′-diaminobenzophenones **355** and *β*-diketones (Scheme 77). Their mutagenicities were investigated (25JCS1493, 62JCS632, 66JME161, 71ZC61, 86JC(P1)1225, 98JPP475).



(353) **(354)**

X = H, H; -CH=CH-; -$(CH_2)_n$-, n = 1-4

Scheme 76.

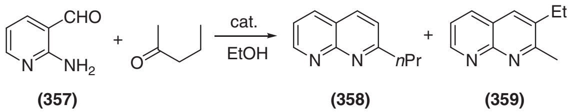


Scheme 77.

7. NAPHTHYRIDINE DERIVATIVES

McWilliams and coworkers (03JOC467) have studied the catalytic activity and selectivity of bases as catalysts in the [1,8]naphthyridine synthesis from 2-aminonicotinaldehyde **357** (69BSB289, 74JOC720, 80T2359, 01MI1, 01SC1573) and 2-pentanone. As shown in Table 4, many different Brönsted and Lewis bases promote this reaction. The best in terms of acceleration and regioselectivity is pyrrolidine (03JOC467).

Table 4. Screening of catalysts for the reaction between **357** and 2-pentanone



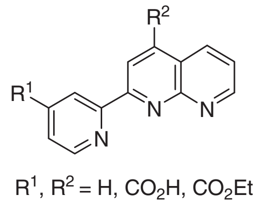
Catalyst	Conversion[a]	Ratio[b]
NaOH	>99	37:63
NaOEt	87	49:51
NH_4OAc	44 (89)	42:58 (40:60)
NH_4OH	18 (28)	44:56 (43:57)
$C_8H_{17}NH_2$	92	55:45
Et_2NH	1 (7)	(74:26)
Piperidine	11 (39)	89:11 (88:12)
Morpholine	8 (26)	78:22 (77:23)
Proline	5 (64)	83:17 (70:30)
Pyrrolidine	97	86:14
Azetidine[c]	98	54:46
2-Methylaziridine	1 (9)	(41:59)
DBU	69 (98)	24:76 (27:73)
Et_3N	0	N/A
H_2SO_4[d]	0	N/A

[a] Mole percent conversion to **358** + **359** after 23 h at 23°C, determined by HPLC. Values in parentheses were recorded after an additional 23 h at 70°C.

[b] Ratio of **358**:**359** after 23 h at 23 °C, determined by GC. Values in parentheses were recorded after an additional 23 h at 70 °C.

[c] Reaction run with 1.1 equiv. of azetidine hydrochloride and 1.0 equiv of DIPEA.

[d] Reaction with 0.05 equiv of H_2SO_4.



(360)

Figure 15.

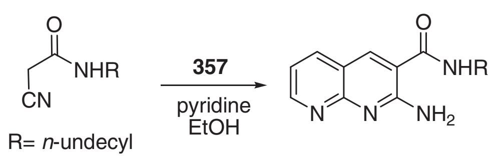
In subsequent studies, a similar interaction between 2-aminonicotinaldehyde and unsymmetrical functionalized alkan-2-ones was reported. This method typically provides substituted [1,8]naphthyridines in high yields under standard hydroxide-catalyzed conditions (01OL1101, 07MC25, 07RCB1911).

2-(Pyrid-2′-yl)-1,8-naphthyridines containing a carboethoxy group appended at the 4- and 4′-positions were prepared as photosensitizers **360** (Figure 15). Complexation with Ru(II) and NaNCS leads to [Ru $(L)_2(NCS)_2$] and subsequent hydrolysis of the ester affords a carboxylic acid dye-sensitizer utilized in solar cells (06IC10131).

2-Aminonaphthyridine **362** is obtained from 2-aminonicotinaldehyde **357** and the cyano amide **361** in 62%. A ratio of 4:1 for **362** and octahydroxypyridine[4]arene in polar media forms a gel-like system (Scheme 78) (02EJO2120).

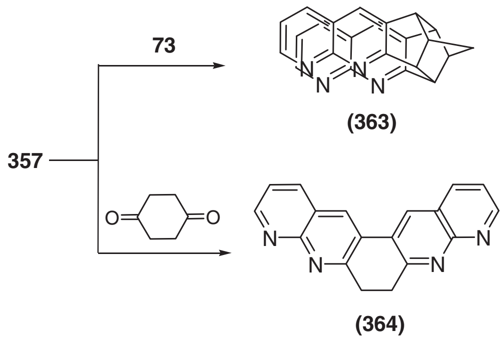
Marchand et al. (88TL6681) and Lim et al. (91JOC1492) have synthesized bis(carboxamidonaphthyridine) **363** in 21% yield from tetracyclo-$[6.3.0.0^{4,11}.0^{5,9}]$undecane-2,7-dione (**73**) and 2 equivalents of *o*-aminonicotinaldehyde **357** followed by treatment with octanoic anhydride. **357** and 1,4-cyclohexanedione yields bis(aminonaphthyridine) **364** in 49% yield (Scheme 79) (75JOC3407).

Application of the Reimer–Tiemann reaction to 2,6-diaminopyridine **365** affords a 26% yield of 2,6-diaminopyridine-3-carboxaldehyde **366** and a small amount (4%) of 2,6-diaminopyridine-3,5-dicarboxaldehyde.



(361) (362)

Scheme 78.

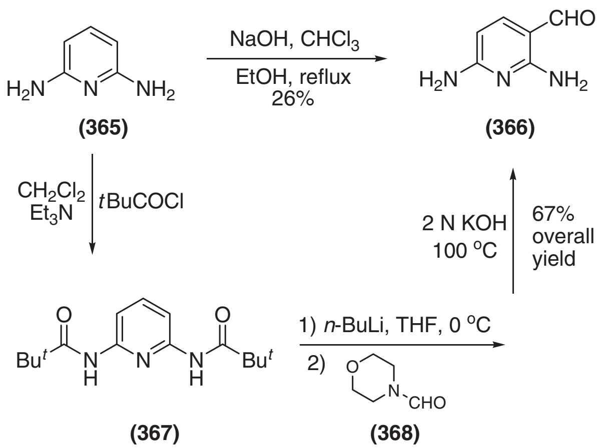


Scheme 79.

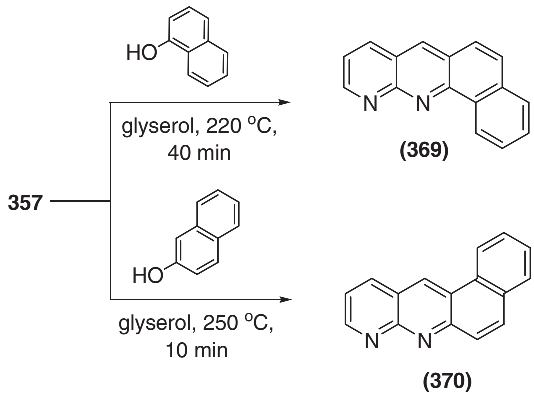
Alternatively, conversion of **365** to 2,6-bis(pivaloylamino)pyridine **367** *via* lithiation with *n*-butyllithium followed by treatment with *N*-formylmorpholine **368** and hydrolysis of the latter produces **366** in 67% overall yield (Scheme 80) (93JOC6625).

Pyridine **366** with a variety of activated and nonactivated ketones yields 2-amino-1,8-naphthyridines (09H(79)411) and bis(2-amino-1,8-naphthyridines) in moderate to good yields, providing a convenient synthesis for useful building blocks utilized as new host–guest and self-assembling systems (93JOC6625).

2-Aminonicotinaldehyde **357** with *α*- and *β*-naphthols leads to naphtho [1,2-*b* and 2,1-*b*][1,8]-naphthyridines **369**, **370** in a single synthetic procedure. The oxidation of the naphthonaphthyridines **369**, **370** with peroxy



Scheme 80.



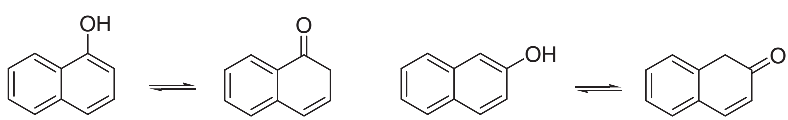
Scheme 81.

acids affords novel products such as a seven-membered oxazepine (Scheme 81) (96H(43)2139). Replacement of 2-aminonicotinaldehyde with 2-aminobenzaldehyde leads to quinoline derivatives (56MI315, 73CCC3862).

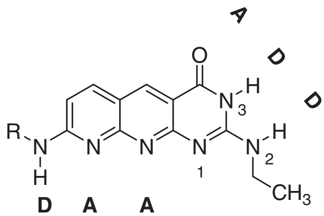
These naphthols tautomerized *via* the Ullmann–Fetvadjian reaction to ketones (Scheme 82) (1903CB1027, 49JCS670, 50JCS1146, 51JCS2871, 67JCS213).

Heterocyclic unit **371** containing complementary donor–donor–acceptor (DDA) and acceptor–acceptor–donor (AAD) hydrogen bonding arrays at an angle of about 60° was also designed to self-assemble into a hexamer. This complex is an especially stable, hexameric, disk-shaped aggregate (**371**)$_6$ containing 18 hydrogen bonds formed by the pairing of self-complementary DDA and AAD sites. Additionally, six (secondary) hydrogen bonds may be present in (**371**)$_6$ because the N2-H group can serve as a long-range donor to N1 (Figure 16) (98JA9092, 02JA13757).

Bell and Liu prepared newly designed heterocyclic receptors *via* the Friedländer condensation and used them in the complexation of urea. For example, urea complex **376** (88JA3673) is at least 10 times more stable than the best crown ether receptor that also solubilizes solid urea in chloroform (Scheme 83) (90AGE931).

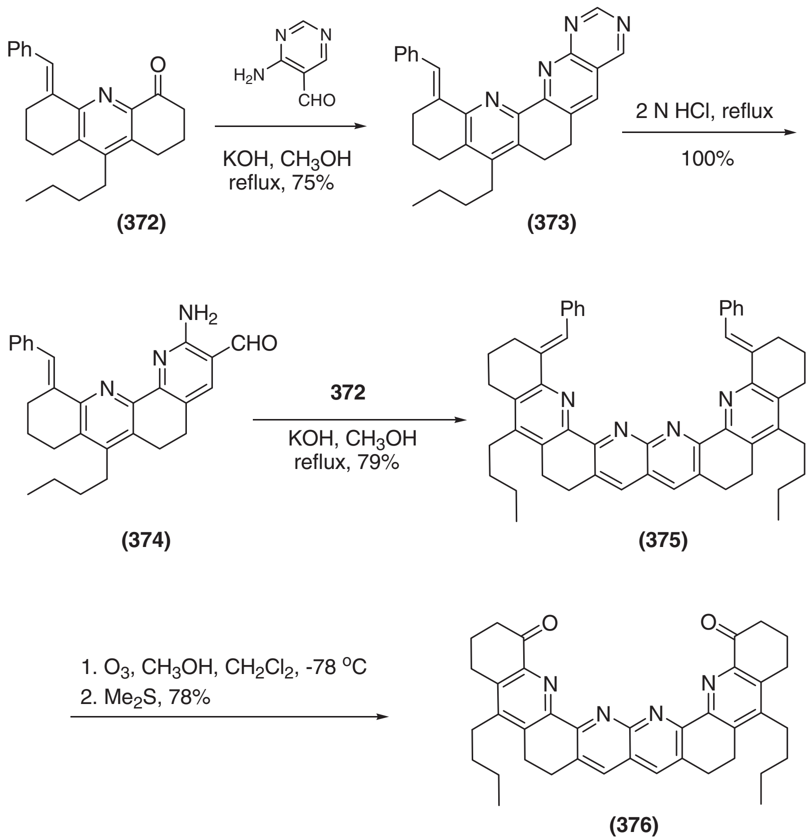


Scheme 82.

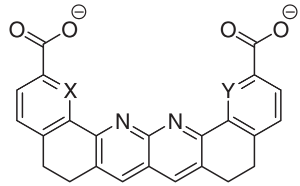


(371)

Figure 16.



Scheme 83.



(377)

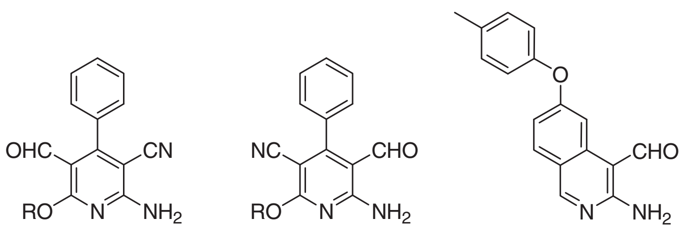
X = Y = N
X = N, Y = CH
X = Y = CH

Figure 17.

In the another attempt, 1,8-naphthyridines **377** were prepared *via* 4-aminopyrimidine-5-carboxaldehyde (Figure 17) and used as receptors in complexation studies with several guanidinium and ammonium guests, including derivatives of the amino acids arginine and lysine (02JA14092).

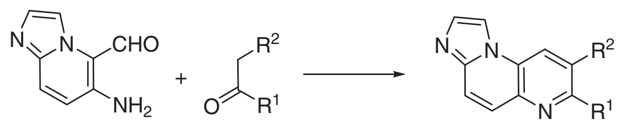
o-Aminoaryl-(heteroaryl)-aldehydes with acetoacetic ester lead to pyridine ring annulation. For example, isomeric *ortho*-aminoformylquinolines (77CR459, 81JHC925, 82JHC1289, 02ASJC1303) and *ortho*-aminoformylpyridines (54JA596, 57JOC138) are used to synthesize various isomeric naphthyridines. 2-Aminonicotinaldehyde **357** and ketones are widely used for the preparation of 1,8-naphthyridine derivatives (74JHC151, 80TL4485, 86JHC689, 93AJC987, 94AF809, 94JCM268, 03EJI3547), in some cases with HCl (77JHC685, 90JOC4744), NaF (03SC3131), NaOH (04JOC1959), pyridine/MW (02IJB215), PTSA (03IJB1170), LiCl/MW (06IJB302), NH_4OAc (06JIB1051), montmorillonite K-10 (06JIB2749), or Al_2O_3 (07JIB1721).

An efficient method prepares a variety of polycondensed 1, 8-naphthyridines from pyridines **378**, **379**, and **380** and cyclic and heterocyclic ketones (Figure 18) (84ACP268, 91AJC481, 95H(41)1001, 96H(43)53, 03EJM265, 08T3446).



(378) **(379)** **(380)**

Figure 18.



(381) (382)

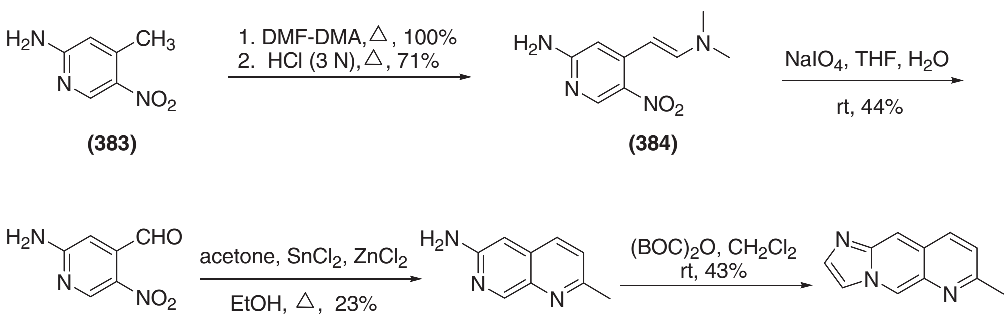
Scheme 84.

Imidazo-[1,2-*a*]-naphthyridines **382** can be obtained from imidazo-[1, 2-*a*]-pyridine **381** in 5–65% yield (Scheme 84). They were evaluated for their antitumor activity by the NCIs *in vitro* human tumor cell line screening panel. Among them, pentacyclic derivatives exhibit *in vitro* activity comparable to the anticancer agent amsacrine (08EJM2505).

Pyridinoimidazo-[1,2-*a*]-pyridine **387** is synthesized from 2-amino-4-methyl-5-nitropyridine **383** through a linear cyclization. A mixture of DMF–DMA forms the agent for vinylamine functionalization **384**. The formation of pyridinoimidazopyridine **387** from **383** involves formation of pyridine-fused naphthyridine by the McNaughton modified method in a single step using $SnCl_2$ and $ZnCl_2$ and finally reductive cyclization of **385** followed by treatment of the resulting adduct **386** with $(Boc)_2O$ (Scheme 85) (07TL8392).

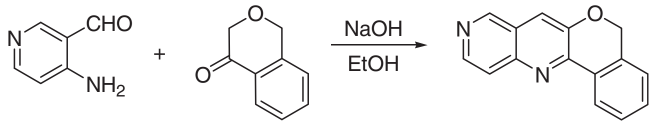
1,6-Naphthyridine **390** is synthesized from 4-amino-3-pyridinecarbaldehyde **388** and isochroman-4-one **389** in 50% yields (Scheme 86) (03CJOC466).

Methcohn and Hayes prepared 2-amino-7-methylquinoline-3-carbaldehyde (Figure 19) and employed it successfully in 1,6-naphthyridine preparations *via* Friedländer's method (82TL1613).



(383) (384) (385) (386) (387)

Scheme 85.



Scheme 86.

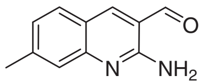


Figure 19.

3-Acetyltropolones **391** reacts with 2-amino-3-pyridinecarbaldehyde to afford the 3-(1,8-naphthyridin-2-yl)tropolone in excellent yields. In a similar manner, 1,6-naphthyridin-2-yl- **392**, 1,7-naphthyridin-2-yl- **393**, 6-pyrido[2,3-*b*]pyrazinyl- **396**, and 1-methyl-6-pyrazolo[5,4-*b*]pyridyl- **398** substituted tropolones are prepared in 72–92% yields (Scheme 87) (96JHC389).

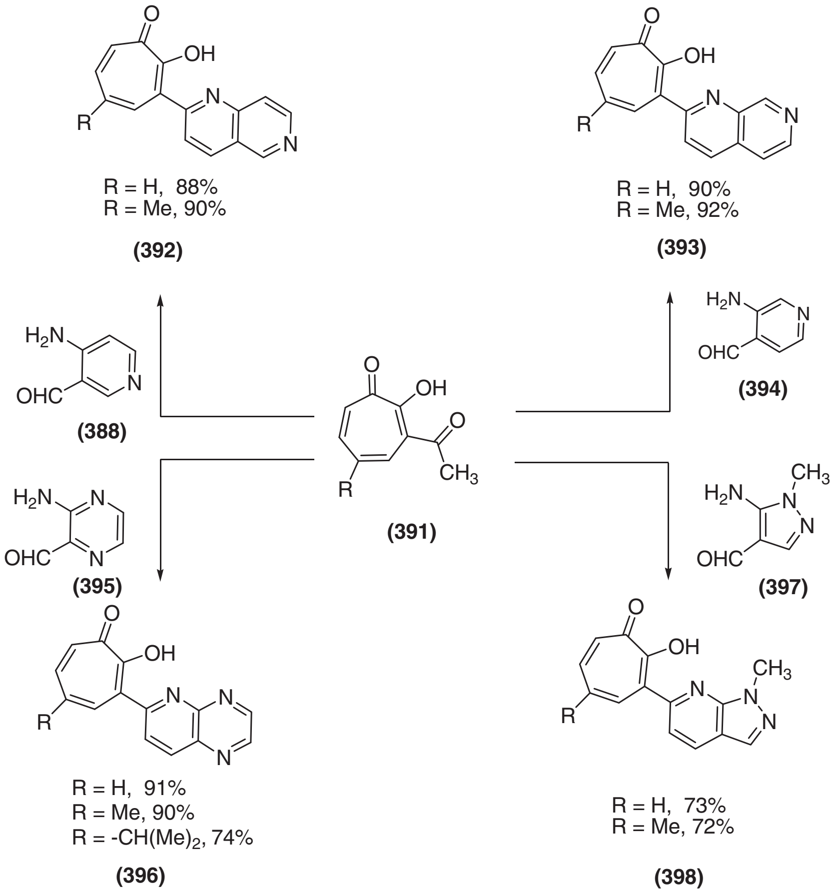
A new class of tricyclic models such as **401** was also based on a Friedländer reaction between chiral piperidine-2,4-diones **400** and azomethine **399** (Scheme 88) with yields of about 70%. Alkylation of the lactam allows the introduction of various pendant arms on the chiral cyclic inducer. Compound **401** is used for the preparation of various enantioselective NADH mimics (03T4911).

Acetyl- and propionylferrocene with various aromatic *o*-amino aldehydes give aza-aryl-substituted ferrocenes. Similarly, 1,1′-diacetyl- and 1,1′-dipropionyl ferrocene analogs **402** provide 1,1′-bis(azaaryl)-substituted derivatives **403** (Scheme 89) (92JOC3780).

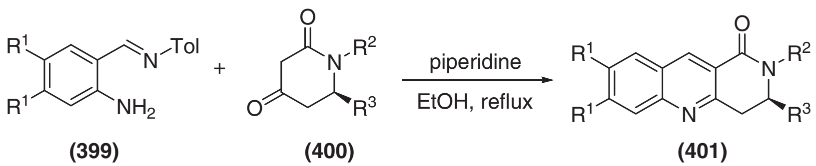
Several 1,5- and 1,7-naphthyridines are prepared from suitable ketone and 2-amino-3-picolinaldehyde **357** or 3-aminoisonicotinaldehyde **394**, respectively (56JCS1045, 60JCS1370, 75CR381). For example, 2,2′-bi[1,7-naphthyridine] **405** is also synthesized from **394** and diacethyl **404** in 40% yields (Scheme 90) (76JHC387).

Queguiner and his group used lithiated pivaloylaminopyridines **407** with aldehydes to synthesize amino ketones. Oxidation of alcohols **408** with CrO_3 or MnO_2 leads to *o*-aminopyridyl ketones **409** (Scheme 91). Amides **409** are hydrolyzed to the amines **410** with HCl. Both **409** and **410** were successfully utilized in Friedländer condensations with various ketones that produced naphthyridine derivatives (89JHC105).

Di-1,8-naphthyridines are synthesized from *o*-aminonicotinaldehyde derivatives **411**, **412**, **413** (79MM803), **379** (08T3446), or diketones **414**



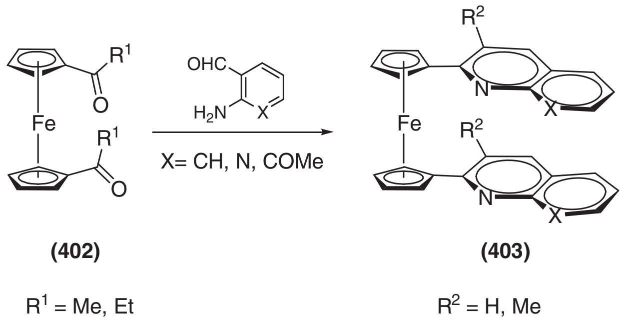
Scheme 87.



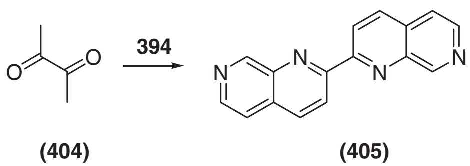
Scheme 88.

(08T3446), **119**, **124**, **415**, **416**, **417** (79MM803), 1,2-cyclohexanedione **418-420** (84JOC2208, 04IC6195), **332** (08IC990), and **86**, **87** (01EJO863) (Figure 20).

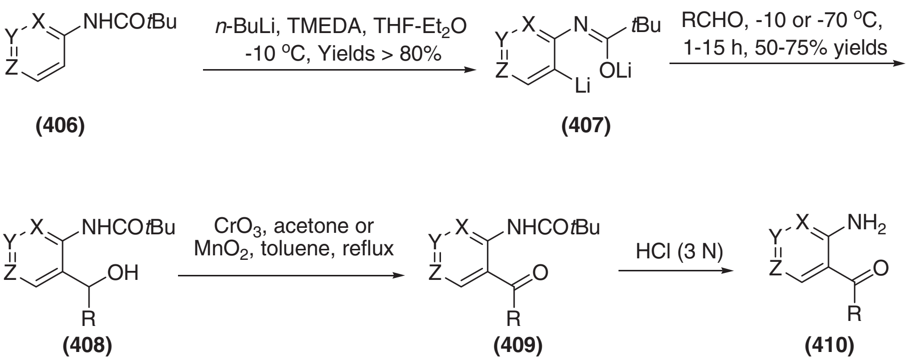
4-Aminopyrimidine-5-carboxaldehyde **421** (67CB3664) and 1,3-cyclohexanedione react in a 1:2 molar ratio to form **422** in quantitative yield.



Scheme 89.



Scheme 90.



Scheme 91.

The transformation of **422** in 2 N HCl results in a very efficient annulation sequence leading to four linearly fused rings **424** in high yield from a monocyclic starting material. **422** in 0.01 N HC1 gives 2-amino-5-oxo-5,6,7,8-tetrahydroquinoline-3-carboxaldehyde **423**. The accumulation of functional groups with this simple molecular framework is noteworthy. Pyrolysis of **422** results in the elimination of 1,3-cyclohexanedione

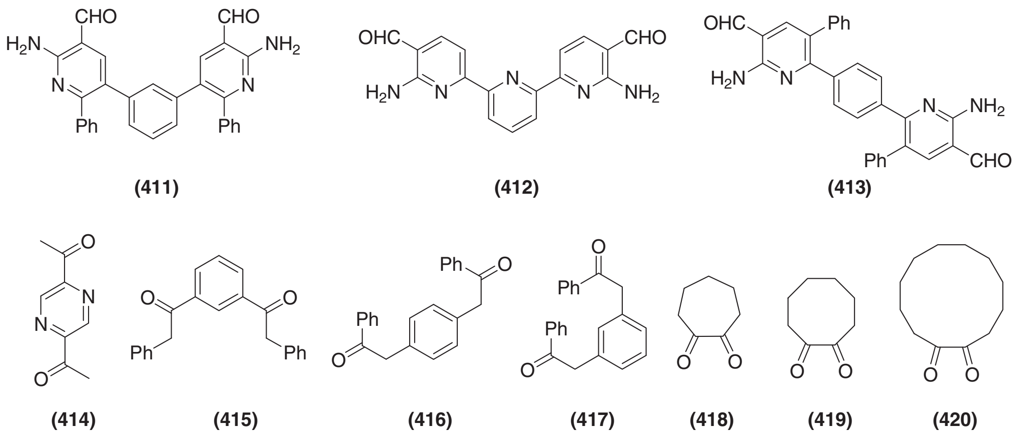
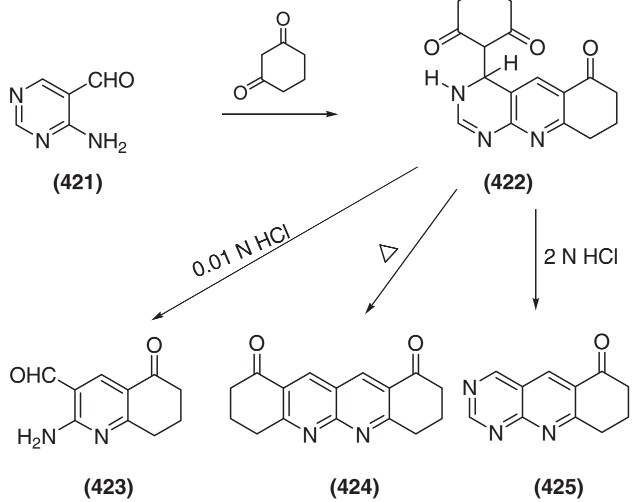


Figure 20.

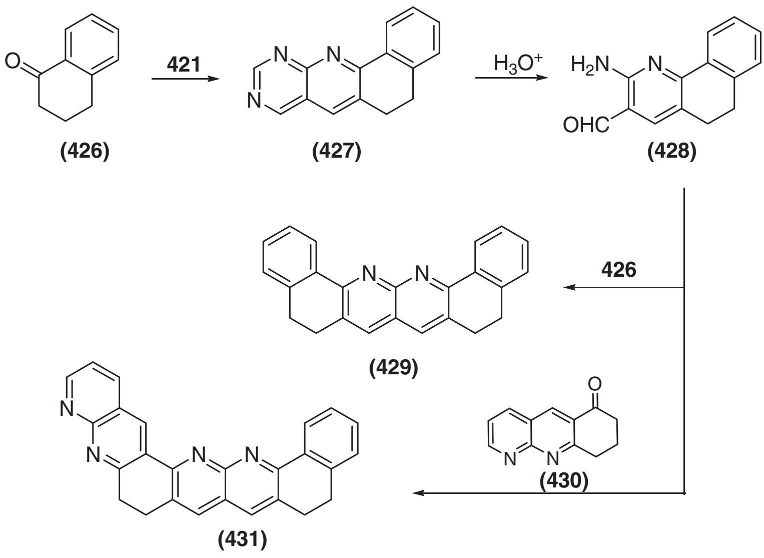


Scheme 92.

with subsequent formation of 6-oxo-6,7,8,9-tetrahydropyrimido[4,5-*b*] quinoline **425** (Scheme 92) (76JOC1058).

ortho-Aminoaldehydes, readily obtained from aromatic cyclic ketones and **421**, react with aromatic ketones and lead to symmetrically **429** and nonsymmetrically **431** fused 1,8-naphthyridines with equal ease (Scheme 93) (75JOC2566).

Three isomeric octacyclic compounds **432**, **433**, and **434**, each composed of three 1,8-naphthyridine moieties, were synthesized from



Scheme 93.

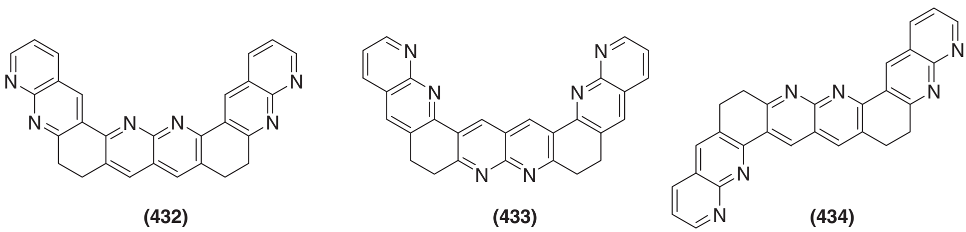
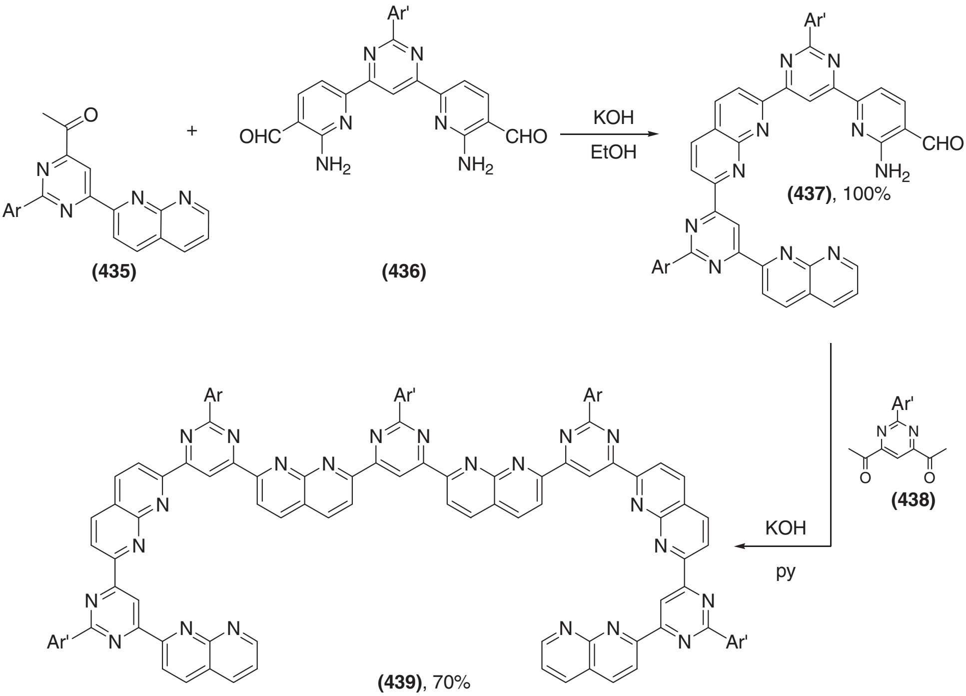


Figure 21.

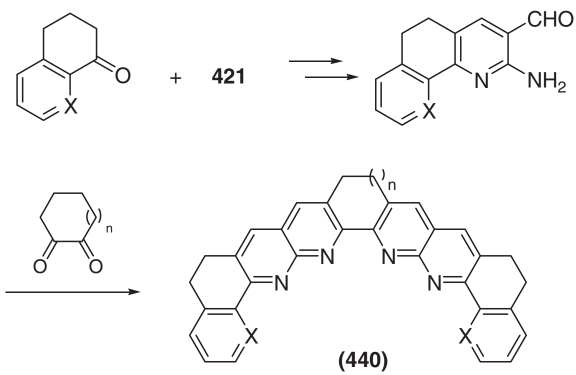
1,3-cyclohexanedione, 2-aminonicotinaldehyde **357**, and 4-aminopyrimidine-5-carboxaldehyde **421** under different conditions (Figure 21) (79JOC531).

Lehn et al. have reported the synthesis and study of a family of foldamers and macrocycles based on 1,8-naphthyridine and pyrimidine units **439**, whose internal cavity is large enough to accommodate ionic substrates. The study focused on the impact of host–guest binding within a cylindrical environment (Scheme 94) (08JOC2481).

Thummel et al. have used 4-aminopyrimidine-5-carboxaldehyde **421** as a useful synthon for the stereochemically controlled introduction of the polycondensed 1,8-naphthyridines **440** (88JA7894) (Scheme 95) (86IC1675).

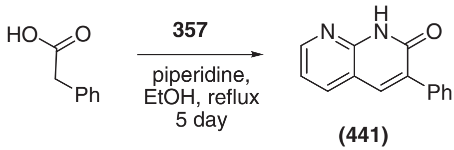


Scheme 94.



X = CH and n = 1, 73%
X = CH and n = 2, 61%
X = CH and n = 3, 78%
X = N and n = 1, 63%
X = N and n = 2, 65%
X = N and n = 3, 88%

Scheme 95.



(441)

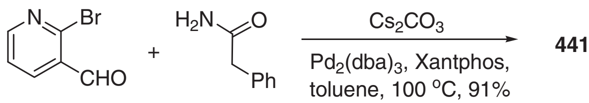
Scheme 96.

3-Phenyl-1,8-naphthyridin-2(1*H*)-one **441** can be synthesized from 2-aminonicotinaldehyde **357** and 2-phenylacetic acid with a catalytic amount of piperidine in ethanol at reflux for 5 days; the yield was about 6% (Scheme 96) (66JOC315).

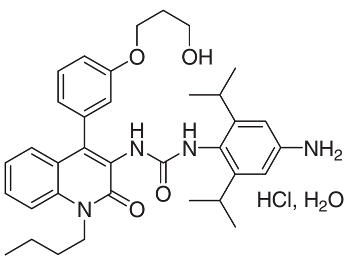
Much later, an alternate synthesis of naphthyridinones and quinolinones was discovered. Palladium-catalyzed amidation of halo aromatic rings with an *ortho* carbonyl group with primary or secondary amides to from substituted naphthyridinones and quinolinones (Scheme 97) (04OL2433).

In the synthesis of SMP-797 **442** as a new potent acyl-CoA cholesterol acyltransferase (ACAT) inhibitor, a modified Friedländer annulation was used (Figure 22) (05T10081).

Oxidation of 3-nitro-4-picoline **443** with selenium dioxide gives 3-nitro-4-pyridinecarboxaldehyde **444**. Condensation of the aldehyde with *p*-toluidine gives *N*-(3-nitro-4-picolylidene)-*p*-toluidine **445**, which is reduced to *N*-(3-amino-4-picoly1idene)-*p*-toluidine **446**. Condensation of the latter with cyclohexanone and acetophenone gives 2,10-diaza-

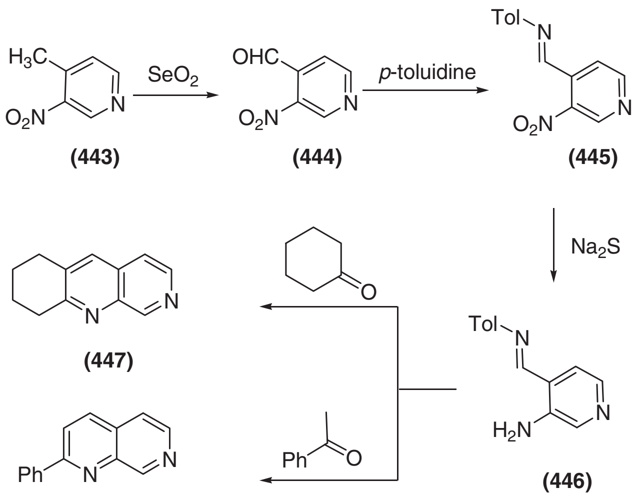


Scheme 97.



(442)

Figure 22.



(443) (444) (445) (447) (446) (448)

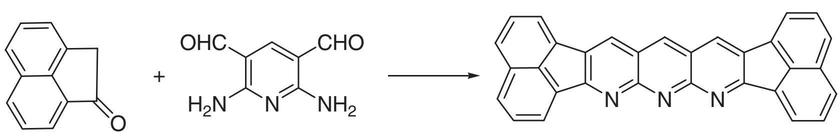
Scheme 98.

5,6,7,8-tetrahydroanthracen **447** and 2-phenyl-1,7-naphthyridine **448**, respectively (Scheme 98) (55JA2438). The imine of **446** undergoes nucleophilic attack by the aldol and then *p*-toluidine serves as the leaving group.

8. ANTHYRIDINE PREPARATION

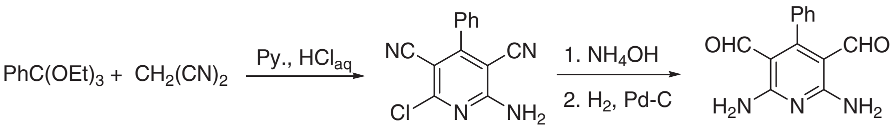
Condensation of 2,6-diaminopyridine-3,5-dicarboxaldehyde with ketones affords substituted and fused 1,9,10-anthyridines. For example, **450** with acenaphthenone **449** gave the fused, fully aromatic diacenaphtho[1, 2-*b*:1′,2′-*i*] 1,9,10-anthyridine **451** in 65% yield (Scheme 99) (77JOC3410).

Murray et al. prepared 2,6-diamino-4-phenyl-3,5-pyridine dicaboxaldehyde **454** as outlined in Scheme 100. Thus, triethylorthobenzoate **452** is heated with two equivalents of malononitrile in pyridine, followed by aqueous hydrochloric acid, to afford **453** in 43% yields (95T635). A similar



(449) (450) (451)

Scheme 99.



(452) **(453)** **(454)**

Scheme 100.

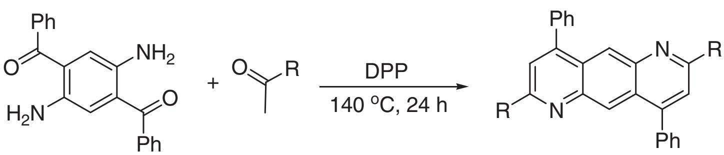
procedure prepares the 4-methylpyridine analog (86JME1080). Aminolysis of **453** with concentrated ammonium hydroxide proceeds in quantitative yield. Catalytic hydrogenation under acidic conditions affords **454** in 32% yield (Scheme 100). Pyridine **454** is used for the preparation of 1,9,10-anthyridines *via* a double Friedländer reaction (95T635). The stability of the hydrogen bonding complexes of these anthyridines was investigated (92JA4010).

9. ANTHRAZOLINES AND POLYANTHRAZOLINES

The synthesis, properties, and electroluminescent device applications of diphenylanthrazoline molecules represented by **456** was reported (Scheme 101). Analogs of **456** emit blue light with fluorescence quantum yields of 58–76% in dilute solution, whereas they emit yellow-green light as thin films. The diphenylanthrazolines are utilized in emissive layers for light-emitting diodes and give off a yellow light with a maximum brightness of 133 cd/m^2 (03JA13548).

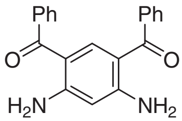
A similar reaction as in Scheme 101 is also catalyzed by polyphosphoric acid in yields ranging from 61% to 88% (09DP218).

The condensation of bisketomethylene monomers with either 2,5-dibenzoyl-1,4-phenylenediamine **455** or 4,6-dibenzoyl-1,3-phenylenediamine **457** (Figure 23) is catalyzed by acid to afford high molecular weight polymers containing the anthrazoline and isoanthrazoline units in the polymer main chain. Base is not an effective catalyst. Phenyl substitution on the anthrazoline and isoanthrazoline units increases the solubility of



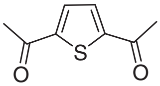
(455) **(456)**

Scheme 101.

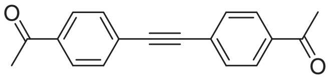


(457)

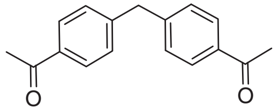
Figure 23.



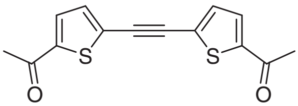
(458)



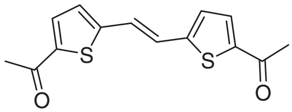
(459)



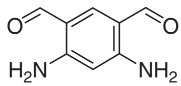
(460)



(461)



(462)



(463)

Figure 24.

these polymers over those in which phenyl substitution is absent. The rod-like character of these polymers, which can be altered by positional isomerism in the chain, has an effect on their solution properties (75PSA2233).

Numerous reports exist where the Friedländer method was employed to synthesize polyanthrazolines (entries 1–8) and polyisoanthrazolines (entries 9–12) from bis-*o*-aminocarbonyl compounds and diketones (Figure 24, Table 5).

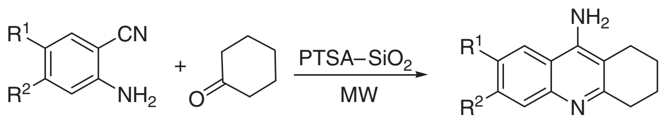
Table 5. Preparation of polyanthrazoline and polyisoanthrazoline

Entry	Bis-*o*-aminocarbonyl	Diketone	References
1	**455**	**100**	(92CM95, 93MM895, 00MM2069)
2	**455**	**104**	(92CM95, 93MM895, 00MM2069)
3	**455**	**106**	(92CM95, 93MM895, 00MM2069)
4	**455**	**458**	(92CM95, 93MM895, 00MM2069)
5	**455**	**459**	(92CM95, 93MM895, 00MM2069)
6	**455**	**460**	(92CM95, 93MM895, 00MM2069)
7	**455**	**112**	(91MM6806)
8	**455**	**461**	(91MM6806)
9	**455**	**462**	(96CM579)
10	**463**	**100**	(69MM286)
11	**463**	**117**	(69MM286)
12	**463**	**124**	(69MM286)

10. TACRINE-LIKE ANALOGS

In 1993 tacrine (9-amino-1,2,3,4-tetrahydroacridine, THA) was the first acetylcholinesterase inhibitor (AChEI) approved in the United States for the treatment of Alzheimer's disease (AD) (99TPS127).

Effects of MW irradiation on the solid-phase synthesis of tacrine and its derivatives were evaluated. Anthranilonitriles **464** with cyclohexanone and PTSA–SiO_2 under MW irradiation gives good to excellent yields of substituted THAs **465** (Scheme 102) (07JHC535).



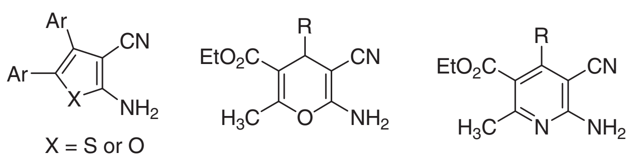
(464) **(465)**

Scheme 102.

Marco and his group reported the synthesis of different tacrine derivatives from functionalized 2-amino-3-cyano-4,5-diarylfurans or thiophene **466** (02AP347), 2-amino-3-cyano-4*H*-pyrans **467** (97BML3165, 02AP347, 04BMC2199), and 6-amino-5-cyanopyridines **468** (01BMC727, 02BMC2077) with cyclic ketones (Figure 25).

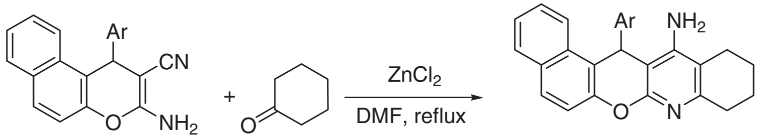
New acridine-type compounds such as **470** can be synthesized from substituted 3-amino-1*H*-benzo[*f*]chromene-2-carbonitrile **469** and cyclohexanone using anhydrous zinc chloride catalyst under reflux (Scheme 103) (08CCL15). Standard reported yields are between 47% and 50%.

Several novel analogs of tacrine **471** and **472** were tested for their ability to inhibit acetylcholinesterase (AChE), butyrylcholinesterase (BChE), and neuronal uptake of 5-HT (serotonin) and noradrenaline (Scheme 104). Changes in the size of the carbocyclic ring of tacrine produced modest potency against cholinesterase enzymes. Addition of a fourth ring resulted in compounds with marked selectivity for AChE over BChE (97JME3516).



(466) **(467)** **(468)**

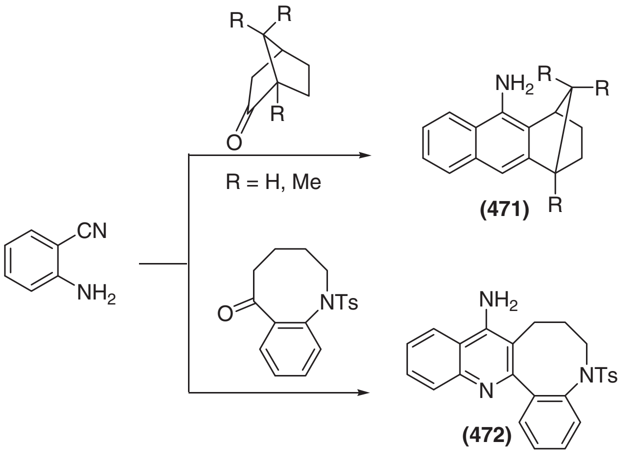
Figure 25.



(469) (470)

Ar = 3-nitrophenyl, 47.2%
Ar = 4-nitrophenyl, 50.4%

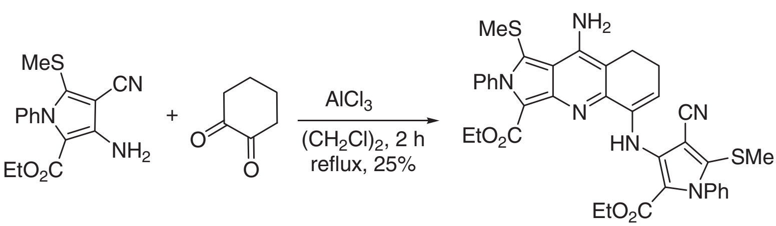
Scheme 103.



(471) (472)

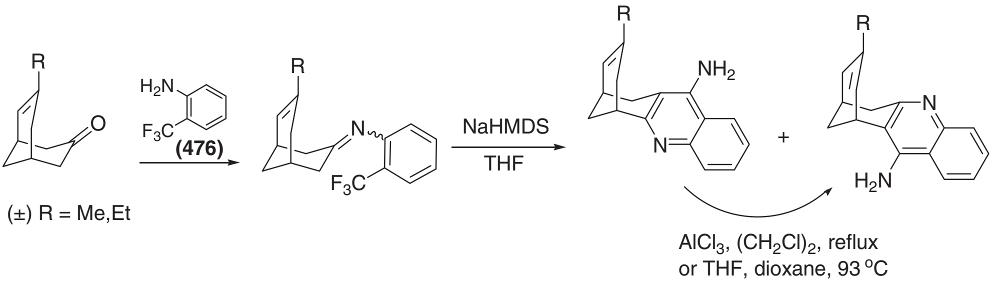
Scheme 104.

3-Amino-4-cyanopyrroles **473** with cyclic ketones are used for the synthesis of substituted 4-azaisoindole tacrine analogs **474** (Scheme 105) (07CJA1). The intermediate ketone is converted to an imine with excess amino nitrile, which is subsequently converted to an enamine.



(473) (474)

Scheme 105.

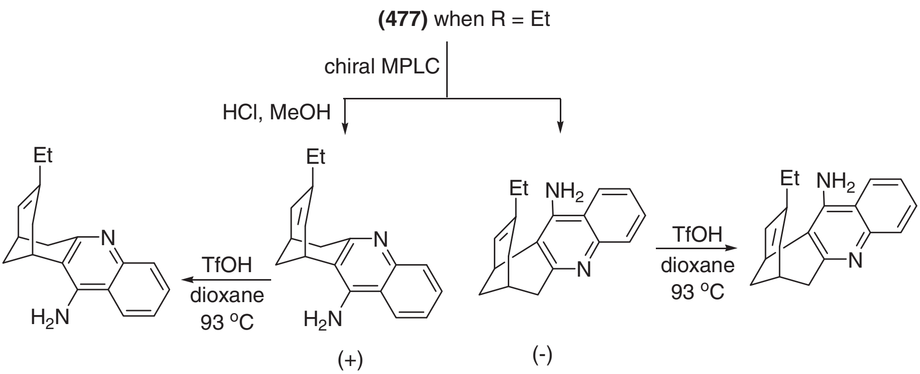


(475) (477)

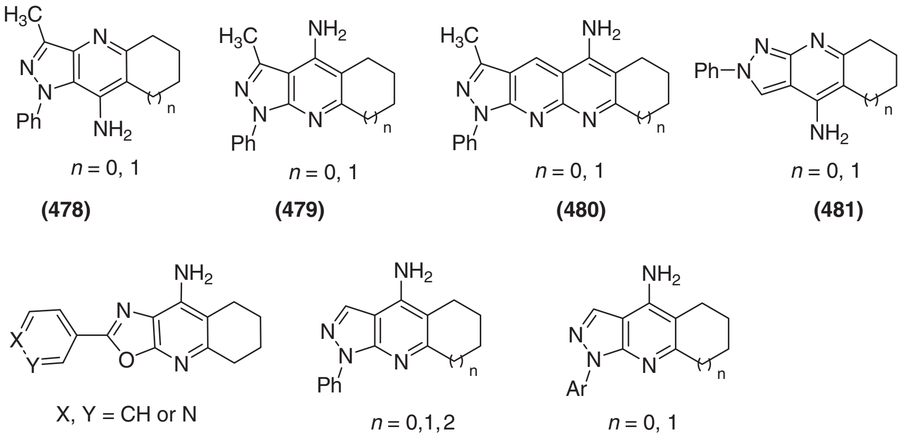
Scheme 106.

(±)-Huprines **477** are easily available *via* the cyclization of racemic enones (±)-**475**, with 2-aminobenzonitrile (94EJM205, 96T5867, 98BMC427, 98TA835, 99JME3227, 00JMC4657, 01JME4733, 09BMC4523) or 2-trifluoromethylaniline **476** (01TA2909) substituted at the fourth- and/or sixth-position. $AlCl_3$ is added as a Lewis acid catalyst and the reagents are refluxed in 1,2-dichloroethane (Scheme 106).

NaHMDS activates the α-carbon of the enamine, which then attacks the labile CF_3 in a benzylic position. The amine group may originate from NaHMDS. The same reactivity of C–F bonds on an aromatic ring was observed in Scheme 30. Racemic 12-amino-6,7,8,11-tetrahydro-7, 11-methanocycloocta-[*b*]quinoline derivatives (*syn*-huprines) substituted at position 9 with an ethyl or methyl group (R = Et or Me) **477** and both *enantio*-enriched forms of the 9-ethyl derivative are readily converted to their *anti*-isomers (huprines). This interconversion is achieved by stereospecific acid-promoted ($AlCl_3$ or triflic acid) isomerization of the endocyclic C=C double bond from the 9(10)- to the 8(9)-position (Scheme 107)



Scheme 107.



(478) **(479)** **(480)** **(481)** **(482)** **(483)** **(484)**

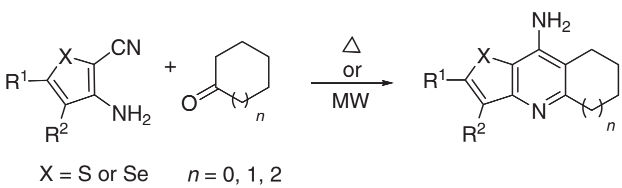
Figure 26.

(01TA2909). The two *anti*-isomers were separated using chiral medium pressure liquid chromatography (MPLC).

A new family of tacrine (THA) analogs **478-481** (03JME1144), **482** (07H(71)2249), **483** (07CCL636), and **484** (08SC4369) containing the azaheterocyclic oxazolo, pyrazolo-[4,3-*d*]pyridine or pyrazolo[3,4-*b*][1,8]naphthyridine systems as isosteres of the quinoline ring of THA was reported (Figure 26).

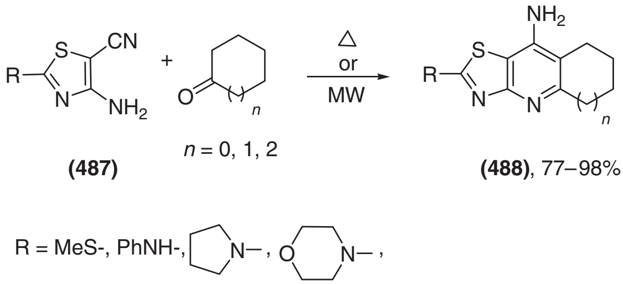
New 3-amino-2-selenophenecarbonitriles (**485** when X = Se) (08S1600) or 3-amino-2-cyanothiophenes **485** when X = S (07S1027, 08JHC853) were condensed with cyclanones to afford, in one-step, tacrine analogs **486** under heating and MW irradiation (Scheme 108).

Kirsch prepared substituted 4-amino-5-cyano-1,3-thiazoles **487** and used them successfully for the synthesis of new substituted [1,3]thiazolo



(485) **(486)**

Scheme 108.



Scheme 109.

[4,5-*e*]pyridines **488** in one-step *via* Friedländer methodology under thermal and MW conditions in good to high yields (Scheme 109) (08T9309).

11. CONCLUSIONS

Since 1882, numerous chemists have focused on the Friedländer annulation as a straightforward method for the synthesis of azaheterocyclic compounds. As has been emphasized, Friedländer method is a well-appreciated tool for the generation of large libraries of related heterocyclic compounds that are to be screened for pharmacological activity etc. This review has dealt with the synthesis of quinolines, pyridines, campthotecines, acridines, tacrines, phenanthrolines, naphthyridines, anthyridines, and anthrazoline derivatives, including several other heterocyclic compounds.

NOT ADDED IN PROOF

After the submission of this manuscript the following references related to Friedlander method have appeared in the literature (09ARK9, 09ARK28, 09BKC3061, 09CJC1122, 09CR1028, 09CRE2652, 09H(78)1573, 09IC6459, 09IJB746, 09JCM649, 09JOM3215, 09M1195, 09M1343, 09MC303, 09MI241, 09MI867, 09MI1122, 09MI1448, 09MI1482, 09S3819, 09SC3293, 09SC3527, 09T2455, 09T8524, 09TL5805, 10ARK305, 10BML3295, 10BSJ672, 10CPB212, 10EJM682, 10EJM2726, 10H(81)689, 10H(81)1923, 10HCA242, 10IJB253, 10JHC287, 10JOC1188, 10JOC3488, 10MI, 10MI95, 10MI100, 10MI137, 10MI140, 10MI875, 10MI1072, 10MI1180, 10MI1250, 10MI1842, 10OBC1894, 10PSS319, 10S1678, 10SC120, 10SL2597, 10T7399, 10T7544, 10TL2342, 10CR1988, 10MI13, 10MI616, 10OL5064, 10T9319, 10JOC7233, 10MI1430).

ACKNOWLEDGMENTS

We thank the Alzahra University and Bu-Ali Sina University, and the Elite National Foundation (Iran) for financial support. Mr. Fitsum Tesfa is thanked for his assistance with obtaining the references in electronic form and Dr. Grant Boyle for proofreading the manuscript.

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CHAPTER 3

Organometallic Complexes of Aminopyridines

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Advances in Heterocyclic Chemistry, Volume 102
ISSN 0065-2725, DOI 10.1016/B978-0-12-385464-3.00003-0

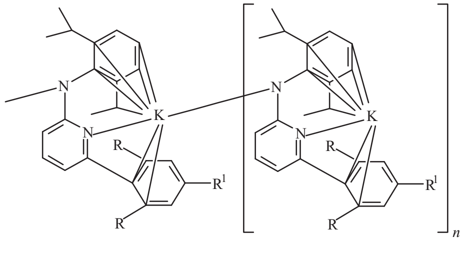
1. INTRODUCTION

This chapter is a continuation of a short series on organometallic compounds represented by the chelates of pyridine derivatives. In the first part, the N(O,S,Se)-chelates were characterized (09AHC(98)225). Herein, organometallic compounds of various amino derivatives of pyridine and its benzannulated forms are considered. The range of ligands includes 2-amino- or 2,6-diaminoderivatives where the amino substituent(s) is/are adjacent to the pyridine nitrogen heteroatom, 3- and 4-aminoderivatives, di- and tri-2-pyridylamines, aminomethyl compounds, carboxamides, and some specific ligands. Only organometallic compounds are considered, those with a metal-carbon bond. A wide scope of coordination compounds is omitted. Material for several ligand groups is systematized by periodic groups, and in conclusion coordination modes are highlighted.

2. DERIVATIVES OF PYRIDINE AND BENZANNULATED PYRIDINES WITH ADJACENT AMINO GROUPS

2.1 Nontransition metals

Deprotonation of (2,6-di-*i*-propylphenyl)(6-(2,4,6-tri-*i*-propylphenyl)pyridin-2-yl)amine and (2,6-di-*i*-propylphenyl)(6-(2,6-dimethylphenyl)pyridin-2-yl)amine using potassium hydride leads to crystalline organometallic polymers of type **1** ($R = R^1 = i$-Pr; $R = Me$, $R^1 = H$) where the aryl substituents also participate in coordination (04EJI3297). 2-Pyridylaniline reacts with trimethylaluminum to yield $[Me_2Al(N(2\text{-}C_5H_4N)Ph)]$ (01JCS(D)2838). The latter with $(Me_3Si)_2NLi$ affords $Li[Me_2Al(N(SiMe_3)_2)(N(2\text{-}C_5H_4N)Ph)]$. The reaction of *t*-butyl lithium with $[Me_2Al(N(2\text{-}C_5H_4N)Ph)]$ in toluene gives $[Li_8(H)(N(2\text{-}C_5H_4N)Ph)_6]^+[Li(Me_2Al(t\text{-}Bu_2)_2)]^-$ (99AGE3367). Zirconium complexes based on the deprotonated amino ligands formed from isoquinoline and 2-aminonaphthalene (L), $[ZrL_2(NMe_2)_2]$, with trimethylaluminum produce chelates **2** (R = Me, $PhCH_2$, Ph) (04OM5885). Lithium salt $(2\text{-}C_5H_4N)(C_6F_5)NLi$ and $[Me_3SbX_2]$ (X = Cl, Br) give monoamides $[Me_3XSb(N(C_6F_5)(2\text{-}C_5H_4N))]$ or bis(amide) $[Me_3Sb(N(C_6F_5)(2\text{-}C_5H_4N))_2]$ where coordination occurs *via* the imine nitrogen atom (07EJI5684).



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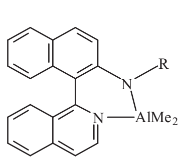

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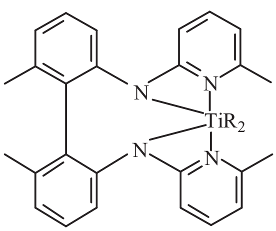
2.2 Titanium group

Reaction of [Ti(L)Cl_2] (H_2L is a biaryl-bridged aminopyridine) with methylmagnesium bromide gives the dimethyl **3** (R = Me) (03OM2972). Dibenzyl **3** (R = CH_2Ph) is prepared from H_2L and [Ti(CH_2Ph)$_4$]. The zirconium(IV) chloride complex of the deprotonated 4-methyl-2-((trimethylsilyl)amino)pyridine (LH) [Zr(η^2(*N,N*)-L)$_3$Cl] reacts with methyl lithium, phenyl lithium, and phenylethynyl lithium to give the σ-alkyl, -aryl, and -alkynyl [Zr(η^2(*N,N*)-L)$_3$Me], [Zr(η^2(*N,N*)-L)$_3$Ph], and [Zr(η^2(*N, N*)-L)$_3$C≡CPh], respectively (96OM1071). In a similar way, [ZrL_3(C≡C-C≡CMe_3Si)] and [ZrL_3Me_2] can be prepared (97CB789). *N*-Adamantyl-2-aminopyridines (HL) readily form [ZrL_2X_2] (X = CH_2Ph, CH_2Bu-*t*) (00CC2099). 2-Arylamino-6-methylpyridine with [Zr(CH_2R)$_4$] (R = Ph, *t*-Bu) yields chelates **4** (04JCS(D)2257). If in 2-arylaminopyridine Ar = 2,4,6-$Me_3C_6H_2$ and X = CH_2Ph, the product is [Zr(L)$_3$(CH_2Ph)]. In contrast, with X = CH_2Bu-*t*, [Zr(L)$_2$(CH_2Bu-*t*)$_2$] is formed. Similarly, pyridin-2-yl-(1,2,3,4-tetrahydronaphthalene-1-yl)amine, 6-methylpyridin-2-yl-(1,2,3,4-tetrahydronaphthalene-1-yl)amine, (1-phenylethylpyridin-2-yl)amine, and (6-methylpyridin-2-yl)(1-phenylethyl)amine (HL) form [ZrL_2R_2] (R = CH_2Ph, CH_2Bu-*t*)$_2$ (04JCS(D)4050). [M(L)(CH_2Ph)$_3$] (M = Zr, Hf, L is a sterically demanding 2-aminopyridinate) is the result of the reaction between 2-aminopyridines with tetrabenzyl zirconium or hafnium (08EJI5088). Further treatment of the products with B(C_6F_5)$_3$ affords the zwitter ionic dibenzyls where the phenyl ring of the B-bound benzyl group is η^6-coordinated. Zirconium(IV) dichloride complexes of (2,6-di-*i*-propyl-phenyl)-(6-(2,6-dimethylphenyl)pyridin-2-yl)amine and (2,6-di-*i*-propyl-phenyl)-(6-(2,4,6-tri-*i*-propyl-phenyl)pyridin-2-yl)amine (LH), [ZrL_2Cl_2] react with methyl lithium to yield alkyls **5** (R = Me, R^1 = H; R = R^1 = *i*-Pr) (07JOM4569). With B(C_6F_5)$_3$ and [PhNMe_2H](B(C_6F_5)$_4$), cationic zirconium-methyl **6** (X = MeB(C_6F_5)$_3$, B(C_6F_5)$_4$) result.



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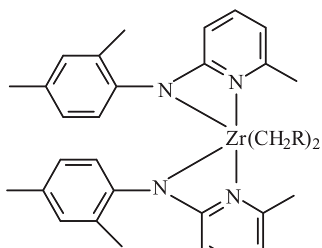

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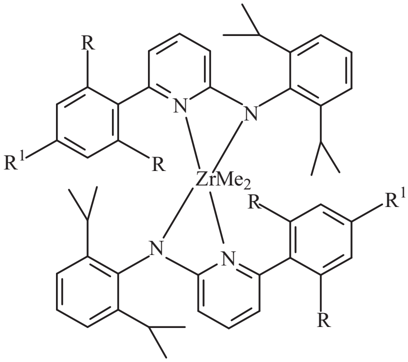

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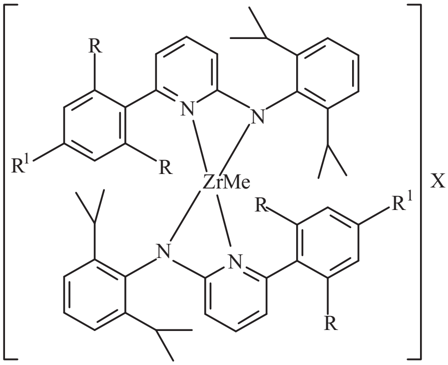

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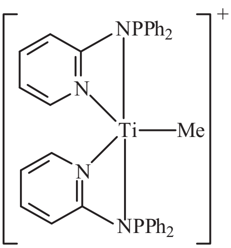
Complex of 2-((diphenylphosphino)amido)pyridine (HL), [Ti(L)$(NEt_2)_2$] in the process of activation by methylalumoxane forms the titanium-methyl cationic, for example, **7** (05OM3255, 07JCS(D)5623). 1,2-Insertion of the double bond of phenylmethylenecyclopropane into the Ti=N bond leads to azatitanacyclobutane **8** (05OM5495). Titanium complex [$TiCl_2L_2$] (LH = 2-((2,6-di-*i*-propylphenyl)amido)pyridine) in magnesium in the presence of bis(trimethylsilyl)acetylene gives the alkyne **9** (08EJI2377). The latter inserts acetone in hexane to yield **10**.



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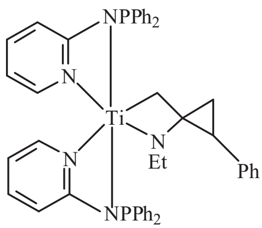

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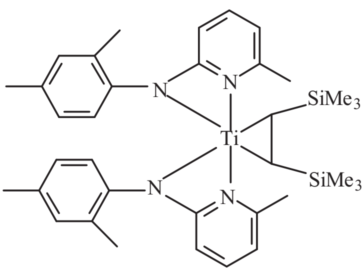

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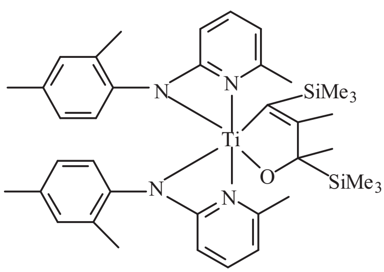

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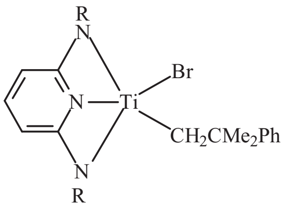
Pyridine diamide ligands L $[2,6\text{-}(RNCH_2)_2NC_5H_3]^{2-}$ (R = 2,6-di-*i*-propylphenyl, 2,6-dimethylphenyl) have been synthesized and give rise to the mono(alkyl) and bis(alkyl) complexes prepared from [$LTiCl_2$] and various Grignard reagents, for example, **11** (96OM5085). Titanium complexes bearing the pyridine diamide ligands $[2,6\text{-}(RNCH_2)_2NC_5H_3]^{2-}$ (R = 2,6-di-*i*-propylphenyl, R = 2,6-dimethylphenyl) have been synthesized. Reduction of the dichloride precursors with sodium amalgam in the presence of alkynes yields titanacyclopentadienes **12** (R^1 = Ph, Et, *n*-Pr, $SiMe_3$) (97OM1491).



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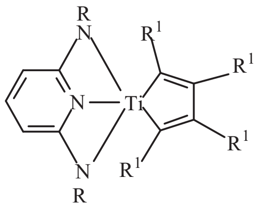

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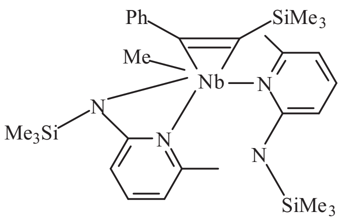
2.3 Vanadium and chromium group

2-Aminopyridine and 2-amino-4,6-dimethylpyridine (HL) react with $[(\eta^5\text{-}C_5H_4SiMe_3)_2NbH_3]$ in tetrahydrofuran (THF) at elevated temperature to yield complexes where the deprotonated aminopyridinate ligands are $\eta^2(N,N)$-coordinated $[(\eta^5\text{-}C_5H_4SiMe_3)_2Nb(\eta^2(N,N)\text{-}L)]$ (00ICA(300)131). Niobium alkyne complexes, for example, **13**, facilitate polymerization of ethene in the presence of methylalumoxane as well as the selective dimerization of 1-butene (98AGE3363, 03CRV283). Lithium salts of (4-methylpyridin-2-yl)(trimethylsilyl)amine and (6-methylpyridin-2-yl)(trimethylsilyl)amine with pentabenzyltantalum afford $\eta^3(N,N,N)$-coordinated tribenzyltantalum(V) followed by toluene elimination (06EJI2683). The lithium salt of (2,6-di-*i*-propylphenyl)(pyridin-2-yl)amine gives such a product with tribenzyltantalum dichloride. $[(\eta^5\text{-}Cp)_2Mo(\eta^2(N,N)\text{-}2\text{-}NHNC_5H_4)](PF_6)$ contains the deprotonated *N,N*-coordinated 2-aminopyridinato ligand (87JOM(320)53). 2-Aminopyridine with $[W(CO)_5(THF)]$ gives $[W(CO)_5(2\text{-}NH_2py)]$ coordinated *via* the pyridine nitrogen atom (01IC1993, 01ICC760) that can be deprotonated with sodium hydride to yield a mixture of the dianionic dinuclear **14** and monoanionic **15** isolated as sodium salts. Photolysis of excess 2-aminopyridine and $[W(CO)_6]$ in THF gives the $\eta^2(N,N)$-coordinated chelate $[W(CO)_4(2\text{-}NH_2py)]$ that can be deprotonated with sodium hydride to give $Na[W(CO)_4(2\text{-}NHpy)]$. The latter with carbon dioxide in acetonitrile (AN) gives the chelated carbamate in $Na[W(CO)_4(OC(O)\text{-}2\text{-}NHpy)]$. 2,6-Diaminopyridine enters into photolysis with $[Cr(CO)_6]$ in toluene to yield $[Cr(CO)_5(\eta^1(N)\text{-}L)]$ where coordination is *via* the nitrogen atom one of the amino groups (04JOM2319).



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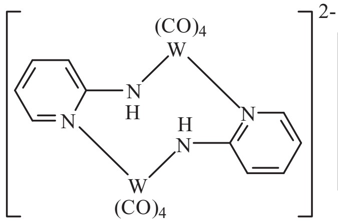

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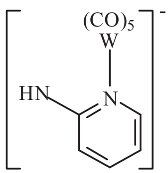

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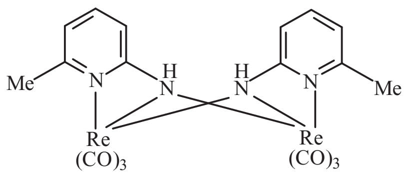
2.4 Manganese group

2-Aminopyridine (LH) with $[(\eta^5\text{-Cp})_2Mn]$ gives the hexanuclear amido cage $[(\eta^5\text{-Cp})_2Mn_3L_4]_2$ (02CC2980). 2-Amino-6-methylpyridine reacts with $[Re_3(\mu\text{-H})_3(CO)_{11}(AN)]$ in refluxing toluene to produce dinuclear **16** (98OM5580). With $[Re_4(\mu\text{-H})_4(CO)_{12}]$ in refluxing 1,2-dichloroethane, the trinuclear anionic cluster **17** is isolated as a triethylammonium salt. Protonation of the product using $(HOEt_2)(BF_4)$ then gives neutral **18** (00EJI1693). The same reaction but in the presence of triphenylphosphine or tri(*p*-tolyl)phosphine also gives the neutral species **19** where one of the CO ligands is substituted with PR_3 (R = Ph, *p*-Tol). Protonation in the presence of an alkyne leads to the neutral alkynyl derivatives **20** ($R = R^1$ = Ph, Et; R = Ph, R^1 = H; R = H, R^1 = Ph). Compound **17** reacts with $[Au(PPh_3)](PF_6)$ in THF to give two isolable heteronuclear clusters **21** and **22** (00OM2043).



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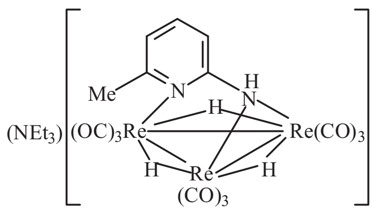

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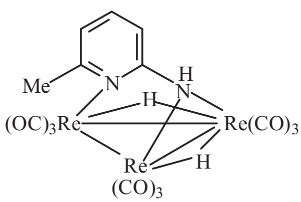
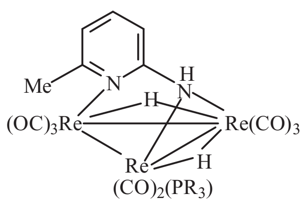
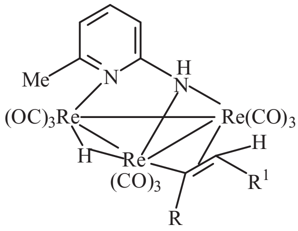




18 **19**



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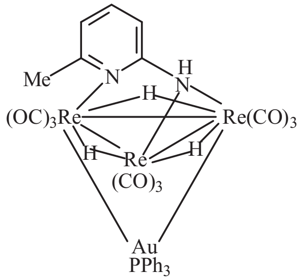

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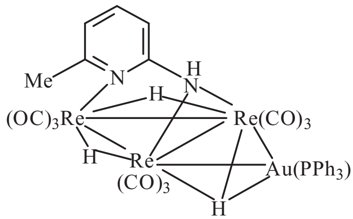

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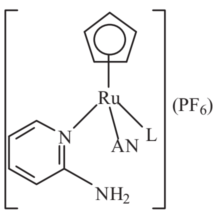
2.5 Iron group

2-Aminopyridine with $[(\eta^5\text{-Cp})Ru(AN)_3](PF_6)$ gives η^1(N)-coordinated **23** (L = AN) (03JCS(D)2329). In the presence of trimethyl and triphenylphosphine, as well as carbon monoxide, **23** (L = PMe_3, PPh_3, CO) can be prepared. For L = PPh_3 and CO, the reaction with diphenylacetylene gives the η^3-allylcarbenes **24** (L = PPh_3, CO). Under oxygen, **23** (L = AN) gives dinuclear **25** with the μ_3-bridging function of aminopyridinates. The same reaction but of 2-*N*,*N*′-dimethylaminopyridine gives the $\eta^6(\pi)$-coordinated **26**. Interaction of $[(\eta^2\text{-H}_2)Ru(H)(PCy_3)_2(\eta^2(N,N)\text{-2-NHpy})]$ with $CH_2{=}CHSiEt_3$ gives complex **27** (96OM3471). 2-Aminopyridine, 2-amino-4-picoline, and 2-(methylamino)pyridine react with $[(\eta^4\text{-cod})Ru(Tp)Cl]$ in the presence of terminal alkynes HC≡CR (R = Ph, *n*-Bu, C_6H_9) to afford the cyclic aminocarbenes **28** (R^1 = H, Me; R^2 = H, Me; R^3 = Ph, *n*-Bu, C_6H_9) (02OM4955).



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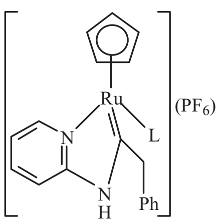

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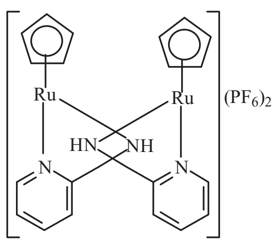

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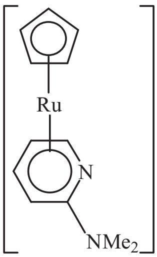

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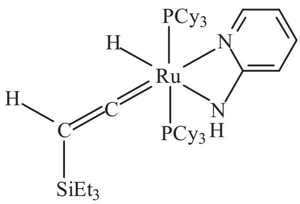

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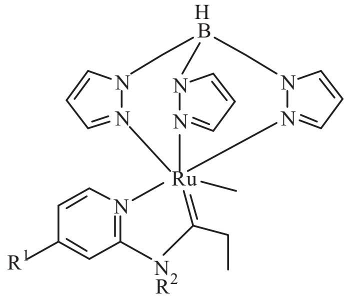

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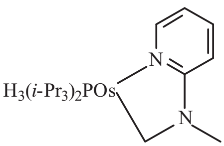
2-Dimethylaminopyridine with $[OsH_6(PPr\text{-}i_3)_2]$ experiences activation of one of the C–H bonds to yield chelate **29** (07OM5140). With $HBF_4{\cdot}Et_2O$, cyclic carbene **30** results. 2-Aminopyridine with $[Os(BCl_2)Cl(CO)(PPh_3)_2]$ gives **31** (E = X = Cl) and **32** (X = Cl) (02OM4862). The latter on dissolution in ethanol forms **32** (R = OEt). Product **31** with sodium bromide yields **31** (E = Br, X = Cl), with sodium borohydride **31** (E = H, X = Cl) and with silver hexafluoroantimonate in wet THF **31** (E = Cl, X = OH). In excess $AgSbF_6$ in acetonitrile cationic **33** follows, while in the presence of carbon monoxide in ethanol cationic **34** is the product. 2-Methylaminopyridine $[Os(BCl_2)Cl(CO)(PPh_3)_2]$ gives, in contrast, exclusively **35** (E = X = Cl). Further reaction with sodium borohydride yields mixture **35** (E = H, X = Cl) and **35** (E = Cl, X = OH), with ethanol **35** (E = Cl, X = OEt), with silver hexafluoroantimonate in acetonitrile/ethanol cationic **36**. 8-Aminoquinoline with $[Os(BCl_2)Cl(CO)(PPh_3)_2]$ gives a base-stabilized borylene complex containing a five-membered chelate ring (00AGE948). 8-Aminoquinoline with $[M(CO)(PPh_3)_2Cl(SiMeCl_2)]$ gives chelates **37** (M = Ru, Os; X = Cl), which in ethanol transform into **37** (M = Ru, Os; X = OEt) (04JOM2511). In contrast, 2-aminopyridine with the osmium precursor gives rise to the product of condensation of the amino NH-functions with two silicon–chlorine bonds in bis-chelate **38**. Bis(8-quinolinyl)amine (HL) forms methyl *trans*-$[Ru(L)Me(PMe_3)_2]$ **39** (08IC11570).



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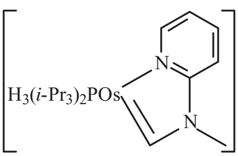

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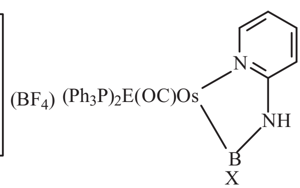

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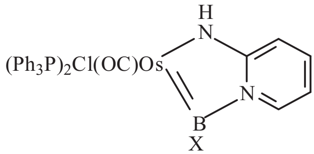

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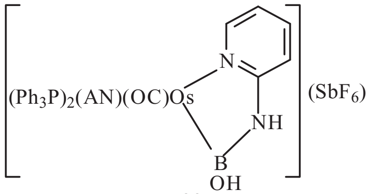

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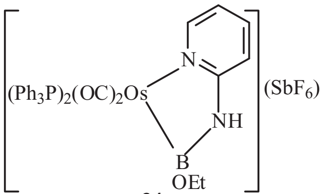

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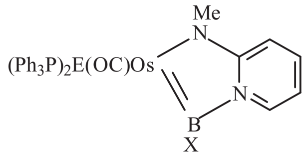

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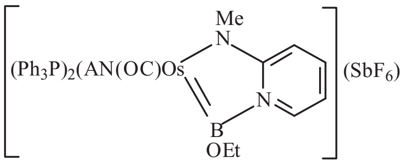

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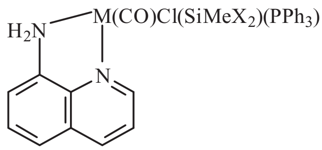

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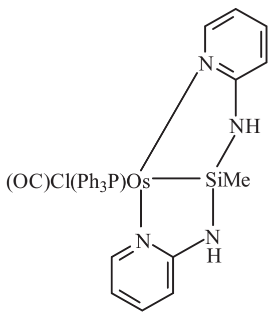

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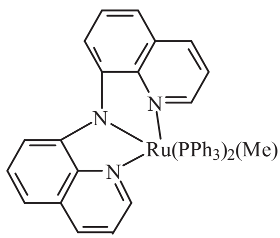

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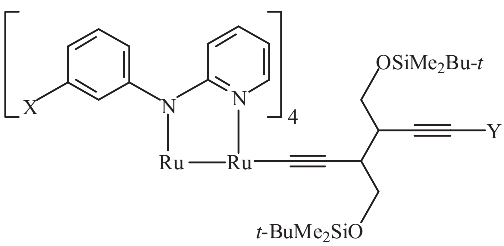
$[Ru_2ClL_4]$ (L is 2-anilinophenylpyridinate) and LiC≡CPh give the dinuclear $[Ru_2(C{\equiv}CPh)L_4]$ (86ICA(113)19) that contains a diruthenium (II,III) unit bridged by four pyridinate ligands. The ruthenium–ruthenium bond is multiple and has order 2.5. 2,3,4,5,6-Pentafluoro-2-anilinopyridinate (L) complex $[Ru_2L_4Cl]$ with LiC≡CPh gives $[Ru_2L_4(C{\equiv}CPh)]$ and $[Ru_2L_4(C{\equiv}CPh)_2]$ (93IC4175, 97IC5449). $[Ru_2L_4Cl]$ with LiC≡CPh leads to Ru_2^{6+} derivatives $[Ru_2L_4(C{\equiv}CPh)_2]$, where L = 2-F-, 2,3-$F_2$-, 2,4-$F_2$-, 2,5-$F_2$-, 3,4-$F_2$-, or 2,4,6-$F_3$anilinopyridinato (04IC4825). $[Ru_2L_4(C{\equiv}CX)]$ and LiC≡CPh also yield bis-alkynyl *trans*-$[(PhC{\equiv}C)Ru_2L_4(C{\equiv}CX)]$ (X = $SiMe_3$, $C{\equiv}CSiMe_3$) (02JOM(655)239). $[Ru_2L_4(C{\equiv}CPh)]$ may also be prepared from $Me_3SnC{\equiv}CPh$ (00JOM(595)300, 02CIC355, 03JOM(670)188, 05OM4854). In a similar manner, $[Ru_2L_4(C{\equiv}CR)]$ (R = $SiMe_3$, CH_2OMe) follow from $LiC{\equiv}CSiMe_3$ and $LiC{\equiv}CCH_2OMe$ (00JOM(596)152). Under desilylation agent n-Bu_4NF, the derivative with R = $SiMe_3$ can be converted to that with R = H. Compounds composed of two tetra(μ-*N*,*N*′-2-anilinopyridinate)diruthenium(II,III) ends bridged by butadiynediyl or ethynediyl ligands are characterized by strong electronic couplings across the carbon bridges (00CC1197). Reaction between $[Ru_2L_4Cl]$ and $LiC{\equiv}CC_5H_4N$ leads to $[Ru_2L_4(C{\equiv}CC_5H_4N)_2]$ (08IC7775). $[Ru_2L_4Cl]$ with $LiC{\equiv}CC{\equiv}CSiMe_3$ produces the monoadduct $[Ru_2L_4(C{\equiv}CSiMe_3)]$ and with excess diacetylide the bis-adduct $[Ru_2L_4(C{\equiv}CC{\equiv}CSiMe_3)_2]$ (01OM2400). In a similar fashion, from $[RuL_4Cl]$ and $LiC{\equiv}CSiPr$-i_3, $[Ru_2L_4(C{\equiv}CSiPr$-$i_3)]$ can be prepared (02OM732). $[Ru_2L_4(C{\equiv}CSiR_3)]$ (R = Me, *i*-Pr) with $LiC{\equiv}CC{\equiv}CSiMe_3$ give $[(Me_3SiC{\equiv}CC{\equiv}C)Ru_2L_4(C{\equiv}CSiR_3)]$ (R = Me, *i*-Pr). The methyl derivative with K_2CO_3 in methanol–THF gives a mixture of $[(HC{\equiv}CC{\equiv}C)Ru_2L_4(C{\equiv}CSiMe_3)]$ and $[(HC{\equiv}CC{\equiv}C)Ru_2L_4(C{\equiv}CH)]$. When R = *i*-Pr, the sole product from sodium hydroxide in methanol–THF is $[(HC{\equiv}CC{\equiv}C)Ru_2L_4(C{\equiv}CSiPr$-$i_3)]$. $[Ru_2L_4Cl]$ with lithium aryl acetylenes gives $[Ru_2L_4(C{\equiv}C$-4-$C_6H_4C{\equiv}CX)]$ (X = $SiMe_3$, H, Ru_2L_4), 1,3-$[Ru_2L_4(C{\equiv}C)_2C_6H_4]$, 1,3-$[Ru_2L_4(C{\equiv}C)_2C_6H_3$-5-$C{\equiv}CH]$, and 1-$[Ru_2L_4(C{\equiv}C)_2C_6H_3$-3,5-$(C{\equiv}CH)_2]$ (02JOM(660)1). $[Ru_2L_4Cl]$ and $LiC_{2n}Li$ (n = 1–4, 6) form polyyndiyls $[Ru_2L_4(\mu$-C,C′$-C_{2n})Ru_2L_4]$ (03JA10057). Application of *E*-hex-3-ene-1,5-diyn-diyl gives **40** (X = Y = H; X = H, Y = Ru_2L_4; X = OMe, Y = Ru_2L_4) (04JA10552). Cross-coupling between $[Ru_2L_4(C_{2(k-1)}H)]$ and $HC{\equiv}CY$ gives $[Ru_2L_4(C_{2k}Y)]$ (k = 2–5, Y = Si(*i*-Pr)$_3$, Ph) along with $[(Ru_2L_4)_2(\mu$-$C_{4(k-1)})]$, the products of homocoupling (05OM3247). Other representatives are $[Ru_2L_4(C{\equiv}CC_6H_4)_n(SCH_2CH_2SiMe_3)]$ (n = 1, 2) (05JA10010, 05JOM4734). A development of these studies is the application of variously substituted aminopyridinate ligands, for example, 2-(3,5-dimethoxyanilino)pyridine and 2-(3-methoxyanilino)pyridine (L) (08JOM1656). As before, $[Ru_2L_4Cl]$ with $LiC{\equiv}CR$ allows the preparation of $[Ru_2L_4(C{\equiv}CC{\equiv}CSiMe_3)]$, $[Ru_2L_4(C{\equiv}CFc)]$, and $[Ru_2L_4(C{\equiv}CC{\equiv}CFc)]$. The reactivity of $[Ru_2L_4Cl]$ (LH = 2-anilino-4-methylpyridine) with carbon monoxide is also of interest (06IC5996). $[Os_2L_4Cl_2]$ (HL = 2-anilinopyridine) readily reacts with $LiC{\equiv}CY$ to yield $[Os_2L_4(C{\equiv}CY)_2]$ (Y = Ph, Fc, $SiMe_3$, $C{\equiv}CSiMe_3$) (05IC5719).



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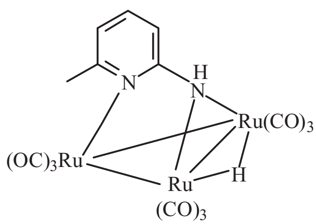
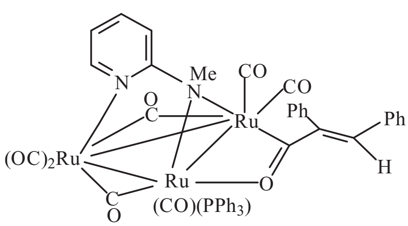
Derivatives of 2-aminopyridine containing substituents in the heteroring (H, 4-Me, 5-Me, 6-Me) and at the amino nitrogen atom (H, Me, Ph when the heteroring is unsubstituted) (HL) with $[Ru_3(CO)_{12}]$ in refluxing toluene give a series of clusters $[Ru_3(\mu\text{-}H)(\mu_3,\eta^2\text{-}L)(CO)_9]$ represented by **41** when the heteroring contains 6-Me (90JCS(D)2201, 02EJI1559). This cluster with triphenylphosphine or diphenylphosphinomethane gives $[Ru_3(\mu\text{-}H)(\mu_3,\eta^2\text{-}L)(CO)_8(PPh_3)]$ (93OM4141) or $[Ru_3(\mu\text{-}H)(\mu_3,\eta^2\text{-}L)(CO)_7(\eta^2\text{-dppm})]$, respectively (90JOM(384)C25, 92JOM(427)363). With excess triphenylphosphine, it gives the ligand-substitution product $[Ru_3(\mu\text{-}H)(\mu_3,\eta^2\text{-}L)(CO)_7(PPh_3)_2]$, which can be protonated by $HBF_4{\cdot}OEt_2$ to yield $[Ru_3(\mu\text{-}H)_2(\mu_3,\eta^2\text{-}L)(CO)_7(PPh_3)_2](BF_4)$ (91IC4611). Further addition of triphenylphosphine to the product gives $[Ru_3(\mu\text{-}H)_2(\mu_3,\eta^2\text{-}L)(CO)_6(PPh_3)_3](BF_4)$. With potassium methoxide, the latter two products lead to $[Ru_3(\mu\text{-}H)_2(\mu_3,\eta^2\text{-}L)(CO)_6(COOMe)(PPh_3)_2]$ and $[Ru_3(\mu\text{-}H)_2(\mu_3,\eta^2\text{-}L)(CO)_5(COOMe)(PPh_3)_3]$, respectively. The reactions of **41** with diphenylphosphine lead to $[Ru_3(\mu\text{-}H)(\mu_3,\eta^2\text{-}L)(CO)_8(PPh_2H)]$ and $[Ru_3(\mu\text{-}H)(\mu_3,\eta^2\text{-}L)(CO)_7(PPh_2H)_2]$ (90JOM(393)C30, 91ICA(186)225). The monophosphine derivative at reflux in THF transforms into the phosphido-bridged $[Ru_3(\mu_3,\eta^2\text{-}L)(\mu\text{-}PPh_2)(\mu\text{-}CO)_2(CO)_6]$ and diphosphine gives $[Ru_3(\mu\text{-}H)(\mu_3,\eta^2\text{-}L)(\mu\text{-}PPh_2)_2(CO)_6]$ (94OM55). Cluster **41** with $HBF_4{\cdot}OEt_2$ also gives the dihydride $[Ru_3(\mu\text{-}H)_2(\mu_3,\eta^2\text{-}L)(CO)_9](BF_4)$ (90JCS(D)3347), which with potassium methoxide yields a mixture of the starting complex and $[Ru_3(\mu\text{-}H)_2(\mu_3,\eta^2\text{-}L)(COOMe)(CO)_8]$ and with PPh_3 affords $[Ru_3(\mu\text{-}H)_2(\mu_3,\eta^2\text{-}L)(CO)_8(PPh_3)](BF_4)$. The latter with potassium methoxide yields the methoxycarbonyl $[Ru_3(\mu\text{-}H)_2(\mu_3,\eta^2\text{-}L)(COOMe)(CO)_7(PPh_3)]$. $[Ru_3(\mu\text{-}H)(\mu_3,\eta^2\text{-}L)(CO)_7(PPh_3)_2]$ reacts with molecular hydrogen or diphenylacetylene by carbon–phosphorus bond scission, the products being $[Ru_3(\mu\text{-}H)(\mu_3,\eta^2\text{-}L)(\mu\text{-}PPh_2)_2(\mu\text{-}Ph)(CO)_6]$ and $[Ru_3(\mu\text{-}H)(\mu_3,\eta^2\text{-}L)(\mu\text{-}PPh_2)(\mu\text{-}\eta^1{:}\eta^2\text{-}PhC{=}CHPh)(Ph)(CO)_5(PPh_3)]$, respectively (93OM1006).



41

Cluster **41** with diphenylacetylene gives the alkenyl-bridged $[Ru_3(\mu_3,\eta^2\text{-}L)(\mu\text{-}\eta^1{:}\eta^2\text{-}PhC{=}CHPh)(CO)_8]$ (92JMC(71)L7, 94OM4352). $[Ru_3(\mu_3,\eta^2\text{-}L)(\mu\text{-}\eta^1{:}\eta^2\text{-}PhC{=}CHPh)(CO)_8]$ adds triphenylphosphine without substitution accompanied by the intramolecular CO-insertion to yield **42** (99OM187). In the second step, ligand substitution yields **43**. Cluster **43** with carbon monoxide gives the ligand-substitution product **44**. Thermolysis of $[Ru_3(\mu_3,\eta^2\text{-L})(PPh_3)_2(CO)_8](BF_4)$ leads to the phenyl derivative $[Ru_3(\mu\text{-Ph})(\mu_3,\eta^2\text{-L})(\mu\text{-PPh}_2)(PPh_3)(CO)_7](BF_4)$ (97OM1743). Protonation of the aminopyridinate product with $HBF_4{\cdot}OEt_2$ gives $[Ru_3(\mu\text{-H})(\mu_3,\eta^2\text{-L})(\mu\text{-}\eta^1{:}\eta^2\text{-PhC}{=}\text{CHPh})(CO)_8](BF_4)$ (94OM4352), a pre-catalyst for the hydrogenation of diphenylacetylene (95SL579, 96OM449, 99MI1). This compound reacts with triphenylphosphine to afford products of CO-substitution containing one and two phosphine moieties and under vigorous conditions three PPh_3 ligands (96JOM(511) 103). With bis(diphenylphosphino)methane (dppm), reductive elimination of *cis*-stilbene occurs however, and the product is $[Ru_3(\mu\text{-H})(\mu_3,\eta^2\text{-L})(\mu\text{-dppm})(\eta^1(P)\text{-dppm})(CO)_7](BF_4)$. Deprotonation of $[Ru_3(\mu\text{-H})(\mu_3,\eta^2\text{-L})(\mu\text{-}\eta^1{:}\eta^2\text{-PhC}{=}\text{CHPh})(CO)_8](BF_4)$ can be achieved by sodium methoxide yielding $[Ru_3(\mu_3,\eta^2\text{-L})(\mu\text{-}\eta^1{:}\eta^2\text{-PhC}{=}\text{CHPh})(CO)_8]$. In contrast, with sodium hydroxide the neutral hydride $[Ru_3(\mu\text{-H})(\mu_3,\eta^2\text{-L})(CO)_9]$ and *cis*-stilbene follow. Compound **41** and its triphenylphosphine-substituted product react with silanes and diphenylacetylene to yield numerous clusters where silane and alkenyl moieties are attached to different ruthenium atoms or only silane moieties are present: $[Ru_3(\mu\text{-H})_2(\mu_3,\eta^2\text{-L})(CO)_8(Si(OMe)_3)]$, $[Ru_3(\mu\text{-H})(\mu_3,\eta^2\text{-L})(CO)_8(SiR_3)_2]$ (R = Et, OMe), $[Ru_3(\mu_3,\eta^2\text{-L})(\mu\text{-}\eta^1{:}\eta^2\text{-PhC}{=}\text{CHPh})(CO)_7(SiR_3)]$ (R = Et, OMe), $[Ru_3(\mu_3,\eta^2\text{-L})(\mu\text{-}\eta^1{:}\eta^2\text{-PhC}{=}\text{CHPh})(CO)_7(PPh_3)]$, and $[Ru_3(\mu\text{-H})(\mu_3,\eta^2\text{-L})(\mu\text{-}\eta^1{:}\eta^2\text{-PhC}{=}\text{CHPh})(CO)_6(SiEt_3)(PPh_3)]$ (93OM2973). $[Ru_3(\mu\text{-H})(\mu_3,\eta^2\text{-L})(PPh_3)_n(CO)_{9-n}]$ ($n = 0$–2) with triethylsilane and tri-*n*-butylstannane give the oxidative substitution products $[Ru_3(\mu\text{-H})_2(\mu_3\text{-2-NH-6-Mepy})(SiEt_3)(PPh_3)_n(CO)_{8-n}]$ ($n = 0$–2) and $[Ru_3(\mu\text{-H})_2(\mu_3,\eta^2\text{-L})(Sn(n\text{-Bu})_3)(PPh_3)_n(CO)_{8-n}]$ ($n = 0$–2) (92OM3334). $[Ru_3(\eta\text{-H})(\mu_3,\eta^2\text{-L})(\mu,\eta^1{:}\eta^2\text{-PhC}{=}\text{CHPh})(CO)_7(PPh_3)]$ with triphenylphosphine leads to the substitution product $[Ru_3(\mu_3,\eta^2\text{-L})(\mu_3\eta^1{:}\eta^2\text{-PhC}{=}\text{CHPh})(CO)_6(PPh_3)_2]$ (94JOM (480)205). $[Ru_3(\mu\text{-H})(\mu_3,\eta^2\text{-L})(CO)_7(PPh_3)_2]$ with diphenylacetylene yields $[Ru_3(\mu_3,\eta^2\text{-L})(\mu,\eta^1{:}\eta^2\text{-PhC}{=}\text{CHPh})(\mu,\text{-PPh}_2)(Ph)(CO)_5(PPh_3)]$.



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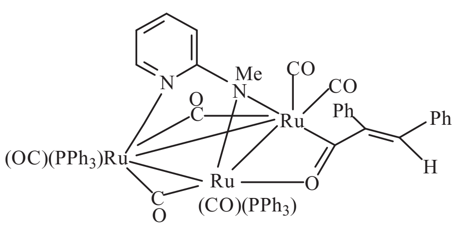

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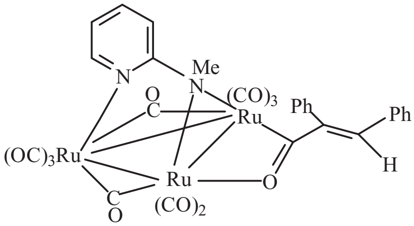
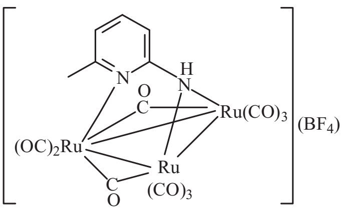


44

Cluster **41** reacts with diphenylacetylene and further with $HBF_4{\cdot}OEt_2$ and carbon monoxide to give cluster **45** with two carbonyl bridges (95IC1620). The product reacts with chloride, iodide, and acetate anions as $(PPN)^+$ or $(n\text{-}Bu_4N)^+$ salts to give $[Ru_3(\mu\text{-}X)(\mu_3,\eta^2\text{-}L)(CO)_9]$ (X = Cl, I, MeCOO), and with $(PPN)(BH_4)$ $[Ru_3(\mu\text{-}H)(\mu_3\text{-}L)(CO)_9]$ (97OM812). $[Ru_3(\mu_3,\eta^2\text{-}L)(CO)_{10}](BF_4)$ reacts with $(PPN)_2[Ru_3(\mu_3\text{-}S)(CO)_9]$ to afford the hexanuclear $[Ru_6(\mu\text{-}H)(\mu_4\text{-}S)(\mu_3,\eta^2\text{-}L)(CO)_{17}]$. Under triphenylphosphine, **45** gives the substitution product $[Ru_3(\mu_3,\eta^2\text{-}L)(CO)_9(PPh_3)](BF_4)$. Reaction of **41** with diphenylacetylene in the presence of R_3SnH (R = n-Bu, Ph) gives $[Ru_3(\mu\text{-}H)(\mu_3,\eta^2\text{-}L)(SnR_3)_2(CO)_8]$ and $[Ru_3(\mu\text{-}H)(\mu_3,\eta^2\text{-}L)(\mu\text{-}\eta^1{:}\eta^2\text{-}PhC{=}CHPh)(SnR_3)(CO)_7]$ (93OM157). Complex **45** with excess sodium hydroxide gives clusters **41**, **46**, and further anionic **47** (95OM3124) while cation **45** and anion **47** give hexanuclear cluster **46**.



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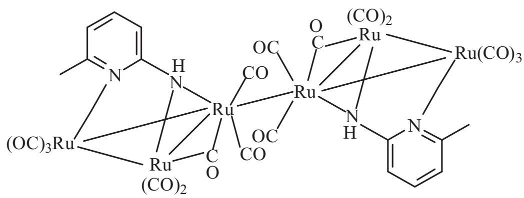

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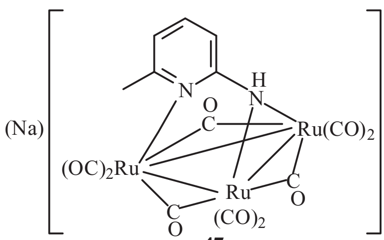
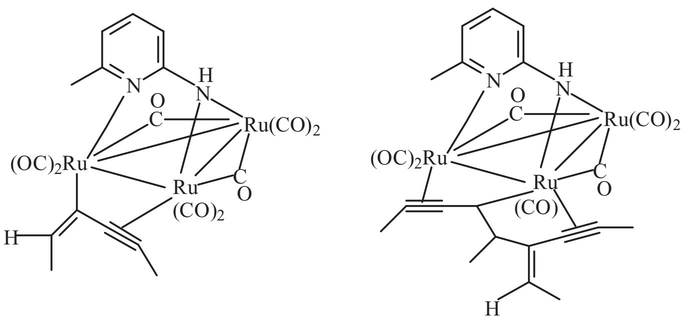


47

Complex **41** with molecular hydrogen gives $[Ru_6(\mu\text{-}H)_6(\mu_3,\eta^2\text{-}L)(CO)_{14}]$ (94OM426, 95JOM(494)169), which adds $P(p\text{-}Tol)_3$ to yield another hexanuclear cluster $[Ru_6(\mu\text{-}H)_6(\mu_3,\eta^2\text{-}L)(CO)_{14}(P(p\text{-}Tol)_3)_2]$. $[Ru_3(\mu_3,\eta^2\text{-}L)(\mu\text{-}\eta^1{:}\eta^2\text{-}PhC{=}CHPh)(CO)_8]$ with molecular hydrogen in the absence of diphenylacetylene gives **41** and **46** (94OM4352). With carbon monoxide the derivative $[Ru_3(\mu_3,\eta^2\text{-L})(\eta^1\text{-PhC=CHPh})(CO)_9]$ follows, which contains a terminal alkenyl ligand. $[Ru_6(\mu_3\text{-H})_2(\mu_5\text{-}\eta^2\text{-L})(\mu\text{-CO})_2(CO)_{14}]$ with diphenylacetylene in toluene gives a mixture, among them $[Ru_6(\mu_5\text{-}\eta^2\text{-L})(\mu_3\text{-CO})(\mu\text{-CO})_2(CO)_{14}]$, $[Ru_6(\mu_5\text{-}\eta^2\text{-L})(\mu_4\text{-}\eta^2\text{-PhCCPh})(CO)_{16}]$, $[Ru_7(\mu_5\text{-}\eta^2\text{-L})(\mu_5\text{-}\eta^4\text{-PhCCPh})(CO)_{17}]$, $[Ru_6(\mu_5\text{-}\eta^2\text{-L})(\mu_5\text{-}\eta^8\text{-PhCCPh})(\mu\text{-CO})(CO)_{13}]$, $[Ru_5(\mu_5\text{-}\eta^2\text{-L})(\mu_4\text{-}\eta^2\text{-PhCCPh})(\mu\text{-CO})(CO)_{12}]$, and $[Ru_5(\mu_5\text{-}\eta^2\text{-L})(\mu_4\text{-}\eta^2\text{-PhCCPh})(\eta^6\text{-PhMe})(\mu\text{-CO})(CO)_9]$ with retention of the coordination mode of the aminopyridinate ligand (05OM665). With diphenylbutadiyne also a mixture of clusters results, $[Ru_6(\mu_5\text{-}\eta^2\text{-L})(\mu_5\text{-}\eta^3\text{-PhCCCCH}_2\text{Ph})(\mu\text{-CO})(CO)_{14}]$, $[Ru_6(\mu_5\text{-}\eta^2\text{-L})(\mu_5\text{-}\eta^4\text{-}trans\text{-PhCCCHCHPh})(\mu\text{-CO})(CO)_{14}]$, and $[Ru_5(\mu_5\text{-}\eta^2\text{-L})(\mu_4\text{-}\eta^2\text{-}trans\text{-PhCCCHCHPh})(\eta^6\text{-PhMe})(\mu\text{-CO})(CO)_9]$ (06OM1492). With arenes at high temperature $[Ru_6(\mu_3\text{-H})_2(\mu_5\text{-}\eta^2\text{-L})(\eta^6\text{-arene})(\mu\text{-CO})_2(CO)_{11}]$ (arene = C_6H_6, C_6H_5Me, 1,4-$C_6H_4Me_2$) are the products of substitution of the arene for the three carbonyl ligands of the apical ruthenium atom (06OM2683). Cycloheptatriene in chlorobenzene at reflux gives a mixture of $[Ru_6(\mu_3\text{-H})(\mu_5\text{-}\eta^2\text{-L})(\eta^5\text{-C}_7H_9)(\mu_3\text{-CO})(\mu\text{-CO})_2(CO)_{11}]$, $[Ru_6(\mu_3\text{-H})(\mu_5\text{-}\eta^2\text{-L})(\mu_3\text{-}\eta^7\text{-C}_7H_7)(\mu\text{-CO})_2(CO)_{11}]$, $[Ru_6(\mu_5\text{-}\eta^2\text{-L})(\mu_2\text{-}\eta^7\text{-C}_7H_7)_2(\mu\text{-CO})(CO)_9]$, and $[Ru_6(\mu_6\text{-C})(\mu_3\text{-}\eta^7\text{-C}_7H_7)_2(\mu\text{-CO})_2(CO)_8]$. Dicyclopentadiene in chlorobenzene at reflux temperature affords $[Ru_6(\mu_3\text{-H})(\mu_5\text{-}\eta^2\text{-L})(\eta^5\text{-Cp})(\mu_3\text{-CO})(\mu\text{-CO})_2(CO)_{11}]$, $[Ru_6(\mu_3\text{-H})_2(\mu_5\text{-}\eta^2\text{-L})(\eta^5\text{-Cp})_2(\mu_3\text{-CO})(\mu\text{-CO})(CO)_9]$, and $[Ru_5(\mu_5\text{-}\eta^2\text{-L})(\mu_4\text{-}\eta^4\text{-C}_{10}H_{10})(\mu\text{-CO})_2(CO)_{10}]$. With indene in chlorobenzene at reflux, the product is $[Ru_7(\mu_5\text{-}\eta^2\text{-L})(\mu_3\text{-H})(\mu\text{-}\eta^9\text{-C}_9H_7)(\mu\text{-CO})_3(CO)_{11}]$ and in excess of indene $[Ru_6(\mu_3\text{-H})(\mu_5\text{-}\eta^2\text{-L})(\eta^5\text{-C}_9H_7)(\mu_3\text{-CO})(\mu\text{-CO})_2(CO)_{11}]$ (07OM1414). With fluorene in decane at reflux the product is $[Ru_6(\mu_3\text{-H})_2(\mu_5\text{-}\eta^2\text{-L})(\eta^6\text{-C}_{13}H_9)(\mu\text{-CO})_2(CO)_{11}]$, with azulene under identical conditions $[Ru_6(\mu_5\text{-}\eta^2\text{-L})(\mu_3\text{-}\eta^{10}\text{-C}_{10}H_8)(\mu\text{-CO})_2(CO)_{10}]$, and with acenaphthylene $[Ru_4(\mu_4\text{-}\eta^2\text{-L})(\mu\text{-}\eta^6\text{-C}_{12}H_8)(\mu\text{-}\eta^4\text{-C}_{12}H_8)(\mu\text{-CO})_2(CO)_5]$, $[Ru_6(\mu_4\text{-}\eta^2\text{-L})(\mu_3\text{-}\eta^{10}\text{-C}_{12}H_8)(CO)_{12}]$, $[Ru_7(\mu_5\text{-}\eta^2\text{-L})(\mu_4\text{-}\eta^{12}\text{-C}_{12}H_8)(\mu\text{-CO})(CO)_{12}]$, and $[Ru_6(\mu_4\text{-}\eta^1\text{-L})(\mu_4\text{-}\eta^{12}\text{-C}_{12}H_8)(\mu\text{-CO})_2(CO)_9]$. The latter case is unusual because the coordination modes of the aminopyridinate ligands change. Heating $[Ru_6(\mu_3\text{-H})_2(\mu_5\text{-}\eta^2\text{-L})(\mu\text{-CO})_2(CO)_{14}]$ in undecane at reflux yields nonanuclear cluster complexes $[Ru_9(\mu\text{-H})_2(\mu_4\text{-}\eta^2\text{-L})_4(CO)_{17}]$ and $[Ru_9(\mu_3\text{-H})(\mu\text{-H})(\mu_4\text{-}\eta^2\text{-L})_3(CO)_{18}]$ (06OM5672). Aminopyridinate ligands are attached to edge-bridging ruthenium atoms *via* the pyridine nitrogen heteroatom, while imido nitrogens cap the metallic triangles. Treatment of $[Ru_6(\mu_3\text{-H})_2(\mu_5\text{-}\eta^2\text{-L})(\mu\text{-CO})_2(CO)_{14}]$ with 2-amino-6-methylpyridine on refluxing in decane leads to the hexanuclear $[Ru_6(\mu_3\text{-H})_2(\mu\text{-H})_2(\mu_4\text{-}\eta^2\text{-L})_2(CO)_{14}]$ and the octanuclear $[Ru_8(\mu\text{-H})(\mu_4\text{-}\eta^2\text{-L})_3(\mu_3\text{-}\eta^2\text{-HL})(\mu\text{-CO})_2(CO)_{15}]$ (06CJC105). The deprotonated aminopyridinate ligand is in a coordination mode when the imido nitrogen atom caps a triangle of ruthenium sites and the pyridine nitrogen is bound to an additional ruthenium atom. The ligand HL in the second cluster caps a Ru_4 square through the imido nitrogen and the pyridine nitrogen is bound to one of these Ru_4 sites. Thermolysis of $[Ru_3(\mu\text{-H})(\mu_3\text{-HL})(CO)_9]$ in decane at 150°C gives nonanuclear $[Ru_9(\mu_3\text{-H})_2(\mu\text{-H})$

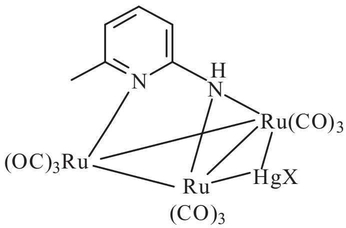
(μ_5-O)(μ_4-L)(μ_3-HL)(CO)$_{21}$], [Ru$_9$(μ_5-O)$_2$(μ_4-L)(μ_3-HL)$_2$(μ-CO)(CO)$_{20}$], [Ru$_9$(μ_5-O)$_2$(μ_4-L)(μ_3-HL)$_2$(μ-CO)$_2$(CO)$_{19}$], and [Ru$_9$(μ_4-O)(μ_5-O)(μ_4-L)(μ_3-HL)(μ-HL)(μ-CO)(CO)$_{19}$] along with hexanuclear [Ru$_6$(μ_3-H)$_2$(μ_5-HL)(μ-CO)$_2$(CO)$_{14}$] and pentanuclear [Ru$_5$(μ_4-L)$_2$(μ-CO)(CO)$_{12}$] (06IC6020). Heating [Ru$_6$(μ_3-H)$_2$(μ_5-η^2-L)(μ-CO)$_2$(CO)$_{14}$] in 1-octene gives [Ru$_7$(μ_3-H)(μ_5-η^2-L)(μ-η^3:η^4-MeC$_7$H$_8$)(μ-CO)$_3$(CO)$_{12}$], in 1-nonene [Ru$_7$(μ_3-H)(μ_5-η^2-L)(μ-η^3:η^4-MeC$_7$H$_7$Me)(μ-CO)$_3$(CO)$_{12}$], in 1-decene [Ru$_7$(μ_3-H)(μ_5-η^2-ampy)(μ-η^3:η^4-MeC$_7$H$_7$Et)(μ-CO)$_3$(CO)$_{12}$] and their isomers (07OM2482). The coordination mode of aminopyridinate ligands is retained.

Cluster **41** with hexa-2,4-diyne gives **48** containing a bridging three-electron hex-2-yn-4-en-4-yl ligand and **49** containing a bridging five-electron 5-methyl-6-(ethyliden)nona-2,7-diyn-4-en-4-yl ligand (01OM4973). With other diynes RC≡CC≡CR in THF the ynenyl [Ru$_3$(μ_3-η^2-L)(μ-η^3-RC≡CC=CHR)(μ-CO)$_2$(CO)$_6$] (R = CH$_2$OPh, R = Ph) follow with 1,4-disubstituted butynen-3-yl ligand attached to two ruthenium atoms (01CEJ2370). With diphenylbutadiyne [Ru$_3$(μ-η^2-L)(μ_3-η^6-PhCC$_5$(C≡CPh)HPh$_2$)(CO)$_7$] containing an η^5-cyclopentadienyl ring and a bridging carbene moiety, a [3 + 2] cycloaddition of 1,4-diphenylbutynen-4-yl takes place in the course of coordination with a triple bond of a second diphenylbutadiyne.



48 **49**

Cluster **41** reacts with HgPh$_2$ to give [Ru$_6$(μ_4-Hg)(μ_3,η^2-L)$_2$(CO)$_{18}$] containing two [Ru$_3$(μ_3,η^2-L)(CO)$_9$] moieties bridged by a mercury atom bonded to the two NH-bridged ruthenium atoms belonging to each Ru$_3$ unit (91JOM(420)431). The product with mercury(II) halides gives **50** (X = Cl, Br, I). The latter (X = Cl) further reacts with dimeric [(η^5-Cp)M(CO)$_3$]$_2$ (M = Mo, W) and [Co$_2$(CO)$_8$] abbreviated as M$_2$L$_{2n}$ to yield the heteronuclear **51** (ML$_n$ = (η^5-Cp)Mo(CO)$_3$, (η^5-Cp)W(CO)$_3$, Co(CO)$_4$) (92JOM(434)123).



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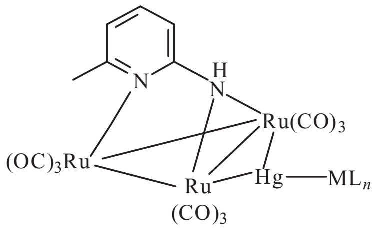
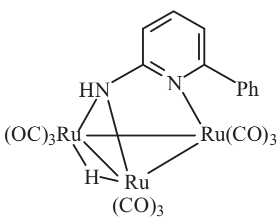


51

Sodium 2-anilinopyridinate (L) with [$Ru_3(CO)_{12}$] followed by metathesis, denoted as (PPN)Cl, gives anionic (PPN)[$Ru_3(\mu_3,\eta^2$-L)$(CO)_9$] and (PPN)[$Ru_3(\mu_3,\eta^2$-L)$(CO)_{10}$] (92OM1351). (PPN)[$Ru_3(\mu_3$-η^2-L)$(CO)_9$] reacts with P(n-Bu)$_3$ or PPh_3 to yield the addition product (PPN)[$Ru_3(\mu_3$-η^2-L) $(CO)_9L^1$] (L^1 = P(n-Bu)$_3$, PPh_3) (96IC755). 2-(Methylamino)pyridine (HL) reacts with P(n-Bu)$_3$ to yield monosubstituted (PPN)[$Ru_3(\mu_3$-η^2-L) $(CO)_8$(P(n-Bu)$_3$)]. The 2-anilinopyridine analog of **41** (90JA8607) inserts benzyl isocyanide to give [$Ru_3(\mu$-PhCH$_2$N=CC(Ph)=CH(Ph)(μ_3,η^2-L)) $(CO)_8$] (94OM4673). 2-Amino-6-phenylpyridine with [$Ru_3(CO)_{12}$] yields trinuclear μ_3-η^2 **52** and μ_3-η^3 **53** as well as hexanuclear μ_5-η^2 **54** and μ_5-η^3 **55** clusters (03JCS(D)2808, 04OM1107, 08OM2878). In contrast, this ligand with [$Os_3(CO)_{10}(AN)_2$] gives only trinuclear μ-η^1 **56**, μ-η^2 **57**, and under UV-induced decarbonylation μ_3-η^2 **58** clusters. Under reflux in toluene **57** transforms to the cyclometalated μ-η^3 **59**. Clusters similar to **54** can be prepared in refluxing xylene from 6-methyl-2-aminopyridine or 2-aminopyridine (04IC5450). 6-Methyl-2-aminopyridinate cluster with triphenylphosphine gives monosubstituted [$Ru_6(\mu_3$-H)$_2(\mu_5$-η^2-L)(μ-CO)$_2$(CO)$_{13}$(PPh$_3$)] and with excess phosphine di- and trisubstituted [$Ru_6(\mu_3$-H)$_2(\mu_5$-η^2-L)(μ-CO)$_2$(CO)$_{12}$(PPh$_3$)] and [$Ru_6(\mu_3$-H)$_2(\mu_5$-η^2-L) (μ-CO)$_2$(CO)$_{11}$(PPh$_3$)$_3$] result. With bis(diphenylphosphino)methane the product is [$Ru_6(\mu_3$-H)$_2(\mu_5$-η^2-L)(μ-CO)$_2$(CO)$_{12}$(μ-dppm)].



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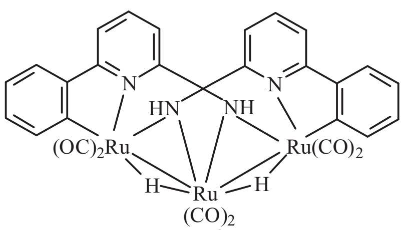

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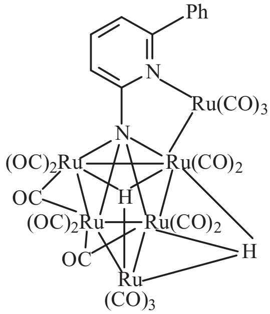

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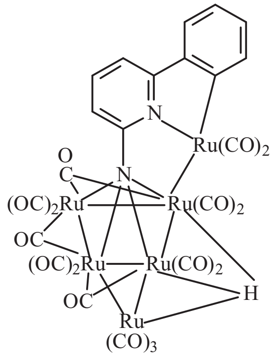

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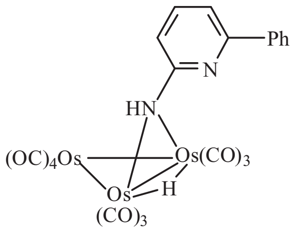

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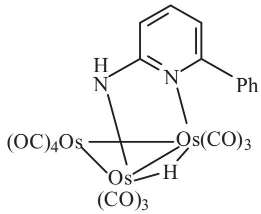

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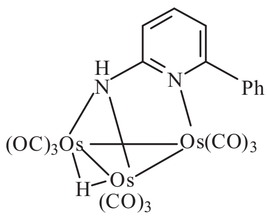

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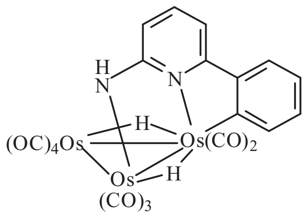
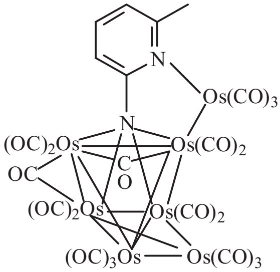


59

2-Aminopyridine and 2-(benzylamino)pyridine react with $[Os_3(CO)_{10}(\eta^2\text{-COE})_2]$ or $[Os_3(CO)_{12}]$ to form $[Os_3H(CO)_{10}(\mu\text{-}NC_5H_4X)]$ ($X = NH$, NCH_2Ph) (82JCS(D)1205). Thermal decarbonylation gives $[Os_3H(CO)_9(\mu_3\text{-}NC_5H_4X)]$. 2-Aminopyridine with $[Os_3(CO)_{12}]$ under more vigorous conditions gives the dinuclear species $[Os_2(CO)_6(\mu\text{-}NC_5H_4NH)_2]$. 2-Amino-6-methylpyridine (HL) with $[Os_3(CO)_{12}]$ in decane at reflux yields trinuclear $[Os_3(\mu\text{-}H)(\mu_3\text{-}\eta^2\text{-}L)(CO)_9]$ (07OM3212). When the reaction is conducted in a twofold excess of $[Os_3(CO)_{12}]$, three products result, **60–62**. 2-Amino-3-bromopyridine with $[Os_3(CO)_{10}(AN)_2]$ and $[(\eta^5\text{-}Cp^*)IrCl_2]_2$ in methylene chloride at reflux yield two heteropolynuclear clusters with different coordination modes of the heteroaromatic ligand, **63** with $\mu_3\text{-}\eta^3$-mode and **64** with $\mu_3\text{-}\eta^2$-mode with deprotonation (08JOM1528).



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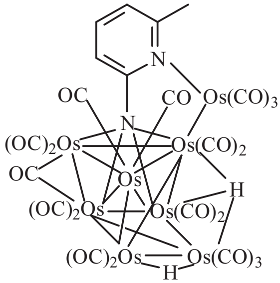

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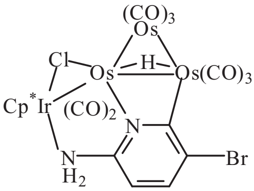

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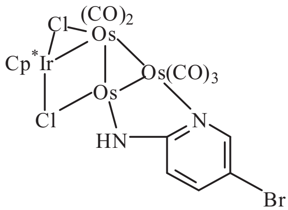
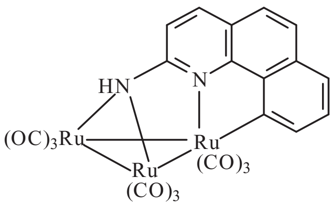


64

2-Amino-7,8-benzoquinoline with $[Ru_3(CO)_{10}(AN)_2]$ in THF gives cyclometalated **65**, and an excess of ligand in refluxing toluene gives μ_3-η^3 bridged **66** where the pyridine nitrogen atom is excluded from coordination (02OM5055). In contrast, with $[Os_3(CO)_{10}(AN)_2]$, the sole product is amido-bridged μ-η^1 **67**.



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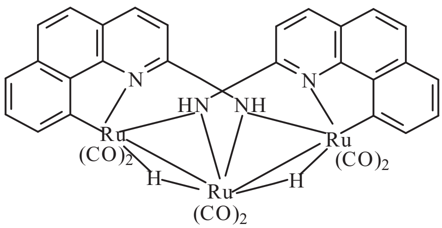

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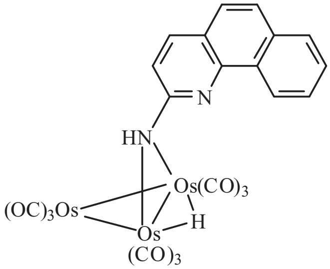
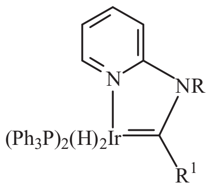


67

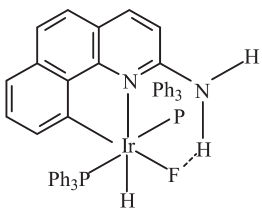
2.6 Cobalt, nickel, and copper group

2-Dimethylaminopyridine reacts with $[H_2Ir(OCMe_2)_2(PPh_3)_2](BF_4)$ to give a cyclic carbene $[H_2Ir(=CHN(Me)py)(PPh_3)_2](BF_4)$ **68** ($R = Me$, $R^1 = H$) *via* a sequence of oxidative addition and reversible α-elimination in the conversion of N–Me to N–CH=Ir by double C–H activation (01CC213, 04JA8795). The same reaction for 2-diethylaminopyridine gives **68** ($R = Et$, $R^1 = Me$), with 2-pyrrolidinopyridine **68** ($R_2 = (CH_2)_4$, $RR^1 = (CH_2)_2$), 2-piperidinopyridine **68** ($R_2 = (CH_2)_6$, $RR^1 = (CH_2)_4$), and 2-hexamethyleneiminopyridine **68** ($R_2 = (CH_2)_6$, $RR^1 = (CH_2)_6$). Tetrakis (μ-2-anilinopyridinato)dirhodium(II,III) $[Rh_2L_4Cl]$ with sodium or lithium acetylides ($C{\equiv}CH$, $C{\equiv}CPh$, $C{\equiv}CC_5H_{11}$-*n*, $C{\equiv}CC_4H_8C{\equiv}CH$) give dinuclear complexes of composition $[Rh_2L_4(C{\equiv}CR)]$ (90IC4033). $[Rh_2L_4Cl]$ and $Li(C{\equiv}C)_2SiMe_3$ in THF give $[Rh_2L_4(C{\equiv}C)_2SiMe_3]$ (01IC2275). $[Rh_2L_4(C{\equiv}C)_2Rh_2L_4]$ can be prepared from $[Rh_2L_4(C{\equiv}C)_2$-$Li]$ and $[Rh_2L_4Cl]$.



68

Lithium pyridinates of composition Li(2-PhNpy) and Li(2-Ph-4-*t*-Bu-py) (LiL) react with $[Rh(Cl)(L_2)]_2$ (L_2 = nbd, tfb, $(CO)_2$) to yield dinuclear $[Rh(\mu\text{-}L)(L_2)]_2$ (87ICA(128)119). Lithium 2-aminopyridinate and lithium 2-anilinopyridinate (LiL) with the dimer $[(\eta^4\text{-cod})Ir(\mu\text{-}Cl)]_2$ give dinuclear iridium(I) $[(\eta^4\text{-cod})Ir(\mu\text{-}L)]_2$ (99ICA(292)244). Cyclometalated complex **69** based on 2-aminobenzoquinoline (96JA13105, 99CC297) is prepared from the ligand and $[(\eta^4\text{-cod})Ir(PPh_3)_2](BF_4)$ in the presence of molecular hydrogen in methylene chloride. Initially $[Ir(L)(PPh_3)_2(H)(H_2O)](BF_4)$ is formed, and on interaction with a fluoride **69** results. It can be protonated at the amino nitrogen atom using $HBF_4{\cdot}Et_2O$ to yield **70** (99OM1615), both species being stabilized by the intramolecular hydrogen···fluorine bond. The aqueous complex formed at the initial stages in 2-hexanone is converted to **71** (00JOM(600)7, 00OM2228).



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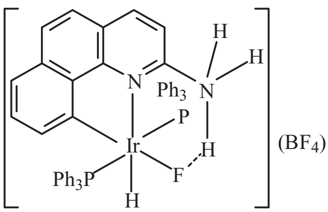

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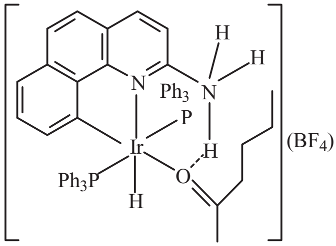
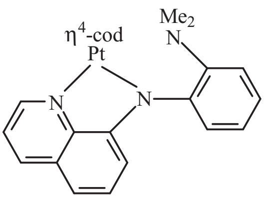


71

o-Dimethylaminophenyl(8-quinolyl)amine with [(η^4-cod)$PtCl_2$] in the presence of triethylamine in methylene chloride gives **72** and 3,5-dimethylphenyl(8-quinolyl)amine affords under these conditions a similar product **73** (01IC5083). Bis(8-quinolyl)imine (HL) reacts with [(η^4-cod) $PtCl_2$] and triethylamine to yield [Pt(L)Cl] (02OM1753). The lithium salt of this ligand with [(η^4-cod)Pt(Me)Cl] gives [Pt(L)Me], which gives [Pt(L) (OTf)] with triflic acid. Heating the product in benzene with of NEtPr-i_2 gives the phenyl complex **74**. 9-Aminoacridine (L) is coordinated *via* the pyridine nitrogen atom in [Pd(C_6F_5)(L)(PPh_3)Cl] (08IC6990). The divalent complex of bis(8-quinolyl)amide (HL) **76** undergoes oxidative addition with methyl iodide to afford octahedral **77** (06IC4316) where the amide nitrogen is readily quarternized with HBF_4 leading to [Pt(H)(L)$PtMe_2I$] (BF_4).



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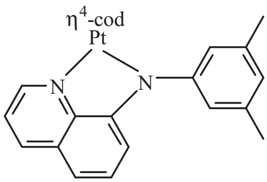

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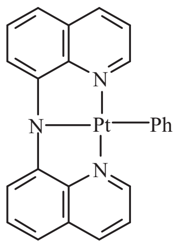

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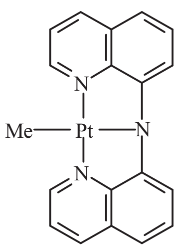

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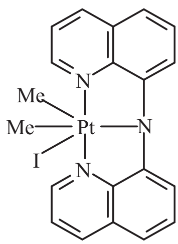
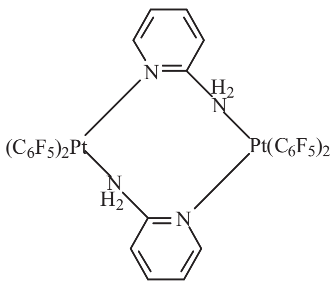


76

2-Aminopyridine bridges two $Pt(C_6F_5)_2$ fragments in dinuclear **77** (08IC8767). The mononuclear anionic $(N(n\text{-}Bu)_4)[Pt(C_6F_5)_3L]$ (L = 2-aminopyridine) is in the chelate mode (05IC9444). 2,6-Bis-pyridyl-bridged bis-azaallyl lithium complexes with $[PdCl_2(PhCN)_2]$ yield metallacyclic **78** (04JOM1230). 6-((Ferrocenylmethyl)amino)-2-picoline with $[Au(C_6F_5)(THT)]$ gives the η^1(N)-coordinated **79**, which on further interaction with $[Au(acac)(PPh_3)]$ gives dinuclear **80** where the amino nitrogen is also engaged in coordination (04EJI4820). With $[Au(C_6F_5)_3(OEt_2)]$ or $[Au(C_6F_5)_2Cl]_2$, the product is gold(III) **81** (R = C_6F_5, Cl). Dimethyl zinc with 2-pyridylamines $HN(2\text{-}C_5H_4N)R$ (R = Ph, 3,5-Xy, 2,6-Xy, $PhCH_2$, Me) (HL) forms $[MeZnL_2]^-$ (01CEJ3696). *N*-2-pyridylaniline with $ZnMe_2$ and further with *n*-BuLi affords $[(Ph(2\text{-}C_5H_4N)N)_2Zn(n\text{-}Bu)Li]$ (02JCS(D)3129). Lithium (2-(6-methyl)pyridyl)trimethylsilylamide (LiL) with diethyl zinc gives $[(ZnEt)_2L_2]$ where the amido groups bridge the metal centers (91JCS(D)2859).



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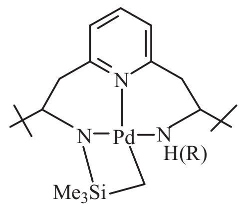

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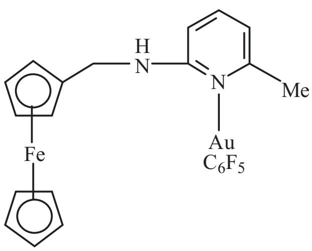

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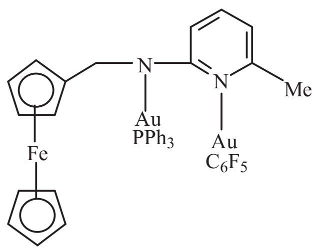

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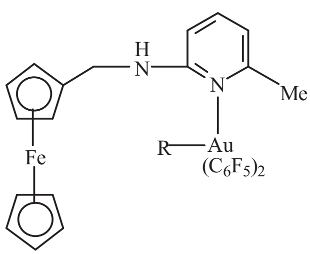

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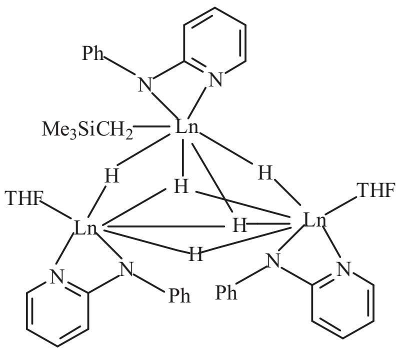
2.7 Rare earth metals

The reactions of bis(alkyl) [Ln(η^2(*N*,*N*)-L)(CH_2SiMe_3)$_2$(THF)] (Ln = Y, Lu; HL = 2-anilino-6-phenylpyridine) with phenylsilane and molecular hydrogen result in trinuclear rare earth alkyl hydrido clusters **82** (08OM2905). *N*-(2-((2,6-Di-*i*-propylphenylimino)methyl)phenyl)quinolin-8-amine (HL) with [Ln(CH_2SiMe_3)$_3$(THF)$_2$] affords bis(alkyl) **83** (Ln = Sc, $n = 0$; Ln = Y, $n = 1$; Ln = Lu, $n = 0$) (06CEJ8969, 07CEJ2764, 08OM5889). Alkylation of [YL_2Cl(THF)] (LH = [6-(2,6-dimethylphenyl) pyridin-2-yl]amine) with $LiCH_2SiMe_3$ in hexane gives the alkyl yttrium [$YL_2CH_2SiMe_3$(THF)] (07OM5770). Treatment of the latter with phenylsilane in toluene affords the product of an intramolecular sp^3-hybridized C−H bond activation, [Y(L)(L-H)(THF)]. The neutral ligand LH with [Y (CH_2SiMe_3)$_3$·2THF] in toluene gives the alkyl [Y(CH_2SiMe_3)(L)(THF)] (L = 2-*t*-butyl-6-(1-(quinolin-8-ylamido)-2-trimethylsilanylethyl) phenoxide) (08EJI4126) while the complex of deprotonated bis(4-methyl-2-pyridylamino)tetramethyldisiloxane (LH) [$L_2NdLi(THF)_n$] with [(η^4-cod)Pd (Me)Cl] gives [$L_2NdPdMe$] (98AGE832, 00AGE468, 01CEJ1630, 03EJI791). The lithium salt of **84** and yttrium(III) chloride in THF give heterodinuclear **85** (01AGE4089) that with copper(I) chloride provides the yttrium–copper **86**.



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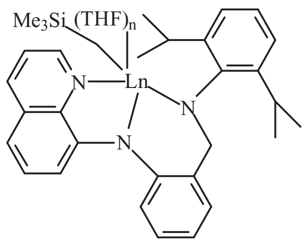

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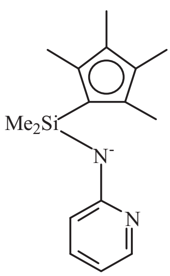

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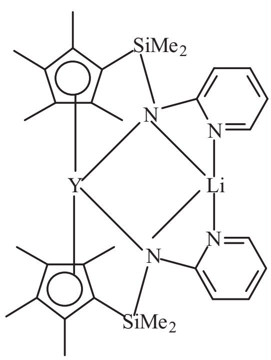

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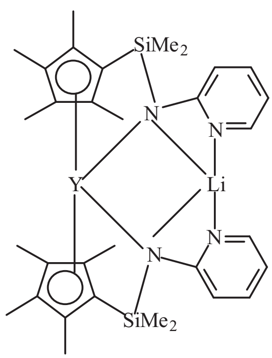

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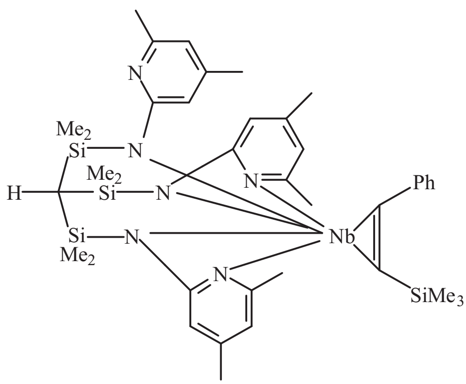
3. DI AND TRI-2PYRIDYLAMINES

Trimethylaluminum with di-2-pyridylamine (LH) yields a mononuclear zwitter ion [$Me_2Al(L)$], dinuclear [$Me_5Al_2(L)$], and tetranuclear [$Me_6Al_4(\mu_3\text{-}O)_2(L)_2$], depending on conditions (97OM4257, 98OM5334). In [Me_2AlL], [Me_2GaPy_2N], dimeric [$Me_2InL]_2$, and polymeric [$Me_2TlL]_\infty$ the metal center is coordinated exclusively through the pyridine nitrogen atoms of the deprotonated ligand (98EJI311). [μ-(2,2′-Dipyridylamido-N^{α},N:N')-μ-(2″,2‴-dipyridylamido-$N^{\alpha'}$,N'':N''')bis{bis(pentafluorophenyl) thallium(III)}] contains a four-membered chelate ring (77ICA(21)L1). Tris (2-aminopyridine) $HC(SiMe_2NHC_5NH_2\text{-}4,6\text{-}Me_2)_3$ as a trilithium salt with [$NbCl_3(DME)(\eta^2\text{-}PhC{\equiv}CSiMe_3)$] gives **87** where two of the three aminopyridinate moieties are $\eta^2(N,N)$ coordinated, and one is coordinated only by the amido-nitrogen center (97OM5585). Di-2-pyridylamine (HL) reacts with [$M(CO)_4(bipy)$] (M = Mo or W) to yield the mixed-ligand [M (CO)$_3$(bipy)(η^1(N)-HL)] (86JCS(D)759). In [$W(CO)_5L$] the di-2-pyridylamine ligand is monodentately coordinated (00AX(C)181). The lithium di-2-pyridyl amido salt (LiL) gives [$Cr_3L_4Cl_2$], which reacts with LiC≡CPh to yield [$Cr_3L_4(C{\equiv}CPh)_2$] (98ICC1). The ligand reveals a bidentate coordination mode as in [$Cr_2L_4(C{\equiv}CPh)_2$] (00IC748). Di-2-pyridylamine reacts with [$Ru_3(CO)_{12}$] or [$Ru_3(CO)_{10}(AN)_2$] to yield the C-metalated **88** (02OM2540). In contrast, in the reaction with [$Os_3(CO)_{10}(AN)_2$] the deprotonated ligand plays the role of a μ_3-η^2-bridge where all the nitrogen donor sites are involved in cluster formation, and the product has structure **89**. On heating or UV-irradiation, the product decarbonylates to give cluster **90**.



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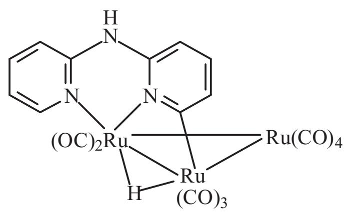

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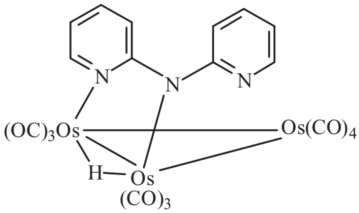

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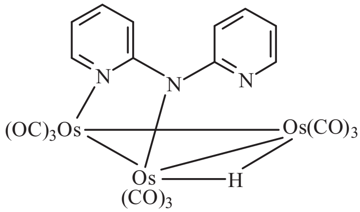

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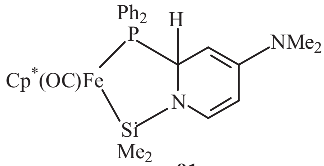
The following mononuclear complexes based on di-2-pyridylamine (HL) are known: [M(L′$_2$)(η^3(*N,N,N*)-HL)](ClO_4) (M = Rh, Ir; L′$_2$ = cod, nbd, tfb; L′ = CO), [M$(CO)_2$(η^3(*N,N,N*)-HL)][M$Cl_2(CO)_2$] (M = Rh, Ir), [(η^4-diolefin)M(Cl)(η^3(*N,N,N*)-HL)] (M = Rh, Ir; diolefin = cod, nbd, tfb), and [(η^4-diolefin)M(η^2(*N,N*)-L)] (M = Rh, Ir; diolefin = cod, nbd, tfb) (86ICA(115)65). In the latter complex only two pyridine nitrogen atoms are coordinated but an amide nitrogen is available as a source of the dinuclear complexes [(η^4-diolefin)Rh(μ-L)Rh(CO)$(PPh_3)_2$](ClO_4) and [(η^4-diolefin)(Cl)Rh(μ-L)Rh(η^4-diolefin)] (diolefin = cod, nbd, tfb). Di-2-pyridylamine reacts with cyclometalated [Ir(2-Phpy)$_2$Cl]$_2$, and further with potassium hexafluorophosphate to yield [Ir(2-Phpy)$_2$(η^2(*N,N*)-L)] (PF_6) where coordination occurs *via* the pyridine nitrogen atoms (06ICA4144). [$Ni_3L_4(AN)_2$]$(PF_6)_2$ with excess phenylacetylene gives [Ni_3L_4(C≡CPh)$_2$] (03JCS(D)3015, 04IC2277). The di-2-pyridylamine (L) complex [Pt(Me)(η^2(*N,N*)-L)(DMSO)](OTf) enters numerous substitution reactions replacing dimethyl sulfoxide by various charged nucleophiles in methanol (98IC5460).

Di-2-pyridylamine (L) with [Pt_2Me_4(μ-SMe_2)$_2$] forms [$PtMe_2$(η^3(*N,N*)-L)], where the ligand is coordinated *via* two pyridine nitrogen heteroatoms (06OM1583) that oxidatively adds methyl iodide to yield platinum(IV) [$PtIMe_3$(L)]. Protonation of [$PtMe_2$(η^2(*N,N*)-L)] using hydrochloric acid gives [Pt(Me)(η^2(*N,N*)-L)X] (X = Cl, OTf). The same process with triflic acid but in the presence of dimethylsulfide gives [Pt(Me)(η^2(*N,N*)-L)(Me_2S)] (OTf). The range of organocopper compounds with η^2(*N,N*)-coordination *via* the pyridine nitrogen atoms includes [Cu(LH)X]Y (X = C_2H_4, C_3H_6, Y = ClO_4; X = C_2H_2, Y = BF_4), [Cu(LH)(CO)(CO_4)] (84IC2813). Di-2-pyridylamine (HL) with [Au(C_6F_5)(THT)] forms [(C_6F_5)Au(μ-HL)Au(C_6F_5)] where only pyridine nitrogen atoms are involved in coordination (03EJI2170).

4. DERIVATIVES OF PYRIDINE AND BENZANNULATED PYRIDINES WITH NONADJACENT AMINO GROUPS

4-(Dimethylamino)pyridine (L) with [(η^5-Cp)$_2Fe_2(CO)_3$(μ-Si(I)(*t*-Bu))] gives [(η^5-Cp)$_2Fe_2(CO)_3$(μ-Si(L)(*t*-Bu))](I) (94JA8575). It also reacts with

$[(\eta^5\text{-Cp})_2Fe_2(CO)_2(\mu\text{-CO})(\mu\text{-Si(I)(CHR}_2))]$ (R = Et, Ph) to yield the η^1(N)-coordinated $[(\eta^5\text{-Cp})_2Fe_2(CO)_2(\mu\text{-CO})(\mu\text{-Si(L)(CHR}_2))](I)$ (97BCJ2193). 4-Dimethylaminopyridine with $[RuCl_2(CO)_2(PPh_3)_2]$ gives the substitution product $[RuCl_2(CO)(L)(PPh_3)_2]$ (98POL2013). 2-Dimethylaminopyridine reacts with $[(\eta^5\text{-Cp}^*)Fe(CO)(\eta^2(Si,P)\text{-}Me_2SiPPh_2)]$ to yield **91** by insertion of the aromatic N–C bond into the Si–P bond (04JA5060). The aromaticity of the pyridine ring is lost.



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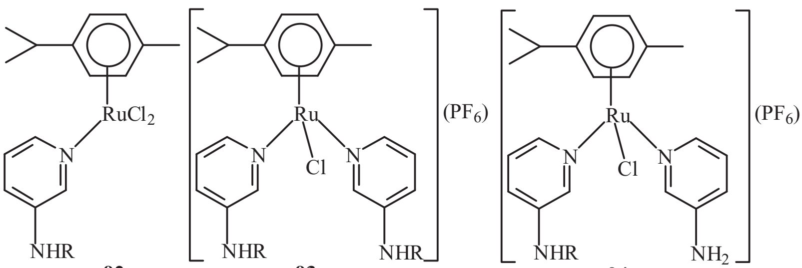
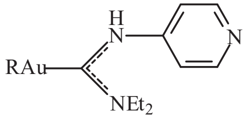
3- and 5-Aminoquinolines (L) react with $[(\eta^4\text{-diene})Rh(Cl)]_2$ (diene = cod, nbd) to yield monomeric $[(\eta^4\text{-diene})Rh(Cl)L]$ monodentately coordinated *via* the pyridine nitrogen atom (99JOM(586)150). 3-Aminopyridine-based ligands react with $[(\eta^6\text{-cymene})(Cl)Ru(\mu\text{-Cl})_2Ru(Cl)(\eta^6\text{-cymene})]$ in acetic anhydride to yield neutral **92** (R = ferrocenyl, anthracenyl), and cationic **93** (R = ferrocenyl, anthracenyl) in the presence of silver hexafluorophosphate (03JOM(666)63). Neutral **92** with 3-aminopyridine in acetic anhydride in the presence of silver hexafluorophosphate gives the mixed-ligand cationic **94** (R = ferrocenyl, anthracenyl). 4-Aminopyridine with $[(\eta^4\text{-1,5-diamino-3-azapentane})Pt(Br)]Br$ in the presence of silver perchlorate gives the η^1(N)-coordinated $[(\eta^4\text{-1,5-diamino-3-azapentane})Pt(4\text{-}NH_2py)](ClO_4)$ (90JCS(D)3271).



92 **93** **94**

An interesting development is based on the neutral gold isocyanide complexes [Au(R)(CNpy-4)] (R = C_6F_5, 2,4,6-$(CF_3)_3C_6H_2$) and $[Au_2(\mu\text{-}C_6F_4C_6F_4)(CNpy\text{-}4)_2]$ (07JCS(D)5339). They enter into nucleophilic attack by NEt_2H, NH_2Me (for mononuclear and dinuclear precursors), and $H_2NC_6H_4NH_2$ at the α-carbon atom of the isocyanide moiety to yield the gold carbene derivatives **95** (R = C_6F_5, 2,4,6-$(CF_3)_3C_6H_2$), **96** (R = C_6F_5, 2,4,6-$(CF_3)_3C_6H_2$), **97** and **98**.



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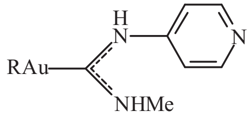

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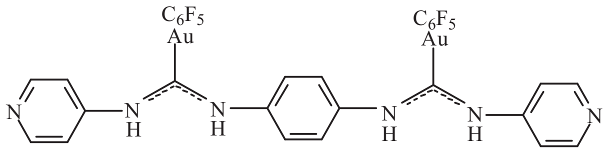

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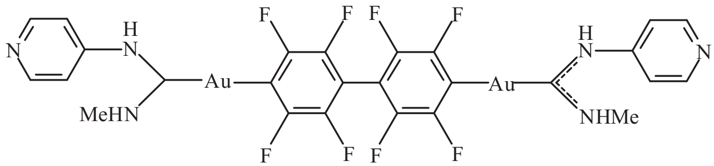

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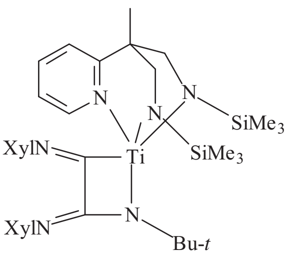
5. AMINOMETHYL DERIVATIVES

5.1 Nontransition metals and titanium group

Thallium amide with $[(2\text{-}C_5H_4N)C(Me)\{CH_2N(Li)SiMe_3\}_2]_2$ substitutes the lithium ions by thallium(I) in two steps, yielding the mixed metal amide $[(2\text{-}C_5H_4N)C(Me)\{CH_2N(Li)SiMe_3\}\{CH_2N(Tl)SiMe_3\}]_2$ and the thallium(I) diamide $[(2\text{-}C_5H_4N)C(Me)\{CH_2N(Tl)SiMe_3\}_2]_2$ (00EJI2577).

Alkylation of the complex of tridentate diamidoamine ligand $[TiCl_2\{MeC(2\text{-}C_5H_4N)(CH_2NSiMe_3)_2\}]$ with RMgCl ($R = PhCH_2$, Me_3SiC_2) yields the mono- and dialkyl $[Ti(Cl)R\{H_3CC(2\text{-}C_5H_4N)(CH_2NSiMe_3)_2\}]$, $[TiR_2\{H_3CC(2\text{-}C_5H_4N)(CH_2NSiMe_3)_2\}]$, and $[TiR_2\{H_3CC(2\text{-}C_5H_4N)(CH_2NSiMe_3)_2\}]$ (97CB1751, 00CC173). Imido titanium compound $[Ti(NBu\text{-}t)(py)(MeC(2\text{-}C_5H_4N)(CH_2NSiMe_3)_2)]$ with 2,6-xylylisocyanide gives the product of a double insertion into the titanium−imido bond, **99** (97CC1555, 00OM4784). Reactions with 2-butyne or 1-phenylpropyne are C–N couplings and also lead to the four-membered titanaazetidines **100** (R = Me, Ph) (98CC2555, 01OM3308). The same products can be obtained in a 2 + 2 cycloaddition with 1,2-butadiene and phenylallene. [2 + 2] Cycloaddition with *t*-BuCP forms the mononuclear **101** (00OM3205). $[Ti(NBu\text{-}t)(L)(py)]$ and $[Ti(N\text{-}2,6\text{-}C_6H_3Pr\text{-}i_2)(L)(py)]$ with phenyl acetylene and *p*-tolyl acetylene give the [2 + 2] cycloaddition products $[Ti(L)(\eta^2\text{-}N$

(Bu-*t*)CH=CR)] (R = Ph, *p*-Tol) and [Ti(L)(η^2-*N*(2,6-C_6H_3Pr-i_2)CH=CR)] (R = Ph, *p*-Tol) (04CC704). Titanium imido complexes [Ti(NBu-*t*)(L)(py)] (L = MeC(2-C_5H_4N)(CH_2N(3,5-$C_6H_3Me_2$))$_2$) with aryl acetylenes afford the [2 + 2] cycloadducts **102** (Ar = Ph, *p*-Tol) (08OM2518). A second equivalent of alkyne affords azatitanacyclohexadienes **103** (07OM5522). The complex with Ar = Ph inserts sulfur and selenium atoms into the Ti–C bond of the azatitanacyclobutene unit to give five-membered metallacycles [Ti(L)(η^2-N(*t*-Bu)CH=C(Ph)E)] (E = S, Se). Isonitriles insert into the Ti–C bonds to give **104** (Ar = Ph, R = Xyl, Cy, *t*-Bu; Ar = *p*-Tol, R = *t*-Bu). Zirconium (1-pyridinio)imido complex [Zr(L)(N=NC_5H_5)(OTf)(py)] reacts with PhC≡CLi to yield alkynyl **105** (08OM172). A similar reaction occurs with a hafnium analog (08JCS(D)6231). The hydrazido complex with the same ligand [Zr(L)($NHNMe_2$)Cl] with $LiCH_2SiMe_3$ gives [Zr(L)($NHNMe_2$)(CH_2SiMe_3)] (08JCS(D)2111).



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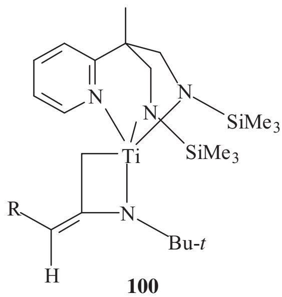

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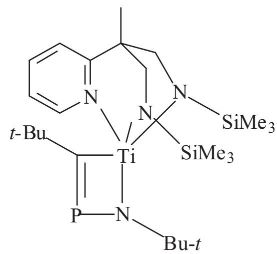

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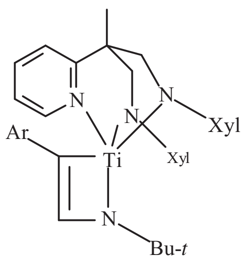

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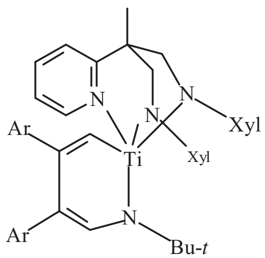

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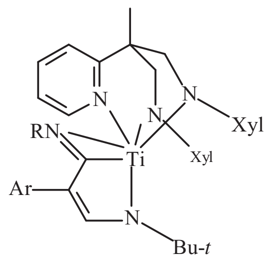

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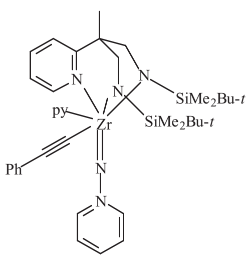

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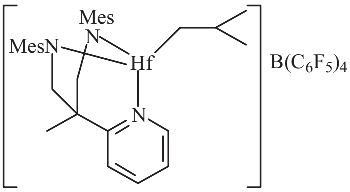
The arylated diamidopyridine ligands $[MeC(2\text{-}C_5H_4N)(CH_2NAr)_2]^{2-}$ (Ar = 2,4,6-$Me_3C_6H_2$, 2,4,6-(*i*-Pr)$_3C_6H_2$) (L) form alkyls of zirconium and hafnium [$ZrLR_2$] (Ar = 2,4,6-$Me_3C_6H_2$, R = Me, $PhCH_2$, Np, *i*-Bu), [Zr(L)(THF)Me_2] (Ar = 2,4,6-$Me_3C_6H_2$), [Li(Et_2O)][$ZrLMe_3$] (Ar = 2,4,6-$Me_3C_6H_2$), [Zr(L)(*i*-Bu)$_2$] (Ar = 2,4,6-(*i*-Pr)$_3C_6H_2$), [$HfLR_2$] (Ar = 2,4,6-$Me_3C_6H_2$, R = Me, Et, *n*-Pr, *n*-Bu, *i*-Bu, *i*-Pr), [Hf(L)(*i*-Pr)Cl] (Ar = 2,4,6-$Me_3C_6H_2$), and [Hf(L)(*t*-Bu)Cl] (00JA7841, 01JA10746, 02OM5785). [$HfLR_2$] (Ar = 2,4,6-$Me_3C_6H_2$, R = *i*-Bu) with (Ph_3C)(B(C_6F_5)$_4$) gives cationic **106** active in 1-hexene polymerization (03OM4569). The dimethyl analog similarly forms dinuclear **107**.



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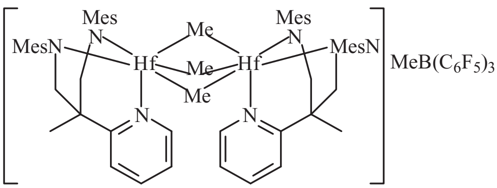
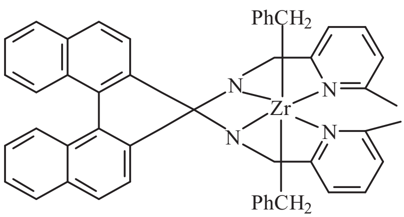


107

Complexes of (*N*,*N*′-di(6-methylpyridin-2-yl)-2,2′-diaminobinaphthalene) (H_2L) [$MLCl_2$] or [ML(OTf)$_2$] (M = Zr, Hf) with Me_2CHCH_2MgBr give [ML(CH_2CHMe_2)$_2$] (M = Zr, Hf) (05OM3335). The ligand with [Zr(CH_2Ph)$_4$] gives [Zr(L)(CH_2Ph)$_2$], **108**. Both can be activated with B(C_6F_5)$_3$ to give [ML($CH_2CHCHMe_2$)](HB(C_6F_5)$_3$) or [Zr(L)(CH_2Ph)($PhCH_2B(C_6F_5)_3$)]. (2-C_5H_4N)CH_2N{$CH_2CH_2N(H)SiMe_3$}$_2$ (H_2L) forms the zirconium dichloride [Zr(L)Cl_2], which with $MgCH_2PhCl$ or $MgCH_2SiMe_3Cl$ forms chloroalkyls **109** (R^1 = Cl, R^2 = CH_2Ph, CH_2SiMe_3) (05OM5586). The product (R^2 = CH_2Ph) with methylmagnesium bromide forms mixed dialkyl **109** (R^1 = Me, R^2 = CH_2Ph). [Zr(L)Cl_2] with methyl lithium, benzylmagnesium chloride, $MgCH_2SiMe_3Cl$, *t*-butylmagnesium chloride yields a series of dialkyls **109** (R^1 = R^2 = Me, CH_2Ph, CH_2SiMe_3, CH_2Bu-*t*). Bis(2-picolyl)(2-hydroxy-3,5-di-*t*-butylbenzyl)amine with [Zr(CH_2Ph)$_4$] gives trialkyl **110** (07IC7199).



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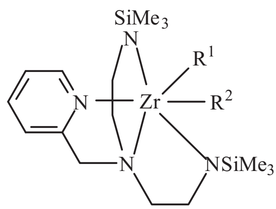

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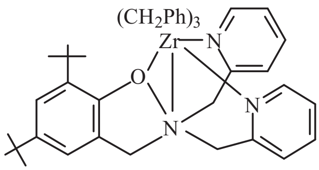
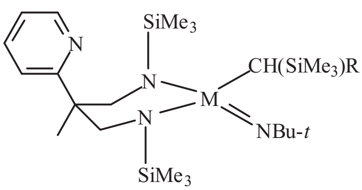


110

5.2 Vanadium group

The reactivity of [M(NR)Cl(η^3(*N,N,N*)-MeC(2-C_5H_4N)(CH_2NSiMe_3)$_2$)(py)] (M = Nb, Ta; R = *t*-Bu, 2,6-C_6H_3(*i*-Pr)$_2$) is of interest and often leads to organometallic compounds (01OM3531). Thus with LiCHR($SiMe_3$) both niobium and tantalum species give four-coordinate **111** (R = $SiMe_3$, H; M = Nb, Ta) without pyridine nitrogen coordination, while tantalum **112** is five-coordinated. The niobium precursor with $PhCH_2MgCl$ gives **113** with involvement of the heteroring, while that with LiC_5H_4Me leads to **114** where the pyridine ring is not coordinated (01CCR(216)65). [Ta(NBu-*t*)Cl (η^3(*N,N,N*)-MeC(2-C_5H_4N)(CH_2NSiMe_3)$_2$)(py)] with methyl lithium gives [Ta(NBu-*t*)(η^3(*N,N,N*)-MeC(2-C_5H_4N)(CH_2NSiMe_3)$_2$)(Me)] and with allylmagnesium chloride [Ta(NBu-*t*)(η^3(*N,N,N*)-MeC(2-C_5H_4N)(CH_2NSiMe_3)$_2$) (η^1-C_3H_5)] (04OM4444). (3,5-$Cl_2C_6H_3HNCH_2$)$_2$C(2-C_5H_4N)(CH_3) (H_2L) is the basis for [$TaMe_3$(L)] (01OM5682).



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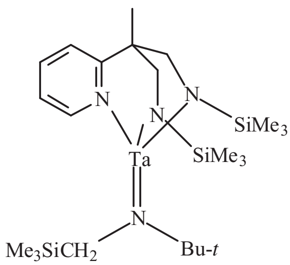

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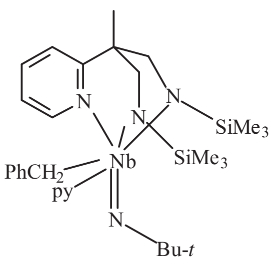

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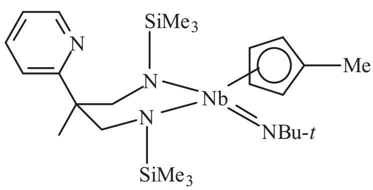
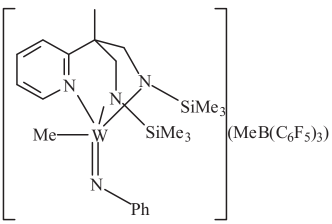


114

5.3 Chromium group

[W(NPh)(L)Cl_2] (L = MeC(2-C_5H_4N)(CH_2NSiMe_3)$_2$) with methylmagnesium bromide or phenylmagnesium chloride affords [W(NPh)(L)R_2] (R = Me, Ph) (03IC4961). [W(L)(NPh)Me_2] reacts with B(C_6F_5)$_3$ to form cationic [W(L)(NPh)Me](MeB(C_6F_5)$_3$) **115** (L = MeC(2-C_5H_4N)(CH_2NSiMe_3)$_2$) (02CC2618). Carbon dioxide cycloadds at the W=NPh bond to yield **116**. With AN and THF, **115** forms adducts **117** (L = AN, THF) (04OM4444). When the reaction with B(C_6F_5)$_3$ is conducted in methylene chloride with 0.5 equivalents of a reagent, the major product is the methyl-bridged dinuclear **118**. Methyl tungsten [W(NPh)(L)Me]$^+$ (L = MeC (2-C_5H_4N)(CH_2NSiMe_3)$_2$) **115** reacts with the heterocumulenes CO_2, CS_2, RNCO, and RNCS (R = *t*-Bu, Ar) by insertion into a W–N bond to give **116** (illustrated for carbon dioxide) and subsequent rearrangement *via* a 1,3-migration of a trimethylsilyl group to give **119** (05ACR839, 05OM2368).



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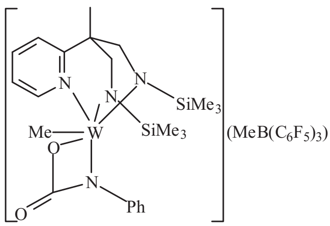

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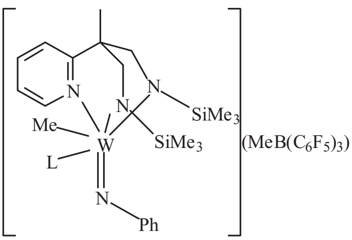

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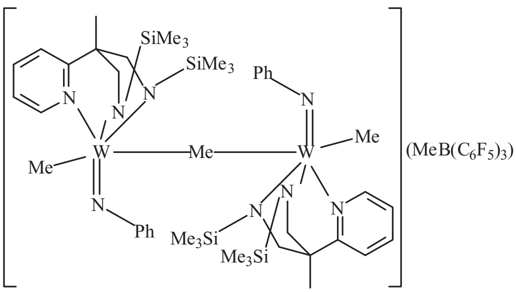

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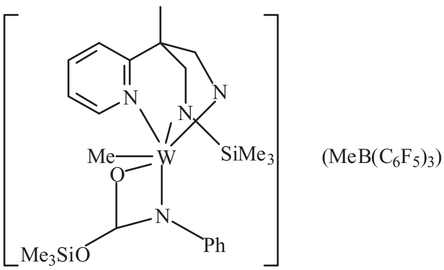
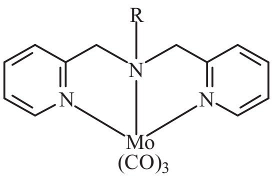


119

Bis(2-pyridylmethyl)amine (L) forms cationic $[W(CO)_2(\eta^3\text{-allyl})(\eta^3(N,N,N)\text{-L})](PF_6)$ (allyl = C_3H_5, 2-MeC_3H_4) and tris(2-pyridylmethyl)amine (L) – $[M(CO)_2(\eta^3\text{-allyl})(\eta^3(N,N,N)\text{-L})](PF_6)$ (M = Mo or W, allyl = C_3H_5 or 2-MeC_3H_4) (92JOM(435)319). In the latter, the N-donor set comprises two pyridyl rings and the exocyclic nitrogen atom while the third pyridyl ring is oriented away from the metal. Bis(2-pyridylmethyl)amines with $[Mo(CO)_6]$ give **120** (R = H, $PhCH_2$) (02EJI1518). Using bromine in chloroform, the product with R = H gives cationic **121**. Tridentate coordination of the potentially tetradentate pyridinophane ligand *N*,*N*′-dimethyl-2,11-diaza[3.3](2,6)pyridinophane (L) occurs in $[Mo(\eta^3(N,N,N)\text{-L})(CO)_3]$ (98EJI1381). The complex of tri-2-pyridylmethylamine (L) $[Cr(L)Cl_2](BPh_4)$ with methyl and phenyl Grignard reagents produces cationic chromium(III) **122** (R = Me, Ph) (03IC6876, 08EJI2633).



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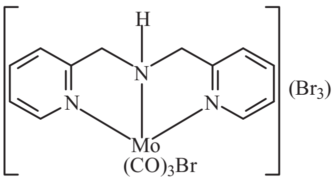

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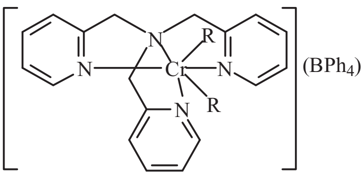
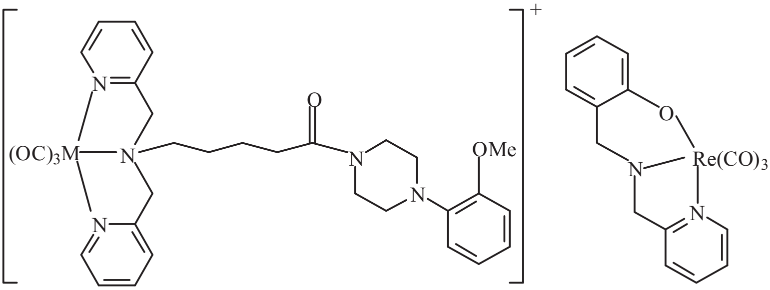


122

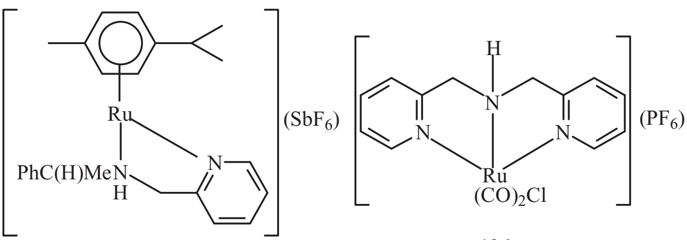
5.4 Manganese and iron group

With substituted bis(2-pyridyl)methylamine and the carbonyl precursor $[M(CO)_3(H_2O)_3]^+$ chelates **123** (M = Re, Tc) form using microwave heating (08IC8213). An illustration of the tripodal *N*,*N*,O-chelates is rhenium(I) **124**, prepared from the corresponding ligand and $[Re(CO)_3(H_2O)_3]Br$ in methanol at reflux (08IC1337). Carbohydrate-appended 2,2′-dipicolylamine ligands, 2-(bis(2-pyridinylmethyl)amino)ethyl-β-D-glucopyranoside, 2-(bis(2-pyridinylmethyl)amino)ethyl-β-D-xylopyranoside, and 2-(bis(2-pyridinylmethyl)amino)ethyl-α-D-mannopyranoside (L) with $(NEt_4)_2[ReBr_3(CO)_3]$ form $[Re(CO)_3(\eta^3(N,N,N)\text{-L})]^+$ (05IC2698).

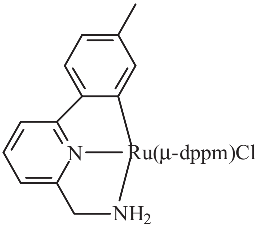


123 **124**

Complexes of 2,6-bis((dimethylamino)methyl)pyridine (L), [Ru(η^3(*N*,*N*,*N*)-L)Cl(PPh_3)](OTf) and [Ru(η^3(*N*,*N*,*N*)-L)(OTf)(PPh_3)](OTf) under carbon monoxide yield [Ru(η^3(*N*,*N*,*N*)-L)Cl$(CO)_2$](OTf) and [Ru(η^3(*N*,*N*,*N*)-L)(OTf)(CO)(PPh_3)](OTf), respectively (98IC1749). *N*-(2-pyridylmethyl)-(R)-1-phenylethylamine, *N*-(2-pyridylmethyl)-(R)-1-naphthylethylamine, *N*-(2-pyridylmethyl)-(R)-1-cyclohexylethylamine, *N*-(2-pyridylmethyl)-(1R2S,4R)-1-bornylamine, and *N*-(2-quinolylmethyl)-(R)-1-naphthylethylamine with [(η^6-arene)Ru(μ-Cl)Cl$]_2$ (arene = *p*-MeC_6H_4Pr-*i*, C_6H_6, C_6Me_6) in methanol-containing sodium hexafluoroantimonate give [(η^6-arene)Ru(L)Cl]; one representative is illustrated as **125** (08JCS(D)3328). Bis(2-pyridylmethyl)amine at reflux with [Ru$(CO)_2Cl_2]_n$ in ethanol gives bis-chelate **126** (06ICA309). 1-(Pyridin-2-yl)methanamine ligands in combination with phosphines afford ruthenium catalysts, for example, **127**, for the reduction of ketones and aldehydes (08EJI4041). Terdentate cyclometalated **128** (R = Cl, OH, H) are catalysts for transfer hydrogenation of acetophenone with 2-propanol (07CEJ7479). 1-Substituted-1-(6-arylpyridin-2-yl)methanamines (R = H, Me, *t*-Bu) react with [Ru$Cl_2(PPh_3)_3$], 1-(1-(dicyclohexylphosphino)ethyl)-2-(diphenylphosphino)ferrocene in the presence of triethylamine to yield cyclometalated **129** (R = H, Me, *t*-Bu; R^1 = H, Me) that serve as catalysts for the asymmetric transfer hydrogenation of alkyl aryl ketones (09CEJ729).



125 **126**



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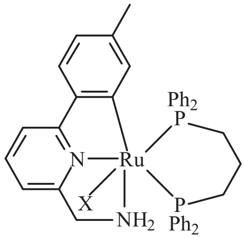

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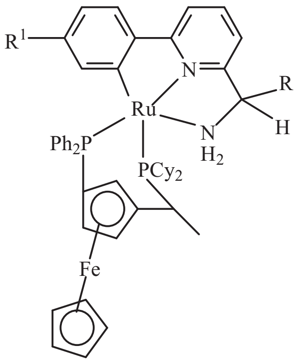
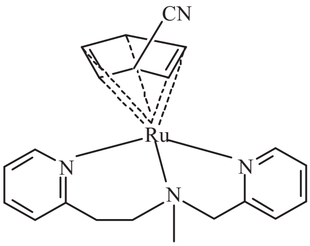


129

(2-(2-Pyridyl)ethyl)(2-pyridylmethyl)methylamine (L) with [(η^6-C_6H_6)$RuCl_2$]$_2$ and ammonium hexafluorophosphate in methanol gives [(η^6-C_6H_6)Ru(η^3(*N*,*N*,*N*)-L)](PF_6)$_2$ (94JCS(D)465). In a similar way (η^5-cyanocyclohexadienyl)ruthenium(II) **130** can be prepared (96IC3431). Identical bis-chelates containing rings of varying size are based on *N*-methyl ((2-pyridyl)ethyl(2-pyridyl)methyl)amine, *N*-methyl-*N*,*N*-bis(2-pyridylmethyl)amine, and *N*-methyl-*N*,*N*-bis(2-pyridylethyl)amine (09ICA483). Another series includes half-sandwich η^6-benzene ruthenium complexes containing η^3(*N*,*N*,O)-ligands, 4-nitro-6-(((2′-(pyridin-2-yl) ethyl) methylamino)methyl)-phenol, 2,4-di-*t*-butyl-6-(((2′-(pyridin-2-yl) ethyl)methylamino)methyl)-phenol, and 2,4-di-*t*-butyl-6-((2′-(pyridin-2-yl)benzylamino)methyl)-phenol (07JOM3248). (2,6-Bis(2-mercaptophenylamino)dimethylpyridine) (H_2L) with Fe(II) salts gives pentacoordinate [FeL], which coordinates CO to give diamagnetic [Fe(CO)(L)] and in a similar way [Ru(CO)(L)], which can be illustrated as **131** (99IC5314). With $NOBF_4$ in methylene chloride **131** (M = Ru) is transformed to the tetrafluoroborate cationic complex, but application of $NaBH_4$ in methanol reverses the process (05ICA1798).



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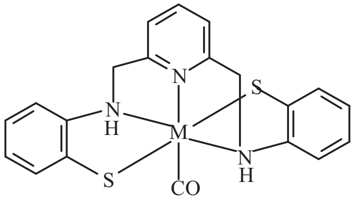
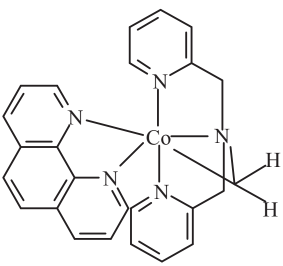


131

5.5 Cobalt group

UV-photolysis of [*N*,*N*-bis(2-pyridylmethyl)amino acidato]phenanthrolinecobalt(III) (amino acidato = glycinato, alaninato, 2-cyclopropylglycinato) leads to the elimination of carbon dioxide from the amino acid based chelate and the formation of Co–C–N metallacycles of the type **132** (00JCS(D)2801, 04JCS(D)150). 2,6-Bis(1′,3′-diamino-2′-methylprop-2′-yl)pyridine (L) forms η^3(*N*,*N*,*N*)-coordinated [Co(L)(Me)]$(NO_3)_2$ and [Co(L)(Me)](S_2O_6) can be prepared from [Co$(NH_3)_5$(Me)]$(NO_3)_2$ and by anion exchange of the product with $Na_2S_2O_6{\cdot}2H_2O$ (99ICA(286)98). Aminomethylpyridine ligands with [(η^5-Cp*)Ir(μ-Cl)Cl]$_2$ in methanol-containing sodium hexafluoroantimonate yield cationic **133** (R = Ph, Nph, Cy), **134**, and **135** (07JCS(D)1911, 08CCR782). These products with silver hexafluoroantimonate in methylene chloride – acetone yield dicationic aqua species similar in structure to **133–135** where the chloride ligand is substituted by a water molecule contained in the acetone and the complex ion acquires charge +2. Dicationic species similar to **133** (R = Ph, Nph) and **134** in acetone at room temperature evolve to cyclometalated **136**, **137**, and **138**. In [(η^5-Cp*)Ir(Cl)(L)](SbF_6) (L = *N*-(2-pyridylmethylene)-1-phenylethylamine, *N*-(2-pyridylmethylene)-1-naphthylethylamine, *N*-(2-quinolylmethylene)-1-naphthylethylamine, *N*-(6-methyl-2-pyridylmethylene)-1-naphthylethyl amine, *N*-(2-pyridylmethylene)-1-cyclohexylethylamine,



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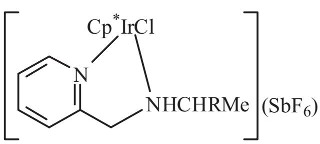
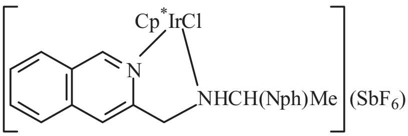


133

N-(2-pyridylmethylene)-1-bornylamine two nitrogen atoms of the imine ligands are involved in coordination (98OM2986). In [(η^5-Cp*)Rh(Cl)(L)] (SbF_6) (L = *N*-(2-pyridylmethylene)-1-phenylethylamine, *N*-(2-pyridylmethylene)-1-naphthylethylamine, *N*-(2-pyridylmethylene)-1-cyclohexylethylamine) the coordination is the same (99OM3364).



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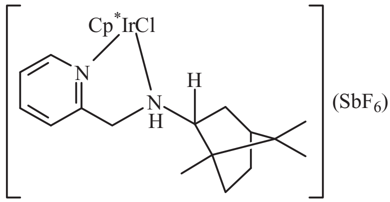

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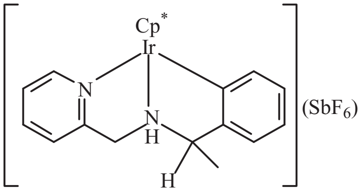

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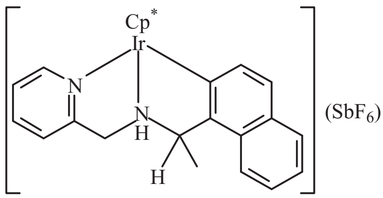

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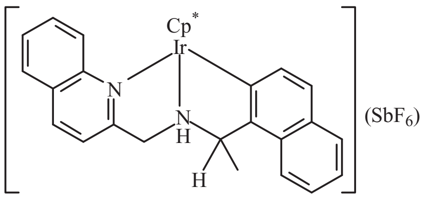
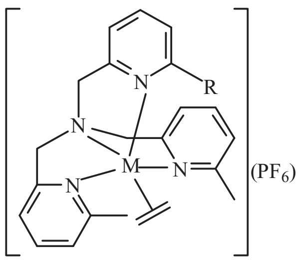


138

Bis(2-pyridylmethyl)amine (L) forms the η^3(*N*,*N*,*N*)-coordinated complex [(η^4-cod)Rh(L)](BPh_4) (99CEJ2921, 03OM3022, 04EJI2385). *N*,*N*,*N*-Tri((6-methyl-2-pyridyl)methyl)amine forms iridium(I)–ethene complex **139** (M = Ir, R = Me) as the hexafluorophosphate salt (02AGE2135). Oxidation using ferrocinium hexafluorophosphate then gives dicationic iridium(II) **140**. In a similar way, the complex with R = H can be prepared (02OM4312). Both **140** (R = H, Me) in acetonitrile enter iridium–carbon coupling yielding the ethene-bridged iridium(III) dinuclear **141**. Under oxygen in acetonitrile, further oxidation to iridium(III) formylmethyl **142** occurs. In acetone, the first step is the formation of the superoxo radical **143**, which on 1,3- or 1,2-intramolecular coupling of coordinated ethene and Ir-O-O moieties may produce radicals **144** or **145** (05JCS(D)879). One-electron oxidation of [(η^2-C_2H_4)Ir^I(L)]$^+$ (L = *N*, *N*,*N*-tri((6-methyl-2-pyridyl)methyl)amine, *N*-(2-pyridylmethyl)-*N*,*N*,-di((6-methyl-2-pyridyl)methyl)-amine results in iridium(II) [(η^2-C_2H_4)Ir^{II}(L)]$^{2+}$ (05JA1895). In acetonitrile, they generate radical [(η^1-C_2H_4)Ir^I(L)(AN)]$^{2+}$.

Oxygenation of **139** (M = Rh, Ir; R = H, Me) in solution gives 2-rhoda(III) and 2-irida(III)-oxetanes **146** (M = Rh, Ir; R = H, Me) (97AGE2064, 00CEJ298, 04AGE4142). Cationic **139** (M = Rh, R = H) with H_2O_2 and NH_4^+ in acetonitrile gives a metallacyclic amide (99AGE219). In the solid phase, **139** (M = Rh, Ir; R = H) enter into dioxygenation with molecular oxygen or air to yield 3-metalla-1,2-dioxolanes represented by isomers **147** and **148** (01AGE2106, 03EJI1072). Cationic $[(\eta^2\text{-}C_2H_4)Rh(L)]^+$ (L = *N*-alkyl-*N*,*N*-di(2-pyridylmethyl) amine, alkyl = R = Me; R = *n*-Bu; R = $PhCH_2$) are oxygenated in acetonitrile by aqueous hydrogen peroxide to 2-rhoda(III) oxetanes $[(\eta^2\text{-C,O-}CH_2CH_2O\text{-})Rh(L)(AN)]^+$ (01CEJ416). Cationic **139** (M = Rh, R = Me) is the result of the reaction of *N*,*N*,*N*-tri((6-methyl-2-pyridyl)methyl)amine and $[(\eta^2\text{-}C_2H_4)_2Rh(\mu\text{-}Cl)]_2$ in methanol in the presence of potassium hexafluorophosphate (02EJI2671). $[(\eta^2\text{-}C_2H_4)Ir^{II}(L)]^{2+}$ (L = *N*,*N*,*N*-tri((6-methyl-2-pyridyl)methyl) amine and ethyl diazoacetate reacts with the Ir-carbenoid radical $[Ir^{III}(CH(COOEt))(AN)(L)]^{2+}$ which then couples with the acetonitrile adduct of the precursor and leads to C–C bond formation in dinuclear tetracationic iridium (III) **149** (08CEJ7594). In contrast, trimethylsilyldiazomethane gives the mononuclear dicationic iridium(III) **150**.



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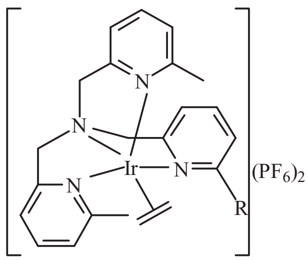

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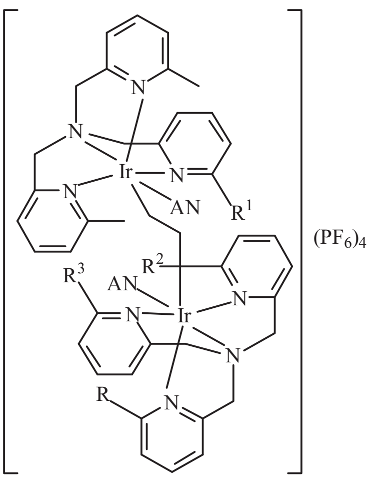

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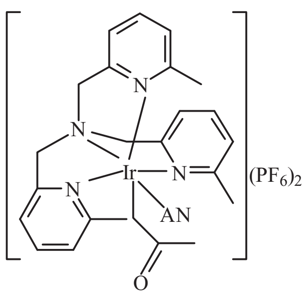

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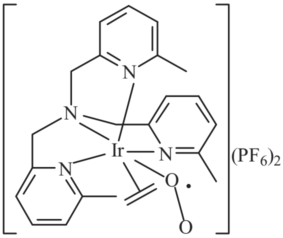

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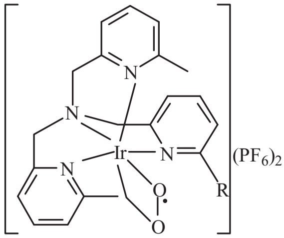

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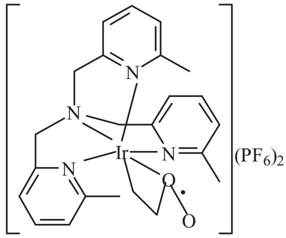

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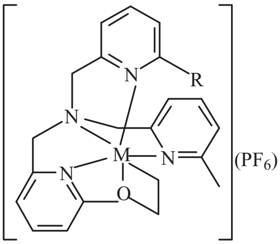

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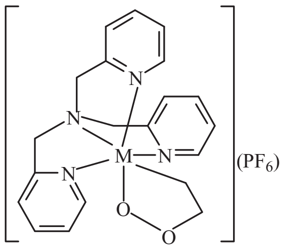

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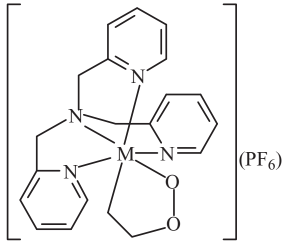

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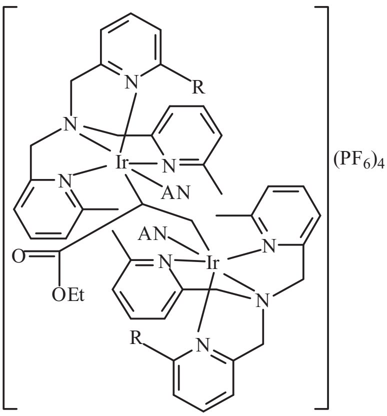

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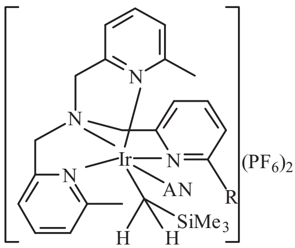
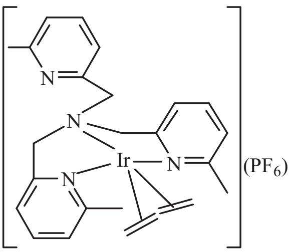


150

N,*N*,*N*-Tri((6-methyl-2-pyridyl)methyl)amine and *N*-methyl-*N*,*N*-bis((6-methyl-2-pyridyl)methyl)amine with [(η^2-C_2H_4)Ir(Cl)] and potassium hexafluorophosphate in methanol give the bis(ethene) iridium(I) **151** and **152**, respectively. Both readily dissociate one ethene molecule. In the case of **151**, the mono-ethene **139** (M = Ir, R = Me) slowly transforms to the cyclometalated **153** in acetonitrile. The rhodium complex reacts with molecular oxygen with displacement of ethene and formation of a peroxo-species. Both iridium mono-ethene species, in contrast, bind molecular oxygen in a peroxo-arrangement without loss of the ethene molecule. *N*-(2-pyridylmethyl)-*N*-(6-methyl-2-pyridylmethyl)amine (L) with $[(\eta^4\text{-nbd})Rh(\mu\text{-Cl})]_2$ and ammonium hexafluorophosphate gives rhodium(I) $[(\eta^4\text{-nbd})Rh(\eta^3(N,N,N)\text{-L})](PF_6)$ (04OM4236) that with silver hexafluorophosphate leads to rhodium(II) $[(\eta^4\text{-nbd})Rh(\eta^3(N,N,N)\text{-L})](PF_6)_2$. Attempted oxidation using ferric chloride in acetone–water leads to rhodium(III) η^1-3-hydroxy- or 3-methoxynonbornenyl dinuclear **154**. Ferrocenium hexafluorophosphate leads to similar isomeric products. Protonation of the rhodium(I) material occurs at the pyridine nitrogen atom to give cationic **155**. Bis(2-picolyl)amine (H_2L) forms $[(\eta^4\text{-cod})Ir(H_2L)]^+$ **156**, which readily undergoes double deprotonation of the amine and one of the methylene protons to yield the anionic $[(\eta^4\text{-cod})Ir(L)]^-$ (08CEJ10932). The latter can be oxidized to the ligand-centered radical **157** even in the air, and in this radical one of the pyridine rings is switched not coordinated and the other partially loses aromaticity.



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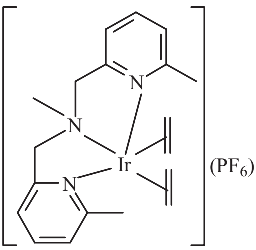

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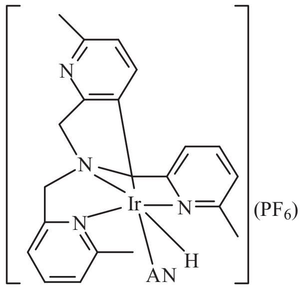

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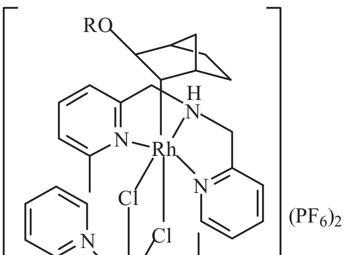

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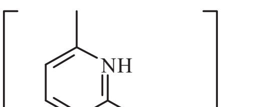

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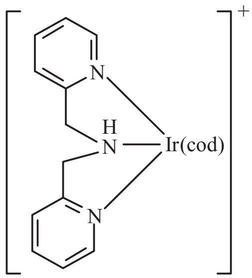

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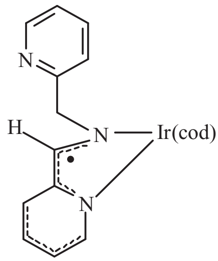
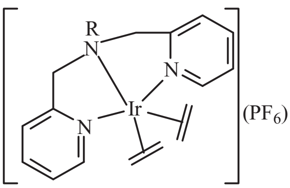


157

N,*N*′-di(2-pyridylmethyl)amine, *N*-methyl-*N*,*N*′-di(2-pyridylmethyl)amine, and *N*-benzyl-*N*,*N*′-di(2-pyridylmethyl)amine (L) react with [(η^2-C_2H_4)$_4$Ir(Cl)] and potassium hexafluorophosphate in methanol to yield iridium(I) cationic [(η^2-C_2H_4)$_2$Ir(η^3(*N*,*N*,*N*)-L)](PF_6) **158** (R = H, Me, CH_2Ph) (05OM5964). Heating **158** (R = H) in acetonitrile gives iridium (III) ethyl **159**. In contrast, heating **158** (R = CH_2Ph) leads to a dinuclear iridium(III) **160** containing a cyclometalated benzylamino moiety and two bridging hydride ligands. In acetonitrile, the product readily transforms to mononuclear iridium(III) hydride **161**.



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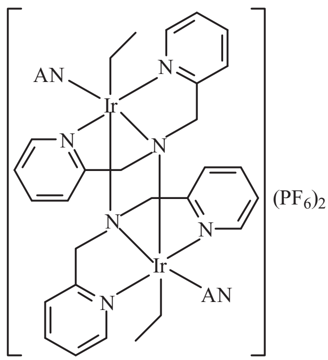

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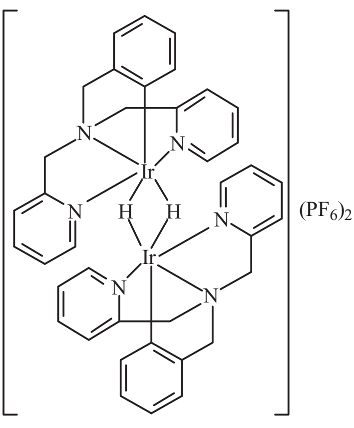

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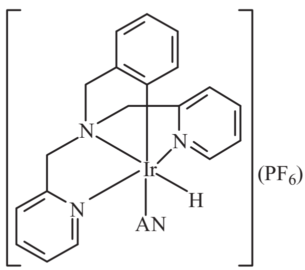
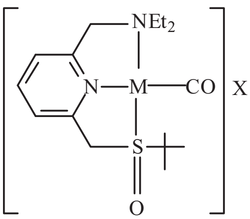


161

2-(Diethylamino-methyl)-6-(*t*-butyl sulfonyl methyl)pyridine (L) reacts with [(η^2-COE)$_4$Rh$_2$Cl$_2$] in acetonitrile to form neutral rhodium(I) [Rh (η^3(*N*,*N*,S)-L)(AN)] (08OM1892). The product with rhodium(I) or iridium (I) precursors gives cationic [Rh(η^3(*N*,*N*,S)-L)(AN)](BF$_4$) and [(η^2-COE)Ir (η^3(*N*,*N*,S)-L)](BF$_4$). These are readily carbonylated to yield **162** (M = Rh, X = PF$_6$) and **162** (M = Ir, X = BF$_4$) and further on deprotonation by *t*-BuOK – dearomatized **163** (M = Rh, Ir). The process can be reverted in acetic acid. Using dimethylaminopyridines, pyridinophane and macrocyclic ligands can be prepared. They react with [(η^4-cod)M(Cl)$_2$] (M = Rh, Ir) and [(η^4-nbd)Rh(Cl)$_2$] to yield chloride cationics **164** and **165**, which can be transformed to PF$_6$, BF$_4$, or BPh$_4$ salts (02EJI457).



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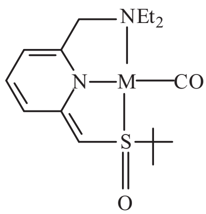

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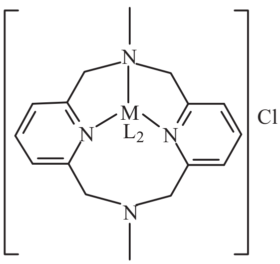

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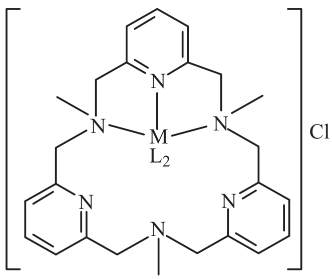
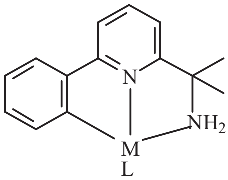


165

5.6 Nickel group

2-Phenyl-6-(2-amino-*i*-propyl)pyridine with K$_2$[PtCl$_4$] and PdCl$_2$ gives cyclometalated **166** (M = Pt, Pd, L = Cl) (04OM4406). Potassium *t*-butoxide acts a deprotonating agent for the platinum complex and leads to heterodinuclear bis-chelate **167**. Species **166** (M = Pt, L = Cl) with molecular hydrogen gives hydride **166** (M = Pt, L = H). 6-Phenyl-2-((2,6-di-*i*-propylphenyl) imino)pyridine (HL) with Pd(OAc)$_2$ and PdCl$_2$(PhCN)$_2$ in benzene yields neutral *ortho*-palladated **168** (X = OAc, Cl) (05EJI4794). The chloride in the presence of NaB(3,5-(CF$_3$)$_2$C$_6$H$_3$)$_4$ reacts with acetonitrile, triphenylphosphine, di(*o*-methoxyphenyl)phosphine (L^1) in methylene chloride to give cationic [Pd(L^1)(η^3(C,*N*,*N*)-L)](B(3,5-(CF$_3$)$_2$C$_6$H$_3$)$_4$).



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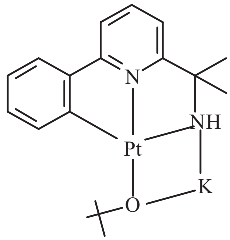

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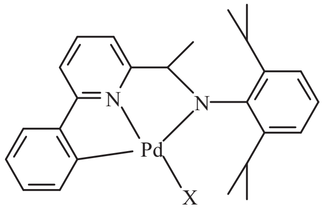
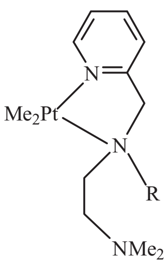


168

Bis(2-pyridylmethyl)amine (L) is η^3(*N,N,N*)-coordinated in [Pt(L)(Me)]Cl and [Pt(L)(η^1-allyl)Me_2]Br (97OM1946). *N,N,N*′-Trimethyl-(*N*′-(2-pyridylmethyl)ethylenediamine, *N*-benzyl-*N,N*′-dimethyl-*N*-(2-picolyl)ethylenediamine with [$PtMe_2$(μ-SMe_2)]$_2$ give the η^2(*N,N*)-chelates **169** (R = Me, CH_2Ph) (03OM787). Oxidative addition of methyl iodide forms cationic bis-chelates of platinum(IV) **170** (R = Me, CH_2Ph) with η^3(*N,N,N*)-coordination. Protonation with HX (X = CF_3SO_3, CF_3COO, BF_4) leads to similar cationic **171** (R = Me, CH_2Ph; X = CF_3SO_3, CF_3COO, BF_4), which reductively eliminate methane to provide platinum(II) **172** with the same set of R and X. The methyl(hydrido)platinum(IV) complex [Pt(H)(CH_3)$_2$(L)]$^+$ (L = di-2-pyridyl methylamine) reductively eliminates methane (99OM4456). 2-Aminomethylpyridine with (n-Bu_4N)[Pt_2(μ-Cl)$_2$(C_6F_5)$_4$] gives chelate **173** (04JCS(D)2733).



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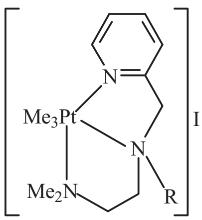

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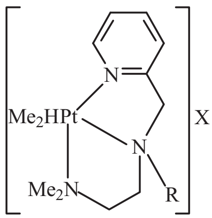

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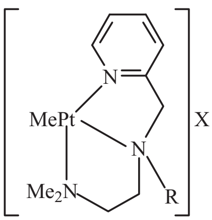

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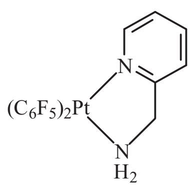
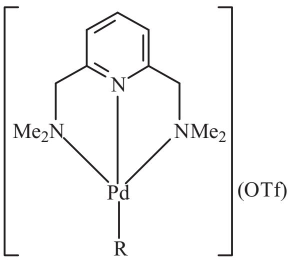


173

2,6-Bis((dimethylamino)methyl)pyridine and *N*,*N*,*N*′-trimethyl-*N*′-(2-picolyl)ethylenediamine (L) are the basis for complexes [Pd(R)(η^3(*N*,*N*,*N*)-L)](OTf), **174** and **175** (R = Me, Ph, 4-$O_2NC_6H_4$, 2-MeC_6H_4, 1-Nph, Mes) (91RCT133, 94OM3244) prepared by the reaction of a ligand with [Pd(I)(Me)(tmeda)] and silver triflate. Bubbling carbon monoxide results in insertion into the palladium−methyl or aryl bond to form acetyl or aroyl compounds (95OM5628).



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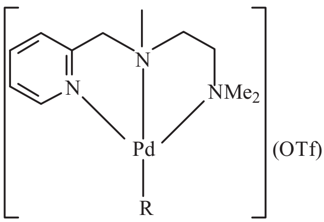
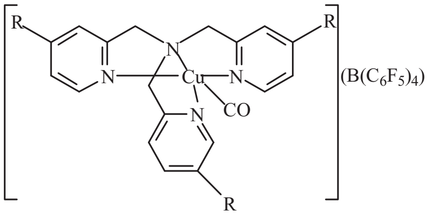


175

5.7 Copper group

Tris(2-pyridylmethyl)amine and similar ligands with $[Cu(AN)_4](B(C_6F_5)_4)$ in diethyl ether saturated with carbon monoxide give cationic **176** (R = H, Me_2N, Me_2O) and **177–179** (01IC4514, 03IC3016, 08IC241). (*t*-Butyldimethylsilyl)(2-pyridylmethyl)amine can be metalated with dimethyl zinc to give dimer **180** (01EJI851, 01ZAAC1141). Metalation of (2-pyridylmethyl)(tri-*i*-propylsilyl)amine using dimethyl zinc gives dimeric **181** (R = *i*-Pr_3Si) (02EJI389). Further addition of dimethyl zinc yields the C–C coupling product **180** (R = *i*-Pr_3Si). Metalation with di-*n*-butyl magnesium gives magnesium bis((2-pyridylmethyl)(tri-*i*-propylsilyl)amide and further methyl magnesium (2-pyridylmethyl)(tri-*i*-propylsilyl)amide. Similar reactions occur when R = *t*-$BuMe_2Si$. Compound **180** (R = *t*-$BuMe_2Si$) reacts with aniline to give **180** (R = Ph) (02OM906). 2-Pyridylmethylamine is metalated by dimethyl zinc to yield methylzinc-2-pyridylmethylamide, trimeric in the crystalline phase (02ZAAC1425). Thermolysis in toluene in the presence of dimethyl zinc then leads to bis-(methylzinc-2-pyridylmethylamido)-*N*- and *N*′-bis(methylzinc)-2,3,5,6 – tetrakis(2-pyridyl)-1,4-diaza-cyclohexane. Bis(2-pyridylmethyl)amide with ZnR_2 (R = Me, $CH(SiMe_3)_2$) gives dinuclear **182**, which can be partially hydrolyzed to **183** when R = $CH(SiMe_3)_2$ (08JOM1027).



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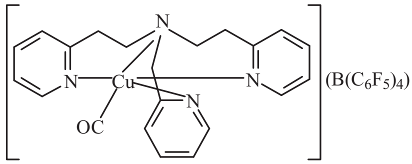

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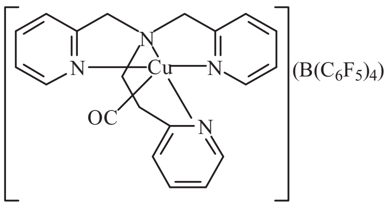

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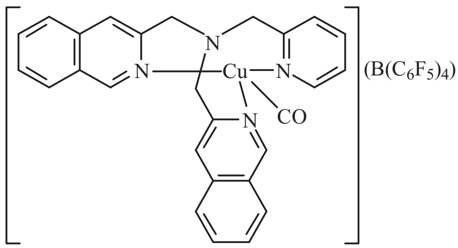

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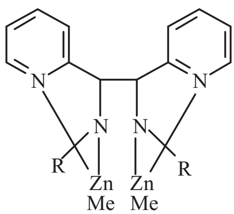

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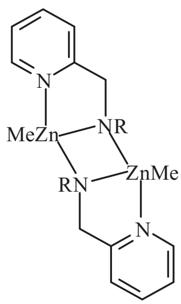

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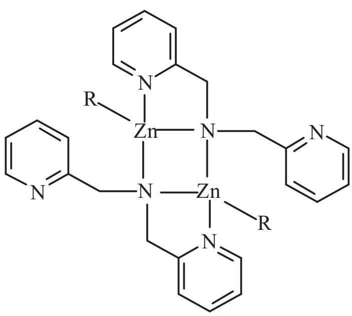

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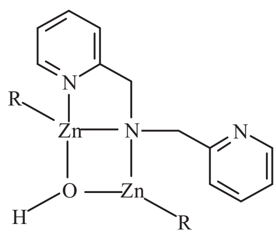
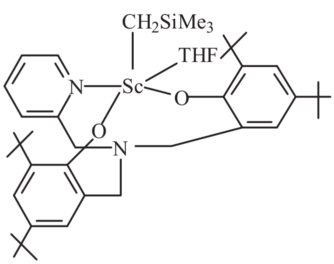


183

5.8 Rare earth metals

Bis(phenoxide) ligand with [Sc(CH_2SiMe_3)$_3$(THF)$_2$] in THF affords **184** (02JOM(647)145). The dilithium salt of (2-C_5H_4N)CH_2N{CH_2CH_2N(H) $SiMe_3$}$_2$ (00CC1167) with $ScCl_3$ gives five-coordinate **185** (R = Cl) (02JCS(D) 1694) that with $LiCH_2SiMe_3$ forms alkyl derivative **185** (R = CH_2SiMe_3). Lithiated diamidopyridines with [ScR_3(THF)$_2$] (R = CH_2SiMe_3, Ph) give series **186** (R = CH_2SiMe_3, R^1 = $SiMe_3$, *p*-Tol, Mes; R = Ph, R^1 = $SiMe_3$) (02JCS(D)4649). 2,6-Bis(((2,6-di-*i*-propylphenyl)amino)methyl)pyridine with [Ln(CH_2SiMe_3)$_3$ (THF)$_2$] (Ln = Lu, Y, Sc) gives series **187** (Ln = Sc, x = 1; Ln = Lu, x = 2, Ln = Y, x = 2) (03OM1212). 2,6-Bis((mesitylamino)methyl) pyridine enters the same type of reaction to yield products with the same type of structure but where x is always 1. Reaction of imino-amido-pyridine with [Ln(CH_2SiMe_3)$_3$(THF)$_2$] gives dialkyls **188** (Ln = Sc, Y, Lu) (08OM4310). The dilithium salt of 2,6-bis(2,6-di-*i*-propylanilidomethyl)pyridine (Li_2L) forms dichloride complexes with thorium(IV), [$ThCl_2$(L)(DME)], which subsequently give dialkyl [Th(CH_2SiMe_3)$_2$(L)] with $LiCH_2SiMe_3$ (07OM692).



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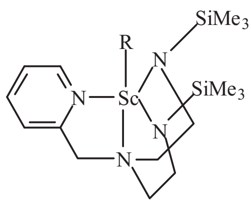

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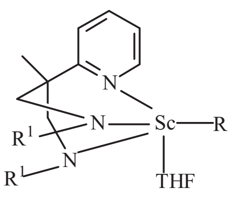

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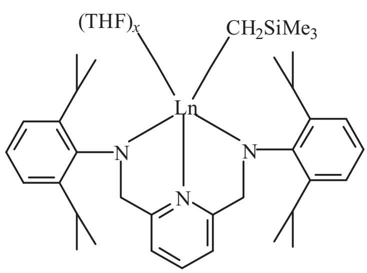

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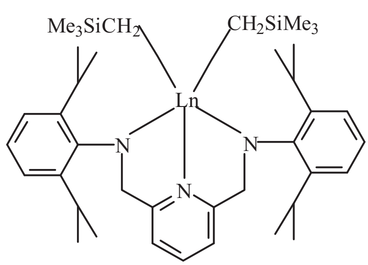
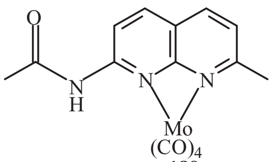


188

6. CARBOXAMIDE LIGANDS

2-Acetamido-7-methyl-1,8-naphthyridine with $[Mo(CO)_6]$ in diglyme forms the mononuclear complex **189** where the nitrogen atom of the acetamido-group is not coordinated (96CB683). Picolylaminediacetic acid (H_2L) with $(NEt_4)_2[ReBr_3(CO)_3]$ forms the tridentately coordinated $[Re(L)(CO)_3]$, but, in contrast, with $(NEt_4)[ReBr_3(CO)_2(NO)]$ tetradentately coordinated [Re(L)(CO)(NO)] **190** follow where one of the CO-ligands is displaced (05IC6082). 4-((Biotinamido)methyl)pyridine and 4-(*N*-((6-biotinamido)hexanoyl)aminomethyl)pyridine interact with $[Re(CO)_3(LL)(AN)](OTf)$ in THF/methanol to yield η^1(N)-coordinated complexes **191**, where LL is a derivative of 1,10-phenanthroline ($R^1 = R^2 = R^3 = H$; $R^1 = H$, $R^2 = R^3 = Me$; $R^1 = Me$, $R^2 = H$, $R^3 = Ph$), or dipyrido[3,2-*a*:2′,3′-*c*)-phenazine, or benzo[*i*]dipyrido[3,2-*a*:2′,3′-*c*]phenazine (02JA9344, 04OM3062). 2-Pyridinecarboxamides (R = H, Me, *i*-Pr, Ph, CH(Me)(Ph)) react with the dimer $[(\eta^4\text{-cod})Rh(Cl)]_2$ in alkali to yield **192** (96JOM(523)179). *N*-(1-Naphthyl)picolinamide with $[Ir(PPh_3)_3Cl]$ and triethylamine in ethanol give cyclometalated **193** (X = H, Cl) (08JOM3281).



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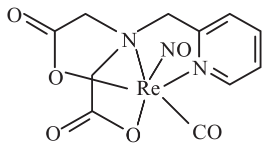

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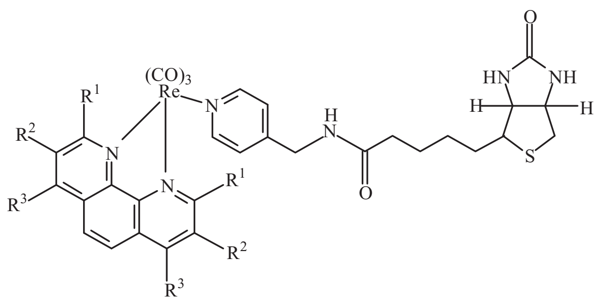

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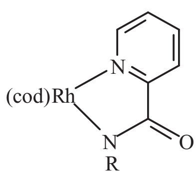

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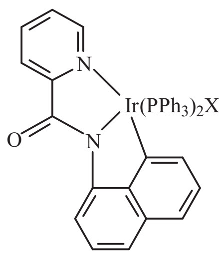

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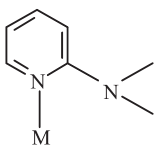
7. CONCLUSION

1. Mononuclear complexes of 2-aminopyridine, its derivatives and deprotonated 2-aminopyridinates may be η^1(N)-coordinated *via* a pyridine as in **194** or amino nitrogen atom in **195**, or η^2(*N,N*) chelated in the neutral **196** or, most typically, deprotonated **197** form. η^6-Coordination **198** *via* the heteroring is a unique mode observed only in a single case. Complex substituents sometimes participate in coordination. Thus, aryl moieties as in NHAr may be coordinated in an η^3-mode. Alkylamine substituents often give rise to aminocarbene structures located around the central atom. Cyclometalation, borylation, and silylation of substituents, cyclometalation of the heteroring, and protonation of an amino group are features of some representatives. Cluster chemistry gives rise to a variety of bridging modes, some of them – μ_2,η^1, **199**, μ_2,η^2, **200**, very common μ_2,η^3, **201**, μ_3,η^2, **202**, and μ_5,η^2, **203** in clusters of high nuclearity. Clusters in many cases contain cyclometalated heteroring.



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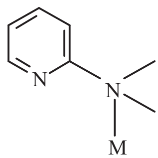

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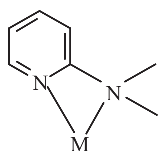

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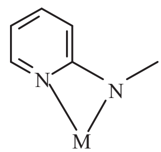

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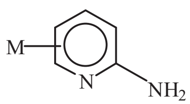

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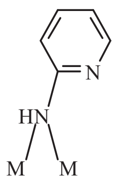

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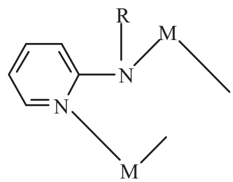

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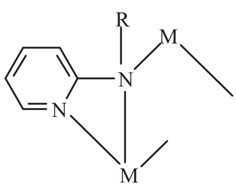

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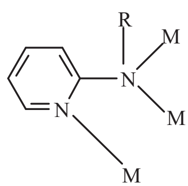

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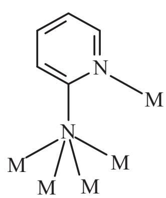

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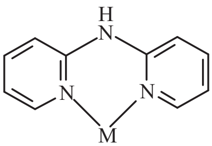
2. Di- and tri-2-pyridylamines reveal η^2(*N,N*) **204** and η^3(*N,N,N*) **205** modes in mononuclear complexes and μ_2,η^3, **206** and **207**, μ_2,η^2, **208** and **209**. Derivatives with nonadjacent amino groups are typically

coordinated *via* the pyridine nitrogen, although silylation and carbene formation by the amino groups sometimes occur.



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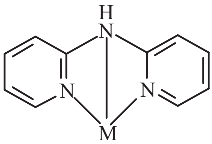

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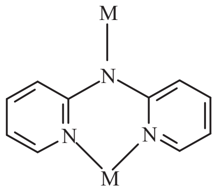

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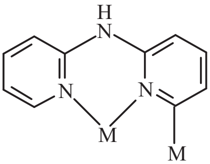

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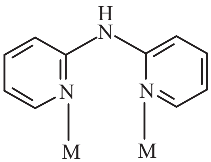

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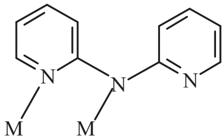

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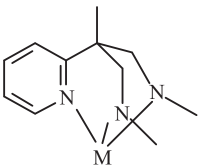
3. Aminomethylpyridine ligands typically coordinate *via* all the available heteroatoms forming bis- and poly-chelates, **210** and **213**. However, at times a pyridine moiety or an amino group is not coordinated.



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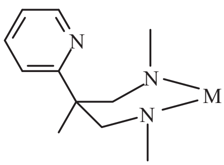

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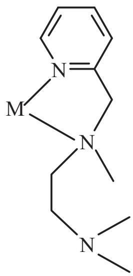

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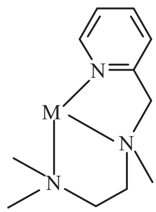

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8. LIST OF ABBREVIATIONS

Ac	acetyl
acac	acetylacetonate
AN	acetonitrile
Bu	butyl
cod	1,5-cyclooctadiene
COE	cyclooctene
Cp	cyclopentadienyl
Cp*	pentamethylcyclopentadienyl
Cy	cyclohexyl

DME	dimethoxyethane
dppm	bis(diphenylphosphino)methane
Et	ethyl
Fc	ferrocennyl
Me	methyl
Mes	mesityl
Np	neopentyl
Nph	naphthyl
nbd	2,5-norbornadiene
Np	neopentyl
OTf	triflate
Ph	phenyl
PPN	$[Ph_3PNPPh_3]^+$
Pr	propyl
py	pyridine
tfb	tetrafluorobenzobarrelene
THF	tetrahydrofuran
THT	tetrahydrothiophene
tmeda	*N*,*N*,*N*′,*N*′-tetramethylenediamine
Tol	tolyl
Tp	tris(pyrazol-1-yl)borate
Xyl	xylyl

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