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ORGANOMETALLIC WAYS FOR THE MULTIPLICATION OF CHIRALITY

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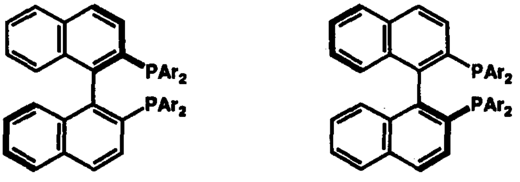
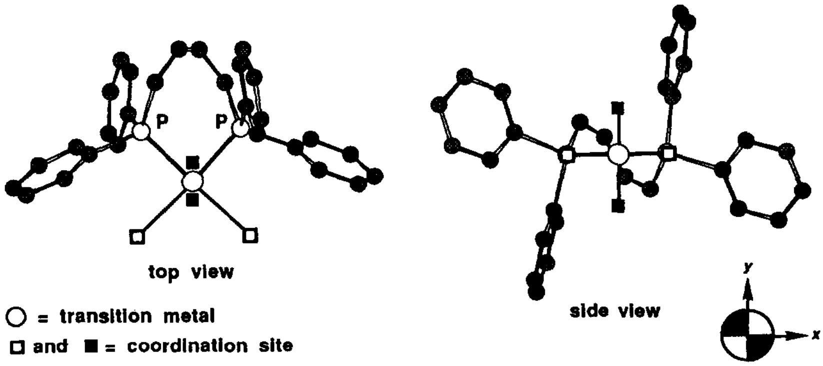
Chirality is a key element in science and technology. A wide range of biological and physical functions result from precise molecular recognition that involves strict matching of chirality. In living systems, enzymes, receptors, and other binding sites interact with enantiomers in decisively different manners. Certain physical properties related to advanced electronics and optics are also obtained by means of highly ordered assemblies of chiral molecules. Therefore truly efficient ways to provide enantiomerically pure compounds constitute genuine challenges for synthetic organic chemists. Stereoselective synthesis of optically active compounds using small quantities of chiral sources is particularly desirable. In this context, suitably designed chiral metal complexes can precisely discriminate between enantiotopic atoms, groups, or faces in achiral molecules and catalyze the production of a broad array of natural and unnatural substances of high enantiomeric purity.[1,2] Certain racemates are also resolved kinetically by reactions with chiral metal complexes. This organometallic strategy, that provides a general principle of asymmetric catalysis to multiply chirality, has tremendously extended the potential for stereoselective organic synthesis. I have intensively investigated this intriguing area since the mid-1960s when we were engaged in the Cu-catalyzed asymmetric carbene reaction,[3,4] and it is now my great pleasure to set forth selected highlights accomplished by my colleagues in these three decades. Some of them are not only useful for the laboratory preparation of optically active compounds but also for the innovative industrial synthesis of significant chiral substances. The chiral efficiency of the chemical means rivals, or sometimes exceeds, that of biological processes, converting the chemist's dream into reality.[5]

Proper combination of the central metals and chiral auxiliaries as well as selection of reaction conditions is crucially important for obtaining a high degree of stereoselectivity. Transition metal complexes containing



(*R*)-BINAP (*S*)-BINAP

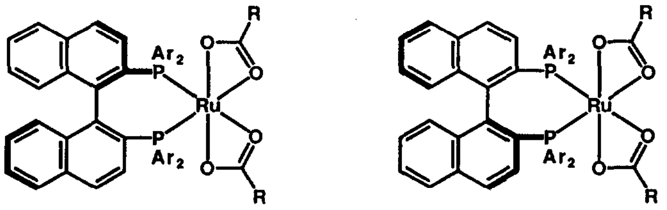
atropisomeric BINAP [2,2'-bis(diarylphosphino)-1,1'-binaphthyl] exhibit exceptionally high chiral recognition in various catalytic reactions.[6] BINAP is a fully arylated diphosphine ligand which exerts paramount steric influence, provides polarizability, and enhances the Lewis acidity of the metal complexes. Axially dissymmetric BINAP element has C_2 symmetry which, in many cases, halves the number of possible diastereomeric intermediates or transition states. The BINAP ligand is conformationally flexible and accommodates various transition metals without a serious increase of torsional strain. The resulting seven-membered BINAP metal chelate rings contain only sp^2-hybridized carbons and in turn are conformationally unambiguous and highly skewed. This feature provides distinct differentiation of the quadrant space sectors in the metal complexes. The *P*-phenyl rings play a key role in transmitting the chirality originally generated by the binaphthyl skeleton to the other metal coordination sites.[7,8] Bond-formation and -breakage occurring in such an extremely dissymmetric environment results in excellent chiral efficiency. By choosing the chirality of BINAP, either antipodal product can be synthesized equally. In addition, the fully aromatic compound possesses high chemical stability and crystallinity, providing another practical advantage.



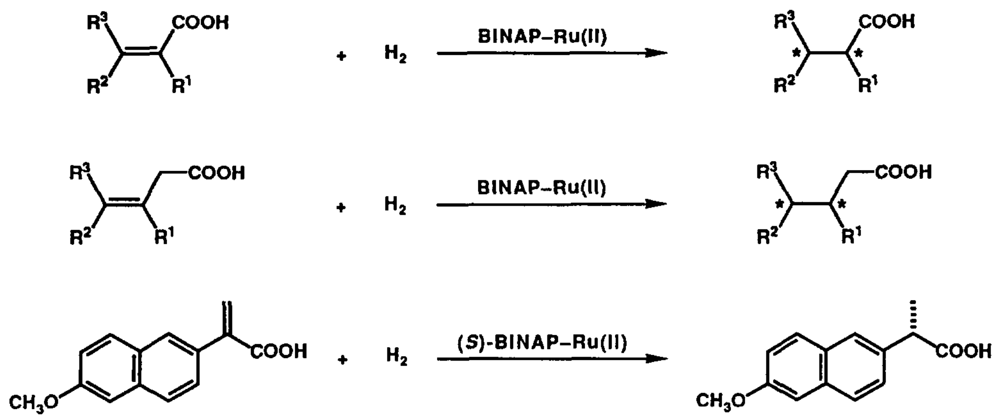
Schematic representation of λ-configurated (*R*)-BINAP transition metal complexes (Ar in BINAP = C_6H_5; naphthalene rings are omitted for clarity)

BINAP–Ru(II) complexes catalyze highly enantioselective hydrogenation of various organic unsaturated substrates. The scope is remarkably wide. The homogeneous hydrogenation can be operated simply on any scale ranging from <100 mg to >100 kg with high (up to 50%) substrate concentration and with a high substrate to catalyst ratio, and the resulting chiral products are, in many cases, easily isolated. Some successful examples follow.

First, BINAP–Ru(II) dicarboxylates[8,9] promote hydrogenation of various α,β- and β,γ-unsaturated carboxylic acids in alcoholic media to give the corresponding optically active saturated acids.[10] Use of some hydroxylated substrates leads to chiral alkylated γ- and δ-lactones. The reaction of tiglic acid with deuterated gas and methanol solvent indicates that the double bond saturation proceeds with cis stereochemistry. Gaseous hydrogen is incorporated in the α position and hydrogens of the protic molecules are introduced to the β position, suggesting the operation of a metal monohydride mechanism where the Ru center remains in the +2 oxidation state throughout the catalytic cycle.[11] An important application is the synthesis of the anti-inflammatory drug naproxen.

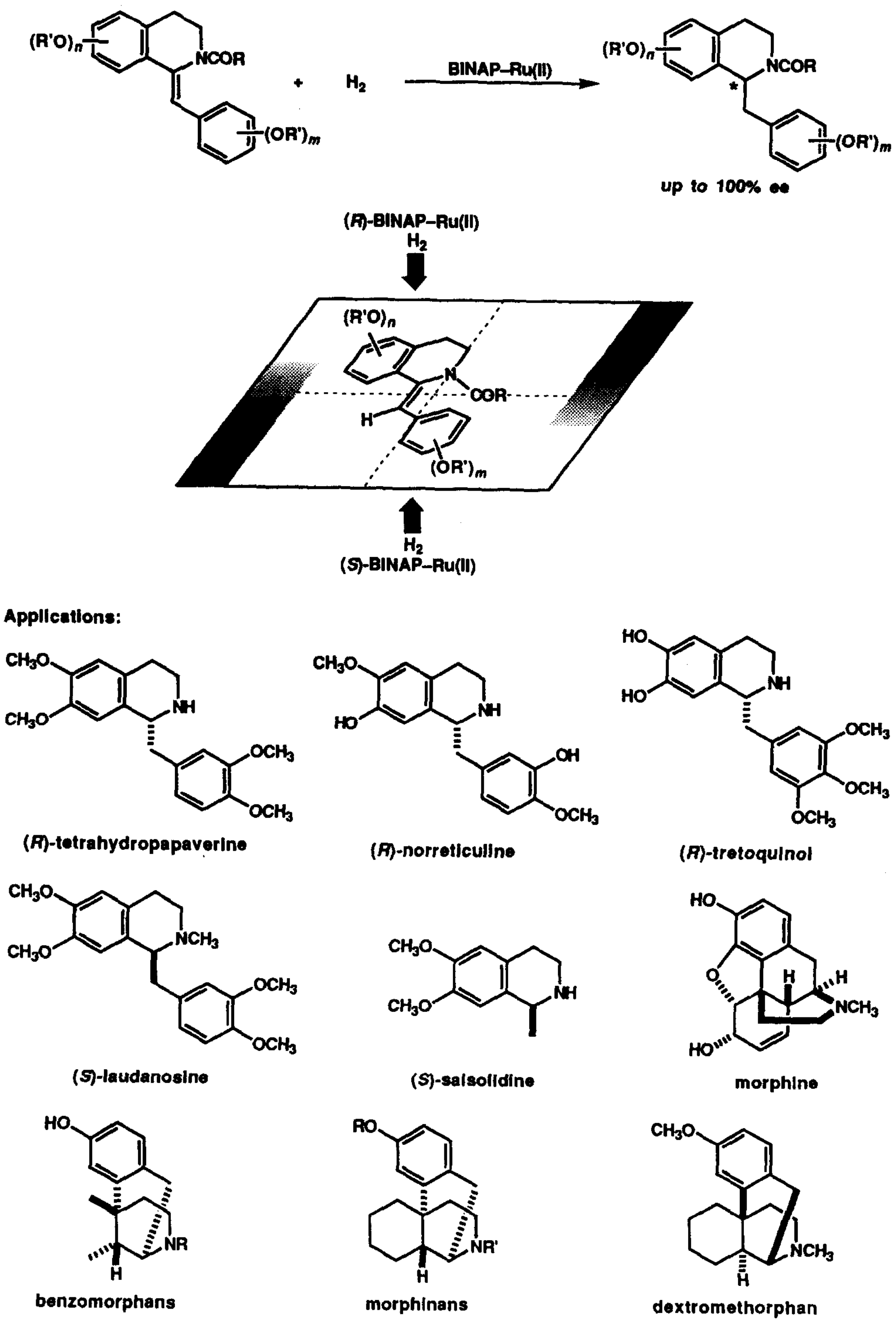


(*R*)-BINAP-Ru(II) complex (*S*)-BINAP-Ru(II) complex

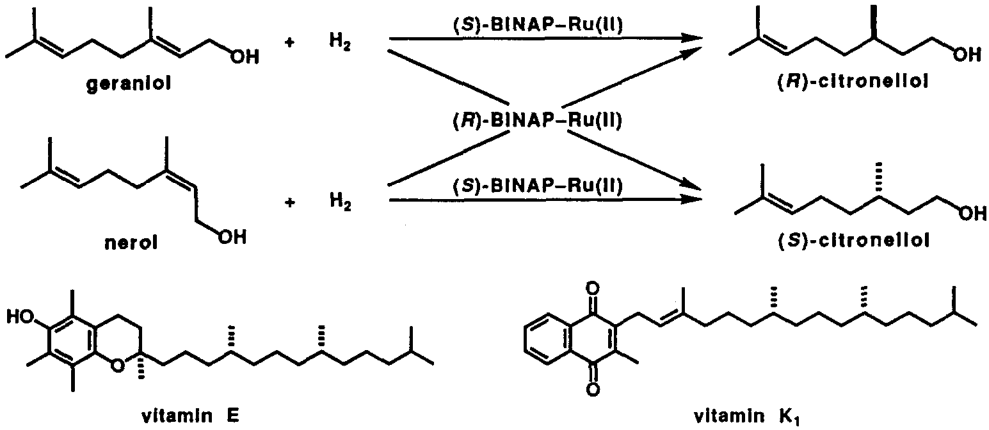


naproxen, 97% ee

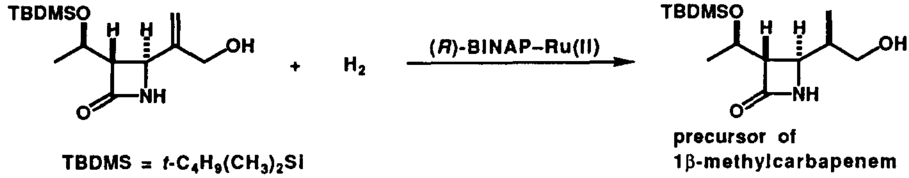
The BINAP–Ru catalysts effect the enantioselective hydrogenation of 2-acyl-(*Z*)-1-benzylidene-1,2,3,4-tetrahydroisoquinolines.[12] This discovery realized a general asymmetric synthesis of isoquinoline alkaloids including morphine, benzomorphans, and morphinans such as the antitussive dextromethorphan.[13] Optically active α- and β-amino acids are also accessible from appropriately amido-substituted olefins.[14]

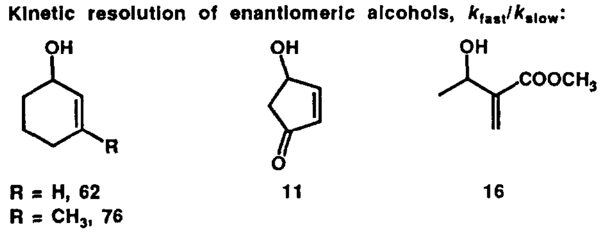

(R'O)n
NCOR
(OR')m
+ H2
BINAP–Ru(II)
(R'O)n
NCOR
*
(OR')m
up to 100% ee
(R)-BINAP–Ru(II)
H2
(R'O)n
N
COR
H
(OR')m
H2
(S)-BINAP–Ru(II)
Applications:
CH3O
CH3O
NH
OCH3
OCH3
(R)-tetrahydropapaverine
CH3O
HO
NH
OH
OCH3
(R)-norreticuline
HO
HO
NH
OCH3
OCH3
OCH3
(R)-tretoquinol
CH3O
CH3O
NCH3
OCH3
OCH3
(S)-laudanosine
CH3O
CH3O
NH
(S)-salsolidine
HO
O
H
H
NCH3
HO
morphine
HO
NR
H
benzomorphans
RO
NR'
H
morphinans
CH3O
NCH3
H
dextromethorphan

Geraniol and nerol are convertible to natural or unnatural citronellol with >96% enantiomeric purity without saturation of the C-6–C-7 olefinic bond.[15] The hydrogenation of (*R*,*E*)-6,7,10,11-tetrahydrofarnesol affords (3*R*,7*R*)-hexahydrofarnesol, a C_{15} side chain of vitamin E (tocopherol) and a part of vitamin K_1. The

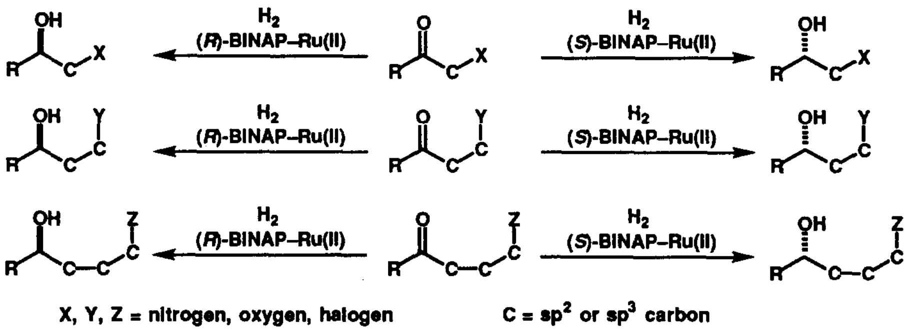


reaction of an allylic alcohol having a chiral azetidinone moiety leads diastereoselectively to a 1β-methylcarbapenem intermediate.[16] Furthermore, certain racemic allylic alcohols can be resolved by the BINAP–Ru catalyzed hydrogenation,[17] providing a practical way to (*R*)-4-hydroxy-2-cyclopentenone, a building block for the three-component prostaglandin synthesis.[18]

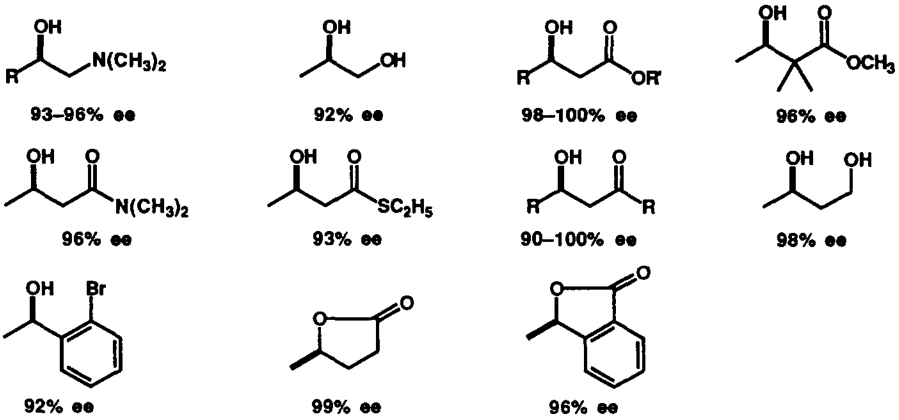




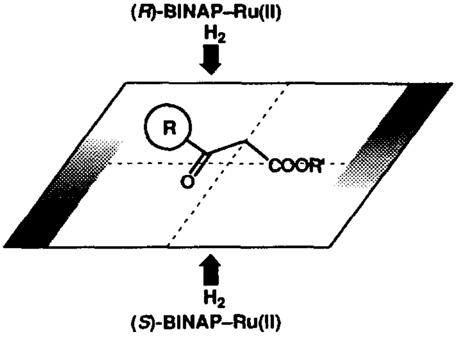
BINAP–Ru(II) complexes containing halide ligands catalyze hydrogenation of a diverse array of functionalized ketones to give the corresponding secondary alcohols with high enantiomeric purity. The sense of asymmetric induction is generally predictable.[19]

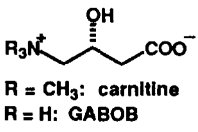


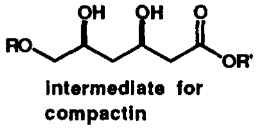
Examples of the (R)-BINAP–Ru(II) catalyzed hydrogenation:



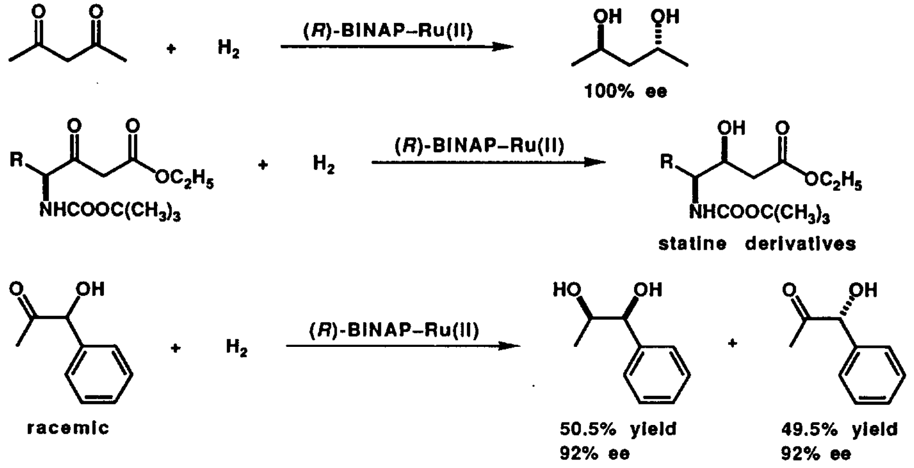
β-Keto esters are particularly good substrates for this stereoselective reaction.[20] A high-temperature, short-period hydrogenation of ethyl 4-chloro-3-oxobutanoate gives the chloro hydroxy ester in 97% ee, which is transformed to carnitine or GABOB.[21] In a like manner, a building block for the synthesis of compactin and analogues, HMG-CoA reductase inhibitors, is also available.



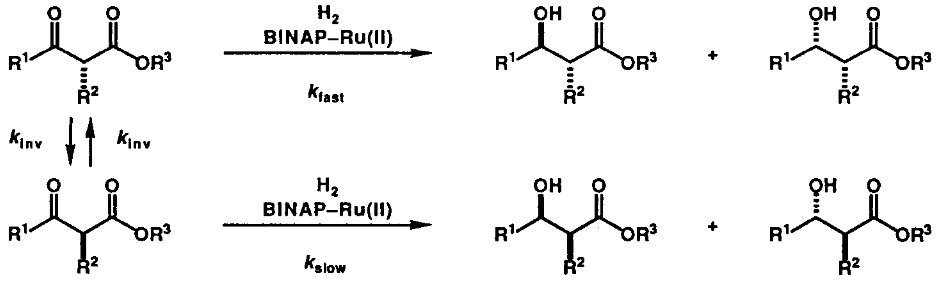


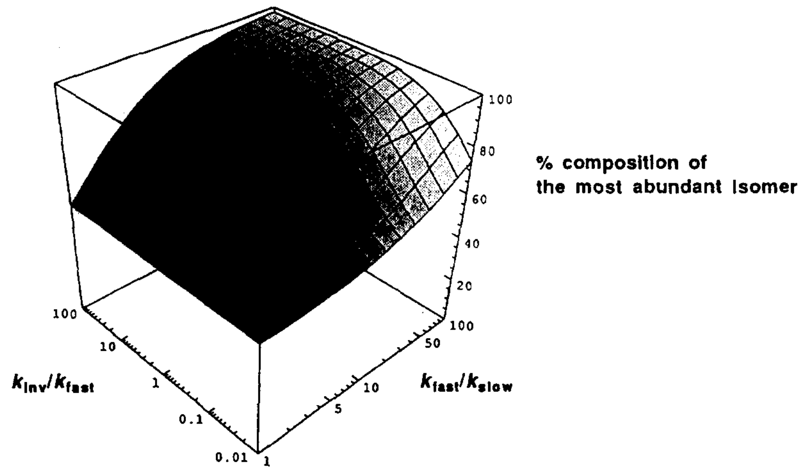


The steric course of the reaction of chiral ketonic substrates is markedly affected by the pre-existing stereogenic centers. The BINAP–Ru catalyzed double hydrogenation of 1,3-diones leads to the anti-1,3-diols of high enantiomeric purity.[20] Furthermore, the hydrogenation of BOC-protected γ-amino β-keto esters selectively affords threo-configurated statine, a component of the aspartic proteinase inhibitor pepstatin, or its analogues.[22] Some racemic α-hydroxy ketones can be resolved by the Ru-catalyzed hydrogenation.



Unlike ordinary kinetic resolution where the maximum yield of the desired enantiomer is 50%, the second-order stereoselective reaction utilizing in situ racemization of chirally labile substates allows for the synthesis of a single stereoisomer among several possible stereoisomers in 100% yield and 100% ee, in principle. Indeed the BINAP–Ru catalyzed hydrogenation of α-substituted β-keto esters leads stereoselectively to one hydroxy ester isomer among four possible stereoisomers.[23] The stereoselectivity is highly affected by the relative reactivities of the enantiomeric substrates, the relative ease with which stereoinversion and hydrogenation take place, the ability of the catalyst differentiating between hypothetical enantiofaces of the keto ester substrate (catalyst control, Ccat), and the diastereoselectivity of the reaction of the chiral substrate using a



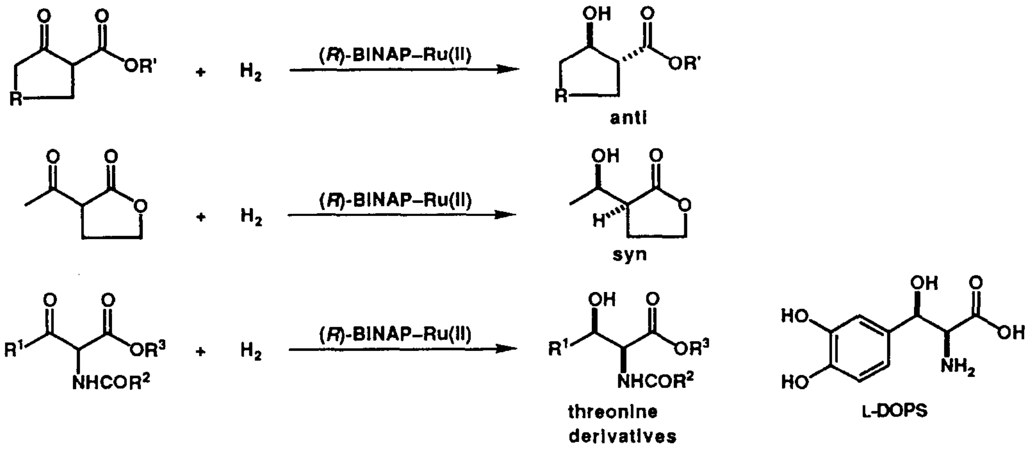


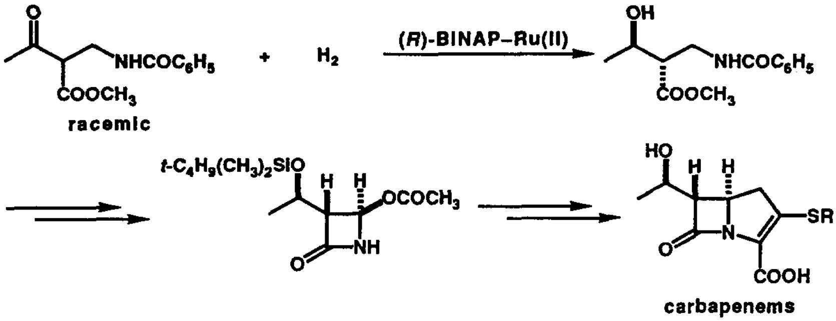
3D-graphic demonstration of k_{inv}/k_{fast}, k_{fast}/k_{slow}, and composition of the most abundant stereoisomer with Ccat = 10 and Csub = 10

hypothetical achiral Ru catalyst (substrate control, Csub). Therefore, the overall efficiency of the dynamic kinetic resolution actually depends on the substrate structures and the reaction conditions.[24]

The C-3 absolute configuration (hydroxy ester numbering) is determined by the chirality of the BINAP catalyst, while the C-2/C-3 relative configuration is controlled by the skeleton and substituent of the ketonic substrates. Racemic 2-alkoxycarbonyl-cycloalkanones are hydrogenated to the corresponding anti products with high diastereo- and enantioselectivity, whereas hydrogenation of 2-acetyl-4-butanolide exhibits a high degree of syn selection.

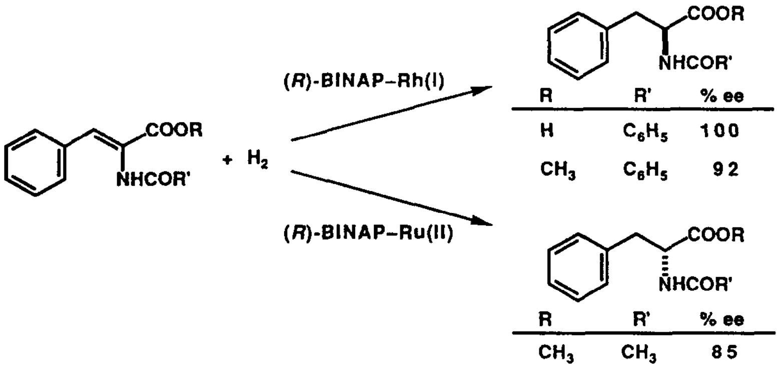
Open-chain β-keto esters containing an α-amido or -carbamate substituent are hydrogenated with excellent syn selectivity providing a convenient way to threonines, including the anti-Parkinsonian L-DOPS.



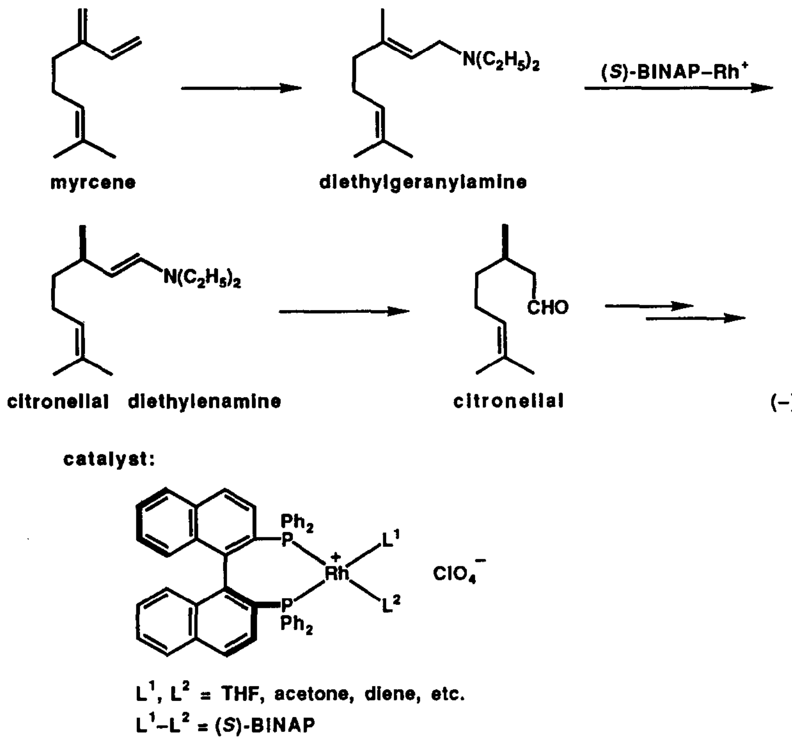


The most significant industrial application of this method is the stereoselective synthesis of a chiral azetidinone, a common synthetic intermediate of carbapenem antibiotics, by the hydrogenation of racemic methyl α-(benzamidomethyl)acetoacetate (120 tons/year).[23,25] The second-order stereoselective hydrogenation with the (*R*)-BINAP catalyst in dichloromethane affords the 2*S*,3*R* threo isomer with 94:6 diastereoselectivity and 99% ee. The computer-aided quantitative treatment indicates that the reaction occurs with a 15:1 enantiomer discrimination (k_S/k_R) and k_{inv}/k_R ratio greater than 90 and with excellent catalyst control (R^*:S^* = 104:1) and substate control (threo:erythro = 9:1).[24]

Cationic BINAP–Rh(I) complexes effect highly enantioselective hydrogenation of α-(acylamino)acrylic acids or esters, giving amino acid derivatives.[26] Interestingly, the Rh(I) and Ru(II) complexes, which have the same BINAP chirality, produce antipodal amino acids as the predominant products.[12,14]

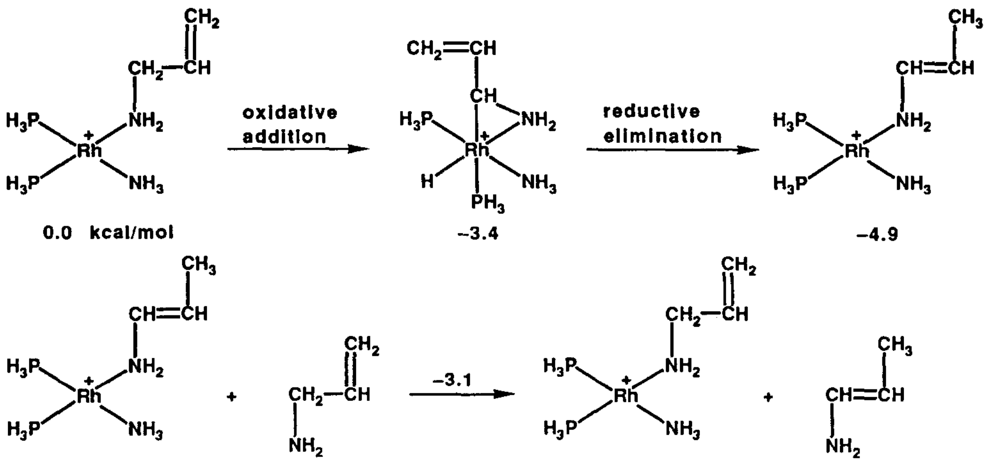


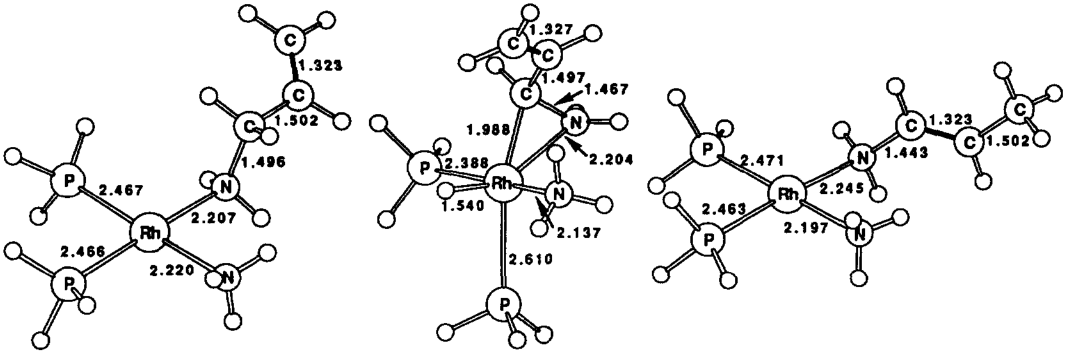
More importantly, the BINAP–Rh(I) complexes[26] catalyze the enantioselective isomerization of allylic amines to enamines.[27] Development of this work has enabled citronellal to be synthesized in 96–99% ee by the Rh catalyzed isomerization of diethylgeranylamine. The optical purity of the synthetic (*R*)-cintronellal is far superior to that of the natural product, ca. 80%. This transformation working on a 9-ton scale represents the



world's largest industrial application of asymmetric catalysis.[25,28] This asymmetric process allows production of (–)-menthol and other terpenic substances totaling ca. 1500 tons per year.

The isomerization of allylic amines has proved to proceed via a unique nitrogen-triggered mechanism.[29] The ab initio MO calculation of the model system indicates that the suprafacial 1,3-hydrogen shift occurs by



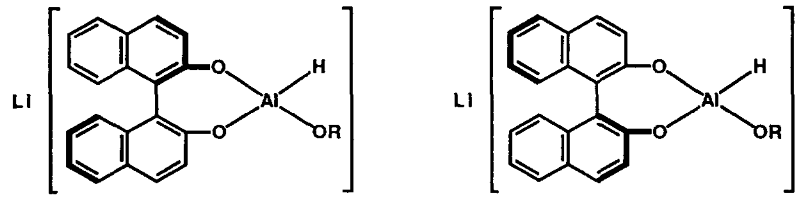


Rh(I)-allylamine complex   Rh(III) intermediate   Rh(I)-enamine complex

oxidative addition of the C-1–H bond to the Rh(I) center in the square planar complex followed by reductive elimination of an enamine from the Rh(III) intermediate accompanied by allylic transposition.[30] With the BINAP–Rh template, the enantiotopic C-1 hydrogens of geranylamine are discriminated clearly in the oxidative addition step.

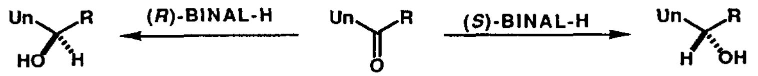
Thus the BINAP chemistry is particularly powerful in the field of pharmaceuticals, agrochemicals, flavors, and fragrances. A diverse array of terpenes, vitamins, antibiotics, amino acids, alkaloids, and other biologically significant compounds are accessible by homogeneous asymmetric catalysis.

Other atropisomeric 1,1'-binaphthyl derivatives are also useful in certain asymmetric synthesis. For example, binaphthol-modified lithium aluminum hydride reducing agent (BINAL-H) exhibits exceptionally high enantioselection in the stoichiometric reduction of a wide range of prochiral carbonyl compounds, where two substituents flanking the carbonyl group are differentiated mainly by differences in their electronic



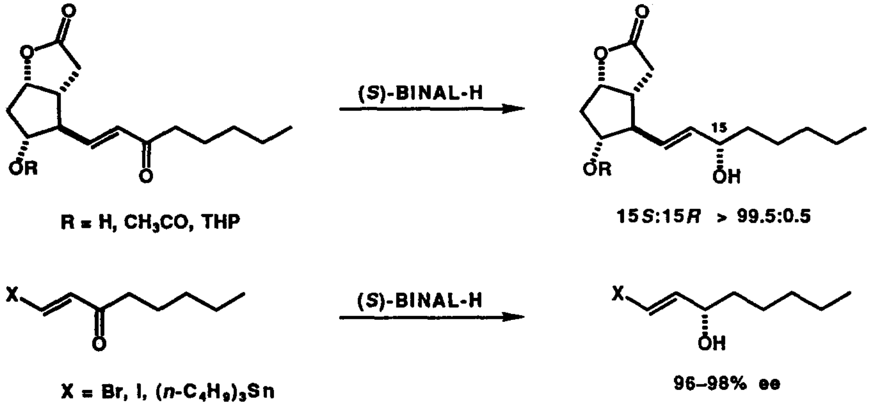
(*R*)-BINAL-H   (*S*)-BINAL-H

General sense:



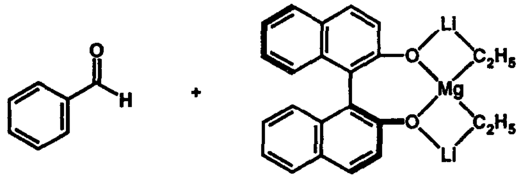
Un = aryl, alkenyl, alkynyl, etc.   R = alkyl, H

Applications:

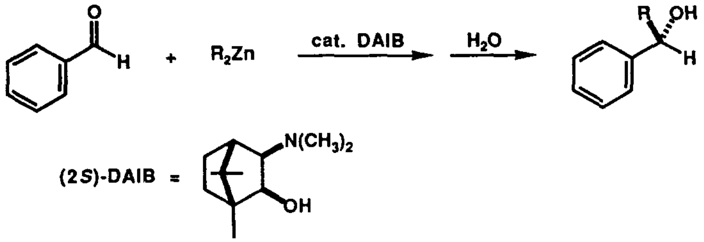


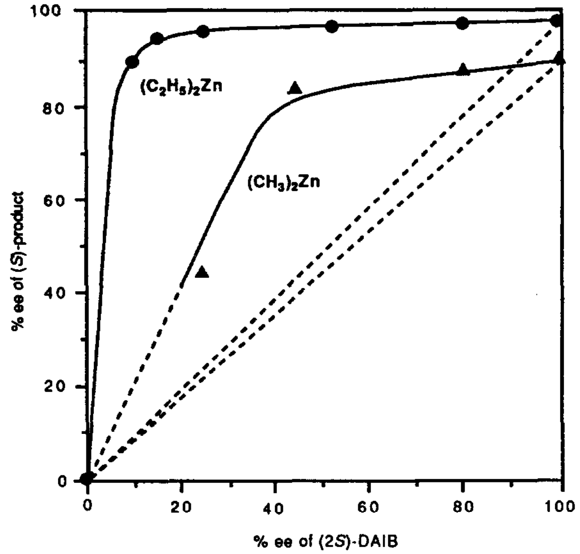
properties.[31] The reagent is remarkably effective in generating the 15*S* configuration of the prostaglandin series, facilitating greatly the current commercial synthesis[32] as well as the three-component coupling synthesis.[18]

In addition, chiral binaphthol acts as an efficient auxiliary in the enantioselective alkylation of aldehydes with magnesium/lithium binary organometallic reagents.[33]



Highly enantioselective addition of dialkylzincs to aldehydes is achievable in the presence of a catalytic amount of (2*S*)-3-*exo*-(dimethylamino)isoborneol (DAIB).[34,35] The alkyl transfer proceeds via a mechanism involving dinuclear Zn complexes.

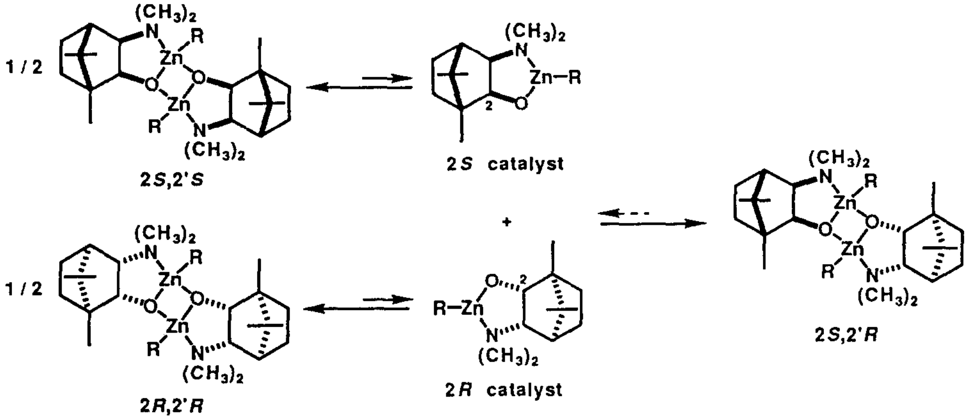




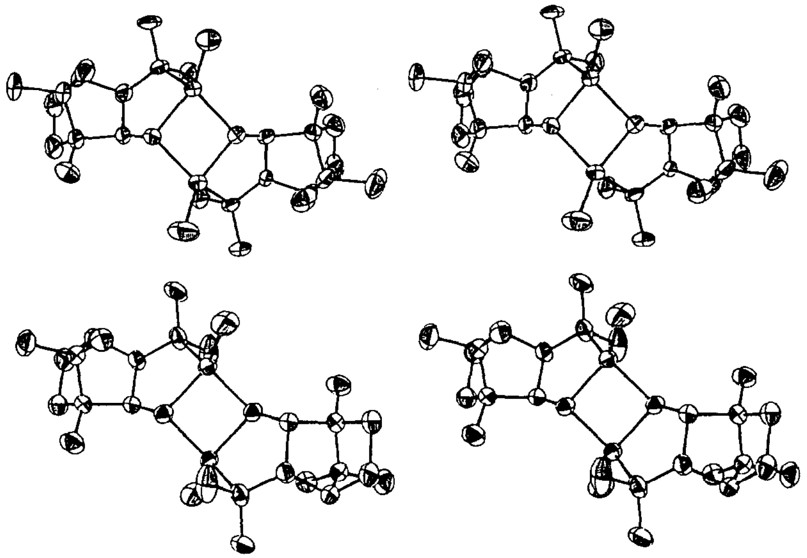
Correlation of the ee of the alkylation product and the ee of the chiral auxiliary

This reaction exhibits a unique, enormous nonlinear effect in terms of enantiomeric purity of the chiral source and products.[34,36] Typically, the reaction using DAIB in 15% ee (2*S*:2*R* = 57.5:42.5) forms the alkylation product in 95% ee, which is close to the 98–99% ee obtained with enantiomerically pure DAIB. The striking chiral amplification arises from the strict matching of chirality, viz., self and nonself recognition of the enantiomeric catalyst. The 2*S* and 2*R* alkylzinc alkoxide formed from dialkylzincs and DAIB are active catalytic species. The self or nonself recognition of these chiral monomers leads to diastereomeric dimeric complexes, where the heterochiral 2*S*,2'*R* complex is overwhelmingly more stable than the homochiral 2*S*,2'*S* or 2*R*,2'*R* dimer. When partially resolved (2*S*)-DAIB is used as a chiral auxiliary, all the minor 2*R* enantiomer is converted to the 2*S*,2'*R* dinuclear zinc complex by taking the same amount of the 2*S* monomer; the remaining 2*S* monomer forms its dimer. The latter tends to dissociate more readily into the true monomeric Zn catalyst, exhibiting a high turnover efficiency. Under some conditions, the chiral efficiency of the enantiomerically pure catalyst system is >600-times higher than that of the achiral counterpart.

The above described accomplishments were attained by the sustained experimental and intellectual efforts of my young, able collaborators at Nagoya, whose names are given in the reference literature. I have also enjoyed very fruitful collaborations with the research groups led by Professors H. Takaya (Institute of Molecular Science/Kyoto University), S. Otsuka and K. Tani (Osaka University), and N. Oguni (Yamaguchi University), and Dr. S. Akutagawa (Takasago Research Institute).



Enantiomer recognition of the chiral trigonal Zn compounds



Stereoview of the 2*S*,2'*S* (upper) and 2*S*,2'*R* dinuclear complex (lower) (R = CH_3)

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BIOGRAPHICAL SUMMARY

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Birth: Kobe, Japan; September 3, 1938.

Family Status: Married (to Hiroko Oshima), two sons (Eiji and Koji).

Education:
Bachelor: Kyoto University, 1961.
Master: Kyoto University, 1963.
Ph.D.: Kyoto University (Professor H. Nozaki), 1967.
Postdoctoral Fellow: Harvard University (Professor E. J. Corey), 1969–1970.

Appointments:
Research Associate, Department of Industrial Chemistry, Kyoto University, 1963–1968.
Associate Professor, Department of Chemistry, Nagoya University, 1968–1972.
Professor, Department of Chemistry, Nagoya University, 1972–present.
Director, Chemical Instrument Center, Nagoya University, 1979–1991.
Director, ERATO Molecular Catalysis Project of the Research Development Corporation of Japan, 1991–present.
Science Advisor, Ministry of Education, Science and Culture, 1992–present.
Professor, Institute for Fundamental Research of Organic Chemistry, Kyushu University, 1993–present.

Publications:
Over 300 papers.

Field of Research:
Organic chemistry including synthetic organic chemistry, main-group and transition metal organic chemistry, homogeneous catalysis, asymmetric synthesis, physical organic chemistry, etc. Synthesis of terpenes, alkaloids, antibiotics, prostaglandins, carbohydrates, nucleosides, nucleotides, and related unnatural compounds.

Memberships:
The Chemical Society of Japan.
The Pharmaceutical Society of Japan.
The Society of Organic Synthetic Chemistry, Japan.
The American Chemical Society.
The Royal Society of Chemistry.
American Association for the Advancement of Science.

Editorial Boards:
Organic Syntheses, Board of Editors, 1983–1988, Advisory Board, 1988–present.
Tetrahedron, Consulting Editor, 1987–present.
Tetrahedron Letters, Consulting Editor, 1987–present.
Comprehensive Organic Synthesis, Pergamon Press, Board of Editors, 1987–1991.
Tetrahedron Computer Methodology, Consulting Editor, 1989–present.
Tetrahedron: Asymmetry, Consulting Editor, 1990–present.
Chemical Reviews, Editorial Advisory Board, 1989–present.
Monatshefte für Chemie, Regional Editor, 1990–present.

Organic Synthesis in Japan: Past, Present, and Future, Society of Synthetic Organic Chemistry, Japan, Editor-in-Chief, 1990–1992.
Synthesis, Honorary Advisory Board, 1992–present.
Houben–Weyl 2000, Thieme, Board of Editors, 1992–present.
International Monograph Series in Organic Chemistry, Thieme, Board of Editors, 1992–present.
Nucleosides and Nucleotides, Editorial Advisory Board, 1992–present.
Accounts of Chemical Research, Editorial Advisory Board, 1994–.
Contemporary Organic Synthesis, The Royal Society of Chemistry, International Advisory Board, 1994–.

Awards and Honors:
The Chemical Society of Japan Award for Young Chemists for 1972.
The Matsunaga Prize, 1978.
Chunichi Cultural Prize, 1982.
The Chemical Society of Japan Award for 1985.
Award from Taipei Prostaglandin Conference and Academia Sinica, 1988.
The Naito Foundation Research Prize for 1988.
1988/89 Centenary Medal and Lectureship from the Royal Society of Chemistry.
The Fluka Prize, Reagent of the Year 1989, Switzerland.
The Toray Science & Technology Prize, 1990.
The Merck–Schuchardt Chair, 1990, Belgium.
The George Fisher Baker Lecturer, 1990, Cornell University.
J. G. Kirkwood Award, 1991, The American Chemical Society/Yale University.
The Asahi Prize for 1992.
Tetrahedron Prize for Creativity in Organic Chemistry, 1993, Pergamon Press, UK.

International Conferences and Lectureships; Invited from or as:
The Robert A. Welch Foundation Conferences on Chemical Research, XVII. Organic-Inorganic Reagents in Synthetic Chemistry, Houston, 1973.
Japan–US Joint Seminar on Prospects in Organotransition-metal Chemistry, Honolulu, 1974.
The Seventh International Conference on Organometallic Chemistry, Venice, 1975.
The New York Academy of Science, Conference on the Place of Transition Metals in Organic Synthesis, New York, 1976.
Second Joint Conference of the Chemical Institute of Canada and the American Chemical Society, Montreal, 1977.
The American Chemical Society, 1978 Inorganic Chemistry Symposium, Inorganic Compounds with Unusual Properties. II. Molecular Catalysis and the Conversion, Production and Storage of Energy, Athens, Georgia, 1978.
A Seminar/Workshop on Homogeneous Catalysis: Metal Ion Activation of Chemical and Biochemical Processes, Canberra, 1979.
Third IUPAC Symposium on Organic Synthesis, Madison, 1980.
Gordon Research Conference on the Chemistry of Heterocyclic Compounds, New Hampton, New Hampshire, 1980.
First IUPAC Symposium on Organometallic Chemistry Directed Toward Organic Synthesis, Fort Collins, Colorado, 1981.
Karl Pfister Visiting Professor at Massachusetts Institute of Technology, Cambridge, 1981.
Pacific Coast Lecturer, USA/Canada, 1982.
Second China–Japan–USA Joint Symposium on Organometallic and Inorganic Chemistry, Shanghai, 1982.
EUCHEM Conference on Methods in Organic Synthesis, Louvain-La-Nouve, Belgium, 1982.
Bürgenstock Conference, Bürgenstock, Switzerland, 1983.
Hoechst Workshop Conference on Selectivity: a Goal for Synthetic Efficiency, Reisensburg, FRG, 1983.
Victor J. Chambers Memorial Lecturer at University of Rochester, Rochester, 1984.
Second Japan–Korea Seminar on Organic Chemistry: Organic Reactions of Synthetic Utilities, Kyoto, 1984.
Fifth Asian Symposium on Medicinal Plants and Spices, Seoul, 1984.
The Nobel Symposium on Asymmetric Organic Syntheses, Karlskoga, Sweden, 1984.
Seventh International Symposium on Organosilicon Chemistry, Kyoto, 1984.
Fifth FECHEM Conference on Organometallic Chemistry, Cap d'Agde, France, 1984.
Visiting Professor at University of Montpellier, France, 1984.

Lecturer of the French Ministry of National Education, France, 1984.
Kyoto Conference on Prostaglandins, Kyoto, 1984.
Princeton–SmithKline & French Lecturer, USA, 1985.
Third International Conference on Chemistry and Biotechnology of Biologically Active Natural Products, Sofia, 1985.
Lecturer of the Hungarian Academy of Sciences, Budapest, 1985.
Lecturer of Shanghai Institute of Organic Chemistry, Academica Sinica, Shanghai, 1985.
Third Japan–Korea Seminar on Organic Chemistry, Dae Jeon, Korea, 1986.
The American Chemical Society, 1986 Ernest Guenther Award Symposium, New York, 1986.
Ischia Advanced School of Organic Chemistry, Ischia Island, Italy, 1986.
EUCHEM Conference on Applications of Transition Metal in Organic Synthesis, Toulon, 1986.
European Council, Intensive Course of Molecular Aspects of the Chemical Processes Involving Transition Metal Complexes, Toulon, 1986.
Syntex Distinguished Lecturer at Colorado State University, Fort Collins, Colorado, 1987.
Organic Syntheses Lecturer at Iowa State University, Ames, Iowa, 1987.
Lecturer of the Polish Academy of Sciences, Warsaw, 1987.
Tenth International Symposium on Synthesis in Organic Chemistry, Cambridge, 1987.
The Taniguchi Foundation, Sixth International Conference on Catalysis, Sanda, Japan, 1987.
The First Princess Chulabhorn Science Congress 1987. International Congress on Natural Products, Bangkok, 1987.
Royal Society Discussion Meeting, The Influence of Organometallic Chemistry on Organic Synthesis: Present and Future, London, 1988.
Western Switzerland 3e Cycle Lecturer, 1988.
Swiss Chemical Society, Symposium on Stereoselectivity in Organic Synthesis, Geneva, 1988.
Lecturer at Academia Sinica for the Celebration of the 60th Anniversary, Taipei, 1988.
Taipei Conference on Prostaglandin and Leukotriene Research, Taipei, 1988.
Tokushima Symposium on Natural Product Chemistry, Tokushima, Japan, 1988.
Gordon Research Conference on Organometallic Chemistry, Newport, Rhode Island, 1988.
Fourth Japan–Korea Seminar on Organic Chemistry, Tokyo, 1988.
Seventh IUPAC Conference on Organic Synthesis, Nancy, 1988.
Visiting Professor at University of Pierre and Marie Currie (Paris VI), Paris, 1988.
The Lemieux Lecturer at University of Ottawa, Ottawa, 1988.
Organic Syntheses Lecturer at Wayne State University, Detroit, 1988.
The Corey Symposium, Harvard University, Cambridge, 1988.
Centenary Lecturer, The Royal Society of Chemistry, UK, 1989.
Fifth International Seminar on Modern Synthetic Methods, Interlaken, 1989.
Merck–Schuchardt Lecturer, FRG, 1989.
Symposium on Progress and Prospects in Organic Synthesis, Lausanne/Champéry, 1989.
Fifth IUPAC Symposium on Organometallic Chemistry Directed Towards Organic Synthesis, Florence, 1989.
William S. Johnson Symposium in Organic Chemistry, Stanford University, Stanford, 1989.
1989 International Chemical Congress of Pacific Basin Society, Honolulu, 1989.
Fifth Japan–Korea Seminar on Organic Chemistry, Dae Jeon, Korea, 1990.
Symposium on Selective Transformations in Organic Chemistry, Royal Netherlands Chemical Society, Wageningen, Netherlands, 1990.
Arthur J. Birch Lecturer at the Australian National University, Canberra, 1990.
Third Symposium on Organic Synthesis via Organometallics, Marburg, 1990.
Third Belgian Organic Synthesis Symposium, Louvain-la-Neuve, 1990.
George Fisher Baker Lecturer at Cornell University, Ithaca, 1990.
Bio-Méga Lecturer at University of Montreal, Montreal, 1990.
Herbert C. Brown Lecturer at Purdue University, West Lafayette, Indiana, 1991.
150th Anniversary Congress, The Royal Society of Chemistry, London, 1991.
Second International IUPAC Symposium, Organic Chemistry: Technological Perspectives, Baden-Baden, 1991.
Morris S. Kharasch Visiting Professor at University of Chicago, Chicago, 1991.
Monsanto Symposium on Current Methods for Enantioselective Synthesis, St. Louis, 1991.
Japan–US Seminar on Selectivity in Synthetic and Bio-organic Chemistry, Tokyo, 1991.
Merck–Frosst Lecturer at University of Toronto, Toronto, 1991.
Visiting Professor at Texas A&M University, Frontiers in Chemical Research Series, College Station, Texas, 1991.
Catalytica Seminar on Advances in Catalytic Technologies, Santa Clara, 1991.

Kirkwood Awardee, The American Chemical Society/Yale University, New Haven, 1991.
Third Haaman & Reimer Symposium on Recent Developments in Flavor and Fragrance Chemistry, Kyoto, 1992.
Merck Centennial Lecturer at University of Minnesota, Minneapolis, 1992.
Rhone–Poulenc Rorer Lecturer at Ohio State University, Columbus, 1992.
Karl Folkers Lecturer at University of Wisconsin, Madison, 1992.
Syntex Distinguished Lecturer at University of Colorado, Boulder, 1992.
Organic Syntheses Lecturer at University of California, Irvine, 1992.
Henri Kagan Science Day at Université de Paris-Sud, Orsay, 1992.
Merck Centennial Lecturer at University of Illinois at Urbana-Champaign, 1992.
Kraft Distinguished Lecturer at Indiana University, Bloomington, 1992.
The JRDC International Symposium on Supermolecules and Molecular Systems, Fukuoka, 1992.
Third Eurasia Conference on Chemical Sciences, Bangkok, 1992.
JSPS–KOSEF Symposium on Asymmetric Synthesis, Seoul, 1993.
Western Switzerland 3e Cycle Lecturer, University of Lausanne, Lausanne, 1993.
Fifth International Conference on the Chemistry of the Platinum Group Metals, The Royal Society of Chemistry-Dalton Division, St. Andrews, 1993.
34th IUPAC Congress, Beijing, 1993.
15th Conference on Isoprenoids, Zacopane, Poland, 1993.
Third Max Tishler Memorial Lecture Meeting, Kitasato Institute, Tokyo, 1993.
The Majima Memorial Symposium on Organic Chemistry, Sendai, 1993.
19th IUPAC symposium on the Chemistry of Natural Products, Karachi, 1994.
Meeting on Reduction in Organic Synthesis, The Fine Chemicals Group of the Society of Chemical Industry, London, 1994.
Merck Lecturer, University of Cambridge, UK, 1994.
Max Tishler Prize Lecturer, Harvard University, Cambridge, 1994.

PUBLICATIONS OF RYOJI NOYORI

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(2) Polycondensation of Xylylene Dibromides by Transition Metals in Water Suspension. K. Sisido, N. Kusano, R. Noyori, Y. Nozaki, M. Simosaka, and H. Nozaki, *J. Polym. Sci. Part A*, **1**, 2101 (1963).

(3) 9,10-Bridged 9,10-Dihydroanthracenes. K. Sisido, R. Noyori, N. Kôzaki, and H. Nozaki, *Tetrahedron*, **19**, 1185 (1963).

(4) Acetolysis of *trans*-1,2-Dibromobenzocyclobutene. H. Nozaki, R. Noyori, and N. Kôzaki, *Tetrahedron*, **20**, 641 (1964).

(5) Reaction of Phenylcarbene Formed from Benzaldehyde Tosylhydrazone in Certain Solvents. H. Nozaki, R. Noyori, and K. Sisido, *Tetrahedron*, **20**, 1125 (1964).

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(7) The Reactions of Phenylcarbene with Polynuclear Aromatic Compounds. H. Nozaki, M. Yamabe, and R. Noyori, *Tetrahedron*, **21**, 1657 (1965).

(8) The Photo-Sensitized Isomerization of *cis,trans,trans*-1,5,9-Cyclododecatriene. H. Nozaki, Y. Nisikawa, Y. Kamatani, and R. Noyori, *Tetrahedron Lett.*, 2161 (1965).

(9) The Reaction of Ethyl Diazoacetate with Styrene Oxide. H. Nozaki, H. Takaya, and R. Noyori, *Tetrahedron Lett.*, 2563 (1965).

(10) Preparation of *cis*-Cyclododecene, Cyclododecyne, and Cyclododecanone. H. Nozaki and R. Noyori, *J. Org. Chem.*, **30**, 1652 (1965).

(11) The Simmons–Smith Reaction of *trans*, *trans*, *cis*-1,5,9-Cyclododecatriene. H. Nozaki, M. Kawanisi, and R. Noyori, *J. Org. Chem.*, **30**, 2216 (1965).

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(20) Reaction of Carbethoxycarbene with 2-Phenyloxirane and 2-Phenyloxetane. H. Nozaki, H. Takaya, and R. Noyori, *Tetrahedron*, **22**, 3393 (1966).

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(27) Reduction of *gem*-Dibromocyclopropanes with Chromium(II) Sulphate. H. Nozaki, T Aratani, and R. Noyori, *Tetrahedron*, **23**, 3645 (1967).

(28) Photochemistry of Certain Non-enolizable β-Diketones. H. Nozaki, Z. Yamaguti, T. Okada, R. Noyori, and M. Kawanisi, *Tetrahedron*, **23**, 3993 (1967).
(29) Photochemical Alkylation of Nitrogen Heteroaromatics by Carboxylic Acids under Decarboxylation. H. Nozaki, M. Katô, R. Noyori, and M. Kawanisi, *Tetrahedron Lett.*, 4259 (1967).
(30) Photochemical Behaviour of Enolic β-Diketones Towards Cycloolefins. H. Nozaki, M. Kurita, T. Mori, and R. Noyori, *Tetrahedron*, **24**, 1821 (1968).
(31) Photochemical Reactions of *trans*-Anethole. H. Nozaki, I. Otani, R. Noyori, and M. Kawanisi, *Tetrahedron*, **24**, 2183 (1968).
(32) The Photochemistry of 1,2,3-Triphenylaziridine. H. Nozaki, S. Fujita, and R. Noyori, *Tetrahedron*, **24**, 2193 (1968).
(33) Photo-Induced Polar Addition of Protic Solvents to 2-Cycloheptenone. H. Nozaki, M. Kurita, and R. Noyori, *Tetrahedron Lett.*, 2025 (1968).
(34) Asymmetric Ring Opening of *gem*-Dibromocyclopropanes Leading to Allenic Hydrocarbons. H. Nozaki, T. Aratani, and R. Noyori, *Tetrahedron Lett.*, 2087 (1968).
(35) Synthesis of Certain [8](2,5)Heterophanes. H. Nozaki, T. Koyama, T. Mori, and R. Noyori, *Tetrahedron Lett.*, 2181 (1968).
(36) Synthetic Studies on Cyclic Compounds by Means of Photochemically Excited Species. H. Nozaki, T. Okada, T. Mori, R. Noyori, and M. Kawanisi, *Nippon Kagaku Zasshi*, **89**, 215 (1968).
(37) Homogeneous Catalysis in the Decomposition of Diazo Compounds by Copper Chelates. Asymmetric Carbenoid Reactions. H. Nozaki, H. Takaya, S. Moriuti, and R. Noyori, *Tetrahedron*, **24**, 3655 (1968).
(38) Photochemical Reactions of 2,6-Cycloheptadienone. H. Nozaki, M. Kurita, and R. Noyori, *Tetrahedron Lett.*, 3635 (1968).
(39) Photochemical Reaction of 2,7-Cyclooctadienone in Protic Solvents. R. Noyori and M. Katô, *Tetrahedron Lett.*, 5075 (1968).
(40) The Reactive Intermediate in the Photoinduced Alcohol Addition of *cis*-2-Cyclooctenone. R. Noyori, A. Watanabe, and M. Katô, *Tetrahedron Lett.*, 5443 (1968).
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(43) Partial Asymmetric Synthesis of Methylenecyclopropanes and Spiropentanes. R. Noyori, H. Takaya, Y. Nakanisi, and H. Nozaki, *Can. J. Chem.*, **47**, 1242 (1969).
(44) Photochemical Reaction of Benzopyridines with Alkanoic Acids. Novel Reductive Alkylation of Acridine, Quinoline, and Isoquinoline under Decarboxylation. R. Noyori, M. Katô, M. Kawanisi, and H. Nozaki, *Tetrahedron*, **25**, 1125 (1969).
(45) Retro-Diels–Alder Reaction Induced by π,π^* Excitation and by Electron Impact. H. Nozaki, H. Katô, and R. Noyori, *Tetrahedron*, **25**, 1661 (1969).
(46) Reaction of Methylenecyclopropanes with Palladium Chloride. R. Noyori and H. Takaya, *Chem. Commun.*, 525 (1969).
(47) A Total Synthesis of Prostaglandin $F_{2\alpha}$ (*dl*) from 2-Oxabicyclo[3.3.0]oct-6-en-3-one. E. J. Corey and R. Noyori, *Tetrahedron Lett.*, 311 (1970).
(48) Total Synthesis of Prostaglandins $F_{1\alpha}$, E_1, $F_{2\alpha}$, and E_2 (Natural Forms) from a Common Synthetic Intermediate. E. J. Corey, R. Noyori, and T. K. Schaaf, *J. Am. Chem. Soc.*, **92**, 2586 (1970).
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(51) Photolysis of 1-Acetylcyclooctene. Direct Observation of Dienol Intermediate in Photochemical Deconjugation of α,β-Unsaturated Ketone. R. Noyori, H. Inoue, and M. Katô, *J. Am. Chem. Soc.*, **92**, 6699 (1970).
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(53) Reaction of α,α'-Dibromo Ketones with Iron Carbonyls in the Presence of 1,3-Dienes. A New Route to Troponoid Compounds. R. Noyori, S. Makino, and H. Takaya, *J. Am. Chem. Soc.*, **93**, 1272 (1971).

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(55) Photo-Induced Cyclization of *cis,cis*-2,8-Cyclononadienone *via* the *cis,trans* Isomer. R. Noyori, Y. Ohnishi, and M. Katô, *Tetrahedron Lett.*, 1515 (1971).
(56) A New Synthesis of α,β-Unsaturated Aldehydes Using 1,3-Bis(methylthio)allyllithium. E. J. Corey, B. W. Erickson, and R. Noyori, *J. Am. Chem. Soc.*, **93**, 1724 (1971).
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