

Recent advances in asymmetric synthesis with chiral imide auxiliaries

David A. Evans and Jared T. Shaw

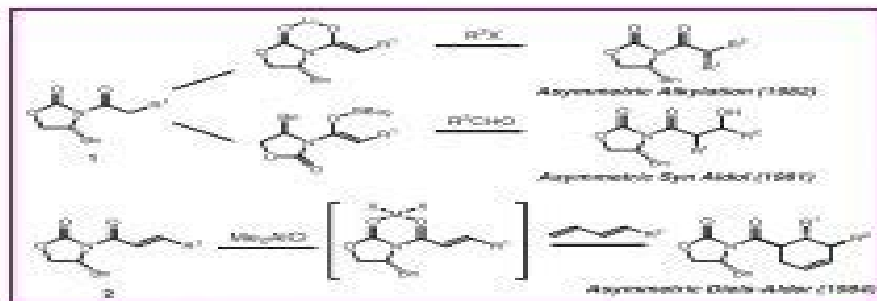
Résumé

Progrès en synthèse asymétrique : utilisation d'imides comme auxiliaires chiraux

Les oxazolidinones chirales et leur dérivés sont des auxiliaires efficaces en synthèse asymétrique. De nouvelles méthodologies ont été développées, notamment des alkylations de composés glycoliques et des réactions d'aldol utilisant une quantité catalytique de métal, produisant les diastéréoisomères anti. Quelques exemples récents et innovateurs d'utilisations de ce type d'auxiliaires en synthèse totale sont aussi abordés.

Mots-clés Key-words

**Auxiliaires chiraux, oxazolidinones, thiazolinethiones, synthèse totale, diastéréosélectivité.
Chiral auxiliaries, oxazolidinones, thiazolinethiones, total synthesis, diastereoselectivity.**



Scheme 1 - Initial reports of asymmetric induction from chiral imides.

The use of chiral *N*-acyloxazolidinone auxiliaries to control absolute stereoinduction has found wide application in a variety of reactions over the last two decades [1]. Despite extensive development by our group and complementary studies by others, new reactions continue to evolve from this useful family of chiral controllers. The most widely employed reactions of these ester surrogates are the asymmetric alkylation, aldol and Diels-Alder reactions (scheme 1). This review covers significant contributions to these areas of development during the last five-year period.

The majority of our work with chiral imides has employed the basic structural elements of oxazolidinone **1** with few exceptions over the last 20 years. Subsequent to our initial reports, many structural variants of **1** have appeared (figure 1). These modified imides have been developed to exhibit different cleavage reactivity or complimentary diastereoselectivity compared

to **1**. The most notable imide analog is the thiazolidine thione developed by Nagao and Fujita, then later used by Crimmins and our group [2]. This auxiliary, unlike many others that have been developed, reacts through different modes of metal binding and thus affords a useful complement to imide **1** (vide infra).

Stereoselective glycolate alkylation

The alkylation of lithium, sodium, and potassium enolates derived from *N*-acyloxazolidinones have proven useful for the preparation of many chiral building blocks. The reaction is generally limited to reactive alkylating agents such as allyl and benzyl halides. The Crimmins group has recently extended the alkylations reaction for use with glycolic acid equivalents such as **3** in the preparation of chiral substrates for a ring-closing metathesis approach to macrocyclic ethers from *Laurencia* Sp (scheme 2) [3].

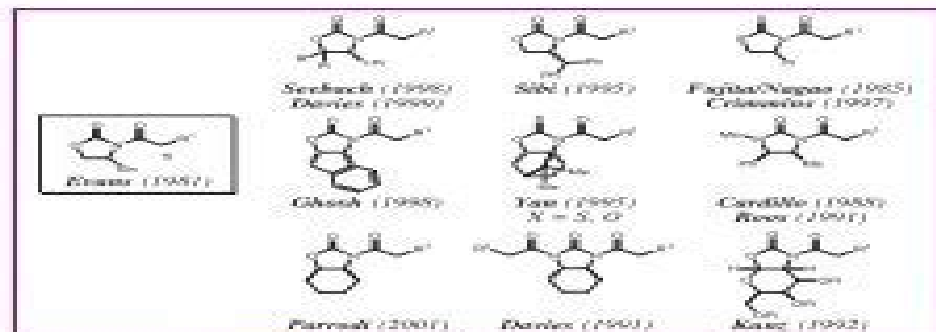


Figure 1 - Selected structural variants of chiral imide **1**.

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and illustrations of 560 schemes and figures **Advances in Asymmetric Synthesis, Vol. 2**, 1997 **Advances in Asymmetric Autocatalysis and Related Topics** Gyula Palyi, Robert Kurdi, Claudia Zucchi, 2017-05-18 **Advances in Asymmetric Autocatalysis and Related Topics** provides various viewpoints on the important developments in asymmetric autocatalysis that have occurred in the past few years also including brand new information in the field Asymmetric autocatalysis is a chemical reaction which leads from achiral starting materials to chiral products and in which the product accelerates its own formation reaction conventional catalysis and promotes the prevalence of its own chiral configuration asymmetric induction The combination of these two effects in the same reaction was unprecedented before 1995 when it was first described by Kenso SOAI at the Tokyo University of Science Since then several new combinations of this effect have been found most intriguingly the possibility of absolute asymmetric synthesis which is the spontaneous formation of the excess of one of the enantiomers of the product a dream of organic chemists for more than a century The book contains expert contributed chapters that describe the most exciting recent developments in the field of the Soai reaction and in related topics ranging from mechanistic studies and theoretical research to very practical problems in chiral syntheses and products Features contributions from global experts including several chapters from Kenso Soai and expert colleagues Focuses on recent developments in the field of asymmetric autocatalysis and newly reported findings Explores the Soai reaction new developments and the light it sheds on homochirality in certain biomolecules **New Frontiers in Asymmetric Catalysis** Koichi Mikami, Mark Lautens, 2007-05-04 A compilation of recent advances and applications in asymmetric catalysis The field of asymmetric catalysis has grown rapidly and plays a key role in drug discovery and pharmaceuticals **New Frontiers in Asymmetric Catalysis** gives readers a fundamental understanding of the concepts and applications of asymmetric catalysis reactions and discusses the latest developments and findings With contributions from preeminent scientists in their respective fields it covers Rational ligand design which is critically dependent on the reaction type reduction oxidation and C C bond formation Recent findings on activation of C H bonds C C bonds and small molecules C O HCN RN C and CO₂ and the latest developments on C C bond reorganization such as metathesis **Advances in chirally economical non linear phenomena racemic catalysis and autocatalysis** Some of the recent discoveries that have led to a renaissance in the field of organocatalysis including the development of chiral Br nstead acids and Lewis acidic metals bearing the conjugate base of the Br nstead acids as the ligands and the chiral bi functional acid base catalysts The book ends with a thought provoking perspective on the future of asymmetric catalysis that addresses both the challenges and the unlimited potential in this burgeoning field This is an authoritative up to date reference for organic chemists in academia government and industries including pharmaceuticals biotech fine chemicals polymers and agriculture It is also an excellent textbook for graduate students studying advanced organic chemistry or chemical synthesis **Advances in Asymmetric Synthesis Using Organoselenium Chemistry** Pete Silks, 2006 **Catalytic Asymmetric Synthesis** Iwao Ojima, 2004-08-13 From the

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Asymmetric Synthesis II Mathias Christmann, Stefan Bräse, 2013-03-19 After the overwhelming success of
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the current developments in the field

Yearbook of Asymmetric Synthesis 1991 K. Zaman, Atta Ur Rahman, M.S.
Shekhani, 2012-12-06 Recent advancements in the field of asymmetric synthesis have been triggered by the challenges this
field has offered to synthetic organic chemists and the importance of preparing optically active compounds of medical value
Newly developed asymmetric organic reactions combined with improvements and novel applications of previously known
reactions have created the need for this current volume Presenting findings reported in 1991 this book covers asymmetric
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and transesterification and miscellaneous asymmetric reactions This book will serve as a useful reference for all researchers

scientists and students working in the field of synthetic organic chemistry

Asymmetric Organocatalysis Lukasz Albrecht, Anna Albrecht, Luca Dell'Amico, 2022-11-10 Asymmetric Organocatalysis Comprehensive resource on the latest and most important developments in the highly vivid field of asymmetric organocatalysis The book provides a comprehensive overview of the most important advancements in the field of asymmetric organocatalysis that have occurred within the last decade It presents valuable examples of newly developed synthetic methodologies based on various organocatalytic activation modes Special emphasis is given to strategies where organocatalysis is expanding its potential by pushing the boundaries and founding new synergistic interactions with other fields of synthetic chemistry such as metal catalysis photocatalysis and biocatalysis The application of different concepts such as vinylogy dearomatization or cascade reactivity resulting in the development of new functionalization strategies is also discussed Sample topics covered within the book include New developments in enantioselective Br nsted acid catalysis with strong hydrogen bond donors Asymmetric phase transfer catalysis from classical applications to new concepts Halogen bonding organocatalysis Asymmetric electrochemical organocatalysis and synergistic organo organocatalysis Immobilized organocatalysts for enantioselective continuous flow processes Mechanochemistry and high pressure techniques in asymmetric organocatalysis Useful tools in elucidation of organocatalytic reaction mechanisms With an overall focus on new reactions and catalysts this two volume work is an indispensable source for everyone working in the field of asymmetric organocatalysis

Asymmetric Synthesis with Chemical and Biological Methods Dieter Enders, Karl-Erich Jaeger, 2007-04-09 Edited by two of the leading researchers in the field this book provides a deep interdisciplinary insight into stoichiometric and catalytic reactions in this continuously expanding area A plethora of top German scientists with an international reputation covers various aspects from classical organic chemistry to process development and from the theoretical background to biological methods using enzymes Throughout the focus is on the development of new synthetic methods in asymmetric synthesis the synthesis of natural and bioactive compounds and the latest developments in both chemical and biological methods of catalysis as well as the investigation of special technical and biotechnical aspects

Asymmetric Synthesis of Natural Products Ari M. P. Koskinen, 2012-06-15 Asymmetric Synthesis of Natural Products 2nd Edition introduces students to this rapidly growing field of organic chemistry The initial chapters present the foundations of asymmetric synthesis including the theory and applications of individual asymmetric reactions This is followed by chapters on each of the major individual classes of natural products their structures biosynthesis and interrelationships as well as examples of asymmetric syntheses and the practical value of these compounds Natural product classes covered include carbohydrates amino acids peptides proteins nucleosides nucleotides nucleic acids polyketides isoprenoids shikamic acid derivatives and alkaloids For this second edition the text has been thoroughly updated and expanded and includes new discussions and examples covering atom and redox economies practical aspects and environmental awareness Organocatalysis has emerged completely in the last ten years and has been

fully integrated into this new edition **Advances in Lewis Acid Directed Asymmetric Synthesis** Christopher R. Sarko, 1995 **Principles of Asymmetric Synthesis** R.E. Gawley, J. Aubé, 1996-11-21

The world is chiral. Most of the molecules in it are chiral and asymmetric synthesis is an important means by which enantiopure chiral molecules may be obtained for study and sale. Using examples from the literature of asymmetric synthesis, more than 1300 references, the aim of this book is to present a detailed analysis of the factors that govern stereoselectivity in organic reactions. It is important to note that the references were each individually checked by the authors to verify relevance to the topics under discussion. The study of stereoselectivity has evolved from issues of diastereoselectivity through auxiliary based methods for the synthesis of enantiomerically pure compounds, diastereoselectivity followed by separation and auxiliary cleavage to asymmetric catalysis. In the latter instance, enantiomers, not diastereomers, are the products and highly selective reactions and modern purification techniques allow preparation in a single step of chiral substances in 99% ee for many reaction types. After an explanation of the basic physical organic principles of stereoselectivity, the authors provide a detailed annotated glossary of stereochemical terms. A chapter on Analytical Methods provides a critical overview of the most common methods for analysis of stereoisomers. The authors then follow the tried and true format of grouping the material by reaction type. Thus, there are four chapters on carbon-carbon bond forming reactions: enolate alkylations, organometal additions to carbonyls, aldol and Michael reactions, and cycloadditions and rearrangements; one chapter on reductions and hydroborations; carbon-hydrogen bond forming reactions; and one on oxidations; carbon-oxygen and carbon-nitrogen bond forming reactions. Leading references are provided to natural product synthesis that have been accomplished using a given reaction as a key step. In addition to tables of examples that show high selectivity, a transition state analysis is presented to explain to the current level of understanding the stereoselectivity of each reaction. In one case, Cram's rule, the evolution of the current theory is detailed from its first tentative 1952 postulate to the current Felkin-Anh-Heathcock formalism. For other reactions, only the currently accepted rationale is presented. Examination of these rationales also exposes the weaknesses of current theories in that they cannot always explain the experimental observations. These shortcomings provide a challenge for future mechanistic investigations.

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