Origin of Catalysis and Selectivity in Lewis Acid-Promoted Diels— Alder Reactions Involving Vinylazaarenes as Dienophiles

Susana Portela and Israel Fernández*



ABSTRACT: The poorly understood factors controlling the catalysis and selectivity in Lewis acid-promoted Diels—Alder cycloaddition reactions involving vinylazaarenes as dienophiles have been quantitatively explored in detail by means of computational methods. With the help of the activation strain model and the energy decomposition analysis methods, it is found that the remarkable acceleration induced by the catalysis is mainly due to a significant reduction of the Pauli repulsion between the key occupied #-molecular orbitals of the reactants and not due to the proposed stabilization of the lowest unoccupied molecular orbital (LUMO) of the dienophile. This computational approach has also been helpful to understand the reasons behind the extraordinary regio—and diastereoselectivity observed experimentally. The insight gained in this work allows us to predict even more reactive vinylazaarene dienophiles, which may be useful in organic synthesis.

INTRODUCTION

It is well known that the Diels—Alder cycloaddition reaction, arguably one of the most useful transformations in organic chemistry. Can be greatly accelerated in the presence of catalytic amounts of a Lewis acid (LA). Typically, the LA-binds the dienophile, resulting in a significant stabilization of the lowest unoccupied molecular orbital (LUMO) of the LA-dienophile complex, which is translated into a more favorable highest occupied molecular orbital (HOMO) (dieno)—LUMO (dienophile) gap, ultimately leading to the observed acceleration. In addition, the LA-catalyzed Diels—Alder reactions are not only faster than their parent uncatalyzed processes but can also proceed with higher regio and stereoselectivities. For instance, recent examples have shown that the inherent enals-selectivity of the cycloaddition can be reversed (i.e., favoring the corresponding exe-cycloadduct) using sterically overcrowded LA catalysts.

In this regard, Hillinski and co-workers very recently reported that the highly inefficient and unselective Diels— Alder reaction involving different dienes such as butadiene or isoprene and vinylpyridines and be transformed into a synthetically useful reaction by simply adding catalytic amounts (0.5 equiv) of the BF, Lewis acid (5cheme 1). The activation of the dienophile via binding of the pyridine lone pair to the LA makes the process not only much faster but also highly regio and endo-diastereoselective, which sharply contrasts with the analogous uncatalyzed cycloadditions. In addition, this synthetic protocol seems general as it was successfully expanded to a good variety of dienes and different virylazaarenes, including 2- or 4-vinylpyridines, quinolines, pyrarines, and pyrimidines.

The observed great acceleration of the cycloaddition was rationalized by invoking the above-mentioned traditional LUMO-lowering concept. In view of the significant stabilization of the LUMO of the dienophile upon binding to BF₂. We have, however, recently demonstrated that this LUMO-lowering concept in slightly related LA-catalyzed Diels-Alder is rather incomplete as it does not consider the impact on the reverse HOMO (dienophile)-LUMO (dieno) interaction, which indeed can offset the favorable HOMO (diene)-LUMO (dienophile) interaction. As a result, we found that the reduction of the Pauli repulsion between the key occupied it-molecular orbitals and not the above orbital

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Selectivity In Lewis Acid Promoted Reactions

Shu Kobayashi

Selectivity In Lewis Acid Promoted Reactions:

Selectivities in Lewis Acid Promoted Reactions Dieter Schinzer, 2012-12-06 The ASI workshop on Selectivities in Lewis Acid Promoted Reactions held in the Emmantina Hotel in Athens Glyfada Greece October 2 7 1988 was held to bring some light into the darkness of Lewis acid induced processes As such the workshop reflects some current trends in organic synthesis where Lewis acids are becoming a powerful tool in many different modern reactions e g Diels Alder reactions Ene reactions Sakurai reactions and in general silicon and tin chemistry. The objective of this meeting was to bring together most of the world experts in the field to discuss the major reactions promoted by Lewis acids Organic synthesis will play a major role in this book connected with some fundamental mechanistic work on allylsilane and tin chemistry Both natural product synthesis and unnatural molecules are presented in the chapters. The book presents all the 15 invited lectures and the contributions of 15 posters I am confident that the material presented in this book will stimulate the chemistry which has been discussed on our meeting around the world The meeting and the book were only possible through a grant of the NATO Scientific Affairs Devision and financial support by the following companies Kali Chemie Hannover W Germany E Merck Darmstadt W Germany Sandoz Basel Switzerland Schering Berlin W Germany Advances in Asymmetric Synthesis ,1995-03-01 Advances in Asymmetric Synthesis Modern Carbonyl Chemistry Junzo Otera, 2008-11-21 The carbonyl group is undoubtedly one of the most important functional groups in organic chemistry both in its role as reactive center for synthesis or derivatisation and as crucial feature for special structural or physiological properties Vast and profound progress has been made in all aspects modern carbonyl chemistry. These achievements are however rather dispersed in the literature and it is often not easy for the researcher obtain a comprehensive overview of a relevant topic Modern Carbonyl Chemistry overcomes this inconvenience by collating the information for appropriate themes In this work internationally renowned experts and leaders in the field have surveyed recent aspects and modern features in carbonyl chemistry such as cascade reactions one pot syntheses recognition or site differentiation Inventing Reactions Lukas J. Gooßen, 2012-10-30 Barry Trost Transition metal catalyzed allylic alkylation Jeffrey W Bode Reinventing Amide Bond Formation Naoto Chatani and Mamoru Tobisu Catalytic Transformations Involving the Cleavage of C OMe Bonds Gregory L Beutner and Scott E Denmark The Interplay of Invention Observation and Discovery in the Development of Lewis Base Activation of Lewis Acids for Catalytic Enantioselective Synthesis David R Stuart and Keith Fagnou The Discovery and Development of a Palladium II Catalyzed Oxidative Cross Coupling of Two Unactivated Arenes Lukas Goo en and K the Goo en Decarboxylative Cross Coupling Reactions A Stephen K Hashmi Gold Catalyzed Organic Reactions Ben List Developing Catalytic Asymmetric Acetalizations Steven M Bischof Brian G Hashiguchi Michael M Konnick and Roy A Periana The De NovoDesign of CH Bond Hydroxylation Catalysts Benoit Cardinal David Karl A Scheidt Carbene Catalysis Beyond the Benzoin and Stetter Reactions Kenso Soai and Tsuneomi Kawasaki Asymmetric autocatalysis of pyrimidyl alkanol Douglas C Behenna and Brian M Stoltz

Natural Products as Inspiration for Reaction Development Catalytic Enantioselective Decarboxylative Reactions of Prochiral Enolate Equivalents Hisashi Yamamoto Acid Catalysis in Organic Synthesis
Main Group Metals in Organic Synthesis
Hisashi Yamamoto, Koichiro Oshima, 2006-03-06 This is the first handbook to cover in detail all aspects of this fascinating field of chemistry In this handy two volume set readers will instantly find the information they need clearly structured according to the individual metals in the main groups hitherto only accessible after much time consuming research The result is in indispensable aid for everyday work in the lab Alongside all the classical organic reactions this book focuses on the modern variations as well as novel current reactions in organic synthesis that are closely linked to main group elements both stoechiometric and catalytic With this work the two prizewinning editors have succeeded in producing a comprehensive compendium of the main group metals as reagents for organic reactions In short this is a must for every organic chemist whether as an efficient introduction to current research for retaining an overview or for looking up detailed information

Methoden der organischen Chemie (Houben-Weyl). ,1997 Stereoselective Synthesis of Drugs and Natural Products Vasyl Andrushko, Natalia Andrushko, 2013-08-16 Brings together the best tested and proven stereoselective synthetic methods Both the chemical and pharmaceutical industries are increasingly dependent on stereoselective synthetic methods and strategies for the generation of new chiral drugs and natural products that offer specific 3 D structures With the publication of Stereoselective Synthesis of Drugs and Natural Products researchers can turn to this comprehensive two volume work to guide them through all the core methods for the synthesis of chiral drugs and natural products Stereoselective Synthesis of Drugs and Natural Products features contributions from an international team of synthetic chemists and pharmaceutical and natural product researchers These authors have reviewed the tremendous body of literature in the field in order to compile a set of reliable tested and proven methods alongside step by step guidance This practical resource not only explores synthetic methodology but also reaction mechanisms and applications in medicinal chemistry and drug discovery The publication begins with an introductory chapter covering general principles and methodologies nomenclature and strategies of stereoselective synthesis Next it is divided into three parts Part One General Methods and Strategies Part Two Stereoselective Synthesis by Bond Formation including C C bond formation C H bond formation C O bond formation C N bond formation Other C heteroatom formation and other bond formation Part Three Methods of Analysis and Chiral Separation References in every chapter serve as a gateway to the literature in the field With this publication as their guide chemists involved in the stereoselective synthesis of drugs and natural products now have a single expertly edited source for all the methods they need <u>Lewis Acids and Selectivity in Organic Synthesis M.</u> Santelli, J.-M. Pons, 1995-11-21 Lewis acids provide inexpensive access to elaborated molecules obtained with high selectivities regio stereo and enantioselectivity Lewis Acids and Selectivity in Organic Synthesis is the first book to deal with these new and promising roles of Lewis acids The book begins with general considerations on Lewis acids and a description

of Lewis acid carbonyl complexes which are involved in most of the reactions described ene reactions allylsilane and allyltin addition to carbonyl compounds addition of nucleophiles to acetals conjugated addition of allylsilanes and allyltins to unsaturated carbonyl compounds Sakurai reaction and Diels Alder reaction Subsequent chapters examine these issue in detail with special attention given to the way Lewis acids induce diastereo and enantioselectivity. The extensive use of schemes approximately 1000 ensures rapid visual uptake of the information Lewis Acids and Selectivity in Organic Synthesis serves as a valuable source of information for all who face the challenge of selectivity in organic synthesis Lewis Acid Promoted Reactions Dieter Schinzer, 1989-09-30 Bulletin of the Chemical Society of Japan Nihon Science of Synthesis: Water in Organic Synthesis Shu Kobayashi,2014-05-14 Water in Organic Synthesis Kagakkai,2004 is essential for the organic chemist in helping gain a thorough appreciation of the latest and most reliable available methods for using water in organic synthesis It illustrates how water can often be a viable and green solvent in the laboratory and provides a detailed introduction to the subject background information evaluated methods practical applications industrial applications special techniques and an overview of the latest trends The reference work also helps in inspiring chemists worldwide to find new approaches and techniques for the application of water in organic synthesis Comprehensive overview of a rapidly progressing field Critical review of aqueous reactions by 47 experts Covering almost all types of organic reactions Including special techniques with water and industrial applications Emphasis on environmental aspects

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Pectenotoxin II. Glenn C. Micalizio, 2001 Activating Agents and Protecting Groups Anthony J. Pearson, William R. Roush, 1999-07-09 Recognising the need for a cost effective reference work that deals not only with the most popular reagents in synthesis but also reaches the widest possible audience of practising organic chemists the editors of The Encyclopedia of Reagents for Organic Synthesis EROS have developed a list of the most important and useful reagents employed in the field conveniently presented in four separate volumes. The reagents included in this volume reflect the fact that protecting groups and activation procedures are often used in combination. There are many instances in the synthesis of natural and unnatural products pharmaceuticals oligosaccharides and oligonucleotides etc where similar tactics must be employed to prevent undesired activation or reaction of functionality Accordingly the most important reagents used to protect amines alcohols carboxyl carbonyl and other reactive functional groups are included in this volume The list of activating agents includes well known reagents that activate functional groups for substitution or elimination reactions as well as less traditional examples e g HMPA used to activate enolates and alkyllithium reagents to increase the nucleophilicity Each article contains all of the information found in EROS as well as expanded related reagents listings and additional references to enable the reader to guickly access a broad range of information that is beyond the scope of the reagent entries themselves This text will prove an invaluable resource *Intramolecular Diels-Alder Reactions of (Z)-substituted-1,3-Dienes* and Studies Directed Toward the Total Synthesis of Superstolide A. Neal A. Yakelis, 2003 **Bulletin of the Korean** Chemical Society, 2006 Additions to C-X ?-Bonds Barry M. Trost,1991 Volume 1 and volume 2 Fluorination in Organic and Bioorganic Chemistry John T. Welch, American Chemical Society. Meeting, 1991 Contains the proceedings of a symposium in Boston April 1990 which explored the use of organofluorine compounds to alter the molecular properties of biological systems for industrial or research purposes Covering theory the synthesis of compounds and biological compounds the 14 papers discuss such topics as bond energies in reactive intermediates perfluorated alkenes and dienes and stereocontrolled synthesis Annotation copyrighted by Book News Inc Portland OR **Modern Catalytic** Methods for Organic Synthesis with Diazo Compounds Michael P. Doyle, M. Anthony McKervey, Tao Ye, 1998-01-19 For the first time in one volume here is a complete and comprehensive text that covers those features of the chemistry of diazocarbonyl compounds that characterise their enduring versatility as intermediates for organic synthesis

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