

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

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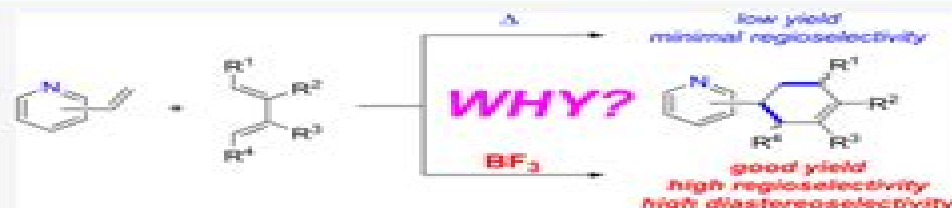
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ABSTRACT: The poorly understood factors controlling the catalysis and selectivity in Lewis acid-promoted Diels–Alder cycloaddition reactions involving vinylazaarenes as dienophiles have been quantitatively explored in detail by means of computational 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 π -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,^{1,2} can be greatly accelerated in the presence of catalytic amounts of a Lewis acid (LA).³ 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) (diene)–LUMO (dienophile) gap, ultimately leading to the observed acceleration.^{4,5} 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.⁶ For instance, recent examples have shown that the inherent *endo*-selectivity of the cycloaddition can be reversed (i.e., favoring the corresponding *exo*-cycloadduct) using sterically overcrowded LA catalysts.⁷

In this regard, Hillinko and co-workers very recently reported⁸ that the highly inefficient and unselective Diels–Alder reaction involving different dienes such as butadiene or isoprene and vinylpyridines⁹ can be transformed into a synthetically useful reaction by simply adding catalytic amounts (0.5 equiv) of the BF_3 Lewis acid (Scheme 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.⁹ In addition, this synthetic protocol seems general as it was successfully expanded to a good variety of dienes and different vinylazaarenes, including 2- or 4-vinylpyridines, quinolines, pyrazines, and pyrimidines.⁹

The observed great acceleration of the cycloaddition was rationalized by invoking the above-mentioned traditional LUMO-lowering concept^{4,5} in view of the significant stabilization of the LUMO of the dienophile upon binding to BF_3 .⁷ We have, however, recently demonstrated that this LUMO-lowering concept is slightly related LA-catalyzed Diels–Alder is rather incomplete as it does not consider the impact on the reverse HOMO (dienophile)–LUMO (diene) interaction, which indeed can offset the favorable HOMO (diene)–LUMO (dienophile) interaction.⁸ As a result, we found that the reduction of the Pauli repulsion between the key occupied π -molecular orbitals and not the above orbital

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