Catalytic Hydrogenation of Carboxylic Acid Derivatives

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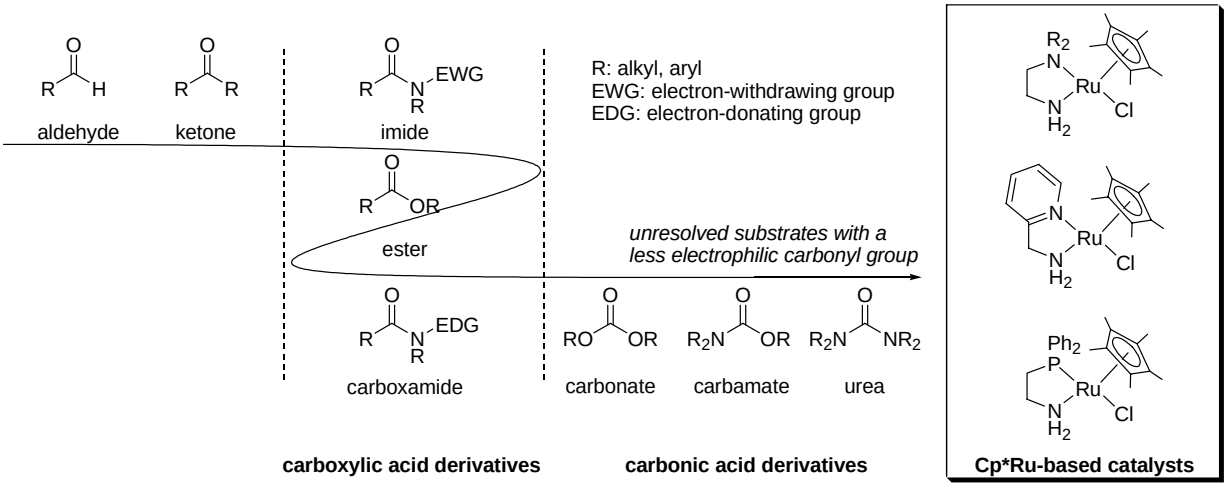
The development of synthetic methods remains a critical issue in chemistry, since there is still a distinct difference of the efficiency in the production of chemical substances between natural providence and art. In this regard, the development of novel catalytic process that provides a direct access to products is potentially beneficial, because it could possibly eliminate a conventional detour requiring sacrificial elements that are not embedded finally in the target molecules, thereby promoting effective utilization of resources and energy.

Straightforward hydrogenation of carboxylic acid derivatives has been one of challenging issues that had hitherto met little success until recently (Scheme).

This lecture will focus on our recent results on the molecular design of Cp*Ru-based catalysts which may provide a direct access to stereochemically well-defined alcohols from ubiquitous carboxylic acid derivatives.

Key findings in our study are (i) heterolytic splitting of H_2 at the coordination sphere of catalyst molecule is greatly facilitated in basic alcoholic media, (ii) electrophilicity of the resulting formal H^+ determines the range of reducible C=O bonds, (iii) basic alcoholic media also facilitates product release from the catalyst molecule, (iv) structural modification of the chelating protic amine ligand results in highly enantioselective hydrogenations.



References

(1) Ito, M.; Sakaguchi, A.; Kobayashi, C.; Ikariya, T. *J. Am. Chem. Soc.* **2007**, *129*, 290. (2) Ito, M.; Koo, L. -W.; Himizu, A.; Kobayashi, C.; Sakaguchi, A.; Ikariya, T. *Angew. Chem. Int. Ed.* **2009**, *48*, 1324. (3) Ito, M.; Kobayashi, C.; Himizu, A.; Ikariya, T. *J. Am. Chem. Soc.* **2010**, *132*, 11414. (4) Ito, M.; Ootsuka, T.; Watari, R.; Shiibashi, A.; Himizu, A.; Ikariya, T. *J. Am. Chem. Soc.* **2011**, *133*, ASAP.

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