A2-11 Photoenzymatic Catalysis – Using Light to Reveal New Enzyme Functions

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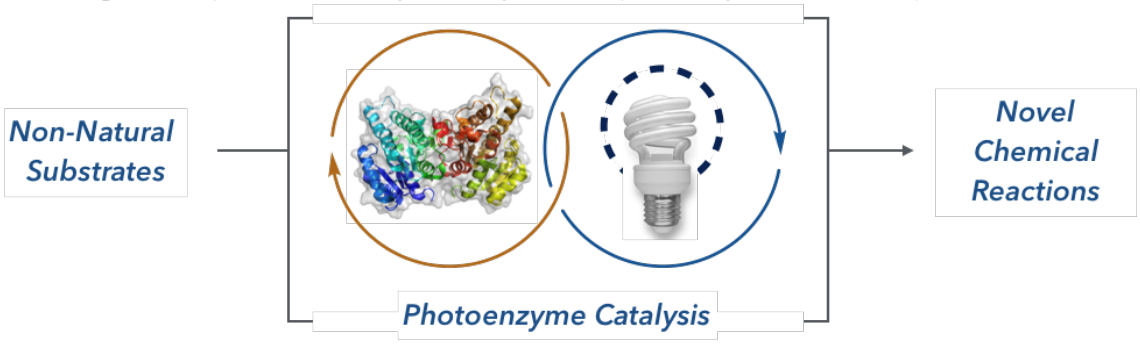
Enzymes are exquisite catalysts for chemical synthesis, capable of providing unparalleled levels of chemo-, regio-, diastereo- and enantioselectivity. Unfortunately, biocatalysts are often limited to the reactivity patterns found in nature. In this talk, I will share my groups efforts to use light to expand the reactivity profile of enzymes. In our studies, we have exploited the photoexcited state of common biological cofactors, such as NADH and FMN to facilitate electron transfer to substrates bound within enzyme active sites. In other studies, we found that enzymes will electronically activate bound substrates for electron transfer. In the presence of common photoredox catalysts, this activation can be used to direct radical formation to enzyme active sites. Using these approaches, we can develop biocatalysts to solve long-standing selectivity challenges in chemical synthesis.



1）Quaternary Charge-Transfer Complex Enables Photoenzymatic Intermolecular Hydroalkylation of Olefins, C. G. Page, S. J. Cooper, J. S. DeHovitz, D. G. Oblinsky, K. F. Biegasiewicz, A. H. Antropow, K. W. Armburst, J. M. Ellis, L. G. Hamann, E. J. Horn, K. M. Oberg, G. D. Scholes, T. K. Hyster. *J. Am. Chem. Soc.* **2021**, *143*, 97-102.

2) Photoexcitation of a Flavoenzyme Enables a Stereocontrolled Radical Cyclization, K. F. Biegasiewicz, S. J. Cooper, X. Gao, D. G. Oblinsky, J. H. Kim, S. E. Garfinkle, L. A. Joyce, B. A. Sandoval, G. D. Scholes, T. K. Hyster *Science* **2019**, *364*, 1166-1169.

3) Inducing Dynamic Stereochemistry for Asymmetric Synthesis, J. S. DeHovitz, Y. Y. Loh, J. A. Kautzky, K. Nagao, A. J. Meichan, M. Yamauchi, D. W. C. MacMillan, T. K. Hyster *Science* **2020,** *369*, 1113-1118.

PROFILE

Todd Hyster（Cornell University, Associate Professor / Department of Chemistry and Chemical Biology）

Todd was born in 1985 in Minneapolis, Minnesota (USA). He received his B.S. in Chemistry (2008) from the University of Minnesota. He joined Tomislav Rovis' group at Colorado State University for his graduate studies where he developed Rhodium-catalyzed C–H activation reactions. During his Ph.D., Todd conducted a Marie Curie Fellow with Thomas Ward at the University of Basel where he prepared an artificial metalloenzyme. After graduating, he joined the group of Frances Arnold at Caltech as an NIH Postdoctoral Fellow. In 2015 he started his independent career at Princeton University and in 2021 moved to Cornell University, where he is currently an Associate Professor of Chemistry and Chemical Biology. Todd was awarded the Searle Scholar Award (2017), Alfred P. Sloan Foundation Fellowship (2018), Junior BIOTRANs award (2021), Amgen Young Investigator Award (2021), and Arthur C. Cope Early Career Scholar Award (2022). Todd's group is focused on developing novel photochemical mechanisms to expand the types of reactions available to enzymes. These mechanisms enable these enzymes to address long-standing selectivity challenges in chemical synthesis.