

Redox biocatalysis and cofactor versatility: Scope, challenges and future directions

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Biocatalysis, the use of enzymes to catalyze chemical reactions converting substrates into products, is increasingly implemented in industry for the manufacture of fine chemicals, pharmaceuticals and biobased products. With current concerns on developing more sustainable and efficient chemical processes, the mild reaction conditions and exquisite selectivity of enzymatic reactions make them attractive for organic synthesis.

Nicotinamide adenine dinucleotide (NAD)-dependent enzymes, in particular oxidoreductases, can catalyze an impressive array of redox reactions with high selectivity (Figure 1), in many cases unattainable with classical chemical catalysts. With the aim to improve the economic viability and tune the reaction rate of NAD-dependent enzymatic reactions, we have been exploring synthetic nicotinamide cofactors (mNADHs) for oxidoreductase-driven reactions.¹ In this lecture I will present our recent studies on (i) understanding acceptance of cofactor analogues in different oxidoreductase families, (ii) identifying a portfolio of oxidoreductases and their catalytic reactions, (iii) future prospects on non-canonical cofactors.

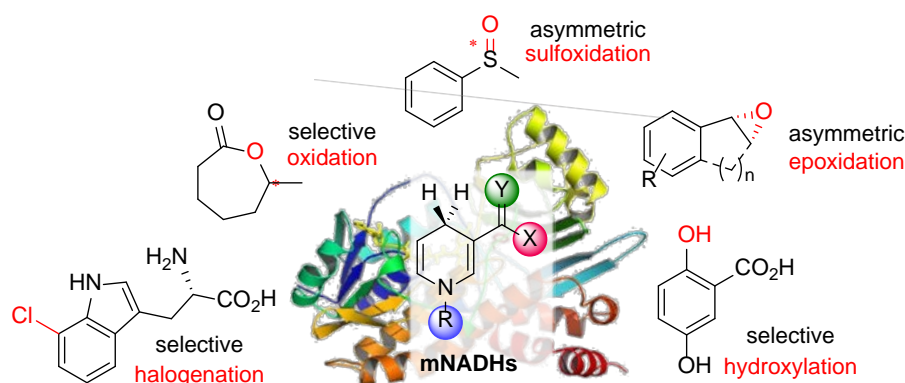


Figure 1. Synthetic cofactors applied to a selection of flavoprotein monooxygenase-catalyzed reactions.

References

- (1) Guarneri, A.; van Berkel, W. J. H.; Paul, C. E. Alternative coenzymes for biocatalysis. *Curr. Opin. Biotechnol.* **2019**, *60*, 63-71.

Caroline E. Paul received her Honours BSc and MSc in Biological Chemistry at the University of Toronto with Prof. M. Nitz on protecting group-free glycosidations, and her PhD degree at the University of Oviedo in the group of Prof. V. Gotor with Prof. V. Gotor-Fernández and Prof. I. Lavandera within the EU FP7 BIOTRAINS project on the development of biocatalytic processes.



After postdoctoral work as a Marie Curie Fellow at TU Delft with Prof. F. Hollmann, she carried out her research interests on biomimetic cofactors for oxidoreductases with a NWO VENI grant at Wageningen University. Since 2018 she is assistant professor at TU Delft in the Department of Biotechnology in Biocatalysis. Her current research interests focus on coenzyme biomimetics for oxidoreductases and other enzyme classes, and exploring non-natural enzymatic reactions for applications in organic chemistry synthesis.