BIOTRANS
LA ROCHELLE 2023

N°1239 / IL
TOPIC(s) : (Chemo)enzymatic strategies / Biocatalytic cascade reactions

ILLUMINATING BIOCATALYSIS: COMBINING CHEMICAL AND ENZYMATIC TRANSFORMATIONS FUELED BY LIGHT

AUTHORS
Sandy SCHMIDT / UNIVERSITY OF GRONINGEN, ANTONIUS DEUSINGLAAN 1, GRONINGEN

PURPOSE OF THE ABSTRACT
Enzyme catalysis and photocatalysis are two research areas that have become of great interest in organic synthesis. This is mainly because both represent attractive strategies for making chemical synthesis more efficient and sustainable. It is therefore not surprising that bio- and photocatalytic approaches are now often combined to exploit the exquisite selectivity of enzymes and the unique chemical transformations accessible to photocatalysis.[1–3]

In recent work, we have investigated the application of photobiocatalytic strategies using different photosensitizers and sacrificial electron donors to drive Rieske oxygenase (RO)-catalyzed hydroxylations in vivo,[4] demonstrating that light-induced electron transfer leads to similar catalytic activities as in whole-cell reactions supplemented with glucose. We are currently exploring the possibility of using this light-induced electron transfer via photosensitizers as a simple way of overcoming the need for the natural redox partners of ROs. This is important because the identification of the physiological redox partner(s) of a given RO is often hampered by the fact that host genomes typically contain numerous candidate genes encoding redox partner proteins, most of which are not located near the RO gene(s). Thus, a light-driven approach to screen for RO activity without the physiological redox partner(s) would provide an effective surrogate electron supply system for functional characterization and/or biocatalytic application of ROs.

In another example, we have shown that the combination of bio- and photocatalysis provides a highly valuable approach to building molecular complexity from simple, cheap and widely available starting materials.[5] By combining photocatalytic C-C bond formation with enzymatic asymmetric reduction, we have demonstrated the direct conversion of simple aldehydes and acrylates or unsaturated carboxylic acids to chiral gamma-lactones. The photochemoenzymatic synthesis of aliphatic and aromatic gamma-lactones was thereby achieved with up to >99% ee and 99% yield.

FIGURES

FIGURE 1

FIGURE 2

KEYWORDS

Biocatalysis | Photocatalysis | Rieske oxygenase | Cascade

BIBLIOGRAPHY

[1] L. Schmermund, V. Jurkaš, F. F. Özgen, G. D. Barone, H. C. Büchsenschütz, C. K. Winkler, S. Schmidt, R. Kourist, W. Kroutil, ACS Catal. 2019, 9, 4115-4144.

[2] W. Harrison, X. Huang, H. Zhao, Acc. Chem. Res. 2022, 55, 1087-1096.

[3] F. F. Özgen, M. E. Runda, S. Schmidt, ChemBioChem 2021, 22, 790-806.

[4] F. Feyza Özgen, M. E. Runda, B. O. Burek, P. Wied, J. Z. Bloh, R. Kourist, S. Schmidt, Angew. Chem. Int. Ed. 2020, 59, 3982-3987.

[5] F. F. Özgen, A. Jorea, L. Capaldo, R. Kourist, D. Ravelli, S. Schmidt, ChemCatChem 2022, 14, e202200855.