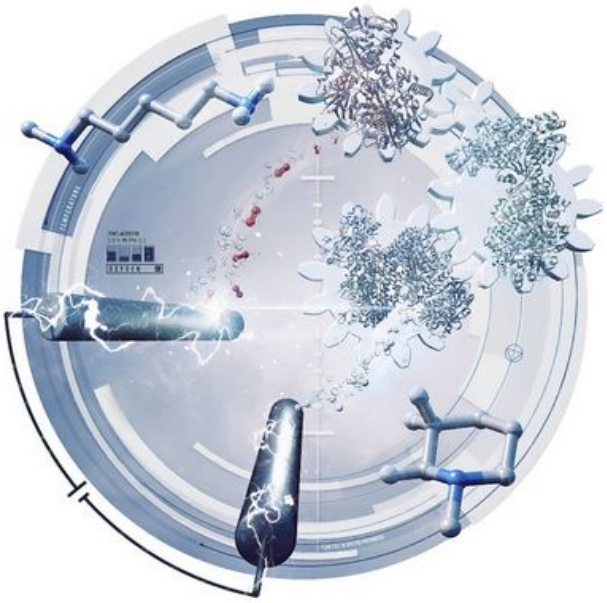
Powering Artificial Enzymatic Cascades with Electrical Energy

Start Time: Monday, March 23, 2020

End Time:



Ammar Al-Shameri, Marie-Christine Petrich, Kai junge Puring, Ulf-Peter Apfel, Bettina M. Nest and Lars Lauterbach

The use of electrical energy to perform chemical synthesis is considered to be clean, easy-to-tune, and sustainable when combined with renewable energy sources. The UniSysCat team of Lars Lauterbach in cooperation with Bettina M. Nestl from the Universitaet Stuttgart and Ulf-Peter Apfel from the Ruhr-Universität Bochum coupled chemical and enzyme catalysts for synthesis of fine chemicals, which is a core objective of UniSysCat.

Here, the authors developed a scalable platform that employs electrolysis for an *in vitro* synthetic enzymatic cascade in a continuous flow reactor. Electrolysis was performed using a pentlandite/Ni catalyst for the hydrogen-evolving reaction (HER) and the oxygen-evolving reaction (OER), respectively. Both H_2 and O_2 were transferred via a gas permeable membrane into the flow system. The membrane enabled the separation of the electrolyte from the biocatalysts in the flow system, where H_2 and O_2 served as electron mediators for the biocatalysts.The system was validated with an enzymatic cascade consisting of an oxidase, imine reductase and hydrogenase for the synthesis of N-heterocycles, which are important building blocks of agrochemicals or pharmaceutics. The O_2-dependent putrescine oxidase converted diamines to the corresponding amino aldehydes. Subsequently a spontaneous

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cyclisation formed the cyclic imines, which were reduced by the NADH-dependent imine reductase to obtain the saturated *N*-heterocycles. The consumed NADH cofactor was regenerated by a O_2 tolerant hydrogenase *via* H_2-oxidation without by-product formation. Production of methylated *N*-heterocycles from diamines with up to 99% conversion yield as well as excellent regio-selective labelling with stable isotopes was possible using this approach.This platform can be applied for a broad panel of oxidoreductases to exploit electrical energy for the synthesis of fine chemicals.

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