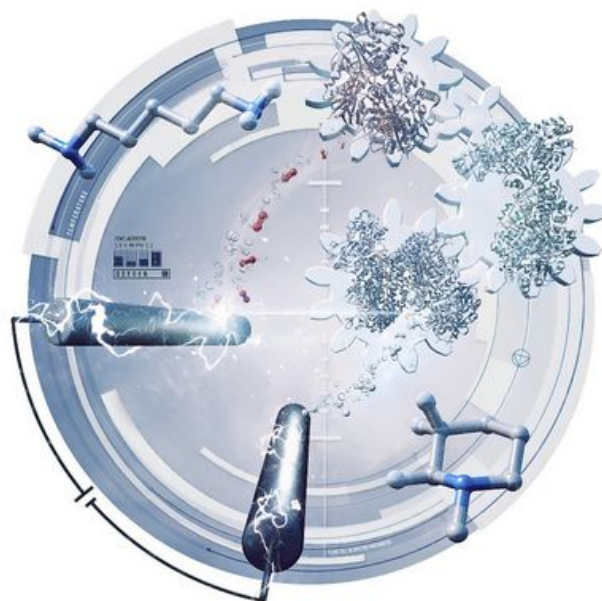


Powering Artificial Enzymatic Cascades with Electrical Energy

Start Time: Monday, March 23, 2020

End Time:



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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 Universität 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 H₂ and O₂ 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 H₂ and O₂ 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 pharmaceuticals. The O₂-dependent putrescine oxidase converted diamines to the corresponding amino aldehydes. Subsequently a spontaneous

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 O₂ tolerant hydrogenase via H₂-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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