



## Swiss Science Concentrates

A CHIMIA Column

Short Abstracts of Interesting Recent Publications of Swiss Origin

### Cooperative Weak Dispersive Interactions Actuate Catalysis in a Shape-selective Abiological Racemase

Y. Wang, M. Rickhaus, O. Blacque, K. Baldrige, M. Juríček\*, J. Siegel\*

*J. Am. Chem. Soc.* **2022**, *144*, 2679-2684.

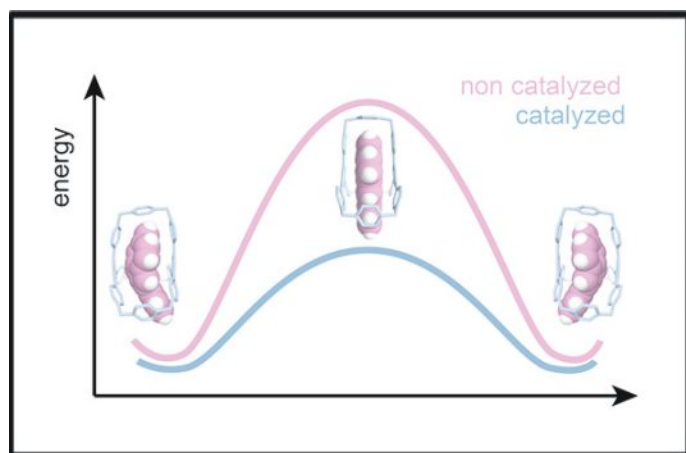
DOI: 10.1021/jacs.1c11032

University of Zurich and Tianjin University

A simple abiological host-guest system is capable of revealing racemase activity with catalytic rate enhancements of  $10^4$  without the need to employ conventional functional groups. Shape-compatibility between the catalyst active site and the reaction transition state is driven by cooperative, weak interactions, similar to the Pauling process with enzymes. As with the Jencks' conjecture about the role of catalytic antibodies, a haptan resembling the planar transition state of the bowl inversion acts as a potent inhibitor of this catalytic process. No substrate/product inhibition is detected, and relatively weak binding of the substrate is observed ( $K_a \approx 10^2 \text{ M}^{-1}$  at 293 K). This 'box-and-bowl' system shows that shape selectivity suffices for the emergence of a catalytic system, with an enzyme-like thermodynamic profile.

#### Authors' comments:

Authors' comments: "This simple, textbook system illustrates that enzyme-like catalysis can emerge from shape complementarity and by using cooperative weak interactions, without the need for a complex array of functional groups."



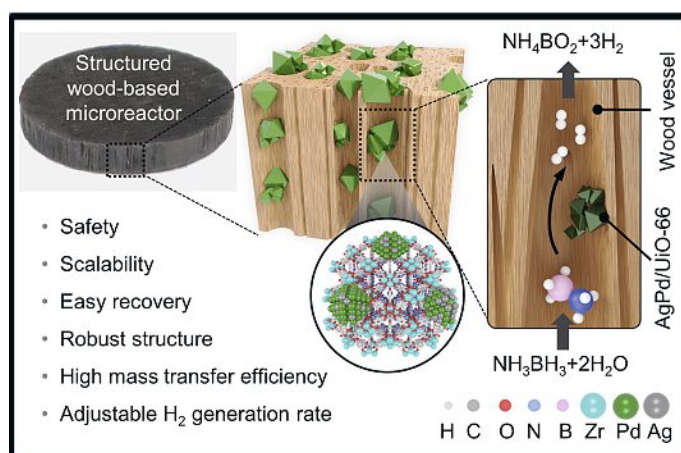
### Natural Wood-based Catalytic Membrane Microreactors for Continuous Hydrogen Generation

K. Tu, S. Büchele, S. Mitchell, L. Stricker, C. Liu, C. Goldhahn, J. Allaz, Y. Ding, R. Günther, Z. Zhang, J. Sun, S. Stucki, G. Panzarasa, S. C. Zeeman, I. Burgert, J. Pérez-Ramírez\*, T. Keplinger\*. *ACS Appl. Mater. Interfaces* **2022**, *14*, 8417-8426. EMPA, ETH Zurich

Green hydrogen ( $\text{H}_2$ ) is a sustainable energy source to replace fossil fuels. Typically,  $\text{H}_2$  generation from solid-state storage chemicals such as ammonia borane is operated in batch mode. Here, a novel and sustainable approach based on wood microreactors has been developed. Firstly, natural wood was functionalized with MOF as a catalyst carrier to stabilize silver-promoted palladium nanoparticles. The wood-based microreactors present an inherent natural oriented microchannels which allowed for continuous flow chemistry. Then, the catalytic microreactors were applied for the continuous generation of  $\text{H}_2$  from ammonia borane, reaching stable productivities. Finally, the structured catalysts showed a good reusability and allowed a fine control of the reaction parameters. Additionally, the wood microreactors can be developed by using different metals and MOF combinations, thus proving the broad applicability of this platform.

#### Authors' comments:

"This work demonstrates strong potential of the structured wood-based microreactor as a scalable platform for catalytic applications in energy- and environmental-related fields."



## Aquimarins, Peptide Antibiotics with Amino-modified C-Termini from a Sponge-derived *Aquimarina* sp. Bacterium

Cora L. Dieterich<sup>+</sup>, Silke I. Probst<sup>+</sup>, Reiko Ueoka<sup>+</sup>, Ioana Sandu, Daniel Schäfle, Michael Dal Molin, Hannah A. Minas, Rodrigo Costa, Annette Oxenius, Peter Sander, and Jörn Piel,<sup>\*</sup> *Nat. Comm. Angew. Chem. Int. Ed.* **2022**, *61*, e202115802.

Institute of Microbiology – ETH Zurich

Marine ecosystems are among the most complex and poorly explored ecosystems on the planet. Previous studies suggested animal-colonizing bacteria as a largely untapped resource of new bio-active natural products. In this paper, the authors have studied the potential of new polyketide-peptide hybrid antibiotics, named aquimarins, from the *Aquimarina* bacterial genus as antibacterial against several important pathogenic strains. Through a multidisciplinary focus, the authors report the discovery of these molecules through a genome guided approach, unravelling the biosynthetic pathway as well as the implicated enzymes; tackled the total synthesis of two natural products and further derivatives in a multi-step approach; and studied the effect and correlation of their bioactivity with the chemical properties, identifying the halogen incorporation as a key for cytotoxicity and the unusual C-terminal amine as key for their antibacterial effect.

### Authors' comments:

“Our complimentary strategy enabled us to discover these novel natural products with new scaffolds and antibiotic activity. We are excited to see possible future applications of aquimarins.”

## Asymmetric Elimination Reaction on Chiral Metal Surfaces

Samuel Stolz, Martina Danese, Marco Di Giovannantonio, José I. Urgel, Qiang Sun, Amogh Kinikar, Max Bommert, Shantanu Mishra, Harald Brune, Oliver Gröning, Daniele Passerone, and Roland Widmer<sup>\*</sup> *Adv. Mater.* **2022**, *34*, 2104481, <https://doi.org/10.1002/adma.202104481>

Empa – Swiss Federal Laboratories for Materials Science and Technology

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Asymmetric catalysis has become of paramount importance in different industrial areas such as pharmaceutical, agricultural or food industry. For a broad class of asymmetric chemical reactions, dehalogenation is an essential reaction step. Therefore, Samuel Stolz and colleagues investigated the thermally activated asymmetric debromination of prochiral 5-bromo-7-methylbenz(a)anthracene (BMA) on the intrinsically chiral PdGa{111} surfaces by different techniques such as temperature-programmed X-ray photoelectron spectroscopy (TP-XPS), scanning tunnelling microscopy (STM), and density functional theory (DFT). They demonstrated that on the chiral PdGa{111} surfaces the halogen elimination process of the two BMA surface enantiomers is separated by up to 46 K, thus achieving an enantiomeric excess of more than 90%. Overall, this work highlights the immense potential of intrinsically chiral crystals in asymmetric catalysis.

### Authors' comments:

“This work exemplarily highlights that intrinsically chiral intermetallic compounds with active site isolation, such as PdGa, are extremely promising candidate materials for asymmetric heterogeneous catalysis.”

