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Swiss Science Concentrates

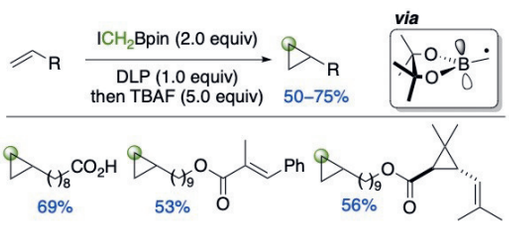
A CHIMIA Column

Short Abstracts of Interesting Recent Publications of Swiss Origin

Cyclopropanation of Terminal Alkenes *via* Sequential Atom Transfer Radical Addition/1,3-Elimination

N. D. C. Tappin, W. Michalska, S. Rohrbach, and P. Renaud,* *Angew. Chem. Int. Ed.* **2019**, doi: 10.1002/anie.201907962. University of Bern

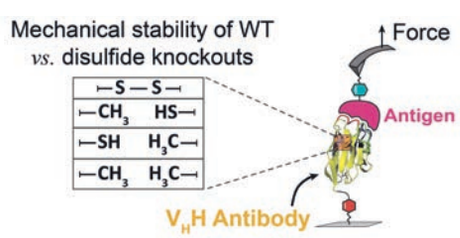
Renaud and co-workers report an operationally simple protocol for the addition of ICH_2Bpin to non-activated terminal alkenes *via* atom transfer radical addition. The resulting 3-iodoalkylboronic esters are of great interest due to their orthogonality in various synthetic transformations. In particular, cyclopropanes are obtained by treatment of this intermediate with a fluoride source, triggering a 1,3-elimination. The mild reaction conditions of this one-pot reaction lead to an excellent functional group tolerance. The high selectivity for the cyclopropanation for non-activated terminal alkenes in the presence of electron-deficient and electron-rich internal alkenes is remarkable since it is difficult to achieve by classical methods.



Removal of a Conserved Disulfide Bond Does Not Compromise Mechanical Stability of a VHH Antibody Complex

H. Liu, V. Schittny, and M. A. Nash,* *Nano Lett.* **2019**, *19*, 5524. University of Basel and ETH Zurich

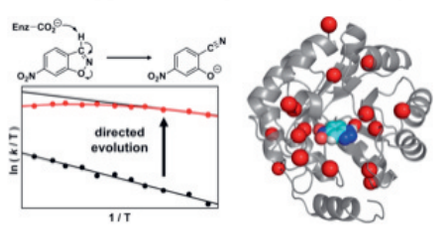
Due to their relative ease of production, small size and high stability compared with conventional full size antibodies, single-domain VHH antibodies are being developed for medical therapy. A conserved disulfide bond within the framework of these affinity proteins is reported to be crucial for their thermal stability. However, its importance for the stability of VHH antibody-antigen complexes toward mechanical stress remained unstudied. Using cysteine-to-alanine mutagenesis and AFMS-SMFS studies, the authors demonstrated that the mechanostability of the VHH:mCherry interaction is not impacted by the removal of this bond and hence is not intimately linked to the mechanical stability of the VHH complex.



Emergence of a Negative Activation Heat Capacity During Evolution of a Designed Enzyme

H. A. Bunzel, H. Kries, L. Marchetti, C. Zeymer, P. R. E. Mittl, A. J. Mulholland, and D. Hilvert,* *J. Am. Chem. Soc.* **2019**, *141*, 11745. ETH Zurich, University of Zurich and University of Bristol

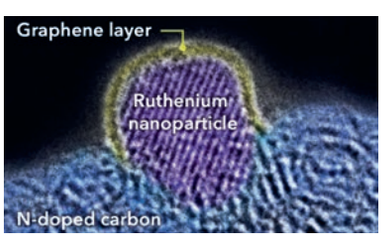
The authors have chosen a computationally designed enzyme, which catalyzes the Kemp elimination, as a model for an evolutionarily naïve catalyst. Then, they optimized its catalytic efficiency by subjecting it to nine rounds of laboratory evolution. As for most natural enzymes and as theorized for primordial enzymes, the rate acceleration achieved by the evolved Kemp eliminases was shown to have enthalpic origins. However, temperature-dependent activation parameters were observed for the most efficient variants of the enzyme. This can be explained by the emergence of an activation heat capacity and suggests strong adaptation to the temperature applied during the evolutionary experiments.



Preserved in a Shell: High-Performance Graphene-confined Ruthenium Nanoparticles in Acetylene Hydrochlorination

S. K. Kaiser, R. Lin, F. Krumeich, O. V. Safanova, and J. Pérez-Ramírez,* *Angew. Chem. Int. Ed.* **2019**, *58*, 12297. ETH Zurich and Paul Scherrer Institute

Ruthenium-based catalysts are cheaper alternatives to gold-based systems for the production of polyvinyl chloride via acetylene hydrochlorination, but they display inferior activity and stability. Pérez-Ramírez and co-workers now identified surface oxidic metallic ruthenium nanoparticles hosted on polyaniline-derived *N*-doped carbon as an efficient catalyst rivalling the performance of gold-based systems. They were able to achieve a 20-fold increased catalyst stability through careful identification and inhibition of the main deactivation modes. The undesired Ru nanoparticle redispersion was inhibited using encapsulation into permeable monolayer graphene shells, whereas O_2 co-feeding allowed the removal of deleterious coke formation at the metal sites.



Prepared by Yann Baumgartner, Nadja Niggli, David Savary, Pierre Thesmar and Olivier Baudoin*

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