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Swiss Science Concentrates

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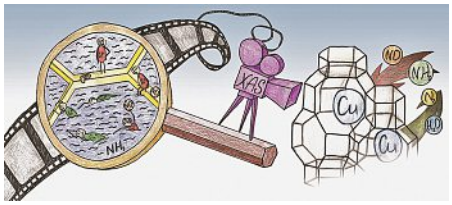
Short Abstracts of Interesting Recent Publications of Swiss Origin

Time-resolved Copper Speciation During Selective Catalytic Reduction of NO on Cu-SSZ-13

Adrian Marberger, Andrey Petrov, Patrick Steiger, Martin Elsener, Oliver Kröcher, Maarten Nachtegaal, and Davide Ferri*, *Nat. Catal.* **2018**, *1*, 221. PSI

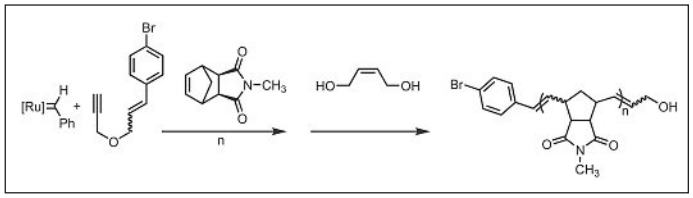


Copper-exchanged zeolites catalyze the selective catalytic reduction (SCR) of NO_x by NH_3 in diesel vehicles. Previously proposed reaction mechanisms were based on spectroscopic observations of the catalyst's structure under steady-state conditions. The group at PSI exploited time-resolved quick-EXAFS and transient experimentation to demonstrate the role of different Cu species in Cu-SSZ-13 under SCR conditions. Cu(I) oxidation was found to be inhibited by NH_3 coordination and represents the rate-limiting step below 250 °C. During the relaxation of inhibition, a fourfold coordinated Cu(II) species was identified as the key reaction intermediate. These results could be transferred to catalytic experiments on the honeycomb catalyst highlighting the power of the reported spectroscopic approach.

Functional Metathesis Catalyst through Ring Closing Enyne Metathesis: One Pot Protocol for Living Heterotelechelic Polymers

Subhajit Pal, Fiorella Lucarini, Albert Ruggi, and Andreas F. M. Kilbinger*, J. *Am. Chem. Soc.* **2018**, *140*, 3181. University of Fribourg.

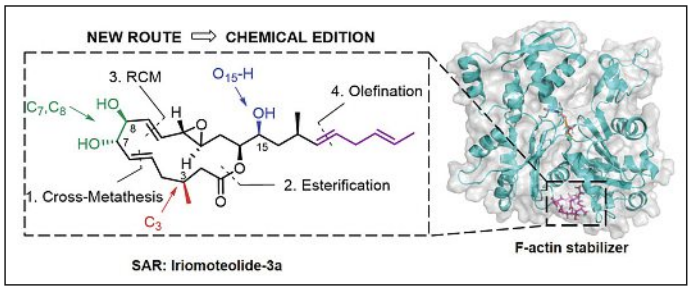
Highly functionalized heterotelechelic polymers are difficult to synthesize yet highly prized for their emerging materials applications. Here the Kilbinger group reports a new 'prefunctionalization agent' (PFA) that is compatible with both generation I and III Grubbs' catalysts. Exploiting the catalysts' selectivity profile for alkynes and multi-substituted olefins, they can now access heterotelechelic polymers with a one-pot protocol without intermediate purification. The scope of the reaction includes methoxy, bromo, triisopropylsiloxy and alkylidene groups on the PFA. End-functionalization was achieved by employing symmetrical olefin chain transfer agents to terminate with a variety of functional groups.



Iriomoteolides: Novel Chemical Tools to Study Actin Dynamics

Andrea Unzue, Riccardo Cribiu, Maria M. Hoffman, Tim Knehans, Karine Lafleur, Amedeo Caflisch*, and Cristina Nevado*, *Chem. Sci.* **2018**, *9*, 3793. University of Zurich

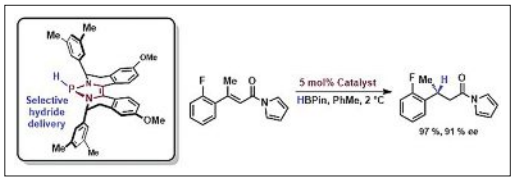
Small molecules that target the actin cytoskeleton have long been recognized as valuable molecular probes and pharmaceutical agents. In this collaborative report, the Caflisch and Nevado groups investigated the cellular targets of iriomoteolide-3a and a collection of related macrolide analogues. A new approach for their synthesis has enabled scaffold-diversification and solved the supply problem. Structure-activity relationships suggest that actin is one of iriomoteolides' primary cellular targets – according to their inhibition of cell migration, induction of morphological changes, reversible cytoplasmic retraction and reduction of F-actin fibers in a time and dose dependent manner. These results demonstrate the potential of iriomoteolide-3a and its related analogues as probes to influence actin dynamics in a reversible and selective manner.



Chiral 1,3,2-Diazaphospholenes as Catalytic Molecular Hydrides for Enantioselective Conjugate Reductions

Solène Miaskiewicz, John H. Reed, Pavel A. Donets, Caio C. Oliveira, and Nicolai Cramer*, *Angew. Chem. Int. Ed.* **2018**, *57*, 4039. EPF Lausanne

Catalytic asymmetric hydrogenation is a key reaction in petrochemical, pharmaceutical, material, and food industries. Nevertheless, there is still a lack of organic catalysts capable of asymmetric hydride delivery under mild reaction conditions. To correct this, Cramer and co-workers reported a new class of structurally rigidified chiral methoxy-1,3,2-diazaphospholene catalysts that perform well in enantioselective 1,4-reductions of α,β-unsaturated carbonyl compounds to give high yields and enantioselectivities of up to 95.5:4.5 e.r. The Cramer group is now investigating these chiral hydride transfer agents for their performance in other enantioselective transformations.



Prepared by Aaron Johnson, Ashkan Karimi, Verena Schreier, and Nathan W. Luedtke*

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