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A CHIMIA Column

Short Abstracts of Interesting Recent Publications of Swiss Origin

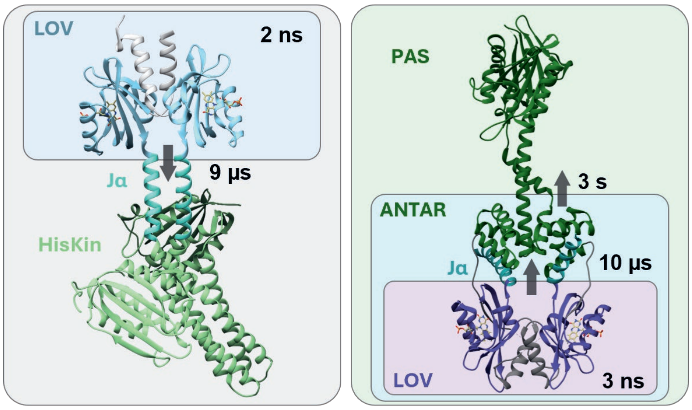
Signal Propagation in LOV-based Multidomain Proteins: Time-resolved Infrared Spectroscopy Reveals the Complete Photocycle of YF1 and PAL

Raoul E. Herzog*, Philipp Janke, Paul M. Fischer, Philipp J. Heckmeier, Chongyao Wei, Probal Nag, Sina J. Hartmann, Matthias Mulder, Fabienne Stierli, Jörg Standfuss, Igor Schapiro*, and Peter Hamm*

Phys. Chem. Chem. Phys. **2026**,
https://doi.org/10.1039/D5CP03982G
Department of Chemistry, University of Zurich, Winterthurerstrasse 190, CH-8057 Zurich, Switzerland.

Light-oxygen-voltage (LOV) proteins are versatile photoreceptors, but how light signals propagate in multidomain LOV proteins is still unclear. The authors of this study demonstrate that time-resolved infrared spectroscopy can resolve the full photocycle dynamics of two multidomain LOV proteins, YF1 and PAL, from picoseconds to hours. Both proteins exhibit nearly identical nanosecond-scale excitation and intersystem crossing, reflecting conserved chromophore–binding pocket interactions, with minor differences attributed to a single phenylalanine residue. Divergence emerges at the microsecond timescale, where cysteine-adduct formation is coupled to global structural changes. In YF1, major conformational changes in the histidine kinase effector domain occur concurrently with adduct formation and are mediated by the Jα-helix. In PAL, adduct formation opens the RNA-binding site and is followed by a later rearrangement of a distal PAS domain. These results highlight topology-dependent roles of the Jα-helix in signal transmission and inform future optogenetic applications.

Authors' comments:
"Transient infrared spectroscopy can provide valuable insights into the signal transduction pathways in multidomain proteins, and reveal the complex, topology-dependent interplay between different domains."



YF1 **PAL**

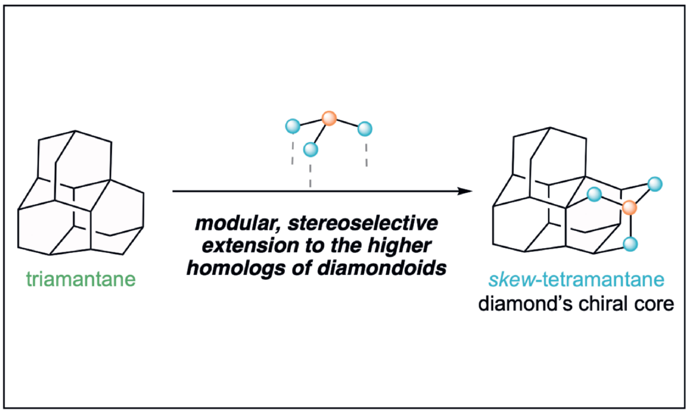
Stereoselective Total Synthesis of Skew-tetramantane

Xiao-Yu Li and Christof Sparr*

Nat. Chem. **2026**, https://doi.org/10.1038/s41557-025-02026-0
Department of Chemistry, University of Basel, 4056 Basel, Switzerland

Diamondoids have attracted long-standing interest due to their extreme rigidity, lipophilicity, non-toxicity, and well-defined three-dimensional structures, yet the selective synthesis of higher chiral diamondoids has remained a major challenge. Skew-tetramantane, the elementary chiral diamondoid and parent σ-helicene, has previously only been identified in trace amounts in fossil fuels, with synthetic efforts proving unsuccessful for decades. Here, a targeted total synthesis of skew-tetramantane was achieved by exploiting recent advances in photocatalysis and transition-metal-catalysed carbene chemistry. A stereoselective adamantalogous cage extension was accomplished through a photocatalytic Giese reaction, followed by highly controlled intramolecular C(sp^3)–H insertions using chiral rhodium catalysis. Subsequent ring expansion, hydration and an unstabilized carbenoid-mediated C(sp^3)–H insertion completed the diamondoid framework. This strategy enables access to isomerically pure skew-tetramantane with catalyst-defined σ-helicity and provides a general, selective route to higher diamondoids.

Authors' comments:
"We discovered that the selective synthesis of higher diamondoids is feasible by the recently developed methods in photocatalysis and C–H insertion chemistry. The stereoselective adamantalogous cage-extension is presented as a conceptual basis to prepare diamondoids – constituents of the most iconic form of carbon."



Prepared by Cesare Berton, James Southwell, Titouan Chetot, Jonas Genz, Stanislav Prytuliak, Fan Liu, Eda Nisli, Deborah Bäcker, Samy Kichou, Dominik Roth, Dilay Kepil, Laura Schüpke, Jan Vetterli, Tamara Trajkovic, Marc Würsch, Henrik Braband, and Jason P. Holland*

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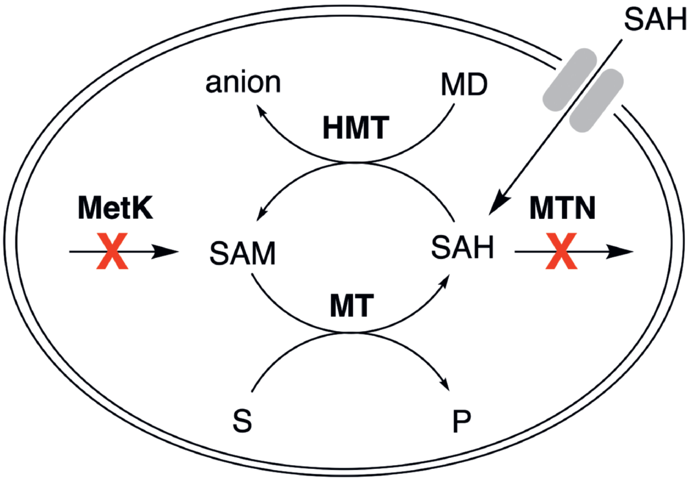
Short-Circuiting the SAM-Cycle in *Escherichia Coli*

Zhong Li, Xiaojin Wen, Seseg B. Bolotova, and Florian P. Seebeck*

J. Am. Chem. Soc. **2025**, *147*, 47690,
https://doi.org/10.1021/jacs.5c17370
Department of Chemistry, University of Basel, Mattenstrasse 22, Basel CH-4002, Switzerland; Molecular Systems Engineering, National Competence Center in Research (NCCR), CH-4058 Basel, Switzerland; Swiss Nanoscience Institute (SNI), Swiss Nanoscience Institute (SNI), CH-4056 Basel, Switzerland

The authors describe an *Escherichia coli* strain constructed with a Short-Circuited SAM-Cycle (SCSC), in which the natural SAM biosynthesis pathway is replaced by a synthetic system. The key achievements include: identifying methyl mesitylene sulfonate (MeMS) as a biocompatible methyl donor; discovering and engineering halide methyltransferases (HMTs) that efficiently methylate S-adenosylhomocysteine (SAH) using MeMS; and creating a metK- and mtn-deficient *Escherichia coli* strain that relies exclusively on HMT activity for SAM regeneration and growth. The strain was successfully used for *in vivo* directed evolution of HMTs, improving their stability and activity, and for the production of fully isotope-labelled natural products, demonstrating the potential of SCSC strains for biocatalysis, metabolic engineering, and libelled compound synthesis.

Authors' comments:
"Replacing a central metabolic pathway in a living organism with a process that we developed in the laboratory was the first exciting venture of our group into synthetic biology."



Dynamics and Control of Active Sites in Cobalt Diselenide Catalysts for Overall Water Electrolysis

Yonggui Zhao*, Nanchen Dongfang, Rolf Erni, Marcella Iannuzzi, and Greta R. Patzke*

Nat. Commun. **2025**, *16*, 10293,
https://doi.org/10.1038/s41467-025-65210-3
Department of Chemistry, University of Zurich, Zurich, Switzerland

Revealing the nature of catalytically active sites under operating conditions is a central challenge in advancing electrocatalyst development. In this work, *operando* surface/bulk-sensitive spectroscopic techniques combined with density functional theory calculations were used to correlate structure and activity in marcasite- and pyrite-type $CoSe_2$ during overall water splitting. Under acidic conditions, marcasite $CoSe_2$ experiences mild surface corrosion, forming disordered Se–Co–Se moieties that promote the hydrogen evolution reaction (HER). In alkaline media, however, the same material undergoes a potential-induced surface transformation, generating metallic Se–Co–Co–Se motifs that act as the active sites for hydrogen production. Such pH-dependent structural evolution is absent in both pristine and sulfur-doped pyrite $CoSe_2$. Additionally, *operando* spectroelectrochemical studies reveal that the formation of highly disordered Co(IV) species during operation is a key feature governing oxygen evolution across chalcogenide catalysts. Overall, the findings underscore how dynamic local coordination environments dictate catalytic reaction kinetics.

Authors' comments:
"Using *operando* spectroelectrochemical methods, we highlight the critical role of phase engineering and heteroatom sulfur substitution in the dynamics of the catalytically active species and sites during water splitting processes."