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Short Abstracts of Interesting Recent Publications of Swiss Origin

Aldehyde-catalyzed epoxidation of unactivated alkenes with aqueous hydrogen peroxide

Ierasia Triandafillidi, Maroula G. Kokotou, Domonik Lotter, Christof Sparr, and Christoforos G. Kokotos, *Chem. Sci.* **2021**, *12*, 10191,
https://doi.org/10.1039/D1SC02360H
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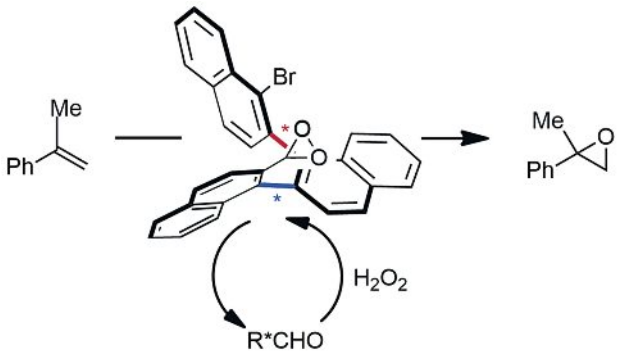
Epoxides are versatile precursors in the synthesis of numerous important molecules. While there are many methods for organocatalytic epoxidation, there has yet to be a selective strategy for this transformation using the environmentally-friendly and inexpensive H_2O_2 as the oxidant. In this publication, the authors report a novel and high-yielding method for the oxidation of cyclic and acyclic alkenes using an atropisomeric two-axis aldehyde. The relative configuration of the stereogenic axes of the catalyst and the resulting proximity of the aldehyde and backbone residues resulted in high catalytic efficiencies, while mechanistic investigations supported a non-radical alkene oxidation by an aldehyde-derived dioxirane intermediate generated from H_2O_2 through Payne and Criegee intermediates.

Authors' comments:
"After observing a surprisingly different reactivity of carbaldehydes within specific atropisomeric multiaxis systems, their catalytic performance was investigated for epoxidation reactions. A mechanism through a unique aldehyde-derived dioxirane is proposed."

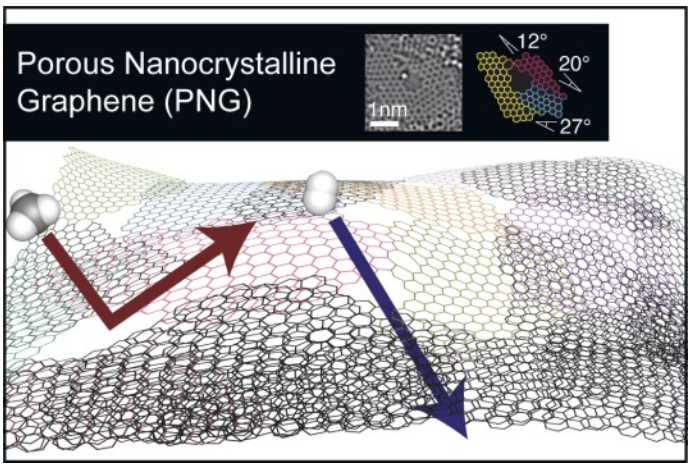


Bottom-up synthesis of graphene films hosting atom-thick molecular sieving apertures

Luis F. Villalobos, Cédric Van Goethem, Kuang-Jung Hsu, Shaoxian Li, Mina Moradi, Kangning Zhao, Mostapha Dakhchoune, Shiqi Huang, Yueqing Shen, Emad Oveisi, Victor Boureau, and Kumar V. Agrawal,* *Proc. Natl. Acad. Sci. USA* **2021**, *118(37)*, e2022201118,
https://doi.org/10.1073/pnas.2022201118
École Polytechnique Fédérale de Lausanne (EPFL)

Single-layer graphene porous films have become highly attractive because of their extraordinary properties and their remarkable applications, notably in the field of high-performance membranes. However, the synthesis of permeable two-dimensional films hosting a high density of molecular-sieving apertures remains a challenge. Herein, the authors report for the first time a direct bottom-up synthesis of a porous nanocrystalline graphene (PNG) hosting a high density (~10^{12} cm^{-2}) of nanopores based on the controlled precipitation and crystallisation of carbon on Ni surface. Results revealed both a high gas permeance and gas pair selectivity comparable to the state-of-the-art graphene membrane prepared by postsynthetic lattice etching. Moreover, this unique nanostructure was functionalised with oxygen and CO_2-philic polymers leading to increased gas selectivity and to attractive carbon capture performance, respectively. Overall, this method improves the scale-up potential of graphene membranes by reducing the processing steps.

Authors' comments:
"The direct synthesis of nanoporous graphene films with a high density of molecular sieving nanopores will accelerate the scale-up of this technology. From a fundamental perspective, the obtained nanopores, formed at the boundary of three or more nanograins, are extremely interesting because their structure is not limited by the lattice of a single graphene grain."



Prepared by Lucia Robustini, Valentina Marchini, Lauriane Pillet, David Lim, and Francesca Paradisi*
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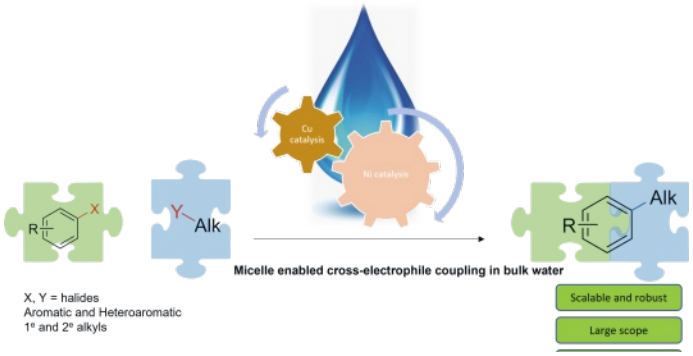
Micelle enabled C(sp²)–C(sp³) cross-electrophile coupling in water via synergistic nickel and copper catalysis

Ning Ye,* Bin Wu, Kangming Zhao, Xiaobin Ge, Yu Zheng, Xiaodong Shen, Lei Shi, Margery Cortes-Clerget, Morgan L. Regnier, Michael Parmentier, Fabrice Gallou,* *Chem. Comm.* **2021**, *57*, 7629, https://doi.org/10.1039/D1CC02885E
Novartis Pharma AG, Switzerland, Suzhou Novartis Technical Development Co., China

In light of the growing demand of the chemistry-in-water technology, a robust nickel and copper co-catalyzed C(sp^2)-C(sp^3) cross-electrophile coupling has been developed, which enables the synthesis of a wide variety of functionalized products under micellar and very mild conditions, using water as the bulk medium, for a cost-effective process. This synergistic nickel and copper effect is an alternative to traditional and previously reported methods to access alkylated arenes, providing good-to-excellent yields and high chemoselectivity. The process has been successfully scaled up to multigram-scale production in a robust manner.

Authors' comments:
"Enabling cross-electrophilic cross-couplings in water using minimal amount of non-noble metals in a robust manner lets us envision a very exciting future for always more efficient modern cross-couplings."



Picomole-Scale Synthesis and Screening of Macrocyclic Compound Libraries by Acoustic Liquid Transfer

Gontran Sangouard, Alessandro Zorzi, Yuteng Wu, Edouard Ehret, Mischa Schüttel, Sangram Kale, Cristina Diaz-Perlas, Jonathan Vesin, Julien Bortoli Chapalay, Gerardo Turcatti, and Christian Heinis* *Angew. Chem. Int. Ed.* **2021**, *60,* 21702, https://doi.org/10.1002/anie.202107815
Institute of Chemical Sciences and Engineering, School of Basic Sciences, Ecole Polytechnique Fédérale de Lausanne (EPFL)

The generation of compound libraries needed for high-throughput screening in drug development typically requires the synthesis of molecules at a milligram or higher scale. In order to generate larger libraries while reducing reagent consumption, reaction volumes, and synthesis time, a new approach was developed in which compounds are combinatorially synthesized at a picomole scale, using acoustic droplet ejection (ADE) technology. ADE allowed contact-free transfer of reagents in nanoliter volumes to discrete wells of 384-microwell plates. The principle was applied for the synthesis of a macrocycle compound library. To test whether ligands could be identified from such libraries, a target-tailored library was generated for the oncology-driving protein–protein interaction (PPI) between p53 and MDM2, and screened. From a library of several thousand macrocyclic compounds, inhibitors with sub-micromolar affinity for the PPI target were identified.

Authors' comments:
"While acoustic liquid transfer has been broadly applied for high-throughput screening, we were happy to find that this powerful technology is also perfectly suited for combinatorial chemical synthesis."