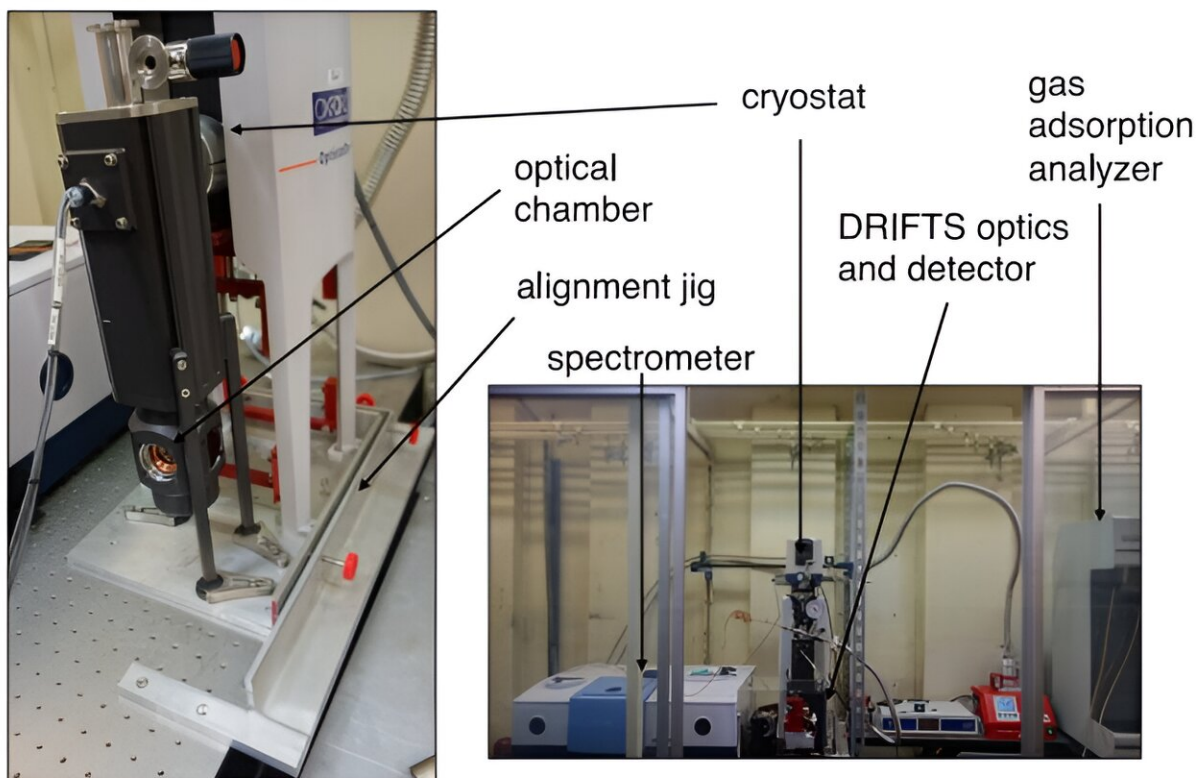


How lab-made catalysts can help convert difficult greenhouse gases

November 7 2023, by Sarah-Lena Gombert



Custom-made in situ DRIFTS apparatus. Credit: *Science* (2023). DOI: 10.1126/science.add7417

Natural gas consists of light hydrocarbons such as methane and ethane. Such gases are more potent greenhouse gases than CO₂, are constantly released into the atmosphere from natural gas wells, and are more

difficult to store than, for example, their corresponding alcohols (methanol and ethanol, respectively).

While large scale facilities exist for transforming natural gas, the excessive cost of building and operating such plants at smaller natural gas wells precludes effective conversion globally. Thus, new cost-effective and benign technologies to solve this problem are highly sought after.

The direct oxidation of hydrocarbon constituents of [natural gas](#) with abundant O₂ as the oxidant at near ambient temperatures and pressure is therefore extremely attractive for the development of new green technologies for hydrocarbon valorization. Scientists have now succeeded in getting the gases under control with a new catalyst. The research team has [published](#) their results in *Science*.

Nature has developed enzymes that are capable of activating dioxygen for selective hydrocarbon oxygenation reactions. One class of non-heme iron containing enzymes is the α -ketoglutarate dependent dioxygenases, such as the well-studied enzyme taurine dioxygenase (TauD). This enzyme utilizes an α -keto acid as a co-substrate to cleave the oxygen–oxygen bond of dioxygen to yield a reactive iron-oxo species (TauD-J) that oxygenates abundant C–H bonds directly to provide the corresponding alcohols.

An international team of researchers led by Prof. Jeffrey R. Long at UC Berkeley, and including researchers from both Mülheim Max Planck Institutes (MPI für Kohlenforschung and MPI for Chemical Energy Conversion), directors Frank Neese and Serena DeBeer, as well as group leaders Eckhard Bill (deceased October 6th, 2022), Daniel J. SantaLucia, Dimitrios A. Pantazis, and Sergey Peredkov, was able to mimic the TauD functionality in a heterogenous catalyst material that is well-suited for solid-gas reactions.

This material belongs to the class of metal-organic frameworks (MOFs), which are crystalline porous materials consisting of organic linkers and metal ions or cluster nodes that feature large surface areas. The structures are highly chemically tunable, and thus allow for well-defined tailoring of new heterogeneous catalysts.

The new materials are capable of catalytic hydrocarbon oxygenation at near ambient temperatures utilizing O₂—reminiscent of enzyme reactivity. The team at the Mülheim Chemistry Campus studied the reactive intermediate generated from the starting MOF and O₂, a high-valent iron-oxo species.

The nature of the material allowed for site-isolation of this reactive iron-oxo species, which was studied with various state-of-the-art spectroscopic techniques, namely variable-temperature variable-field Mössbauer spectroscopy and Fe K β X-ray emission spectroscopy (collected at the PINK X-ray beamline at BESSY II at the Helmholtz-Zentrum Berlin für Materialien und Energie), as well as state-of-the-art [computational methods](#), which confirmed structural and electronic similarities with TauD-J, namely that the intermediate is in a high-spin state.

Significantly, this is the first non-enzymatic system that oxidizes light hydrocarbons with dioxygen akin to metalloenzyme reactivity via a fully characterized high-spin iron-oxo intermediate.

More information: Kaipeng Hou et al, Reactive high-spin iron(IV)-oxo sites through dioxygen activation in a metal–organic framework, *Science* (2023). [DOI: 10.1126/science.add7417](https://doi.org/10.1126/science.add7417)

Provided by Max Planck Society

Citation: How lab-made catalysts can help convert difficult greenhouse gases (2023, November 7) retrieved 2 May 2024 from

<https://phys.org/news/2023-11-lab-made-catalysts-difficult-greenhouse-gases.html>

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