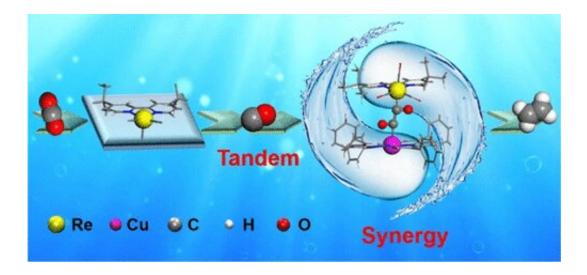


Tandem photocatalysis of CO2 to C2H4 achieved

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Graphical abstract. Credit: *Journal of the American Chemical Society* (2023). DOI: 10.1021/jacs.3c02370

The photocatalytic conversion of CO_2 into C_{2+} products such as ethylene is a promising path toward the carbon neutral goal, but it remains a big challenge due to the high activation barrier for CO_2 and similar reduction potentials of many possible multi-electron transfer products.

In a study published in the *Journal of the American Chemical Society*, the research group led by Prof. Cao Rong and Prof. Huang Yuanbiao from Fujian Institute of Research on the Structure of Matter of the Chinese Academy of Sciences reported a tandem photocatalysis strategy to



support conversion of CO_2 to ethylene.

The researchers developed this tandem photocatalysis strategy by the construction of the synergistic dual sites in rhenium-(I) bipyridine fac-[Re^I(bpy)(CO)₃Cl] (Re-bpy) and copper-porphyrinic triazine framework (PTF(Cu)). With these two catalysts, they produced a large amount of ethylene at a rate of 73.2 μ mol g⁻¹ h⁻¹ under visible light irradiation. Ethylene cannot be obtained from CO₂ by the use of either component of the Re-bpy or PTF(Cu). Only monocarbon product CO is produced under similar conditions with a single catalyst.

In the tandem photocatalytic system, the researchers found that the CO generated at the Re-bpy sites was adsorbed by the nearby Cu single sites in PTF(Cu), and this was followed by a synergistic C-C coupling process which ultimately produces ethylene.

Based on the analysis of the in situ diffuse reflectance infrared Fourier transform spectroscopy (DRIFTS) spectra and density functional theory (DFT) calculations, the researchers photoreduced CO_2 to Re-bpy-CO* over the Re-bpy, from which some of the CO can be desorbed and moved to the nearby PTF(Cu) where it is adsorbed by the single-atomic Cu site to form PTF(Cu)-CO*.

Additionally, the C-C coupling occurred in the synergy of the intermediates PTF(Cu)-CO* and Re-bpy-CO* to form Re-*CO-CO*-Cu, which was finally reduced and released as <u>ethylene</u> with multistep proton-coupled electron-transfer (PCET) processes. DFT calculations demonstrated that the coupling process between PTF(Cu)-*CO and Re-bpy-*CO to form the key intermediate Re-bpy-*CO-*CO-PTF(Cu) is vital to the C_2H_4 production.

This study provides a new way for the design of efficient photocatalysts for photocoversion of CO_2 to C_2 products via a tandem process driven



by visible-light under mild conditions.

More information: Rui Xu et al, Tandem Photocatalysis of CO_2 to C_2H_4 via a Synergistic Rhenium-(I) Bipyridine/Copper-Porphyrinic Triazine Framework, *Journal of the American Chemical Society* (2023). DOI: 10.1021/jacs.3c02370

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