

CATALYTIC ACTIVITY OF Cu/Mo₂CT_x: HYDROGENATION OF CO₂ AND CO TO METHANOL

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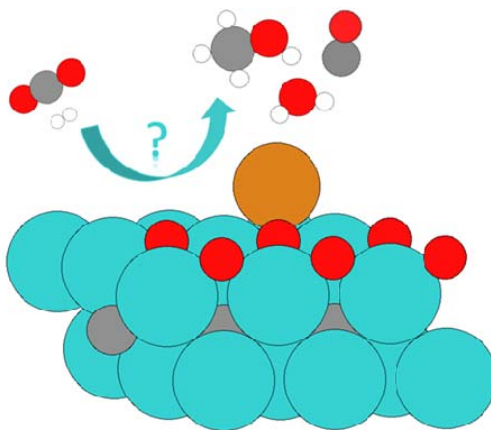
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CO₂ emissions are harmful to the environment, given their role in climate change and ocean acidification. A keyway to harness CO₂ is to use it as a precursor to high-energy density materials ^[1] to facilitate the energy transition. Hydrogenation of CO₂ to methanol is a promising option for this purpose.

The silica-supported Cu/Mo₂CT_x (MXene) catalyst shows higher activity than the industrial reference system Cu/ZnO/Al₂O₃. To better understand CO₂ hydrogenation in Cu/Mo₂CT_x, we performed DFT calculations using the VASP package ^[2] to evaluate the reaction mechanism using a previously calibrated theoretical model benchmarked against the experiment. ^[3]

At a mechanistic level, our results ^[4] show the crucial role played by the Cu/Mo₂CT_x interface in providing a low-energy pathway to facilitate the hydrogenation of CO₂ to methanol. Both the Cu atom and the Mo₂CT_x support participate in the reaction mechanism. Both allow the successive heterolytic cleavages of molecular hydrogen (H₂) necessary to form HCOO*, H₂COO*, and H₂COOH* species, simultaneously with adsorbed H*. CH₃OH is readily formed together with CO under reaction conditions. In addition, the formation of CO through the reverse water-gas shift reaction (RWGS) facilitates its hydrogenation to methanol. These findings open new avenues for the hydrogenation of CO₂ and CO by exploiting metal-support interactions and considering the role of Cu/MXenes interfaces.



[1] Álvarez A. et al. Chem. Rev., 2017, 117, 9804–9838.

[2] Kurlov, A., Deeva, E.B., Abdala, P.M. et al. Nat. Commun., 2020, 11, 4920.

[3] G. Kresse and J. Hafner, Phys. Rev. B 47, 558 (1993); ibid. 49, 14251 (1994). G. Kresse and J. Furthmüller, Comput. Mat. Sci. 6, 15 (1996). G. Kresse and J. Furthmüller, Phys. Rev. B 54, 11169 (1996).

[4] Zhou, H., Chen, Z., Vidal, A. et al. Nat. Catal., 2021, 4, 860–871.