Highly Active and Stable Single-Atom Cu Catalysts Supported by a Metal–Organic Framework

Ali M. Abdel-Mageed,1,# Bunyarat Rungtaweeveranit,2,3,# Magdalena Parlinska-Wojtan,4 Xiaokun Pei,2,3 Omar M. Yaghi,2,3,* and R. Jürgen Behm,1,*

1Institute of Surface Chemistry and Catalysis, Ulm University, D-89069 Ulm, Germany
2Department of Chemistry and Kavli Energy NanoSciences Institute, University of California, Berkeley, CA 94720, USA
3Materials Sciences Division, Lawrence Berkeley National Laboratory, CA 94720, USA,
4Institute of Nuclear Physics, Polish Academy of Sciences, Krakow 31-342, Poland

ABSTRACT: Single-atom catalysts are often considered as the ultimate design principle for supported catalysts, due to their unique geometric and electronic properties, and their highly efficient use of precious materials. Here, we report a single-atom catalyst, Cu/UiO-66, prepared by a covalent attachment of Cu atoms to the defect sites at the zirconium oxide clusters of the metal–organic framework (MOF) UiO-66. Kinetic measurements showed this catalyst to be highly active and stable under realistic reaction conditions for two important test reactions, the oxidation of CO at temperatures up to 350 °C, which makes this interesting for application in catalytic converters for cars, and for CO removal via selective oxidation of CO in H2-rich feed gases, where it shows an excellent selectivity of about 100% for CO oxidation. Time-resolved operando spectroscopy measurements indicate that the activity of the catalyst is associated with atomically dispersed, positively charged ionic Cu species. Density functional theory (DFT) calculations in combination with experimental data show that Cu binds to the MOF by OH/OH2 ligands capping the defect sites at the Zr oxide clusters.


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# These authors contributed equally
* Corresponding authors, emails: yaghi@berkeley.edu, juergen.behm@uni-ulm.de