Water Promoted Direct Methane Oxidation to Methanol on a CeO_2-Cu_2O catalyst

Scientific Achievement

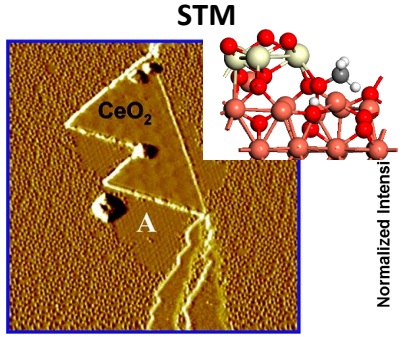
Utilized well defined CeO_x-Cu_2O model catalyst to enable direct methanol synthesis from CH_4 at 70% selectivity. Applied AP-XPS, Density Functional Theory (DFT) and Kinetic Monte Carlo (KMC) simulation to elucidate key mechanistic steps.

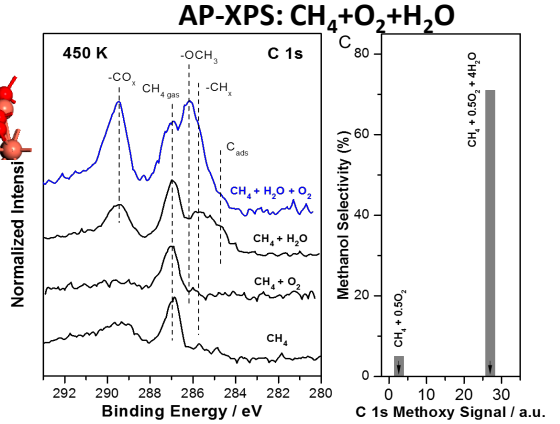
Significance and Impact

Converting C-H bonds in natural gas directly into liquid fuels like methanol is as yet unrealized. There is potential for transforming fuel production by conversion from gas to liquids. Our studies help to establish design principles in supported catalysts to use mixture of CH_4+H_2O+O_2 to close catalytic cycle for the synthesis of methanol. We explain the precise role of each reactant (O_2, H_2O and CH_4) over the surfaces of atomically precise using experiments and theory.

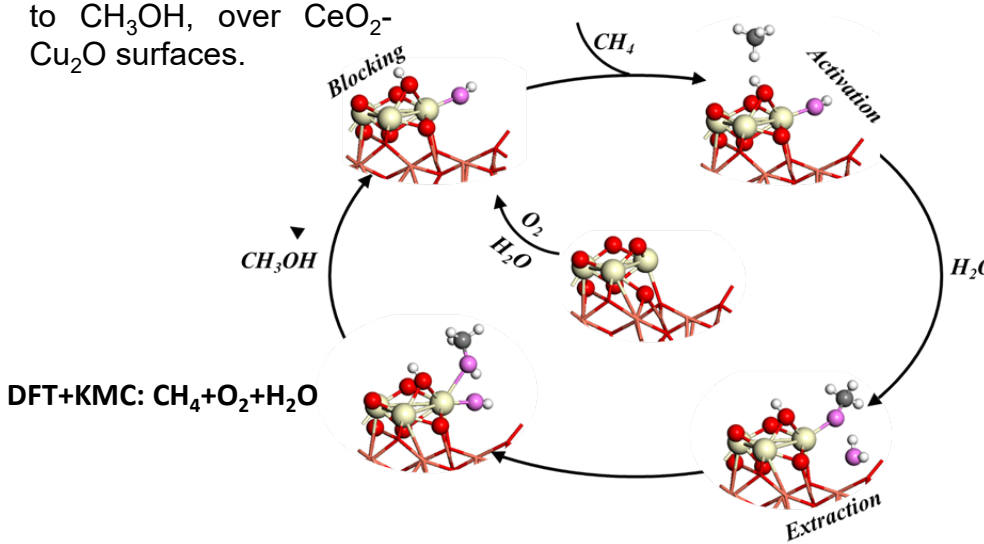
Research Details

Reaction between CH_4, O_2 and H_2O can produce methanol at yields of 70% selectivity, produced at 450K. In situ AP-XPS and DFT-KMC were used to search for complex mechanism. Water plays key triple role in enabling direct conversion: 1. to block sites preventing O_2 dissociation 2. provide O by forming OH at the interfacial sites and 3. extracting agent for driving methanol from surface.





DFT, reactor testing, and AP-XPS characterization to unravel steps that can selectively convert CH_4 to CH_3OH, over CeO_2-Cu_2O surfaces.



Z. Liu, E. Huang, I. Orozco, W. Liao, R. M. Palomino, N. Rui, T. Duchon, S. Nemsak, D. Grinter, M. Mahapatra, Ping Liu, J. A. Rodriguez, S. D. Senanayake, Science, 2020 Vol. 368, Issue 6490, pp. 513-517