



Liquid Fuel Synthesis of Methane along Carbon-Hydrogen Bond Activation

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INTRODUCTION

Direct conversion of methane to liquid C1-2 oxygenates is an interesting approach to sequestering carbon in transportable value-added chemicals while reducing global warming. Existing approaches using tandem steam methane reforming and conversion of methane to liquid fuels by Fischer-Tropsch synthesis are energy and capital intensive. Transition metal catalysts and stoichiometric catalysts using harsh oxidants and reaction conditions do not perform well with randomized product distributions. The development of catalysts that can cleave the C-H bond of methane to selectively produce useful chemicals under mild conditions is essential for commercialization. Single-atom catalysts significantly lower the activation energy due to the induced electrostatic polarization of the C-H bonds, facilitating reaction rate acceleration at low reaction temperatures. Strong metal-support interactions can stabilize intermediates and prevent peroxidation of reaction products. The chemistry of catalytic sites, the effects of metal support interactions, and the stabilization of intermediates on the catalyst to minimize peroxidation are detailed with a forward-looking perspective on improving catalytic performance.

DESCRIPTION

Simple but powerful monatomic catalysts with atomically dispersed active metal centers are of increasing interest in a variety of catalytic reactions. A specific example is the excellent performance in the chemical conversion of carbon dioxide. This report presents his recent efforts to develop single-atom catalysts for his carbon dioxide conversion in thermo catalysis and electro catalysis. The correct choice of support is critical for stability, activity, and selectivity in single-atom catalysis, as the support not only stabilizes but also interacts with a single active site. Three different strategies for single-atom catalyst ar-

chitecture (lattice-matched oxide supports, heteroatom-doped carbon anchors, and mimetic ligand chelates) are focused on support design of single-atom catalysts in different reaction environments. Such monatomic catalysts are very important for the conversion of carbon dioxide because the coordination environment helps to activate carbon dioxide. Due to the very high energy barrier to dissociation of the second C-O bond of carbon dioxide at the single-atom site, these catalysts can provide high selectivity towards CO or formate products.

The abundance of cheap natural gas is changing the energy landscape, opening new opportunities for sustainable chemical technologies and impacting people who rely on traditional fossil fuels. The main component, methane, is underutilized and wasted, causing anthropogenic global warming. Historically, the manipulation of methane 'clavis aurea' has been an insurmountable but challenging task and the subject of intense research, primarily due to the ability to selectively dissociate C-H bonds in methane. This requires large amounts of energy and is an essential requirement for the direct conversion of methane into various value-added products. The discovery of such a process promises the energy harvesting use of natural gas to benefit several important chemical processes related to C1 chemistry.

CONCLUSION

The step of C-H bond activation, once thought to be almost impossible, is now observed at low temperatures and can be observed in oxides, metal oxides, and highly prone to collections of metal-carbide systems. This is a step towards easy liquid fuel synthesis. Highlighting the latest in these results, active site configurations developed from surface science studies that enable such simple reaction events by careful manipulation of the last surface layer of atoms found in catalytic structures.

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