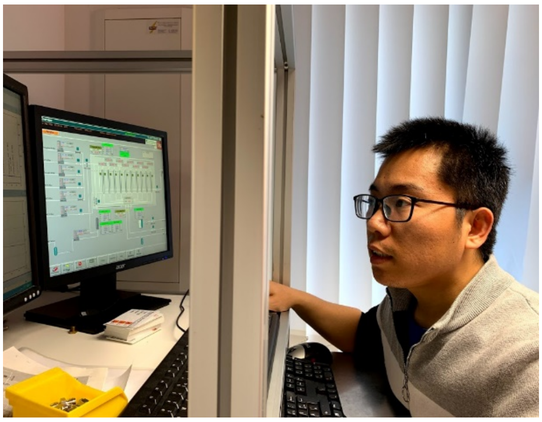
Perovskites for Alkane Selective Oxidation: Performance and Mechanism

Jinhu Dong



The oxidation of light alkanes is a promising energy-efficient process to high-valuable olefins and oxygenates. However, its application is limited by the poor selectivity resulting from the facile over-oxidation to stable CO_x species. Till now, various transition metal oxides catalysts (V, Mo) are proved to be effective to control the partial oxidation reaction.[1]

Perovskites with ABO_3 formula, consisting of network BO_6 octahedra surrounding with A-site cations, have been extensively studied in heterogeneous catalysis. Due to its structural and chemical versatility, perovskites could be an ideal catalyst matrix to describe the complex correlation between oxides structure and catalytic performance. But detailed understanding of oxidation reactions on perovskites is still missing.[2]

Perovskites $AMnO_3$ (A=La, Sm) with various A-deficient content have been applied in the oxidative dehydrogenation of propane in previous studies also in our group. Although these perovskites are well-known for total combustion, they all shows ultralow onset temperature (below 200 ^{o}C) and partial selectivity to propylene under these conditions. No oxygenates are formed except the total oxidation product CO_2. The aim of the present project is to change the product distribution through several modification methods, such as co-feed with additives, surface reconstruction, or bulk substitutions. By combination of kinetic experiments with advanced in-situ or operando characterization of these catalysts (Raman, XPS, IR, conductivity), we will systematically investigate surface structure, reaction intermediates and mechanism, to analyze the kinetic network on perovskites. The goal is to identify structural descriptors that are related to selectivity in hydrocarbon oxidation.

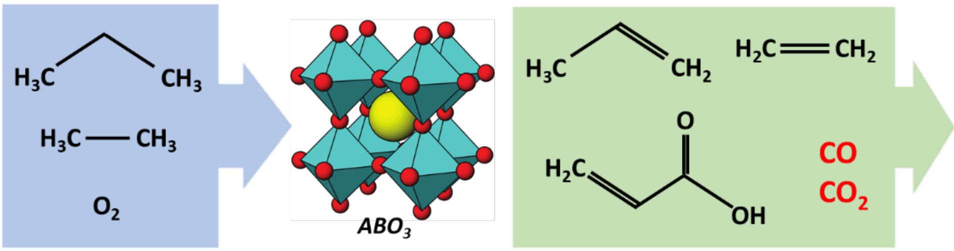


Figure 1. Selective oxidation of alkanes to olefins and oxygenate using perovskites.

[1]Carrero, C. A.; Schlögl, R.; Wachs, I. E.; Schomäcker, R., *ACS Catalysis* **2014,** *4*, 3357-3380.
[2]Royer, S.; Duprez, D.; Can, F.; Courtois, X.; Batiot-Dupeyrat, C.; Laassiri, S.; Alamdari, H., *Chemical Reviews* **2014,** *114*, 10292-10368.