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Study of water–gas shift (WGS) and methanation reaction on supported group VIIIB catalysts

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*Corresponding author (fabio@purdue.edu) Our research is directed to developing fundamental understanding of the water–gas shift (WGS) process for the production of hydrogen for fuel cell applications. Hydrogen (H_2) with low CO concentration is necessary for efficient and long–term performance of proton exchange membrane fuel cells (PEMFCs). The conventional process for hydrogen production involves fuel reforming followed by hydrogen enrichment using the WGS reaction. The industrial Cu–based low temperature WGS (LT–WGS) catalyst has an adequate rate per unit of volume but is unstable under frequent shutdown and startup situations. Thus, an alternative robust and active catalyst for WGS is an important challenge in generating hydrogen for fuel cell applications.

Our approach involves a systematic study to determine the rates, reaction orders and activation energies for WGS and methanation reaction on the various supported Pt group catalysts studied under realistic fuel processing conditions. Previous literature results show that Pt and Pd catalysts supported on various oxides are stable for WGS. We observed, as have several other groups, that the turnover rate (TOR) on a Pt catalyst was 40 times higher when supported on ceria (CeO_2) than on alumina (Al_2O_3). There are at least two issues on these catalysts: low rate of reaction and reaction selectivity. Because of the large concentration of H_2 present during WGS, hydrogen-consuming side reactions such as methanation need to be suppressed. For alumina supported catalysts, WGS rate per gram decreases as Pt >> Pd, Ni and methanation rate per gram decreases as Ni > Pd > Pt. We found that the addition of basic oxides to Pt–Al_2O_3 catalyst suppressed the methanation TOR by a factor of at least 5 and increased the WGS TOR by a factor of ~2. Similarly, addition of basic oxides to Pd–Al_2O_3 catalyst resulted into suppression of methanation TOR by at least a factor of 2 and increased the WGS TOR by at least 8 times. We also observed that addition of acidic oxide such as niobium (Nb) to Pd–Al_2O_3 increased the reaction rate for WGS by 4 times and for methanation by 7 times. Thus, group VIIIB metals show opposite trends in reaction rate per gram for WGS and methanation. Additions of basic oxides to Pt and Pd catalysts help suppress methanation while addition of acidic oxide increases the methanation TOR.