460d The Effects of Nickel Loading on a Ni/$Ce_{0.75}Zr_{0.25}O_2$ Autothermal Reforming Catalyst

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Hydrogen production by catalytic reforming of liquid hydrocarbon fuels continues to be an active area of research. Applications that might require on-demand hydrogen include automotive exhaust treatment, fuel cell based auxiliary power units, and fuel cell hydrogen propulsion systems. Autothermal reforming is a flexible reforming process that combines partial oxidation and steam reforming. Heat requirements are substantially lower than for simple steam reforming, while the hydrogen yield is higher than partial oxidation. Previous work in our group demonstrated that Ni/$Ce_{0.75}Zr_{0.25}O_2$ catalysts are active for autothermal reforming of iso-octane, a model compound for gasoline. The work presented here explores how catalyst properties and reforming activity are affected by nickel loading in the range of 1 to 15 wt%. The objective of this work is to optimize the Ni/$Ce_{0.75}Zr_{0.25}O_2$ catalyst formulation for hydrogen yield and durability against coking. Utilization of low steam to carbon ratios is of particular interest to mobile applications, but generally leads to carbon deposition and catalyst deactivation or reactor pressure drop problems. Achieving a balance between activity and durability is key to the successful development of mobile fuel processors fueled by liquid transportation fuels. Temperature programmed reduction of the catalysts reveals multiple reduction features, the size of which correlate to nickel loading. Chemisorption experiments using hydrogen and carbon monoxide give contradictory results. Carbon monoxide chemisorption indicates that nickel dispersion is highest at the lowest nickel loadings. For these low loadings, hydrogen chemisorption uptake is much lower than the CO uptake, indicating that hydrogen adsorption is suppressed. The $Ce_{0.75}Zr_{0.25}O_2$ support is active for hydrocarbon oxidation over the temperature range studied. The addition of nickel increases the iso-octane conversion and the hydrogen yield, while decreasing C_2 and C_3 products. At lower steam to carbon ratios, catalysts with high nickel loadings suffered from carbon deposition and deactivation. Low nickel loadings gave more stable behavior and less carbon deposition.