

460e Understanding the Change in Reaction Pathways during Catalyst Deactivation in Diesel Fuel Steam Reforming

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Use of fuel cell as a truck auxilliary power unit is being considered for the reduction in CO₂ emission levels, particularly while the vehicle is parked and the diesel engine would only be operated to generate electricity for housekeeping functions. Because diesel fuel is available, one can envision a commercially viable system in which an onboard diesel fuel reformer is used to generate the hydrogen required to run the fuel cell APU.

Experiments are performed in the fixed bed micro-reactor system available in our laboratory. Reactor feeds are preheated to a temperature of about 600°C in a pre-heater and then fed to a tubular reactor for further reaction. The reactor is maintained at temperatures between 600 – 900°C and atmospheric pressure, providing GHSV of the order of 20,000 hr⁻¹. Gaseous products pass into a condenser, where condensable materials are separated and collected for later analysis using a Shimadzu GC2010 instrument. Gas samples are sent to a second Shimadzu GC2010 for direct gas injection and analysis by a pulsed discharge ionization detector. Gas samples are also obtained using a Pfeiffer instruments residual gas analyzer.

In the present paper we are evaluating the ongoing reaction pathways occurring within the system, including cracking, steam reforming, water-gas shift, coke formation and methanation reactions. Hexadecane is used as a surrogate for diesel. It becomes vital to understand how these reactions proceed with time in order to evaluate the changing catalyst activity. Steam reforming reaction experiments were performed with and without the presence of catalyst. Some of these experimental results at steam to carbon of 3 depict that, in the absence of catalyst, a part of the organic feed is cracked in to smaller hydrocarbons such as methane, acetylene, ethylene, ethane, propane and coke with around 50% of the exit gas stream constantly being hydrogen. In the presence of catalyst, most of the organic feed is reformed to hydrogen providing high yields. As time progresses, catalyst activity decreases due to coke formation, leading to higher amount of unreformed hydrocarbons and thus decreasing yields of hydrogen. At steam to carbon of 10, without catalyst, the output is stable and no change in the exit gas composition is observed. Catalytic steam reforming at these conditions provides a nearly constant 95% yield of hydrogen, presumably due to complete reforming of the cracked products at the catalyst surface with lower amounts of coke deposition. This study implies that hexadecane steam reforming is not a single step process but a combination of initial cracking followed by steam reforming of cracked products.