343c Kinetics of Catalyst Deactivation in a Novel Diesel Steam Reforming System

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The recent discussion regarding the coming hydrogen economy has provided unique opportunities to investigate available technologies for the delivery of hydrogen. Current fuel distribution systems are based on hydrocarbon fuels, such as diesel and natural gas, not hydrogen. Therefore, wide spread use of fuel cells will require development of a hydrogen distribution infrastructure or an efficient process to produce hydrogen from hydrocarbon fuels. While hydrogen generation from existing resources, such as natural gas, gasoline and diesel fuel, is a viable alternative for the near-term, the presence of sulfur in the fuel leads to rapid deactivation of the reforming catalyst.

This paper describes our efforts to understand the performance of a Pd based monolith catalyst in a novel diesel steam reforming system. The unique design of the reformer allows the reforming reaction and combustion reaction on each side of the monolith foil. This design provides excellent heat transfer between reforming and combustion. The present study describes the effect of steam to carbon ratio (S/C) , temperature (T) and sulfur content (S) on the deactivation of the catalyst. The data is evaluated using first order deactivation and a first order kinetic model is

$$-\ln(1-X)=K_0\tau\exp(-k_d t)$$

A statistical analysis of the experiments has been completed to determine the combined effect of these three parameters on the rate of catalyst deactivation. The obtained model for the deactivation constant and rate constant are

$$k_d(hr^{-1})=0.0555+51.6T^{-1}[K]+0.000221(S)-0.0169(S/C)$$

$$k_0(min^{-1})=23935-30.2x10^6T^{-1}[K]-17.2(S)+1980(S/C)$$

Novel catalysts in which base metal oxides have been incorporated into the wash-coat have also been evaluated for deactivation, and shown to provide superior sulfur tolerance. A second statistically designed experiment has been used to evaluate the performance of the catalyst at various loadings of base metal oxide additive. The excellent energy integration of the reforming reaction has been evaluated in the novel diesel fuel reformer.