UNIVERSITY OF MICHIGAN

Department of Chemical Engineering Seminar Series

Tuesday, October 13, 2009
Presentation: 1:30 p.m., 1017 H.H. Dow
Refreshments to Follow: 3158 H.H. Dow (Pod Room)

Galen B. Fisher

Adjunct Professor, Chemical Engineering
University of Michigan – Ann Arbor, Michigan

On-Board Hydrogen Generation for Vehicle Emissions Control:

Spatially Resolved Species and Temperature Profiles Along a Reformer Catalyst

Emission standards for vehicles in the coming decade will require near-zero emissions in Europe and the US, particularly for nitrogen oxides (NOx). The more stringent standards coming for engines with both stoichiometric and lean exhaust will require novel systems. The advantages and disadvantages of the leading technologies for the near-term in lean exhaust will be briefly reviewed (e.g., NOx adsorbers, urea SCR, and HC-SCR). Tradeoffs that are made in meeting these pollutant standards and reducing regulated greenhouse gas emissions (CO_2, CH_4, N_2O) will be discussed.

One novel approach for emission reduction is to use hydrogen as a reductant in systems that control NOx and particulate emissions. Hydrogen can be generated by partial oxidation of various fuels on-board with compact reformers. These reformers turn on very fast (<5 sec in a video) and can be used to supply solid oxide fuel cells (SOFCs) that power auxiliary power units (APUs) in large trucks.

To better understand and model the chemistry in fuel reformers the first step was to measure the spatial profiles of temperature and species (reactants & products) within an operating Rh-based fuel reformer catalyst - even at their high peak operating temperatures (~1000 ºC). The axial profiles along a 25 mm monolith catalyst during methane and propane partial oxidation were measured with a resolution of 0.2 mm using quartz capillaries translated along the catalyst for species determination [i.e., Spatially Resolved Capillary Inlet Mass Spectrometry (SpaciMS)]; thermocouples and fiber-coupled pyrometry were used for temperature measurements. This approach can probe species and temperature distributions in a range of applications. Here we found that these methods also worked well, even at the extreme peak temperatures found in the channels of a reformer catalyst.

The next step was to provide a clear distinction between two competing models, direct and sequential, for hydrogen generation using the SpaciMS and temperature data. At the front portion of the monolith (0-5 mm), we found most of the hydrocarbon undergoes total oxidation consuming all of the oxygen available and producing CO_2, H_2O and a significant exotherm. Further downstream, the total oxidation products and the released heat produce H_2 and CO, mainly by steam reforming and the water-gas shift reaction. The data support a sequential reaction scheme where exothermic combustion reactions are followed by largely endothermic reactions generating hydrogen and carbon monoxide. Microkinetic modeling using Chemkin for both the surface and gas phase chemistry provides good agreement with the data. A general summary of our understanding will also include recent data showing the impact of various sulfur species on reformer output with a range of fuels.

The Chemical Engineering Seminar Series is sponsored in part by the Dow Chemical Company

MichiganEngineering