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Title: FUEL COMPOSITION EFFECTS ON FUEL PROCESSOR DYNAMICS

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Fuel Composition Effects on Fuel Processor Dynamics

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Introduction

This report describes our progress in examining fuel effects on hydrogen generation technology research for the specific purpose of hydrogen production for proton-exchange-membrane (PEM) fuel cells. The goal of this research is to explore the effects of various fuels, fuel constituents and fuel impurities on the performance of on-board hydrogen generation devices and consequently on the overall performance of a PEM fuel cell system utilizing a hydrocarbon fuel.

Fuels and fuel impurities have effects on fuel processor operation, efficiency, reformate gas composition, transient performance, lifetime, and durability. These effects are being investigated through experiments with various fuel processor hardware in Los Alamos National Laboratories' fuel processor test facility. The fuel effects ultimately determine the performance and operation of the fuel cell system from their effect on the anode feed stream composition.

In this report, we describe our progress of examining fuel composition effects on fuel processing, including the use of various fuel constituents, fuel impurities, reaction catalysts, fuel processor test reactors and reactor test systems.

Approach: Fuel Processor Test Facility

The fundamental tools for our research on the effects of fuels and fuel impurities on on-board hydrogen generation technology are LANL's fuel processor test facility and the reactor test beds in which the fuel effects are monitored. The fuel processor test facility provides the balance-of-plant framework, the necessary analytical instrumentation and the supervisory control to characterize operation and performance of different fuel processors and fuel components. This facility includes four different fuel processing reactor systems. The different reactors serve complementary roles in our laboratory, including integrated and modular fuel processing systems, heterogeneous and homogeneous oxidation, and adiabatic and isothermal test beds.

The Fuel-Flexible Fuel Processor (F^3P) from Hydrogen Burner Technology is an integrated fuel processor incorporating a partial oxidation reformer (auto-thermal reformer), a high-temperature shift reactor, a zinc oxide sulfur-removal bed, a low-temperature shift reactor, integral steam generator, and combustion-driven fuel vaporizer. This fuel processor is being used for studying the effects of fuels and fuel impurities on a thermally integrated system where the interactions between components are close-coupled. The fuel processor, shown in Figure 1, was received from Hydrogen Burner Technology. Initial operation of this system was conducted on vapor fuels, methane and natural gas, after which liquid fuel testing was initiated.

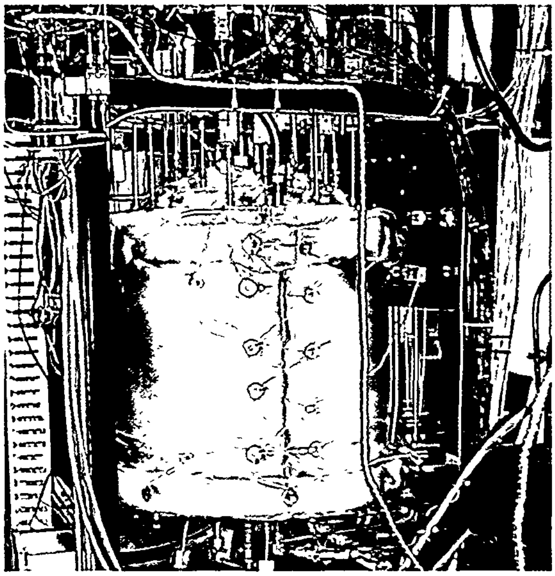

Figure 1: Hydrogen Burner Technology integrated Fuel Flexible Fuel Processor

An adiabatic reactor for use in evaluating fuel components and the fuel component tendency for carbon formation is shown in Figure 2. This reactor incorporates ease of catalyst replacement, *in situ* carbon formation monitoring, gas and temperature measurements with radial and axial profiles. Figure 4 is a schematic showing laser detection for *in situ* carbon formation. An argon ion laser will be used with extinction monitoring to observe the onset of carbon formation. Scattering will used to observe relative carbon particle size distributions, and spectral detection will allow for fluorescence detection of PAHs (poly-cyclic-aromatic-hydrocarbons).

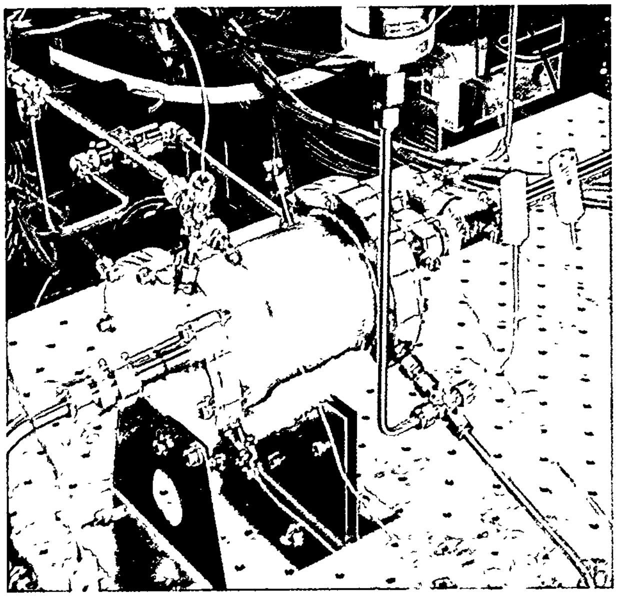

Figure 2: Adiabatic partial oxidation reactor

A micro-scale isothermal catalyst test bed for use in fuel and catalyst measurements under well defined and controlled conditions uses a furnace to keep the reactor under isothermal conditions. This reactor is constructed of a quartz tube with typical catalyst samples of 300 – 500 milligrams.

Results: Fuel Constituent Effects

Initial operation of the HBT reactor was conducted with vapor fuels, specifically natural gas and pure methane. The transition between natural gas and pure methane is shown in Figure 3. As the fuel to the fuel processor was changed from natural gas to methane, the reactor control temperature (not shown in Figure 3) decreased. The control software increased the relative O/C ratio, which resulted in a higher auto-thermal outlet temperature. The higher temperature resulted in a higher conversion of methane, thus lower methane composition out of the reactor. It appears this is the result of differences in heat of combustion of the fuels. Even though the natural gas composition included 97% methane, calculating the heat of combustion shows significant differences at low O/C. The stoichiometric combustion of methane and natural gas have heat of combustions of 212.8 and 213.9 kJ/mol – less than a 0.5 % difference, however the heat of combustion of the fuels at O/C = 1 is 35.65 and 37.43 which is over 5 % different. The natural gas composition was 97.067% methane, with small amounts of ethane (1.479%), propane (0.203%), butanes (0.0622%), pentanes (0.0197%), C_6 (0.0144%), N_2 (0.172%) and CO_2 (0.978%).

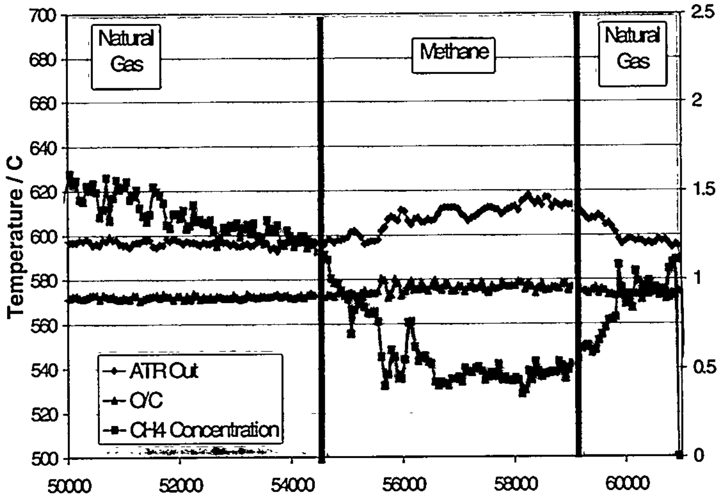

Figure 3: Operation of HBT reactor showing transition of operation between pure methane and natural gas.

Figure 4 shows hydrogen production from iso-octane and iso-octane doped with 100 ppm sulfur. This test was conducted with a nickel supported on alumina catalyst. The initial hydrogen production, shown in Figure 6 as ml/min, was similar for both fuel compositions. However, the iso-octane with 100 ppm sulfur within five hours saw reduced hydrogen production, which was the result of much lowered iso-octane conversion. The pure iso-octane fuel has consistent hydrogen production over the test period of 24 hours. The sulfur effect on this catalyst appears to have greatly inhibited both the oxidation and steam reforming kinetics, thus lowering the iso-octane conversion. This effect has been observed to be less severe on nickel catalysts supported on ceria.[1]

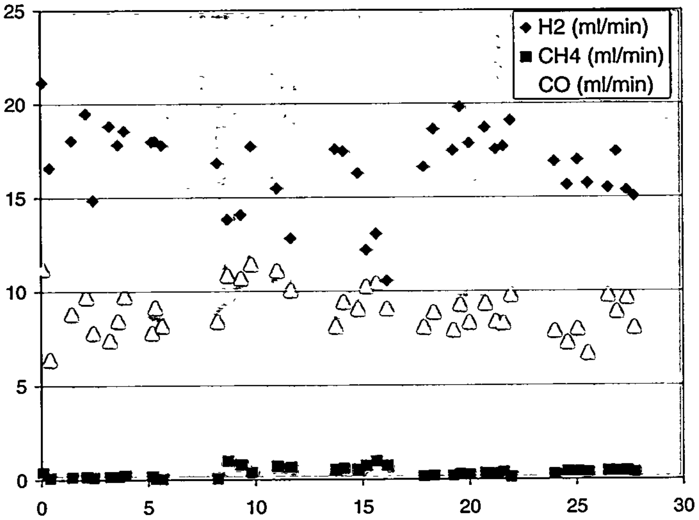

Figure 4: Hydrogen production from iso-octane and iso-octane with 100 ppm sulfur