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Autothermal reforming of JP8 on a Pt/Rh catalyst: Catalyst durability studies and effects of sulfur

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ABSTRACT

Autothermal reforming (ATR) of commercial grade JP8 was performed on a Pt/Rh catalyst deposited on a monolith. This study investigated catalyst performance under three test conditions: (i) 120 startup and shutdown cycles, (ii) 80 h of continuous operation with sulfur-free fuel, and (iii) 370 h of testing with JP8 containing 125 ppm of sulfur. Axial reactor temperature profiles and gas composition data showed that startup and shutdown cycling had no impact on catalyst performance. When durability testing was done with fuel containing 125 ppm of sulfur, the catalyst deactivated initially, which was reflected by a decrease in H_2 concentration and decrease in fuel conversion. However, after 250 h of operation the activity stabilized at 66% fuel conversion and product concentrations were constant for the remaining 120 h of testing. The presence of sulfur resulted in higher CO selectivity, lower H_2 concentrations, and lower fuel conversions compared to data with sulfur-free fuel. The data suggests that the presence of sulfur primarily affects steam reforming reactions, and CO oxidation. Regeneration was attempted with air and with fuel-lean combustion but initial H_2 yields and carbon selectivity were not achieved.

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1. Introduction

The production of hydrogen or synthesis gas on a distributed scale is being investigated for a variety of applications. Possible uses range from power production in fuel cells, combustion enhancements for higher power or emission reductions, or chemical synthesis. Synthesis gas can be obtained from the thermal or catalytic decomposition of a wide range of organic materials such as hydrocarbons, biomass, or waste. For the special case of distributed applications, a liquid fuel is most convenient due to the presence of a well-developed infrastructure for its delivery and its high energy density compared to other fuels. JP8 (Jet Propulsion 8) is a commonly used liquid fuel and is therefore widely distributed, making it a good candidate for portable power applications.

The main catalytic methods for generating synthesis gas from liquid fuels, such as JP8, are partial oxidation (POX), steam reforming (SR), and auto-thermal reforming (ATR). Partial oxidation is an exothermic reaction, where the oxygen (usually from air) to fuel ratio is sub-stoichiometric for combustion, and produces H_2/CO ratios dictated by the H/C ratio of the fuel itself. POX reactors produce very high temperatures and have some degree of complete oxidation which reduces the hydrogen yield [1]. SR can produce much higher H_2/CO ratios compared to POX since hydrogen is added to the fuel via water [2]. Additionally, the product stream has a higher energy density since N_2 dilution (from the addition of air) does not exist. However, steam reforming is an endothermic reaction that, combined with the evaporation of the steam, can have high energy requirements [2]. ATR combines POX and SR by introducing a mixture of oxidant, fuel, and steam into a catalytic reactor. This enables tuning of the H_2/CO ratio with a higher degree of freedom compared to POX and SR (by adjusting both steam to carbon and oxygen to carbon ratios) and eliminates the need for external heating. The overall process can be simplified by designating three types of reactions: oxidation reactions, water consuming reactions, and fuel cracking reactions. Oxidation reactions include partial and complete oxidation (see Eqs. (1) and (2)). Water consuming reactions include steam reforming of fuel and water gas shift (WGS) reaction (see Eqs. (3) and (4)). Cracking of the fuel has been shown to take place during ATR of large hydrocarbons [3,4]. The reactions shown here use $C_{11}H_{21}$ as a representative model for JP8.

$$C_{11}H_{21} + \frac{11}{2}O_2 \rightarrow 11CO_2 + \frac{21}{2}H_2 \quad \Delta H^\circ = -966.8\,\text{kJ/mol} \tag{1}$$

$$C_{11}H_{21} + \frac{65}{4}O_2 \rightarrow 11CO_2 + \frac{21}{2}H_2O \quad \Delta H^\circ = -6618\,\text{kJ/mol} \tag{2}$$

$$C_{11}H_{21} + 11H_2O \rightarrow 11CO + \frac{43}{2}H_2 \quad \Delta H^\circ = +1694\,\text{kJ/mol} \tag{3}$$

$$CO + H_2O \rightarrow CO_2 + H_2 \quad \Delta H^\circ = -41.2\,\text{kJ/mol} \tag{4}$$

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