



Research article

Coke suppression of kerosene by wall catalytic steam reforming

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ABSTRACT

A new method is developed to decrease coke formation during the thermal cracking of kerosene with applications in air-breathing jet engines. Endothermic catalytic steam reforming of kerosene is tested to investigate the potential for consuming excess heat and decreasing coke deposition rates. A plate reactor is used to simulate an engine-cooling channel and to compare the coke deposition with and without use of a reforming catalyst. The reactor temperature and the blending proportions of water fed to the reactor are varied across experiments. Spraying catalyst on the channel's inner surface prevents the production of filamentous coke, the production of which is catalyzed by the metal surface. The generation of a certain amount of hydrogen from the catalytic steam reforming of kerosene favors the reverse direction of the Diels–Alder and polymerization reactions, consuming the aromatic coke precursors and decreasing the coke formation rate. The effects of varying water content and reactor temperature on coke formation have been investigated during the thermal cracking of kerosene with catalysis. The coking rate decreases with an increase in water content. However, the suppression of coking become weaker as the fuel temperature is increased.

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1. Introduction

Rapid development of air-breathing aerospace engines has led aircraft speeds to increase from supersonic to hypersonic. However, hot air generated by the ram effect cannot cool the engine at these higher Mach numbers. Therefore, it is necessary for the fuel itself to be used as a coolant. The thermal cracking of hydrocarbon fuels is endothermic and can significantly contribute to the consumption of excess heat while also producing smaller hydrocarbon molecules that are beneficial in the subsequent combustion process. However, thermal cracking also generates solid products such as coke deposits, which can adversely affect heat transfer and thermal and combustion efficiencies, potentially shutting down the engine [1].

Coke formation is an inevitable side reaction in the thermal cracking of hydrocarbon fuels and was initially studied by researchers in the chemical industry. Albright et al. [2] summarized that three types of coke formation occur in the hydrocarbon fuels: filamentous coke can be formed from metal-catalyzed reactions; condensation coke can be formed from the polymerization and dehydrogenation of simple aromatics; and deposition coke can be formed from the reactions of small-molecular hydrocarbon with free radicals on the existing coke surface. Geem et al. [3] demonstrated that coke was deposited on alloy surfaces primarily via a metal-catalyzed pathway and a free-radical mechanism. Wickham et al. [4] found that condensation coke was the main product of Diels–Alder reactions between different dienes. Other

researchers highlighted that polyolefins and polycyclic aromatic hydrocarbons (PAHs) are precursors for coke formation in the thermal-cracking process [5,6].

Yang et al. [7] adhered Al_2O_3 powder to the surface of a metal tube to decrease the production of filamentous coke. An alumina passivating layer was deposited inside the channels of stainless steel reactors by Gong et al. [8] to deactivate the metal surface for the purpose of coke suppression. Trimm [9] found that adding sulfide to the surface of nickel-based alloy was very effective at suppressing coke formation. Dhuyvetter et al. [10] found that coke formation could be minimized in the process of naphtha steam cracking by continuous dimethyl disulfide (DMDS) addition into the feedstock. Wickham et al. [4] confirmed that 2, 4-diaminotoluene decreased coke deposition during n-heptane cracking by suppressing the Diels–Alder reactions of butadiene and ethylene. Emily et al. [11] found that some hydrogen donor molecules increased the high-temperature stabilization of jet fuels and similar hydrocarbon mixtures. Croswell et al. [12] showed that fuel-hydrogenation decreased the production of aromatic and alkene species during thermal cracking and limit coke deposition at high temperatures. However, although these works show that additives can decrease coke formation during thermal cracking, they cannot eliminate it, especially at surfaces that are exposed to high heat fluxes.

Attempts to decrease coking by employing endothermic catalytic reforming of hydrocarbon fuels to produce high levels of hydrogen production has recently attracted significant attention [13–16]. Korabelnikov et al. [17,18] and Kuranov et al. [19] studied the catalytic conversion and output characteristics of steam reforming based on methane, heptane, and decane. Those works indicate that coke deposition can be alleviated

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with the addition of steam-reforming catalyst in a thermal cracking regime. Hou et al. [20,21] reported on the benefits of endothermic catalytic steam reforming of aviation kerosene.

Despite the range of work that has demonstrated that the catalytic reforming reaction can significantly inhibit coke formation and decrease coke deposition, mechanistic explanations of these phenomena have remained uncertain. There is also little work published that details the factors influencing catalytic steam reforming coke formation during the thermal cracking of kerosene.

This work, therefore, experimentally investigates the effect of catalytic steam reforming on alleviating coke formation in the thermal cracking process of kerosene. A series of results reveal the variation of coking deposition rate and characteristics of gas and liquid products under different operating conditions and leads to an analysis of mechanisms for coke suppression.

2. Experimental system and methods

The catalytic reforming reaction takes place between hydrocarbon vapor and steam on the catalytic surface as follows:



Because this reaction is endothermic, it serves as a thermal protection measure against coke formation. The reaction also generates easily combustible gaseous species, such as hydrogen, carbon monoxide, and light hydrocarbons [21].

All of the experiments are carried out in the setup shown in Fig. 1. High purity nitrogen gas is used to remove residual fuel before and after every experiment. RP-3 Chinese kerosene (critical pressure and temperature of 2.5 MPa and 404 °C, respectively), which is almost entirely made up of cycloalkanes and paraffins, is used as the test fuel. Water and kerosene, together with 0.3% emulsifier, are fed into the fuel tank in advance and mixed to form an emulsion fuel using a gear pump and a static in-pipe mixer. The in-pipe mixer contains static mixer elements and intermediate chambers. The static mixer element contains an 180° twisted blade. Owing to the multi-dimensional movement (horizontal, tangential and radial), the mixing of the emulsified fuel is further improved. A metering pump then pressurized the fuel,

which is directed into a pre-heater and then the reactor. A Coriolis mass flow meter (capacity pressure 10 MPa; flow rate limit of 3 g/s; measurement error $\pm 0.2\%$) measures and controls the fuel flow rate.

The electrically heated reactor consisted of a stainless steel plate with a curving channel as shown in Fig. 2. The path through the reactor consists of five loops with a total length of 2865 mm and a rectangular cross-section of 4 mm \times 2 mm. A total of eleven thermocouples measures the fuel temperatures at the reactor inlet and outlet and the outer wall temperature along the reactor flow path. For the catalytic tests, a nickel-based steam reforming catalyst (Sinopec Qilu Company, PR China) is coated onto the inner wall of the reactor by plasma spraying, which is controlled to within 0.2 mm. Elemental nickel is added as an active component using calcium aluminate as the carrier using an impregnation method. The two sides of the reactor are covered with electric heating boards with heat fluxes simulated by gradually regulating the power output voltage. Most of the heat is absorbed by fuel, which promotes thermal cracking and catalytic reforming reactions.

After exiting the reactor, the reaction products are cooled in a condenser and sent to a gas-liquid separator. Gaseous products are analyzed by gas chromatography (GC SP-3430; Beijing Analysis Instrument Co. Ltd., PR China) while liquid products are collected and analyzed with a gas chromatography-mass spectrometer (GC-MS DSQ, Thermo Fisher Scientific Inc.). Identification of each compound is based on retention times and matching the mass spectrum recorded with those in the spectral library (NIST-11). The yield of each gaseous product is calculated on a per-unit-of-fuel basis as follows:

$$Y_i = ((\omega_i/100) \times V_i/m_{oil}) \times (1000/60), \quad (2)$$

where Y_i is the yield of gaseous component i (L/kg); ω_i is the content of gaseous component i as measured by GC; m_{oil} is the fuel mass flow rate (g/s); and V_i is the volumetric flow rate of all gaseous products (L/min).

The coke deposited on the inner surface of the reactor is analyzed quantitatively using a pressure difference transmitter (ROSEMONT 3051S, pressure drop limit of 60 kPa, error $< \pm 0.025\%$) based on Eqs. (3) and (4). The pressure drop under cold conditions is measured before and after hot experiments. Because under cold conditions the fluid flow is laminar, the friction coefficient is not influenced by the roughness of

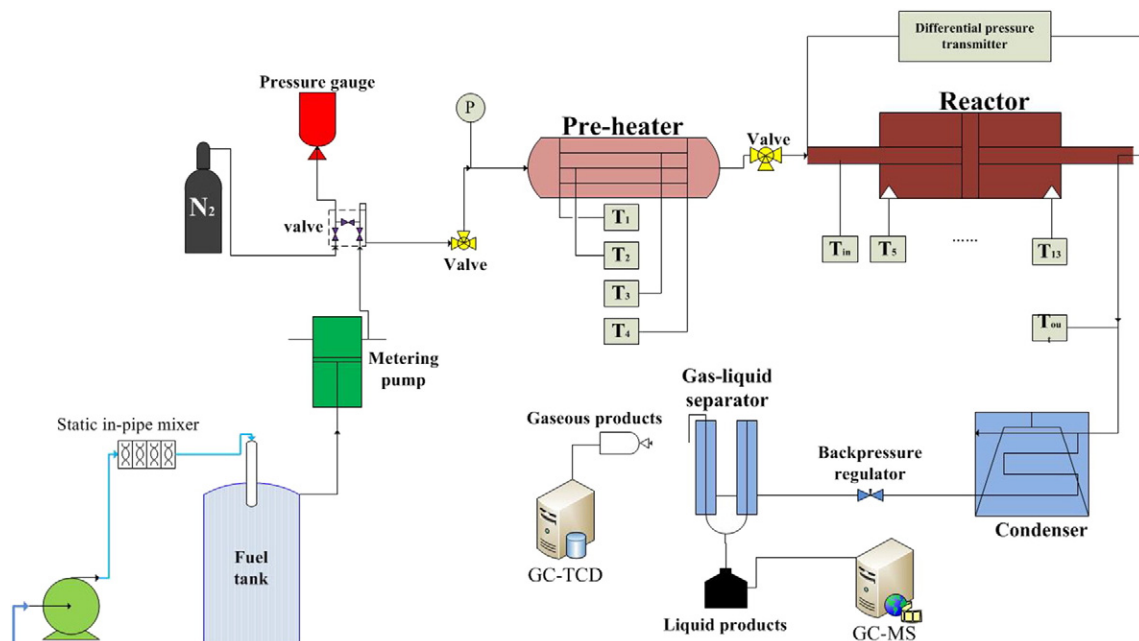


Fig. 1. Experimental setup.

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