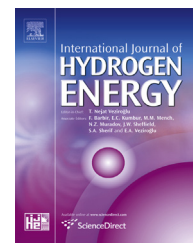




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Long-term stability at fuel processing of diesel and kerosene

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ABSTRACT

The long-term stability at autothermal reforming of diesel fuel and kerosene was studied using Juelich's autothermal reformer ATR 9.2, which is equipped with a commercial proprietary RhPt/Al₂O₃–CeO₂ catalyst. The experiment was run for 10,000 h of time on stream at constant reaction conditions with an O₂/C molar ratio of 0.47, a H₂O/C molar ratio of 1.9, and a gas hourly space velocity of 30,000 h⁻¹. Kerosene produced via the gas-to-liquid process and diesel fuel synthesized via the bio-to-liquid route were used. Both fuels were almost free of mass fractions of sulfur and aromatics. The trends for the desired main products of autothermal reforming H₂, CO, CO₂, and CH₄ were almost stable when kerosene was used. When the fuel mass flow was switched to diesel fuel however, different modes of catalyst deactivation occurred (active sites blocked by carbonaceous deposits, sintering processes), leading to a decrease in the concentrations of H₂ and CO₂ with a simultaneous increase in the CO content. This paper defines carbon conversion as the decisive criterion for evaluating the long-term stability during autothermal reforming of kerosene and diesel fuel. Carbon conversion was diminished via three different pathways during the long-term experiment. Undesired byproducts found in the gas phase leaving the reactor had the strongest impact on carbon conversion. These byproducts included ethene, propene, and benzene. Furthermore, a liquid oily residue was detected floating on the condensed unconverted mass flow of water. This happened once during the whole experiment. Finally, undesired organic byproducts were dissolved in the mass flow of unconverted water. These were found to be straight-chain and branched paraffins, esters, alcohols, acids, aldehydes, ketones, etc. Nevertheless, at the end of the long-term experiment, carbon conversion still amounted to more than 98.2%.

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Introduction

Fuel processing is the umbrella term for a variety of chemical processes aiming to provide a hydrogen-rich gas mixture to be fed into the anode of a fuel cell. The most important of these processes are desulfurization in the liquid or gas phase, reforming of liquid or gaseous fuels, water–gas-shift reaction, preferential oxidation, and catalytic combustion. At Juelich, all of these processes have been investigated. This paper concentrates on the autothermal reforming of synthetic liquid fuels, such as gas-to-liquid (GTL) kerosene and bio-to-liquid (BTL) diesel fuel, with a special emphasis on the experimental evaluation of the long-term stability of this process using Juelich's autothermal reformer ATR 9.2. In the literature, there are several papers dealing with the long-term stability of reforming processes.

O'Connell et al. [1] developed and evaluated a reactor based on micro-channel technology for the steam reforming of diesel fuel. They conducted an experiment for 38 h of time on stream at temperatures above 750 °C and different H₂O/C molar ratios. The hydrogen molar flow leaving the reactor was equivalent to an electrical power of 5 kW. Almost 100% conversion of diesel fuel was observed except for a slight deterioration after 35 h of time on stream. At this point, conversion decreased to 98.3%. In parallel, the concentrations of byproducts in the gas phase rose. For hydrocarbons with two C-atoms (C₂), they amounted to approx. 3000 ppmv, for those with three C-atoms (C₃) the concentrations were in the range of 2000 ppmv.

In their paper, Aicher et al. [2] presented results from experimental tests with their 20 kW_e autothermal diesel reformer. They used diesel fuel with a sulfur mass fraction of less than 10 ppm. The authors conducted a long-term experiment for 300 h of time on stream at a H₂O/C molar ratio of 1.5 and a λ-value of 0.37. The temperature in the reactor was kept above 800 °C at all times. The catalyst performance deteriorated slightly. The H₂ concentration (on a dry basis) decreased from approx. 35 vol.% to 32 vol.% within 300 h. Byproducts, such as ethene, ethane, propene, or propane, were not detected at the beginning of the experiment when the temperature was kept above 800 °C. In another paper, Lenz and Aicher [3] investigated the autothermal reforming of jet fuel and took a close look at the occurrence of byproducts in the gas phase leaving their reactor while varying the λ-value and the gas hourly space velocity. The run time of this experiment was less than 100 h of time on stream. They did not find a significant influence of the λ-value on the concentrations of ethane, ethene, propane, propene, butane, and pentane. The mole fraction of ethene, for example, was in the range of 500 ppmv, that of propene amounted to approx. 1000 ppmv. However, when the gas hourly space velocity was increased from 50,000 h⁻¹ to 300,000 h⁻¹, the concentrations of these byproducts increased remarkably. For ethene, for example, a rise from 500 ppmv to more than 6000 ppmv was observed.

Yoon et al. [4] described a kerosene fuel processor in their study, which was operated for 250 h of time on stream at an O₂/C molar ratio of 0.68, a H₂O/C molar ratio of 2, and a gas hourly space velocity of 12,500 h⁻¹. During this time span, the reforming efficiency decreased from 65% to 55%. The authors

explain that this degradation in reforming performance was caused by carbon deposition on the catalyst due to inhomogeneous mixing of reactants. In parallel, a strong increase in the concentration of ethene in particular was detected during the experiment. Yoon et al. [5] also reported about a diesel fuel processor with an autothermal reforming section for solid oxide fuel cell systems. This fuel processor was successfully run for about 2500 h of time on stream with a negligible degradation rate. Ethene, ethane, propene, and propane were detected in the product gas stream throughout the whole experiment. Their concentrations increased with time.

Pasel et al. [6] investigated the autothermal reforming of Jet A-1 using three different reformer generations in the 5 kW_e class at an O₂/C molar ratio of 0.47, a H₂O/C molar ratio of 1.9, and a gas hourly space velocity of 30,000 h⁻¹. ATR 5 was operated for 86 h with concentrations of benzene in the range of 200 ppmv and a value for total organic carbon (TOC) of 64.4 ppmw at the end of the experiment. Traces of C₃, C₄ and C₅ olefins were also detected. With ATR 7, even 500 h of time on stream were reached. No byproducts were found in the gas phase, and the TOC value was only 0.6 ppmv. The experiment was stopped after 500 h due to blocking of the nozzle for fuel injection. With ATR 8, the operation time was extended to 2000 h of time on stream. The TOC value was in the range of 100 ppmv and a liquid oily residue was detected floating on the condensed unconverted water. Porš et al. [7] studied the autothermal reforming of Ultimate Diesel fuel using two different reformer generations in the 5 kW_e class at an O₂/C molar ratio of 0.47, a H₂O/C molar ratio of 1.9, and a gas hourly space velocity of 30,000 h⁻¹. With ATR 7, a run time of 485 h of time on stream was set. No byproducts were found in the gas phase, and the TOC value was 2.0 ppmv at the end of the experiment. With ATR 8, the operation time was prolonged to 1000 h of time on stream. The TOC value was in the range of 130 ppmv. Byproducts in the gas phase were found to be propene, 1,3-butadiene, and benzene with concentrations at the end of the run of 25 ppmv, 13 ppmv, and 55 ppmv, respectively.

In their paper, Lindström et al. [8] introduced four reactor generations for diesel fuel reforming named M2 – M5. The best results were obtained with generation M5 operating at a λ-value of 0.29 and a H₂O/C molar ratio of 3.0. The total concentration of non-methane hydrocarbons (NMHC) in the product gas flow was lower than 10 ppm. The reported test time was always shorter than 10 h. Lindermeier et al. [9] investigated two different reformer concepts for the catalytic partial oxidation of diesel fuel at a λ-value of 0.40 and a H₂O/C molar ratio of 0. The testing time was in the range of only 5 h. With concept B, they observed H₂ and CO concentrations in the range of 21 vol.% to 22 vol.%. The amounts of unconverted hydrocarbons in the gas phase were low with values in the range of 0.1 g/m³. Rautanen et al. [10] reported about the autothermal reforming of Swedish Environmental Class 1 diesel fuel in combination with a solid oxide fuel cell stack. The test was run for 1200 h of time on stream. The reformer was able to achieve high conversion of diesel fuel with total concentrations of byproducts in the gas phase of only 7 ppm. Roychoudhury et al. [11] studied the performance of a fuel processor in the 2 kW_e power class for more than 1100 h of time on stream. The autothermal reforming reaction

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