



Elimination of by-products of autothermal diesel reforming



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HIGHLIGHTS

- Conventional road diesel fuel was utilized successfully for autothermal reforming.
- Injection of diesel fuel was improved to ensure complete evaporation.
- Neglectable concentrations of problematic by-products in the reformat where achieved.

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ABSTRACT

This paper outlines the design optimization and operation of the Forschungszentrum Jülich's ATR 13 autothermal reformer with conventional diesel fuels. The main objective was to eliminate undesirable by-products that can amass in the reformat. The reforming of conventional, highly aromatic diesel fuels with a high final boiling point presents a particular challenge – these fuels are more difficult to evaporate and are prone to coking; tendencies that can damage both the reformers themselves and downstream components. Here, we present several constructive and operational measures to generate a high-quality reformat from the Swedish Test Diesel SD 10 and, moreover, to prevent the formation of short-chain hydrocarbons and aromatic compounds in the reformat. This will enable the stable, long-term operation of fuel cell-based APUs utilizing conventional diesel fuels on board trucks.

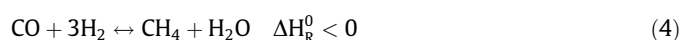
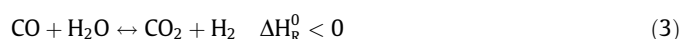
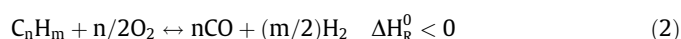
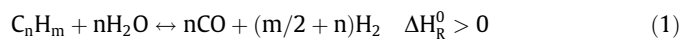
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1. Introduction

The electrical efficiency of conventional auxiliary power units (APUs) on board trucks when the engine is idling is usually below 11% [1–3] compared to an efficiency of 30%–40% in normal driving mode [1,2]. The exhaust emissions of diesel trucks are also considerably higher during idling than during normal operation. Brodrick et al. [1] measured up to twenty times more hydrocarbons, two-and-a-half times more nitrogen oxides and three times more carbon monoxide emitted during idling compared to normal engine operation. The use of biofuels instead of conventional fuels in diesel internal combustion engines (ICEs) is not expected to improve efficiency. They will hardly reduce the pollutants emitted by conventional systems and actually increase the emission of nitrogen oxides [4]. Fuel cell-based APUs are a promising alternative for on-board power supply in trucks during idling. With such a fuel cell-based fuel processing (FP) system, an electrical efficiency of 30%–40% can be achieved during idling and pollutant emissions

can be drastically reduced [5–7]. A fuel cell APU is even expected to be superior to a diesel ICE-based APU of a comparable power class in terms of both efficiency and cost [8]. Here, the hydrogen needed for the fuel cell is produced in the form of a hydrogen-rich gas mixture (reformat) from liquid hydrocarbons, such as the diesel fuel on board trucks, by means of reforming.

The basic reactions for this are endothermic steam reforming (HSR) (Eq. (1)), exothermic partial oxidation (POX) (Eq. (2)) and autothermal reforming (ATR) as a combination of POX and HSR in a reaction chamber. Other thermodynamically controlled reactions are the water-gas shift reaction (Eq. (3)) and methanation of carbon monoxide (Eq. (4)), which create the four main products: hydrogen, carbon dioxide, carbon monoxide and methane.



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Nomenclature

APU	auxiliary power unit	e	eccentricity of swirl ducts [mm]
ATR	autothermal reformer/autothermal reforming	GHSV	gas hourly space velocity [h^{-1}]
BTL	bio to liquid	L	length [mm]
CAB	catalytic burner	n	amount of substance [mol]
ESR	electro-slag remelting process	N	number of inlet channels to swirl chamber
FCGEN	Fuel Cell Based On-board Power Generation	P	power [W]
FP	fuel processing	X	conversion [%]
GC	gas chromatograph(ic)	Φ	convergence angle of swirl chamber [$^\circ$]
GTL	gas to liquid	ϑ	temperature [$^\circ\text{C}$]
HSR	heated steam reforming		
ICE	internal combustion engine		
PEFC	polymer electrolyte fuel cell		
POX	partial oxidation		
PrOx	preferential oxidation		
SEM	scanning electron microscopy		
TOC	total organic carbon		
WGS	water-gas shift reactor		

List of symbols

A	area [mm^2]
D	diameter [mm]

Indexes

DP	dew point
e	electrical
o	orifice
p	inlet port to swirl chamber
s	swirl chamber
th	thermal
V	volume related
W	weight related

The fuel cell stack of an APU must be supplied with a high-quality reformat containing almost no impurities, which might be caused by incomplete conversion of the fuel. Otherwise, the catalytic active sites in the anode of the fuel cell would be irreversibly poisoned and no longer accessible for the reactants hydrogen and oxygen. Incomplete conversion is often due to unfavorable operating conditions of the reformer [9] or a damaged catalyst [10,11]. Possible impurities include ethene, propene and benzene. These by-products are a precursor of carbon deposition [12–14] and have negative impacts on the activity of the autothermal reforming catalyst itself and downstream components such as the water-gas shift reactor (WGS) [15,16] and other purification stages [17], as well as on the fuel cell stack [18–23].

Specchia [24] provides an overview of a large number of fuel processing activities today in a review article. The article also highlights the relevance of the FP system for increasing the efficiency of the on-board power supply in trucks during idling.

Shekhawat et al. [25] investigated the influence that various compounds and their structures, as found in diesel fuels, have on reforming. The fuel components that were easiest to reform were n-alkanes, while aromatics caused poorer conversion, increased coke formation and an increase in the concentration of olefins (particularly ethene but also propene) in the product gas. The influence of different components on coke formation under the same test conditions was also described by Shamsi et al. [26]. The ATR tests were performed for $\text{O}_2/\text{C} = 0.30$ and $\text{H}_2\text{O}/\text{C} = 1.5$. The space velocity (GHSV) was $50,000 \text{ h}^{-1}$.

González et al. [27] present the results of ATR tests with catalysts made of RhPt on $\delta\text{-Al}_2\text{O}_3$ and $\text{CeO}_2\text{-ZrO}_2$. The tests were performed with various diesel fuels for molar ratios $\text{O}_2/\text{C} = 0.3\text{--}0.49$ and $\text{H}_2\text{O}/\text{C} = 2.5$ at a space velocity of $\text{GHSV} = 8000\text{--}13,000 \text{ h}^{-1}$. Using synthetic fuels and a conventional premium fuel, conversions of $X = 97\%$ and $X = 98.5\%$, respectively, were achieved. Using a standard diesel in accordance with DIN EN 590, the conversion was $X = 92\%$. By-products in the reformat are not described but based on the slightly lower conversions, a higher diesel slip (unconverted fuel components condensable at ambient conditions) and a large fraction of by-products in the reformat can be expected. In a post mortem characterization [28] of the catalysts,

the largest coke deposition in the catalyst was found shortly after the catalyst inlet. This directly follows the very fast POX reaction that occurs at the front of the catalyst. The reason for this was given as the very high temperature in the absence of oxygen.

Grote et al. [29] describe HSR investigations for thermal inputs of $2 \text{ kW}_{\text{th}}\text{--}6 \text{ kW}_{\text{th}}$. As a fuel, they used a diesel surrogate with $<2 \text{ ppm}_w$ sulfur and 20% aromatics. At a temperature of $650 \text{ }^\circ\text{C}$ leaving the catalyst and $\text{H}_2\text{O}/\text{C} = 4$, a fuel conversion of $X = 86.2\%$ was achieved. Under these conditions, several hundred ppm ethene and propene were measured in the product gas. Increasing the reaction temperature both increased the conversion and decreased the concentration of by-products. More recent publications [30,31] describe the integration with a polymer electrolyte membrane fuel cell (PEFC). In the reformer, the molar ration of $\text{H}_2\text{O}/\text{C}$ was at 5–6 during the experiments. With a first catalyst, the residual hydrocarbon concentration in the reformat increased during the experiment to 1000 ppm (C_3H_8 equivalent) and rapid fuel cell degradation was observed. With a second catalyst, the residual hydrocarbon concentration was maintained at below 100 ppm (C_3H_8 equivalent) and stack degradation was no longer observed. The duration of the experiment is not given, but approx. 6 h are shown. A test lasting approx. 20 h with a logistic diesel fuel resulted in a residual hydrocarbon concentration of up to 1800 ppm (C_3H_8 equivalent); rapid degradation of the PEFC was also observed here.

An et al. [32] and Kim et al. [33,34] investigated the importance of a proper mixture formation to ensure a high long-term reforming performance. It was highlighted that a low size of droplets in the fuel spray is an essential measure to achieve a good reforming performance and adequate suppression of impurities in the reformat.

The geometric characterization of a pressure swirl atomizers and spray characteristics has been published by numerous authors and is extensively documented in the literature. Lee et al. [35] investigated air core characteristic by visualizing diesel and kerosene sprays. Liu et al. [36] focused on the effect of geometric parameters on the spray cone angle and present an empirical formula to determine the SMD (Sauter mean diameter) of the spray. Durdina et al. [37] aim in their publication on pressure swirl

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