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A diesel fuel processor for fuel-cell-based auxiliary power unit applications

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HIGHLIGHTS

• Fuel processing with autothermal diesel and kerosene reforming for fuel cells.

- \bullet Integrated system design for auxiliary power units in the 5–10 kW_e class.
- Focus on water-gas shift reactor stability under operation with real reformate.
- Optimized and validated operation strategy to enable stable system operation.
- Maximum CO concentration of 1.5% at shift outlet under extreme conditions.

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ABSTRACT

Producing a hydrogen-rich gas from diesel fuel enables the efficient generation of electricity in a fuelcell-based auxiliary power unit. In recent years, significant progress has been achieved in diesel reforming. One issue encountered is the stable operation of water-gas shift reactors with real reformates. A new fuel processor is developed using a commercial shift catalyst. The system is operated using optimized start-up and shut-down strategies. Experiments with diesel and kerosene fuels show slight performance drops in the shift reactor during continuous operation for 100 h. CO concentrations much lower than the target value are achieved during system operation in auxiliary power unit mode at partial loads of up to 60%. The regeneration leads to full recovery of the shift activity. Finally, a new operation strategy is developed whereby the gas hourly space velocity of the shift stages is re-designed. This strategy is validated using different diesel and kerosene fuels, showing a maximum CO concentration of 1.5% at the fuel processor outlet under extreme conditions, which can be tolerated by a high-temperature PEFC. The proposed operation strategy solves the issue of strong performance drop in the shift reactor and makes this technology available for reducing emissions in the transportation sector.

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1. Introduction

Middle distillates such as diesel and kerosene currently represent a significant share of fuels for the transportation sector. Despite energy conversion systems becoming more efficient and international efforts to decarbonize the energy sector in general and transportation in particular, different forecasts continue to assign a decisive future role for diesel fuel and kerosene. The Annual Energy Outlook 2016 from EIA shows that in 2015, 90% of

* Corresponding author. E-mail address: r.c.samsun@fz-juelich.de (R.C. Samsun). large trucks (classes 3–8) in the US were operated with diesel fuel [1]. In their reference case, fuel consumption increases by a factor of 25.5% through 2050, with diesel fuel having a share of 85%. In their Phase 2 standards case, in which new regulations are considered that take effect in 2021, fuel consumption decreases slightly, by 1.8% by 2050, while diesel fuel still has a share of 88%. The World Oil Outlook 2015 from OPEC estimates a global increase in the share of diesel from 29.7% in 2014 to 32.0% in 2040, with product demand increasing from 27.1 mb/d to 35.1 mb/d [2]. A similar trend is also apparent for jet fuel, with its share increasing from 7.4% to 8.4% in the same time-frame, whereas the product demand increases more strongly, from 6.7 mb/d to 9.2 mb/d. The German Energy Agency







estimated a share of diesel fuel of 66-76% for all road traffic according to different scenarios for the year 2025. In this prognosis, the demand for liquid fuels for engines will remain over 95%. The alternatives (electricity, hydrogen and natural gas) account for less than 3% in their forecasts [3]. According to the Mobility and Fuels Strategy of the German Government, the alternative fuel options and/or innovative drive technologies will not be available in the short to medium term, or will only be available to a limited degree. with diesel remaining the dominant fuel [4]. Therefore, it can be assumed that diesel fuel for heavy-duty trucks and kerosene-type jet fuel for aircraft will continue to play an important role in the next few decades. One possibility to reduce fuel consumption and emissions in these means of transportation is to introduce alternative systems to avoid the idling operation of trucks during layovers and ground operation of gas turbine-based aircraft auxiliary power units. A fuel cell system that includes diesel or kerosene reforming utilizes the already available liquid fuel more effectively and covers the auxiliary power supply [5-16]. In this way, the liquid fuel can be fossil-based or renewable-based. In the latter case, the biomass-to-liquid (BtL) route using second generation biomass resources or the power-to-fuel (PtF) route using hydrogen produced from renewable sources via electrolysis and carbon dioxide from industry and accompanying fuel synthesis can be applied. In the future, the utilization of synthetic fuels from renewable sources will contribute to international attempts to reduce CO₂ emissions, while still exploiting the advantages of middle distillates in the transportation sector.

However, the reforming of middle distillates such as diesel and kerosene for fuel cell systems still remains a big challenge and recent publications have focused on this aspect [17-27]. Long-term stability and the full conversion of fuel are the main issues for diesel fuel processors. Recently, stability issues with the water-gas shift reactor during the operation of diesel fuel processors were reported by Samsun et al. [28,29]. A literature review of possible reasons for catalyst deactivation in the water-gas shift reactor is given by Krekel et al. [30]. During steady state operation and shutdown, the formation of carbonates or formates and an over-reduction of the washcoat are widely discussed as typical reasons for the loss of performance of the noble metal shift catalysts. Furthermore, the sintering of catalyst particles, washcoat particle growth and the loss of interaction between catalysts and washcoats are additional possible reasons for deactivation under steady-state operation [30]. Unfortunately, the operation of water-gas shift reactors with real reformates from logistic diesel fuel reforming for longer time periods has not yet been extensively discussed in the literature. In most papers, a synthetic reformate was used and the tests were only carried out for a limited period. Catalyst stability and activity, in combination with the operating strategy of the system, was investigated in detail by Krekel et al. [30,31]. These papers show that higher quantities of hydrocarbons in the reformate, e.g., 220 ppmv, do not have a negative effect on catalyst stability. In addition, the optimization of start-up and shut-down procedures improved fuel conversion in the reformer, which can have a positive effect on the stability of the shift catalyst during transients. Finally, an alternative commercial shift catalyst was identified that showed higher activity and stability under all operating conditions in comparison to the one used in previous work.

Utilizing the previous fuel processor generation developed at Jülich, a performance loss in the shift reactor was observed, which could only be partially recovered by adjusting the operational parameters. In Samsun et al. [29], the CO concentration at the shift reactor outlet was 5.1 (vol.)% at full load and 1.7 (vol.)% at partial load using Ultimate diesel as fuel, a premium diesel containing aromatics and having a high boiling end point (see Sec.2.2). Both levels are higher than the target CO concentration of 1.0 (vol.)% in

the wet reformate (1.25 (vol.)% in the dry reformate), which is set as the optimum concentration for the operation of a hightemperature PEFC stack at 160 °C in this work. This type of fuel cell can tolerate higher CO concentrations, up to 3 (vol.)% [32], however the higher CO concentrations lead to a stronger drop in cell voltage [33]. If the CO concentration is increased from 1 (vol.)% to 2.5 (vol.)% at 160 °C, the drop in cell voltage is doubled independent of current density [33].

In the present work, the next-generation diesel fuel processor in the 28 kW thermal power class with the new shift catalyst is presented and the system is operated using the new start-up/shutdown strategies that build upon the results of recent findings reported in Krekel et al. [30,31]. Considering the requirements of later application as an auxiliary power unit, the fuel processor system is characterized with a focus on the shift reactor, which represented the biggest issue in previous system generations.

2. Experimental methods and approach

In this section, the fuel processing system used in the experiments is introduced and information on the fuels utilized is provided. Finally, the analysis method for the reformate gas is explained.

2.1. Fuel processor system layout

The fuel processor was designed with the aim of producing a reformate gas from diesel and kerosene to feed a high-temperature PEFC. Thus, no CO fine cleaning is required in the system. The key components are an autothermal reformer, a water-gas shift reactor and a catalytic burner. Commercial noble metal catalysts coated on cylindrical monoliths are used in all components. The system layout is presented in Fig. 1.

The fuel processor is built using the heat integration strategy already validated in previous work [29,34]. According to this strategy, heat exchangers are integrated in the reformer and catalytic burner to recover process heat in order to produce superheated steam for the reforming process. At the design point, 70% of the reformer's water demand is injected onto the hot deflecting surface of the catalytic burner. Upon heat exchange with the hot off-gases produced by the catalytic combustion, the injected water completely evaporates. This stream is then mixed with the remaining 30% of the water demand. The saturated steam mixture is then evaporated and superheated in the reformer, as the heat is removed from the hot reformate gas. The new design of the tubes in the shell-and-tube heat exchanger integrated into the reformer

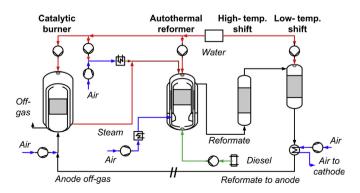


Fig. 1. Layout of the fourth generation fuel processor system for diesel and kerosene. The main components are an autothermal reformer, a water-gas shift reactor with high- and low-temperature stages and a catalytic burner. The layout includes the integrated steam preparation for reforming.

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