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## Further development of a microchannel steam reformer for diesel fuel

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### ABSTRACT

This paper presents results from the ongoing optimisation of a microchannel steam reformer for diesel fuel which is developed in the framework of the development of a PEM fuel cell system for vehicular applications. Four downscaled reformers with different catalytic coatings of precious metal were operated in order to identify the most favourable catalyst formulation. Diesel surrogate was processed at varying temperatures and steam to carbon ratios (S/C). The reformer performance was investigated considering hydrogen yield, reformat composition, fuel conversion, and deactivation from carbon formation. Complete fuel conversion is obtained with several catalysts. One catalyst in particular is less susceptible to carbon formation and shows a high selectivity.

To optimise the reformer geometry, a fifth downscaled reformer was implemented with an increased channel height. In operation the reformer shows advantages in performance and pressure losses in comparison to the original geometry.

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## 1. Introduction

Fuel cell systems comprising of PEM fuel cells and fuel processors have high potential in auxiliary power generation for stationary and mobile applications. By using a fuel with an existing infrastructure like diesel, the fuel cell technology can be established in the market before the installation of a hydrogen refuelling infrastructure. In addition, liquid fuels like diesel bring the advantages of high energy densities and easy storage which are key requirements for vehicular applications like auxiliary power units (APUs).

To allow the use of fuels with complex compositions like diesel, fuel processors are applied to generate a hydrogen-rich reformat gas in a reforming process. Three different approaches in reforming complex fuels are known from literature: catalytic partial oxidation (CPO) [1,2], autothermal reforming (ATR) or oxidative steam reforming [3–5], and

steam reforming (SR) [6–14]. In combination with a PEM fuel cell a fuel processor based on SR is particularly suitable in comparison to other reforming processes since the highest hydrogen yield is achieved. Furthermore, there is no dilution of the hydrogen-rich reformat with atmospheric nitrogen. Cutillo et al. [15] compared two diesel oil fuel processors (FP) based on SR and ATR in order to be coupled to a PEM fuel cell. They concluded that the steam reforming process is more suitable in terms of FP efficiency, APU efficiency, and hydrogen concentration fed to the fuel cell. However, the SR process scheme is more complex and water recovery is more difficult.

In steam reformer development two concepts have prevailed in the past: externally heated tubular reactors filled with catalytically coated pellets and catalytically coated heat exchangers that thermally couple the endothermic reforming reaction with a catalytic combustion. In the power range up to

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20 kW heat exchangers washcoated with highly active catalysts are favourable due to the enhanced heat transfer and the low-pressure drop. Gritsch [16] analysed different configurations of a folded-sheet reactor for methane steam reforming providing up to 15 kW hydrogen in the reformat stream. By implementing steam reformers as plate heat exchangers, the heat transfer can further be improved, in particular by reducing the channel height between the heat exchangers plates. When channel heights fall below 1 mm, the channels are classified as microchannels [16].

Steam reforming in microchannels was investigated under isothermal conditions in several publications in order to identify active catalyst formulations using diesel surrogates [7,11,12] and domestic heating oil [7,8]. A catalyst deactivation study and a catalyst screening in microchannels was presented by Maximini et al. [8], studying steam reforming of sulphur containing domestic heating oil. Microchannel steam reformers for diesel fuel as part of a diesel fuel processor were realised as catalytically coated plate heat exchanger by some research groups [6,7,11,13]. A diesel steam reformer with S-shaped microchannels was presented by Thormann [11]. Wruck [7] reports from the operation of a steam reformer with a thermal power feed up to 10 kW<sub>th</sub> using diesel surrogate. Engelhardt et al. [9] reported from the 500-h operation of the same reformer design processing 6 kW<sub>th</sub> domestic heating oil. In further investigations the reformer was operated with diesel surrogate and was coupled to a LT PEM fuel cell [9,21]. Kolb et al. [3] and O'Connell et al. [22] developed a microstructured diesel steam reformer based on plate heat exchanger. Small amounts of air were introduced into the steam reforming process in order to avoid carbon formation. The steam reforming process in the presence of air at O/C ratios less than 0.7 is called oxidative steam reforming [22].

### 1.1. Objective

In order to combine a diesel steam reformer with a PEM fuel cell, the objective of steam reformer development is to obtain the maximum hydrogen (H<sub>2</sub>) yield while the concentrations of carbon monoxide (CO), methane (CH<sub>4</sub>), and unconverted hydrocarbons should be minimised. Therefore, one main task in the ongoing steam reformer development remains finding an active and stable catalyst that is not susceptible to deactivation from carbon/coke formation. The catalyst deactivation is influenced by the catalyst formulation, the operating conditions and the heat transfer inside the reformer.

Another objective in the optimisation of a microchannel steam reformer is to minimise pressure losses without impairing performance or heat transfer. Thus, the parasitic power consumption of the balance-of-plant equipment can be reduced resulting in higher fuel cell system efficiency.

This paper presents results from the ongoing experimental work on the optimisation of a microchannel steam reformer for diesel fuel. In the following section the principles of diesel steam reforming are described in addition to the materials used for the experiments. Section 3 describes the experimental setup for the investigations. In Section 4, results of the reformer catalyst testing are presented and discussed.

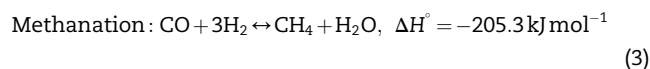
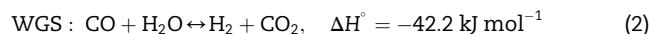
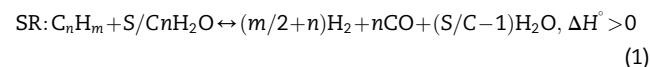
Additionally, results are presented of a steam reformer with increased microchannels. Section 5 discusses the results found and concludes for further action.

## 2. Materials and methods

### 2.1. Diesel steam reforming

Diesel steam reforming is challenging in comparison to the reforming of pure substances since diesel fuel is a complex composition of some hundred hydrocarbon species, some few organic compounds of sulphur, and additives. The hydrocarbon species mainly comprise of paraffins, naphthenes, and aromatic compounds, the latter may comprise of 20 vol. % of the mixture. The carbon number of diesel may vary as it depends on the distribution of the hydrocarbon species [17]. For the presented work a diesel surrogate was obtained from the mixture of Shellsol A 100 (20 wt. %) and Shellsol D 100 (80 wt. %). For this surrogate the chemical formula C<sub>14.41</sub>H<sub>27.08</sub> was calculated from the carbon and hydrogen mass fractions. The hydrocarbon fractions of the mixture obtained and the resulting sulphur content are summarised in Table 1.

The general chemical reaction for steam reforming (SR) of hydrocarbons is given by Eq. (1) where S/C is the steam to carbon ratio. It has to be taken into consideration that during steam reforming numerous side reactions occur, the main of these are listed below: water-gas-shift reaction (Eq. (2)), methanation (Eq. (3)) and carbon forming reactions (Eqs. (4)–(7)). As a result the product of steam reforming contains CO<sub>2</sub>, CH<sub>4</sub>, C and further species. Carbon, however, can lead to the deactivation of the catalyst by blocking the active centers of the catalytic coating and thereby can constrain desirable reactions. Furthermore, carbon can cause physical disintegration of the catalyst support pores and can promote further undesirable side reactions. Carbon formation can be avoided when the reformer is operated outside the thermodynamic carbon formation region [16]. It was also observed that high S/C decrease carbon formation [6,7,11].



**Table 1 – Diesel surrogate composition [6].**

Fraction	Value	Method of analysis
Paraffins	48 wt. %	GC
Naphthenes	32 wt. %	GC
Aromatics	20 wt. %	SMS 2728
Benzenes	5 wt. ppm	GC
Sulphur	<2 wt. ppm	SMS 1897

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