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Catalytic burner with internal steam generation for a fuel-cell-based auxiliary power unit for middle distillates

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ABSTRACT

A catalytic burner (CAB) was developed, which utilizes the anode off-gas of a high temperature polymer electrolyte fuel cell (HT-PEFC). This CAB has two functions within the HT-PEFC-system: It has to convert completely all combustible components including methane and carbon monoxide, even in the low ppm range and it has to provide steam to the autothermal reformer (ATR). Thereby it increases the system's overall efficiency.

Using computational fluid dynamics and experiments with a simple glass model, two catalytic burners (CAB 2 and CAB 3) were designed and constructed for a high temperature PEFC system with thermal powers of 18 kW and 28 kW, respectively. The burners were characterized experimentally in detail. Close attention was given to the steam generation capacity and the thermal behavior.

The constructed burners allowed complete conversion of low calorific fuel gases and a reformate in part load of the ATR was burned reliably as well. Superheated steam was generated free of oscillation. Experimental findings with CAB 2 resulted in an improved reactor generation with a reduced specific weight and geometric changes.

1. Introduction

At the Institute of Energy and Climate Research – Electrochemical Process Engineering (IEK-3), a fuel cell system for use in an auxiliary power unit (APU) is currently being developed by the Fuel Processing and Systems group [1,2].

Middle distillates, such as diesel or kerosene, are converted with air and superheated steam into a hydrogen-rich gas mixture [3,4]. Depending on the gas quality required by the fuel cell, the gas mixture is cleaned. The hydrogen-rich reformate is then fed to the anode of a fuel cell to generate electric power.

The fuel cell's anode off-gas is a low-calorific fuel gas with hydrogen as the component that contributes most to the calorific value. The actual composition of the anode off-gas depends on the operating conditions of the APU, the fuel used and the type of fuel cell. Jülich's APU concepts use diesel or kerosene Jet A-1 as a fuel and a PEFC or HT-PEFC as the fuel cell type. The main differences in terms of the composition of the anode off-gas are caused by the different CO tolerances of these fuel cell types: approx. 100 ppm for a PEFC and up to 3 vol.% for an HT-PEFC. A typical value for HT-PEFCs is around 1 vol. % [5,6]. The relatively low CH_4 content is not that interesting from a thermal point of view, but from a pollution

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point of view it should not be neglected. Methane has a GWP_{100} of 25, which means it is important to remove it from the off-gas [[7], pp. 213].

Several authors have looked at the catalytic combustion of low-calorific synthesis gases with different compositions. The objectives are to curb the environmental impact of any hydrocarbons, NOx and CO that have not been fully converted and to thermally exploit the gas mixtures [8–17].

The chemically bound energy contained in the fuel cell's anode off-gas is used thermally in the catalytic burner. Superheated steam, which is required for the operation of the autothermal reformer in the APU system, is produced using an integrated evaporator in the burner. If there is no evaporator, then the steam must be produced in another way, e.g. using an electrically operated heating system, which reduces the overall efficiency of the fuel cell APU.

Lee et al. [18] designed a catalytic burner for a stationary molten carbonate fuel cell (MCFC) which uses methane. The burner is operated at a space velocity of GHSV = 24,000 h^{-1} and is used to recycle the anode off-gas for the cathode reaction. No other use of the thermal energy generated in the burner is described.

Sarioglan et al. [19] constructed a 5 kW_{th} catalytic burner for a PEM fuel cell as an after-treatment system. They used a precious-metal-based catalyst on a monolithic structure with 600 cpsi. Mixtures of natural gas and hydrogen were used as fuel at space velocities of GHSV = 22,000 h^{-1}–101,500 h^{-1} and air ratios from λ = 3.65–17.4. The gas composition used as fuel for the burners differed significantly from a real PEFC anode off-gas. A utilization of thermal energy was not described.

An integrated steam reformer/burner module for a PEFC-based APU, fueled with propane, was described by Dokupil et al. [20]. The nominal power of the fuel processor was 1 $kW_{th}(H_2)$. In the steady state, the burner was fed with a synthetic anode off-gas and propane at lambda values λ = 1.35–1.43 to heat the reformer and maintain the steam reforming process.

Gardemann et al. [21] described a 3 kW_{th} non-catalytic burner for a fuel-cell-based residential combined heat and power supply system (CHP) to achieve a low pollutant emission. In the steady-state, a mixture of the anode off-gas and propane was utilized, generating heat for a propane steam reformer. For system start-up, propane alone was combusted.

2. Experimental

2.1. Reactor development

First experiences with catalytic combustion were gathered in Jülich with a catalytic radiant burner for the oxidation of methanol (CH_3OH) and the utilization of heat for the evaporation of methanol [22]. At a later date, the first experiments were performed with the CAB 1 catalytic burner, which was mechanically and thermally coupled to the ATR 5 autothermal reformer [23]. At a space velocity of 37,500 h^{-1} and an air ratio of λ = 1.3, the hydrogen in a PEFC anode off-gas was fully converted. Not all of the methane was fully converted. The off-gas temperature was 350 °C–410 °C.

Fig. 1 shows a simplified flow chart of the Jülich APU concept. It can be seen that a compact set-up of this type of overall system is possible. The autothermal reformer (ATR) is fed with a liquid hydrocarbon. Depending on the sector in which the system is used, this fuel is either diesel (in the automobile sector) or kerosene Jet A1 (in the civil aviation sector). The fuel is converted together with air and superheated steam to a hydrogen-rich gas mixture. Gas treatment involves different reactors depending on the type of fuel cell and fuel. However, a two-stage water–gas shift reactor (WGS) is planned in all cases. The CO concentration in the fuel gas is reduced to around 0.5 mol.% – 1.0 mol.% by a reaction between steam and CO forming CO_2 and hydrogen. Depending on the expected sulfur content in the fuel, desulfurization may be required. In the case of a polymer electrolyte membrane fuel cell (PEFC), CO ultrapurification is necessary. However, if a high-temperature PEFC (HT-PEFC) is used, there is no need for CO ultrapurification.

The fuel produced in this manner is fed to the anode of the fuel cell and most of the hydrogen contained in it is converted. The anode off-gas is a low-calorific gas, whose calorific value can be used thermally. One of the functions of the catalytic burner is to convert hydrogen (H_2), carbon monoxide (CO) and methane (CH_4) to carbon dioxide (CO_2) and water (H_2O). The combustible components must be fully converted in the burner before they are released into the atmosphere. Methane must be converted to protect the climate, carbon monoxide is toxic and hydrogen has a high calorific value and a broad explosion limit. The thermal energy produced by the catalytic burner is also used to generate steam for the reforming process and ultimately to improve the overall efficiency of the fuel cell APU. An important requirement is that superheated steam can be produced for the process continuously with no pressure surges in a mobile APU system without the use of a carrier gas such as air. Differences and similarities for two

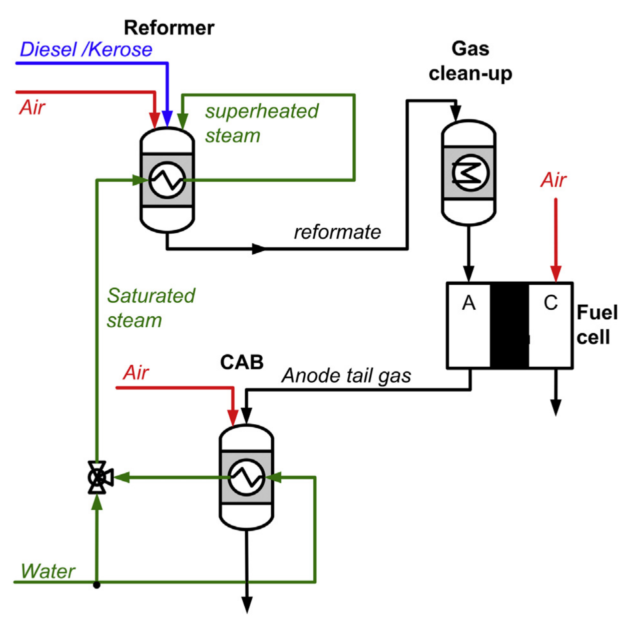

Fig. 1 – Simplified flow sheet of an APU based on a (HT)-PEFC system.