Short communication

Fuel processors for fuel cell APU applications

T. Aicher [a,*], B. Lenz [a], F. Gschnell [a], U. Groos [a],
F. Federici [b], L. Caprile [b], L. Parodi [b]

[a] *Fraunhofer ISE, Heidenhofstr. 2, D-79110 Freiburg, Germany*
[b] *Ansaldo Fuel Cells S.p.A., I-16152 Genova, Italy*

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Abstract

The conversion of liquid hydrocarbons to a hydrogen rich product gas is a central process step in fuel processors for auxiliary power units (APUs) for vehicles of all kinds. The selection of the reforming process depends on the fuel and the type of the fuel cell.

For vehicle power trains, liquid hydrocarbons like gasoline, kerosene, and diesel are utilized and, therefore, they will also be the fuel for the respective APU systems.

The fuel cells commonly envisioned for mobile APU applications are molten carbonate fuel cells (MCFC), solid oxide fuel cells (SOFC), and proton exchange membrane fuel cells (PEMFC). Since high-temperature fuel cells, e.g. MCFCs or SOFCs, can be supplied with a feed gas that contains carbon monoxide (CO) their fuel processor does not require reactors for CO reduction and removal. For PEMFCs on the other hand, CO concentrations in the feed gas must not exceed 50 ppm, better 20 ppm, which requires additional reactors downstream of the reforming reactor.

This paper gives an overview of the current state of the fuel processor development for APU applications and APU system developments. Furthermore, it will present the latest developments at Fraunhofer ISE regarding fuel processors for high-temperature fuel cell APU systems on board of ships and aircrafts.
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1. Introduction

Fuel cell based auxiliary power units (FC APU) are a promising alternative to conventional motor/generator based technologies. FC APUs promise slightly higher efficiencies with significantly reduced emissions and lower noise generation. The system efficiency remains almost constant even at turn-down operation down to less than 50% of full capacity. Another interesting feature of fuel cell APUs is that the water produced in the fuel cell can be used in other parts of the system, e.g. on board of an airplane.

Several fuel cell technologies are appropriate for use in APU systems: low temperature proton exchange membrane fuel cells (PEMFC) and high-temperature fuel cells, e.g. molten carbonate fuel cells (MCFC) and solid oxide fuel cells (SOFC). Since high-temperature fuel cells can tolerate carbon monoxide (CO) in the feed gas, the reformer reactor product gas can be supplied directly to the fuel cell. In PEM fuel cells on the other hand, carbon monoxide poisons the catalysts in the electrode of the anode, and therefore, the CO content in the reformer reactor product gas needs to be reduced prior to entering the fuel cell. This gas treatment comprises a shift reactor and a final CO removal step, by preferential oxidation, preferential methanation, or metal membranes.

Most vehicles use liquid hydrocarbons like gasoline, kerosene, or diesel to run their engines. Therefore, it is apparent that the fuel for the APU is the same fuel in order to keep fuel logistics simple. However, one class of compounds present in liquid hydrocarbons needs to been taken into account: organic sulfur compounds. They pose a major problem for all catalysts in the system, i.e. the catalysts used for reforming, shift and preferential oxidation/preferential methanation/membrane and the catalyst in the fuel cell. Most suppliers for fuel cells (PEMFC, MCFC, and SOFC) recommend total sulfur to stay well below 1 ppmw in the feed gas. This requires sulfur concentrations in the hydrocarbon feed to the reformer to be in the range of 1–10 ppmw.

The removal of sulfur from liquid hydrocarbons down to the aforementioned levels is not trivial and requires severe process conditions, especially for desulfurization of diesel. With vehicles

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traveling throughout the world, e.g. ships and airplanes, it has to be guaranteed for every refuelling that the sulfur level of the fuel is below 1–10 ppmw. This brings up a difficult problem, which requires tremendous efforts to cope with. Here, national and international politics is demanded to even out the way for international use of FC APUs.

For large vehicles, e.g. ships, the on-board desulfurization seems feasible. However, already with airplanes it does not appear to be feasible to remove sulfur in-flight, let alone trucks and cars. Here, the favorable solution is the removal of sulfur in the refineries. In fact, sulfur levels in fuels used for transportation in Europe have decreased steadily over the last decades.

Set aside the sulfur "thread", this paper presents a brief overview over activities by industrial researchers in the field of fuel processors and APU development for mobile applications. Finally, two FC APU prototype systems will be presented in detail, for which Fraunhofer ISE has developed fuel processors: one diesel and one kerosene autothermal reformer. The latest developments of gasoline powered reforming/fuel systems for APU applications have been reported previously [1] and will not be mentioned in this paper.

2. Overview over development of FC APUs

There are many R&D groups in industry, research organizations, and universities working world wide on the development of fuel processors and entire fuel cell APU systems for various mobile applications. In this short overview only recent efforts are summarized. It is not meant to be a comprehensive review of this topic by any means.

2.1. Reforming systems

For a general review of the various reforming processes used in fuel cell systems the extensive work by Larminie and Dicks can be referred to [2]. For high-temperature fuel cells pre-reforming followed by internal reforming in the fuel cell is possible. Some authors also report reformer product gas treatment systems which can comprise water gas shift reactors and CO removal units, e.g. preferential oxidation reactors, methanation reactors or palladium-alloy membranes for PEM FC systems.

Piwetz et al. [3] of Haldor–Topsoe describe a project that demonstrated the use of marine diesel fuel for application in a MCFC system. Their process comprises a one-stage hydrotreater unit (HDS) and a downstream zinc oxide bed to absorb the hydrogen sulfide generated in the HDS. The diesel contained about 0.2 wt.% sulfur components. The pre-treated diesel feed is then supplied to an adiabatic pre-reforming reactor. The HDS was carried out at 45 000 hPa and 380 °C, the pre-reforming at 25 000 hPa and a temperature of 480 °C. Both, the HDS reactor and the pre-reforming reactor used proprietary catalysts. A pilot plant that generated fuel gas for a 32 kW(el) fuel cell system was built and operated. The pilot plant ran for about 2200 h. Minimal catalyst poisoning was observed.

Steinfeld et al. [4] describe the development of a 625 kW(el) power plant for marine applications based on fuel cell energy's

direct carbonate fuel cell technology. The system comprises a high-pressure HDS unit to treat 1 wt.% sulfur diesel fuel. The hydrogen sulfide generated in the HDS reactor is absorbed in a zinc oxide bed. The pre-treated diesel fuel is fed to an adiabatic pre-reformer where it is reacted with steam to a methane rich product gas. This gas is then expanded through a turbo generator, reheated and directed to the anodes in the fuel cell. The turbo expander generates 50 kW(el) of AC power. The diesel pre-treatment section of the unit has been tested in a lab-scale plant. The 625 kW(el) power plant was tested in 2003.

External reforming generates a synthesis gas, which in case of high-temperature fuel cells can be delivered directly to the fuel cell. There are some manufactures, however, that prefer a methane rich gas over a hydrogen rich gas, because of easier control of temperatures inside the SOFC. With PEMFC external reforming with downstream CO shift and CO removal is a must – as described above.

Scoles and Perna [5] developed and built a naval distillate fuel autothermal reformer that runs on marine diesel. The catalyst used could tolerate sulfur in the feed up to 1 wt.%. The unit generates hydrogen for a 20 kW(el) Ballard Power Systems PEM fuel cell and is a precursor to a large 500 kW(el) demonstration plant. The sulfur in the feed is reduced in the reformer to hydrogen disulfide and removed downstream of the reactor by cycling regenerable sorbent beds and a polishing sulfur sorbent bed. The sulfur content in the product gas achieved by this method is 1 ppmv. The relative system size was 571 kW^{-1}, the system weight 18 kg kW^{-1}.

Pereira et al. [6] describe an autothermal reforming process for various diesel types containing up to 1 wt.% of diesel. Their paper addresses critical issues of the autothermal reforming of diesel fuels, like degradation of catalysts, impact of sulfur in the feed and coke formation. High temperatures improve conversion, resulting in a better system efficiency, but reduce material stability. They do not report the impact that sulfur has on catalyst life and catalyst degradation.

Krumpelt et al. [7] investigate the autothermal reforming of gasoline. They report experimental results with a Gd (gadolinium) doped ceria catalyst and also Ruthenium catalyst. The first catalyst was tested for sulfur tolerance by adding 50 ppmw sulfur in form of dibenzothiophene to the gasoline feed. They report that sulfur slightly increases the hydrogen yield. Their test runs were performed for 80 days with numerous starts and stops.

Palm et al. [8] measure product gas compositions for autothermal reforming over a precious metal catalyst for various hydrocarbons. They reach about 95% of the thermodynamic equilibrium. They add a sulfur-containing component to the feed to investigate the influence of sulfur on the reforming reaction. They observe that sulfur decreases the hydrocarbon conversion over run times of 120 h. This is explained by deactivation of the catalyst. Their findings contradict those reported by Krumpelt et al. [7] (see above). In a subsequent paper published by the same team, Pasel et al. [9] report the integration of a water gas shift reactor into an ATR fuel processor. They perform experiments with a diesel surrogate fuel (C13–C15) screening various shift catalysts.

The German Oel-Wärme-Insitut gGmbH develops fuel processors for liquid hydrocarbons based on the cool flame technology for mixture preparation and catalytic partial oxidation for fuel conversion [10]. Their fuel processors are designed for mobile and stationary fuel cell applications in the power range of 1–50 kW(el).

Goebel et al. [11] of General Motors Corporation report the development of a gasoline fuel processor including an autothermal reformer, two water gas shift (WGS) reactors and two preferential oxidation (PrOx) reactors. By utilizing direct water injection into two start-up burners that were used to preheat all the reactors they significantly reduce the system start-up time. In addition, hydrogen was introduced during start-up for catalyst light-off.

Commercial companies, especially in the US and Japan, have also reported successful developments of fuel processor systems for liquid hydrocarbons suitable for or already adapted to APU applications, e.g. Idatech LLC, Nuvera Fuel Cells Inc., Precision Combustion Inc. (PCI), and NexTech Materials Ltd.

2.2. APU systems

Many developments of APU systems have been reported in the past years. Many of these systems are based on methanol as a fuel. The advantage of methanol is that the reforming reaction takes place at temperatures between 200 and 400 °C, as supposed to 800–900 °C required for reforming hydrocarbons. This lower reformer temperature allows for easier design and less expensive material selection. In addition, CO concentration of the methanol reformer product gas is much lower than from reforming reactors that convert hydrocarbons allowing the CO shift reactor to be omitted. The main disadvantage of methanol is that it requires additional tanks in the vehicle and a separate supply infrastructure. Methanol based APUs in the range of up to 5 kW(el) have been developed by Ballard Power Systems Inc. (Canada), Idatech LLC (USA), P21 GmbH (Germany), DaimlerChrysler AG (Germany), to name a few.

The following paragraphs summarize developments of FC APU that use liquid hydrocarbons as fuel. Some insight into the US funding of high-temperature fuel cell systems for distributed power generation, which includes APU systems, is given by Williams et al. [12]. Henne and Winkler give a brief overview about projects within the European FP5 program [13].

Delphi Corporation, USA, started developing an SOFC based APU for mobile applications together with BMW AG, Germany, and Global Thermoelectric Inc., Canada in April 1999. In February 2001 the first gasoline based SOFC APU could be presented, and in December 2002 the second generation SOFC APU. The latter had a power output of 5 kW(el), a size of 44 l and a weight of 70 kg [14]. This system consisted of a SOFC stack by global thermoelectric and catalytic partial oxidation (POX) gasoline reformer. Two different reformer designs were developed by Delphi, a tubular reactor and a plate reactor. No problems with soot formation have been observed. Hydrogen and CO concentrations of the product gas are both in the order of 23 vol.%. Start-up time is in the order of 1 h. Tests of the reforming reactors with diesel yielded about 20 vol.% hydrogen and carbon monoxide, respectively [15]. Since 2004 BMW AG started to investigate the technical feasibility of a PEM fuel cell as basis for their 5 kW(el) APU. This work is done in cooperation with UTC Fuel Cells, USA.

In fall 2004 Volkswagen AG of Germany announced a cooperation with IdaTech LLC to design and manufacture a diesel fuel processor system for a fuel cell APU system. The fuel processor uses a palladium-alloy membrane for hydrogen purification rejecting contaminants such as sulfur compounds and unsaturated hydrocarbons at the same time.

In May 2003 Idatech LLC introduced a 2 kW(el) APU system. It is based on Ballard's NexaTM PEM fuel stack and on Idatech's proprietary methanol steam reformer. CO removal was accomplished by a metal membrane. Later in 2003 they also presented fuel processors for hydrocarbons, also based on steam reforming and metal membranes for gas clean up. At the Hannover Fair in April 2004 they presented steam reformers for diesel, propane and methane for APU systems up to 5 kW(el). They all operated at elevated pressure to be able to use metal membranes for hydrogen purification. In a publication Löffler et al. [16] reveal insights into their reactor design which consists of a pre-reformer that is thermally coupled with the steam reforming reactor.

SOFCo-EFS Holdings LLC developed a 10 kW(el) SOFC APU for propane and methane. The system can be utilized for RVs, trucks and back-up power supply. The start-up time currently is about 30 min, predicted to drop to 15 min with further development [17]. The reactor is a catalytic partial oxidation reformer that has been tested successfully with propane (500 h) and natural gas (85 h) without soot formation [18]. Reported efficiencies are 77% (natural gas) and 73% (propane). The sulfur content in the feed streams was 20 ppm (natural gas) and 185 ppm (propane). Diesel is planned to be tested in the near future, which requires the addition of a little water to avoid soot formation.

In Germany, Webasto Thermosystems GmbH, Germany, is developing a gasoline/diesel APU with a 1 kW(el) SOFC together with H.C. Starck GmbH, Germany and Fraunhofer IKTS, Dresden, Germany. Their system is based on a partial oxidation reactor that converts diesel into a synthesis gas. The start-up time of their current prototype system which includes an SOFC stack is in the order of 3 h. The goals of their product development are a 5 kW(el) system with 50 kg weight and a volume of 50 l.

Besides their partnership with BMW AG, Delphi Corporation developed SOFC based APU systems during the last 10 years. Besides stationary applications Delphi also develops systems for passengers cars, commercial and military vehicles, expecting their first prototypes to be available by late 2005. Their systems comprise fuel processors for diesel or gasoline generation of a synthesis gas which is supplied directly to the SOFC.

Both major manufacturers for commercial airplanes, Boeing and Airbus, have started the development of fuel cell based APUs to replace the turbine APU located in the tail cone of the airplanes. They teamed up with various partner from industry and academia for the realization of their systems.

3. 20 kW(el) diesel APU with MCFC

Fraunhofer ISE has developed a 20 kW(el) autothermal diesel reformer as pilot plant for a molten carbonate fuel cell (MCFC) by Ansaldo Fuel Cells S.p.A. in Genova, Italy. The motivation for this development is the technical demonstration of a diesel powered APU for ships based on a MCFC. In a second step, this pilot plant will be scaled up to a diesel powered 350 kW(el) MCFC APU system for a marine application. While the feed-stock to the 20 kW(el) pilot was 'sulfur-free' (<10 ppm) diesel, the technology demonstrator will be supplied with NATO standard F-76 diesel fuel, containing high sulfur levels up to 1 wt.%. Therefore, the technology demonstrator plant will include a desulfurization unit.

As a first step towards the pilot plant, test runs with noble metal catalysts for autothermal reforming of diesel were performed in a test rig at Fraunhofer ISE. The goal of these tests was to obtain necessary design information, e.g. product gas composition, space velocity, steam-to-carbon ratio (S/C), air number (λ), and temperature distribution in the reactor honeycomb. A proprietary dual-fluid nozzle in combination with a proprietary reactor head design allowed for good mixing of the diesel droplets with the steam/air mixture feed. Thus, the formation of soot and temperature peaks in the honeycomb could be avoided. Fig. 1 shows experimental results obtained with the test rig for one of the catalysts at a steam-to-carbon of 1.4, an air number of 0.33, and a pressure of 4000 hPa. It can been seen that with increasing capacity (diesel feed to the reactor) the temperature in the reactor increases. The temperature spread throughout the reactor honeycomb is fairly small and amounts to about 60 K at all times. This is a sign of uniform mixing upstream of the reactor honeycomb and efficient use of the catalyst volume. The temperature downstream of the catalyst honeycomb is between 630 and 670 °C which is the optimal inlet temperature for the subsequent MCFC. The temperature upstream of the catalyst honeycomb is above 850 °C. This indicates that partial oxidation of diesel
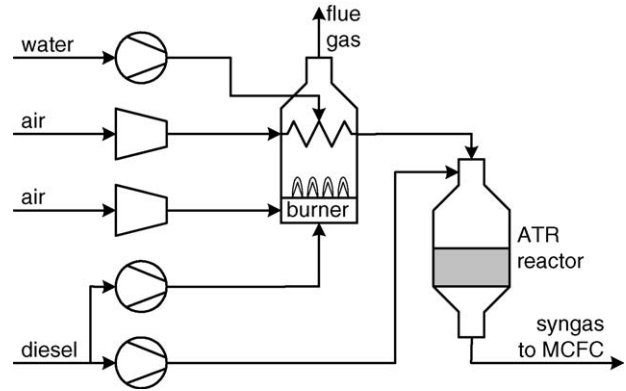

Fig. 2. PFD of the 20 kW(el) ATR diesel reformer.

compounds takes place. This is not surprising because the oxidation reaction is much faster than the subsequent exothermic steam reforming reaction. The latter takes place inside the reactor honeycomb which can be seen by the temperature decrease over the length of the reactor.

The product gas composition is almost constant over the capacity range under investigation. The (dry) hydrogen content is about 30 vol.%, carbon monoxide is in the range of 11–13 vol.% (dry).

Based on the results with the test rig, the diesel reformer pilot plant was designed. The design parameters were: capacity 40–100 kW (based on LHV of diesel), steam-to-carbon ratio between 1.0 and 1.5, and the air number ranging from 0.32 to 0.40. In Fig. 2, a process flow diagram of the ATR diesel reformer pilot plant is shown. The plant consists of two major pieces of equipment, the autothermal reforming reactor itself and a diesel-fired evaporator. The latter became necessary because no heat integration with the fuel cell part of the plant was intended. The diesel burner provides the necessary heat duty to heat up the water and air feed streams to about 350 °C. At the inlet to the reactor, this air/steam mixture is mixed thoroughly with
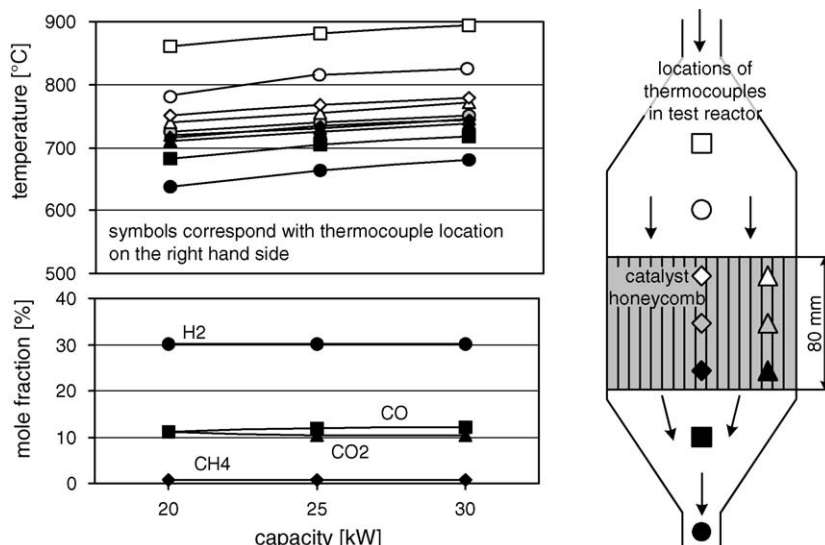

Fig. 1. Experimental results of catalyst screening for S/C = 1.4, λ = 0.33, p = 4000 hPa – reactor temperatures and (dry) product gas composition.
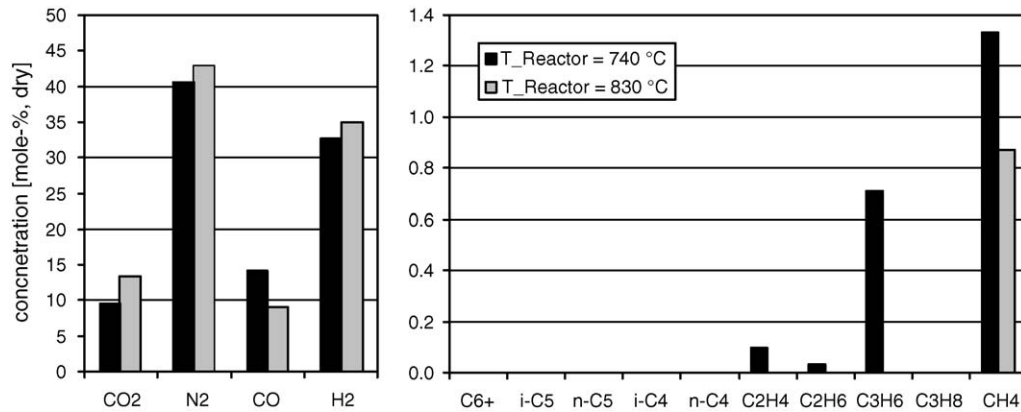
Fig. 3. Measured dry gas composition, at the beginning of the test run, S/C = 1.5, λ = 0.37, p = 4000 hPa. T_Reactor denotes the product gas temperature at the outlet of the reactor honeycomb.

the liquid diesel stream in a proprietary dual fuel nozzle. The reformer product gas is cooled down and sent to gas analysis – this is not shown in Fig. 2. Ultimately, the product gas will be supplied to the MCFC.

After mechanical completion, the pilot plant was started up and operated for over 300 h. During operation it was observed that it is crucial to keep temperatures in the catalyst honeycomb above 800 °C at all times to avoid soot in the product gas and to reduce the concentration of higher hydrocarbons to the single digit ppm range. This is in accordance with observations from Liu et al. [19] and Wunderlich and Stelter [20]. The first observed carbon formation during autothermal reforming of dodecane and hexadecane as diesel surrogates at temperatures below 700 °C.

In addition, a steam-to-carbon ratio of 1.5 is sufficient to avoid soot formation in the partial oxidation step. In Fig. 3 reformer product gas compositions determined by gas chromatography (Agilent 6890N) are presented as dry compositions. The product gas composition was measured for two different average reactor temperatures of 740 and 830 °C. It can be seen that at the higher temperature the amount of higher hydrocarbons is significantly reduced. To obtain the amount of higher hydrocarbons in the product gas, the product gas was cooled down to about 20 °C and the liquid phase separated from the gaseous phase in a flash drum. Detailed analysis of the liquid phase yielded that the amount of higher hydrocarbons (C6+) in the product gas was in the range of 20 ppm. As can be noted from Fig. 3, the hydrogen concentration in the reformer product gas in to about 35 mol%, while the concentration of CO decreases to about 9 mol%.

While the measurements shown in Fig. 3 were taken towards the beginning of the long-term test run, Fig. 4 presents the reactor product gas composition (dry basis) taken after 50, 150, and 300 h of operation. The reactor outlet temperature was kept above 800 °C at all times. The S/C ratio was 1.5, the air number 0.37 and the operating pressure 4000 hPa. It can be noted that the catalyst performance deteriorates slightly over time, with the hydrogen and CO concentration decreasing by about 7 and 3%, respectively. The methane concentrations, on the other hand, triples at the same time. However, all three concentrations have started leveling out, suggesting that the deterioration rate decreases with increasing time.
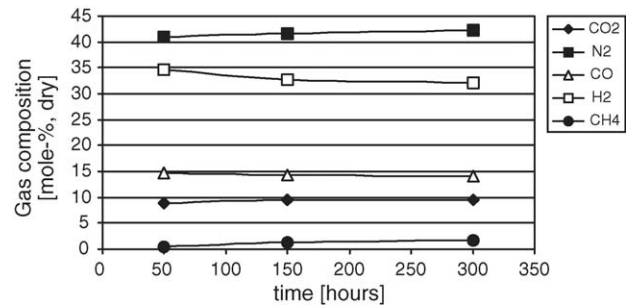

Fig. 4. Measured product gas composition (dry basis) at different run times, S/C = 1.5, λ = 0.37, p = 4000 hPa, T_Reactor above 800 °C at all times.

4. 5 kW(el) kerosene APU with SOFC

In a project funded by the European Union, Fraunhofer ISE developed a compact autothermal kerosene reformer for an aerospace application. Other project partners involved in the development of the SOFC APU system are Liebherr Aerospace, Lindenberg (Germany) and DLR Stuttgart (Germany). The reformer produced hydrogen and carbon monoxide from desulfurized Jet A-1 Fuel for the use in a 5 kW(el) SOFC. Again, for purpose of investigations at Fraunhofer ISE desulfurized kerosene (sulfur content of about 6 ppm) was utilized. The core of the pilot plant was an autothermal reforming reactor, consisting of three catalyst honeycombs (monoliths). The temperature profile along the channels of the reactor was measured at six different locations in each monolith. It was observed, that the (exothermic) partial oxidation of kerosene takes place in front of and inside the first monolith. Subsequently, the temperature decreases in the second and third monolith because of the endothermic steam reforming reaction. After the third metal monolith the chemical equilibrium is almost reached. The steam-to-carbon ratio (S/C) was varied between 1.5 and 2.5. The goal was to minimize S/C in order to reduce the amount of water that needs to be evaporated upstream of the reactor and condensed downstream of the fuel cell. The temperature inside the reactor is influenced by the amount of air provided for the partial oxidation reaction. The maximum temperature inside the reactor was
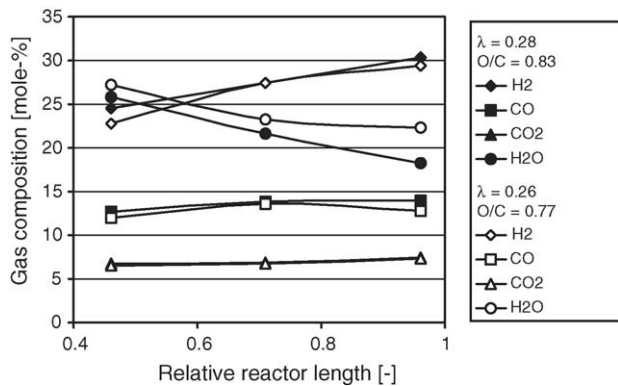
Fig. 5. Gas composition along reactor axis, measured with S/C = 1.5.

kept below 950 °C to minimize accelerated deterioration of the catalyst due to sintering. This was achieved with air numbers of less than 0.30. The capacity of the plant could be varied between 5 and 15 kWth (based on the LHV of kerosene). Pressure in the system was slightly above ambient pressure. For more details please refer to Lenz and Aicher [21]

The product gas was drawn after each catalyst monolith and analyzed in a gas chromatograph (Agilent 6890N). Gas compositions measured downstream of each reactor segment are shown in Fig. 5.

For evaluation of the system efficiency the volume flow, pressure, and temperature of the product gas was measured. Typical efficiencies were in the order of 70–80% (i.e. energy content of synthesis gas at reactor outlet divided by energy content of kerosene feed). The reactor outlet temperature was about 650–750 °C depending on operating conditions. Since the following SOFC requires an inlet temperature of at least 800 °C a booster heater is required.

5. Summary

Several R&D groups in industry, research organizations and universities are currently working on fuel cell based APU systems. At Fraunhofer ISE, a diesel and a kerosene autothermal reformer were developed in collaboration with industrial partners and research organizations. Both plants have been operated successfully for several hundreds of hours, with only slight catalyst degradation. Soot formation was not observed, and trace amounts of higher hydrocarbons were found in the reformer product gases only.

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