

Catalytic autothermal reforming of Jet fuel

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Abstract

Aircraft manufacturers have to reduce the emissions and the specific fuel consumption of their systems. Fuel cell use in a ‘more electric aircraft’ can be one possibility. To keep the technology simple only one fuel (Jet A, Jet A-1) shall be used on board the aircraft. Therefore, the catalytic reforming of Jet A-1 fuel was examined in this paper, although the use of fossil fuels causes the production of greenhouse effect promoting gases like carbon dioxide CO₂.

The autothermal reforming of desulphurised kerosene is examined with a 15 kW (based on the lower heating value of Jet fuel) test rig. The experiments are performed at steam to carbon ratios of S/C = 1.5–2.5 and air to fuel ratios of $\lambda = 0.24$ –0.32, respectively. The composition of the product gas, the volumetric flow rate of the product gas at standard conditions and the temperatures in the catalyst are determined as a function of the operating variables. The gas hourly space velocity (GHSV) is varied between 50,000 and 300,000 h⁻¹.

The influence of sulphur containing feed streams (real Jet fuel) on reforming behaviour is investigated as well as the influence of the hydrogen concentration on the hydrodesulphurisation process. Another simple way of desulphurisation is the adsorption of liquid sulphur containing hydrocarbons, the influence of the variation of the liquid hourly space velocity (LHSV) is measured at a temperature of 150 °C. © 2005 Elsevier B.V. All rights reserved.

Keywords: Autothermal reforming; Jet A-1; Desulphurisation; Aircraft application

1. Introduction

Since 1968, the number of passengers in air traffic increased from 131 to 693 million in the year 2001. In 2010, 1.6 billion passengers are expected. The increase of demand in world air traffic is 4–6% per year. It is expected that the percentage of air travel from the whole passenger volume will increase to 36% in the year 2050 [1].

Emissions of aircraft are released in higher atmospheres, and therefore, are even more harmful for the environment.

In 2008, the OECD is planning to establish a compensation fee for aircraft depending on their actual emissions.

Therefore, fuel cell auxiliary power units (APUs) using on-board hydrogen production from liquid energy carriers have attracted much attention. The most important objectives for reformer development in an aircraft are low-weight, good

dynamic response and delivering a gas quality that contains as much hydrogen as possible. An autothermal reformer is the best solution to meet these requirements.

The autothermal reforming reactor is designed for a gaseous hourly space velocity (GHSV; [2]) of 100,000 h⁻¹. The design point for the water and air feed is: air to fuel ratio $\lambda = 0.3$ and steam to carbon ratio S/C = 1.5.

1.1. Jet fuel

In today’s passenger planes, kerosene-type fuels are chosen as fuel as having the best combinations of properties. Wide-cut Jet fuel (Jet B) is still used in some parts of Canada and Alaska because it is suited to cold climates. But kerosene-type fuels (Jet A and Jet A-1) predominate in the rest of the world.

Jet A is used in the United States, while most of the rest of the world uses Jet A-1. The important difference between the two fuels is that Jet A-1 has a lower maximum freezing

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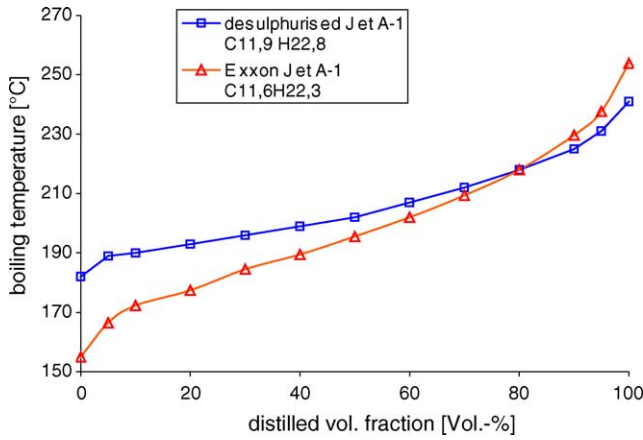


Fig. 1. Distillation curves of aviation fuel Jet A-1 (measured).

point than Jet A (Jet A: -40°C , Jet A-1: -47°C) [3]. The lower freezing point makes Jet A-1 more suitable for long international flights, especially on polar routes during the winter.

Jet A-1 is a mixture of low volatile hydrocarbons in the boiling range of $150\text{--}300^{\circ}\text{C}$. Due to international guidelines [4], the allowed sulphur content in Jet fuel is extremely high (3000 ppm), the actual measured sulphur content is 290 ppm.

In this paper, the reformer tests described were carried out with desulphurised kerosene (sulphur content 6 ppm, obtained from MiRO Mineraloelraffinerie Oberrhein GmbH & Co. KG) and Exxon Jet A-1, obtained from Freiburg airport.

Rachner [5] gives the fraction of aromatics in Jet fuel with between 14 and 20%. The average totals formula of the MiRO fuel is $\text{C}_{11.9}\text{H}_{22.8}$ [6], of the Exxon Jet A-1 fuel it is $\text{C}_{11.6}\text{H}_{22.3}$ [5].

Fig. 1 shows the distillation curves of those fuels. The distillation behaviour is needed because the kerosene is evaporated for the hydrodesulphurisation (HDS) process, described below.

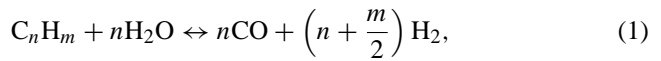
Basis for the design of the autothermal reforming reactor is the lower heating value (LHV) of Jet fuel. Rachner gives the LHV of Jet fuel with 43.26 MJ kg^{-1} . Table 1 presents an overview of properties of Jet fuel in literature.

There are several additives mixed to Jet fuel to tune its properties. Table 2 gives the type, function and influence on reforming performance.

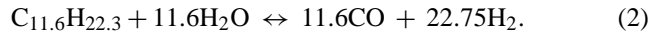
1.2. Autothermal reforming

Autothermal reforming is known as the simultaneous conversion of hydrocarbons with steam and oxygen. The en-

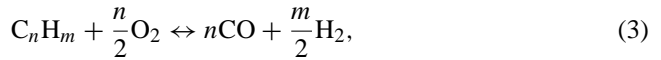
dothermic steam reforming reaction is generally given by:



and for Jet fuel particularly:



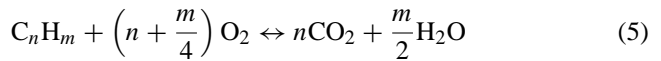
The energy for this endothermic reaction is delivered by the partial oxidation reaction (POX), generally given by:



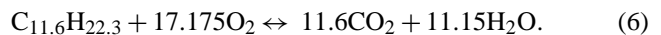
and for Jet fuel:



Parallel to the POX reaction, part of the hydrocarbon is always totally oxidised (TOX):



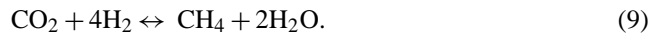
and for Jet fuel:



Also, the shift reaction takes place in the autothermal reforming reactor:



At lower temperatures also the methanation reactions have to be considered:



1.3. Reactor design

The reactor consists of three segments, each one containing a metallic monolith structure, which is coated with a washcoat (Al_2O_3) and precious metal catalysts (Pt and Rh). The benefits of coated metallic structures compared to a fixed bed catalyst are the lower pressure drop, better thermal conductivity and lower thermal mass.

Fig. 2 shows a schematic description of the assembled reforming reactor. After each segment two pipes are integrated in the flanges to draw product gas samples and to access the reactor with thermocouples. The head of the reactor consists of a mixing chamber, with integrated pipes for the kerosene, evaporated water and air feed streams. The temperature profile along the reactor is measured at six different locations in each honeycomb structure. Fig. 3 shows a cross section of

Table 1
Properties of aviation Jet fuel surrogates

Literature source	Molecular formula	Molecular weight (g mol^{-1})	LHV (kJ mol^{-1})	Boiling point ($^{\circ}\text{C}$)
Rachner [5]	$\text{C}_{11.6}\text{H}_{22.3}$	161.9	7238.3	155 (initial boiling point)
Brown [7]	$\text{C}_{15}\text{H}_{30}$	210	9030	245 (mean boiling point)

Table 2
Additives of aviation fuel Jet A-1 [8]

Additive type	Used chemical	Influence on reforming
Antioxidant	2,6-Ditertiary butyl-4 methyl phenol	No influence containing C, H, O
Metal deactivator	<i>N,N'</i> -Disalicylidene-1,2-propane diamine	No influence containing N, C, H, O
Electrical conductivity/static dissipater	Stadis [®] 450 containing 40–50% toluol, 0–5% isopropylalkohol [8]	No influence containing C, H, O
Corrosion inhibitor/lubricity improver	Naturally occuring compounds in Jet fuel (sulphur containing)	Sulphur compounds poison for reforming catalyst
Fuel system icing inhibitor	Di-ethylene glycol monomethyl ether (di-EGME)	No influence containing C, H, O

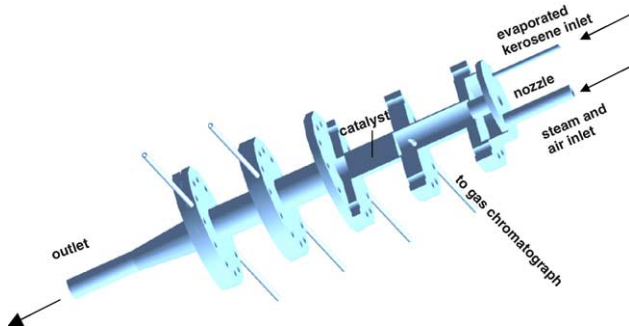


Fig. 2. Schematic description of the reforming reactor.

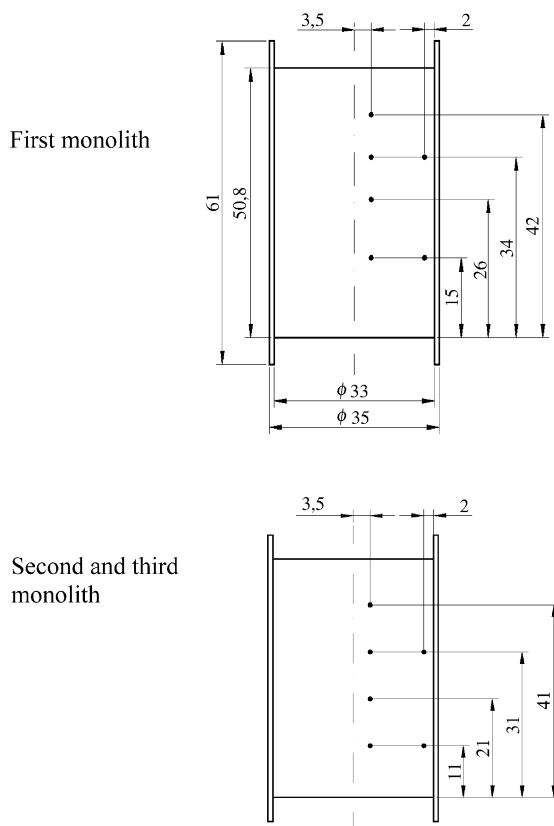


Fig. 3. Location of thermocouples.

the monoliths to illustrate the position of the thermocouples. The black dots denote the location of the thermocouples.

Because a catalyst screening is planned, the prototype of the reactor was designed for easy catalyst mounting and change.

The catalysts consist of a mixture of precious metals (platinum (Pt) and an element from the eighth group of the transition metals), proprietary of UMICORE and Suedchemie. Table 3 gives an overview on the used catalysts. It is assumed that Pt catalyses mainly the oxidation reaction and Rh the steam reforming reaction [9].

Fig. 4 shows a schematic description of the laboratory test rig. Jet fuel and water is metered and pressurised to 3 bar with membrane dosage pumps. The membrane dosage pumps (Grundfos) are used for the dosage of the liquid components. The air flow is controlled by mass flow controllers from MKS. The temperature in the reactor is controlled by the amount of oxygen that is available to maintain the exothermic combustion of Jet fuel. Water is mixed with air and continuously evaporated in a coiled tube heat exchanger. The evaporator is electrically heated with five heating tapes from Horst (each with 1.18 kW). The temperature of the evaporator is controlled with a four-channel PSG controller. The superheated mixture of air and water enters the reforming reactor via an annular gap. The pressurised Jet fuel is sprayed into the reactor with a nozzle. To reduce heat losses to the ambient, the reactor is insulated. Temperatures are measured at the described points with type K thermocouples, all data are recorded with an Agilent data logger (34,907 A).

All components of the plant including mass flow controllers, pressure and temperature measurement, valves and dosage pumps are connected to a PC by GPIB and RS 232 interfaces. The plant is controlled by the instrumentation software LabView (National Instruments).

During reforming, sulphur containing hydrocarbon compounds are converted into hydrogen sulphide H_2S , which also

Table 3
Overview on the used proprietary precious metal catalysts

	UMICORE	Suedchemie
Active component	Precious metal	Precious metal
Sulphur tolerance	<10 ppm	<50 ppm in kerosene
Carrier material	Al_2O_3	Al_2O_3
Maximum GHSV (h^{-1})	ca. 100,000	ca. 100,000

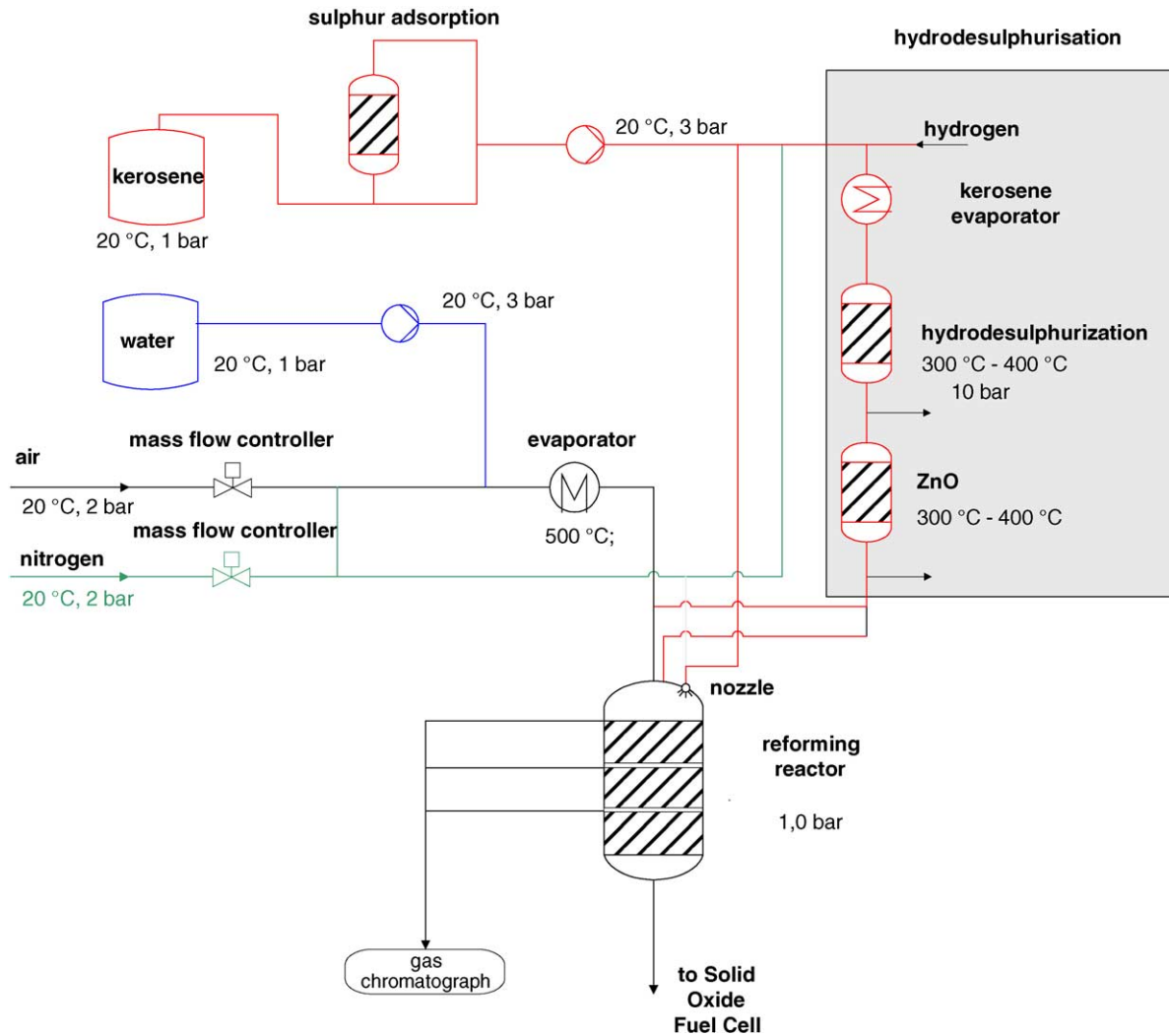


Fig. 4. PFD of the laboratory test rig.

has poisoning effect on the fuel cell. Matsuzaki and Yasuda [10] observed a large temperature dependence of the degradation of the SOFC and calls for desulphurisation at operating temperatures of 750 °C.

A very simple way of cleaning fuels from sulphur is the selective adsorption for removing sulphur (SARS). The SARS process can effectively remove sulphur compounds in liquid hydrocarbons at room or slightly elevated temperatures and ambient pressure. Ma et al. [11] proposed to use the SARS process in future refineries in combination with hydrodesulphurisation of the concentrated sulphur fractions.

The measurements in this paper were carried out with adsorbing material (nickel/nickel monoxide mixture) at 150 °C with a volume flow of real Jet A-1 fuel of 1.5 l h⁻¹. The material was obtained from Suedchemie. The kerosene dosage pump sucks the kerosene through the adsorber. The adsorptive desulphurisation reaction works more efficient at higher temperatures (100–200 °C). Because of the high reactivity of the adsorbent, it is not possible to regenerate it.

For hydrodesulphurisation, a hydrogen flow is mixed to the Jet fuel and preheated to 350 °C in the electrically heated kerosene evaporator. Then, it enters the reactor where the hydrodesulphurisation (HDS) reaction takes place. The gas flow then is purged from hydrogen sulphide in the ZnO-reactor. The HDS reactor contains a CoO and MoO₃ coated catalyst. It is necessary to first reduce CoO and MoO₃ to cobalt and molybdenum sulphide, by treating with H₂S at elevated temperatures for a few hours. The HDS process works at pressures of less than 10 bar and is the state of the art desulphurisation process used in refineries.

The efficiency of the reforming process is calculated by equation (10):

$$\eta = \frac{(\dot{n}_{\text{H}_2} \text{LHV}_{\text{H}_2} + \dot{n}_{\text{CO}} \text{LHV}_{\text{CO}})}{\dot{n}_{\text{C}_x\text{H}_y} \text{LHV}_{\text{C}_x\text{H}_y}} \quad (10)$$

The lower heating values of carbon monoxide and hydrogen are given in Table 4.

Table 4
Lower heating values [12]

Species	LHV (kJ mol^{-1})
Hydrogen	241.83
Carbon monoxide	282.00

Table 5
Columns of the gas chromatograph

Column 1	PLOT, separates C_6^+ , butene, butane, pentane and pentene
Column 2	PLOT, separates C_6^+ , butene, butane, pentane and pentene
Column 3	HP PLOT Q (light hydrocarbon in stabilised crude oils including pentanes) separates ethane, ethene, propane, propene, CO , H_2O and H_2S
Column 4	Porapak Q80/100 separates H_2 , N_2 , O_2 , CO and CH_4
Column 5	Molecular sieve separates O_2 and N_2
Column 6	Porapak Q80/100 separates H_2 , N_2 , O_2 , CO and CH_4

1.4. Gas analysis

The product gas from the reformer is analysed in a gas chromatograph (Agilent 6890). For detection of the molar gas composition the gas chromatograph consists of two detectors (thermal conductivity detectors, TCD). The separation is accomplished by six columns:

Table 5 gives an overview of the function of the columns.

The carrier gas for the back detector is argon, for the front detector it is helium. The thermal conductivity of helium is very similar to the one of hydrogen (helium: $0.142 \text{ W m}^{-1} \text{ K}^{-1}$, H_2 : $0.168 \text{ W m}^{-1} \text{ K}^{-1}$ at 20°C); therefore, the back detector is designed for the use of Argon ($0.016 \text{ W m}^{-1} \text{ K}^{-1}$ at 20°C) as carrier gas. The gas flow in the GC is controlled by four pneumatically driven valves. To avoid condensation of water, the pipe, connecting the reformer to the GC, and the valves are electrically heated to 150°C at any time.

Fig. 5 shows a typical chromatogram, the first peak represents hydrogen with a retention time of 1.007 s, followed

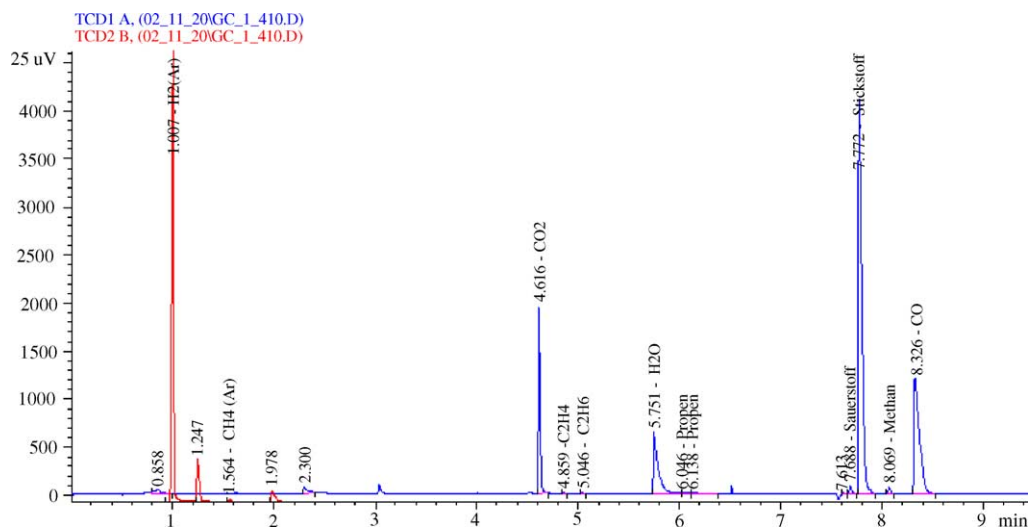


Fig. 5. Product gas analysis with gas chromatograph $S/C = 1.5$, $\lambda = 0.3$, $\text{GHSV} = 50,000 \text{ h}^{-1}$, desulphurised Jet A-1 from MiRO.

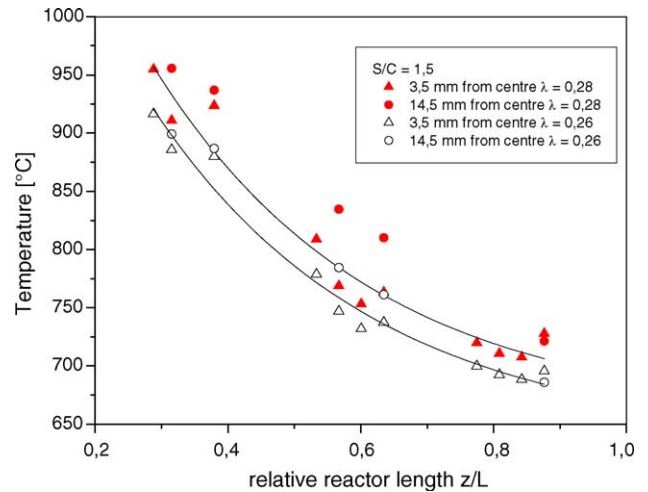


Fig. 6. Temperature profile along the reactor axis ($p = 1 \text{ bar}$, $\text{GHSV} = 50,000 \text{ h}^{-1}$, $S/C = 1.5$), desulphurised Jet A-1 from MiRO.

by CO_2 , C_2H_4 , C_2H_6 , water, propene, propane, nitrogen, methane and CO . Unconverted hydrocarbons ($>\text{C}_6$) show a peak at retention times smaller than 0.9 s.

2. Results and discussion

Fig. 6 shows the temperature profile along the reactor axis. The oxidation reaction takes place in the front part of the first monolith structure, where the peak temperature of 950°C is reached. The high temperature is needed for the conversion of aromatics contained in Jet fuel.

The oxidation and the steam reforming reactions in the hot part of the reactor are mass transfer limited. While the temperature decreases along the reactor axis the steam reforming reaction in the colder part is kinetically limited [13].

The difference between the centre and the exterior is smaller than 50°C . For a lower air to fuel ratio the radial

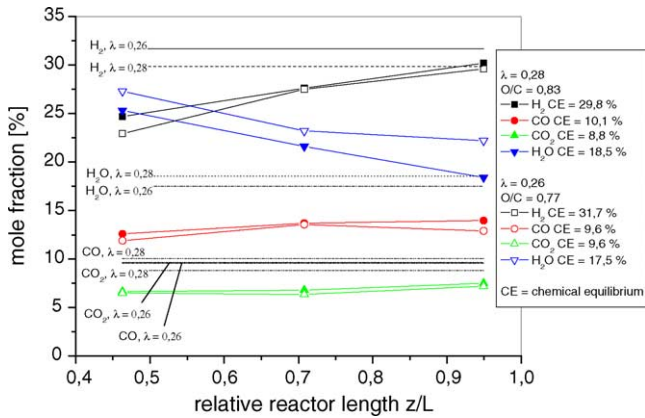


Fig. 7. Measured gas composition (containing water) accomplished with the 5 kW (el.) reformer ($T_{out} = 700\text{ }^{\circ}\text{C}$, $p = 1\text{ bar}$), desulphurised Jet A-1 from MiRO.

temperature profile is nearly constant. As can be seen from Fig. 6, a small increase in the air to fuel ratio λ has a significant impact on the temperature in the reactor. The difference between the temperatures in the centre and the outside of the reactor is caused by an inhomogeneous air distribution in the premixing zone of the reactor. In front of the first catalyst segment, we measured temperatures of $320\text{ }^{\circ}\text{C}$, at this temperature level the Reynolds number of the feed gas mixture in front of the catalyst is $Re = 3270$. At $Re > 2300$, the flow is turbulent.

Fig. 7 shows the resulting molar gas composition along the reactor axis. At the outlet, hydrogen and water have almost reached the chemical equilibrium concentration at the air ratio $\lambda = 0.28$. The chemical equilibrium data are calculated with the software package Chemcad, a process engineering tool from Chemstations GmbH, simulating the autothermal reformer as an adiabatic CSTR (continuously stirred tank reactor). It is not taken into account that the temperature in the autothermal reactor decreases from 950 to $700\text{ }^{\circ}\text{C}$ along the reactor axis. The calculated gas composition is given in the diagram as horizontal lines. The deviation from the product gas composition to the chemical equilibrium for carbon monoxide and carbon dioxide is caused by mass transfer limitation of the water–gas shift reaction (7) in the autothermal reforming reactor. The water content at the reformer outlet, measured at an air to fuel ratio of $\lambda = 0.26$ differs significantly by 20% from the calculated chemical equilibrium values. At this temperature level the conversion of mainly the aromatic compounds in Jet fuel is constricted.

Fig. 8 affirms this, at lower air to fuel ratios, the efficiency (which is the ratio of Jet fuel that is converted to hydrogen and carbon monoxide) decreases. The best process efficiency is reached at an air to fuel ratio of $\lambda = 0.28$, a steam to carbon ratio of $S/C = 1.5$ and a gaseous hourly space velocity of $50,000\text{ h}^{-1}$ with 78.5%.

Fig. 9 shows the measured energy balance at this mode of operation based on the temperature level of $800\text{ }^{\circ}\text{C}$. $H_{dot,chem}$ describes the energy content of the gas flows of the several

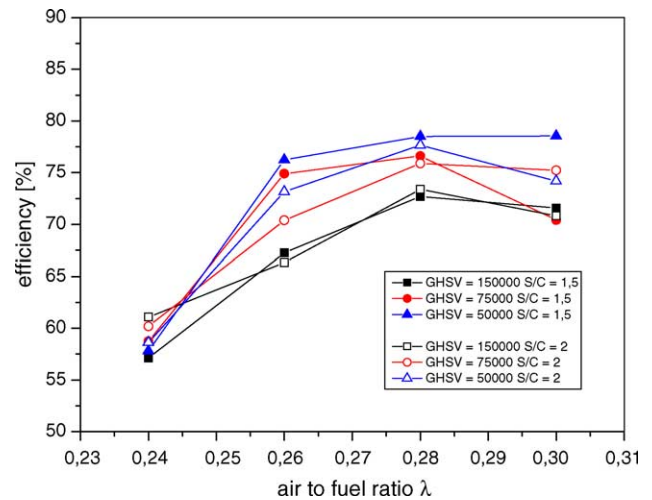


Fig. 8. Efficiency of reforming process, data obtained with UMICORE catalyst ($p = 1\text{ bar}$), desulphurised Jet A-1 from MiRO.

species contained in the product gas, based on the lower heating value. $Q_{dot,therm}$ describes the thermal heat losses of the reactor at the reference temperature $800\text{ }^{\circ}\text{C}$.

The heat losses of the reactor are found to be 1.14 kW , the preheating of the educts from $500\text{ }^{\circ}\text{C}$ in the evaporator to $800\text{ }^{\circ}\text{C}$ in the reactor needs a heat flux of 1.68 kW .

Table 6 gives the gas composition at the outlet of the reformer. Suzuki et al. [14] reports a hydrogen content of 60.2–60.8% on a commercial Ni catalyst and between 63.6 and 70.2% on a Ru based catalyst at an S/C ratio of 3.5 during steam reforming of kerosene. The chemical conversion amounts to 97 and 99.5% after 24 h on the Ru catalysts. The conversion found with autothermal reforming of kerosene is 92.5% at a steam to carbon ratio of $S/C = 1.5$ and an air to fuel ratio of $\lambda = 0.28$ and 88% at an air to fuel ratio of $\lambda = 0.26$, both values at gaseous hourly space velocities of $50,000\text{ h}^{-1}$.

Figs. 10 and 11 show the concentration of larger hydrocarbons in the product gas. The measurements were taken after times, smaller than 100 h after the catalyst has been changed. An increase of methane in the product gas composition could not be detected after this period of time. Aicher et al. [15] re-

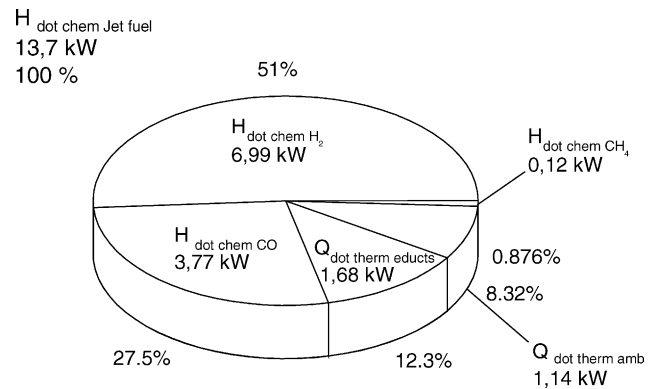


Fig. 9. Energy balance of the Jet fuel reformer at $S/C = 1.5$, $\lambda = 0.28$, $GHSV = 50,000\text{ h}^{-1}$.

Table 6

Gas composition in autothermal reforming of Jet fuel (GHSV = 50,000 h⁻¹, S/C = 1.5, p = 1 bar)

	Gas composition (containing water) (%)				Gas composition (dry basis) (%)			
	H ₂	N ₂	CO	CO ₂	H ₂	N ₂	CO	CO ₂
$\lambda = 0.28$	30.2	30.3	13.96	7.5	37.0	37.1	17.1	9.2
$\lambda = 0.26$	29.6	29.7	12.9	7.2	38.0	38.2	16.6	9.3

port an increase of methane in the product gas from 0.36% after 50 h to 1.2% after 150 h and to 1.6% after 300 h during autothermal reforming of diesel.

At higher temperatures (larger air to fuel ratios), a decrease of hydrocarbons smaller than C₆ (methane, ethane, ethene, propane, propene, butane and pentane) can not be found. However, the content of hydrocarbons larger than C₆ decreases with increasing temperature.

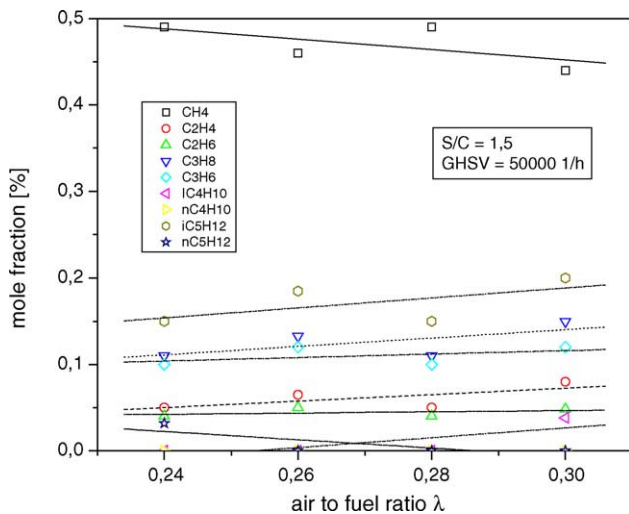


Fig. 10. Measured gas composition (with water) at S/C=1.5 and GHSV = 50,000 h⁻¹ with UMICORE catalyst ($T_{out} = 700^\circ\text{C}$, $p = 1$ bar), desulphurised Jet A-1 from MiRO.

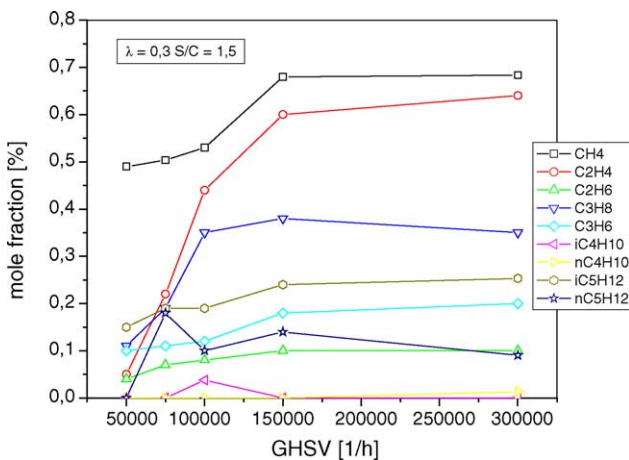


Fig. 11. Measured gas composition (with water) at S/C=1.5 and $\lambda = 0.3$ with UMICORE catalyst ($T_{out} = 700^\circ\text{C}$, $p = 1$ bar), desulphurised Jet A-1 from MiRO.

As can be seen from Fig. 11, the content of the detected hydrocarbons increases with growing space velocities (GHSV). The measurements were taken at steam to carbon ratios of 1.5 and air to fuel ratios of $\lambda = 0.3$ at a pressure of 1 bar absolute. This confirms the assumption, that the steam reforming reaction is kinetically limited [9]. Also, the increase of the ethene concentration with growing space velocities is remarkable. It is assumed, that the path of the reforming process includes the formation of C₂H₄ as an intermediate product. It is also remarkable, that the content of alkenes in the product gas is always higher than the content of the corresponding saturated alkanes.

2.1. Measurements with sulphur

To examine the influence of sulphur compounds in Jet fuel, a test sequence was performed where first the reactor was run with desulphurised kerosene and after steady state conditions were achieved, the pumps were switched to sulphur containing kerosene (290 ppm). These tests were performed at a steam to carbon ratio of S/C = 3 and an air to fuel ratio = 0.34 on a precious metal catalyst proprietary of Suedchemie. The production of hydrogen diminishes instantaneously as can be seen in Fig. 12, while the temperature temporarily climbs up to 980 °C. Due to the adsorption of sulphur by the catalyst, the activity decreases, less hydrogen is produced, and therefore, less heat used for the endothermic steam reforming reaction. The poisoning influence on the CO content in the product gas is negligible although the mole

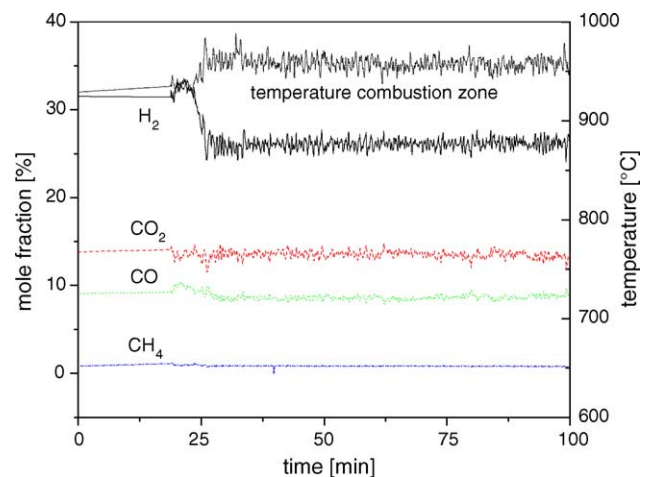


Fig. 12. Measured gas composition (dry basis) with deterioration caused by sulphurous poisoning at S/C=3 and $\lambda = 0.34$ with Suedchemie catalyst ($p = 1$ bar), desulphurised Jet A-1 from MiRO and Exxon Jet A-1.

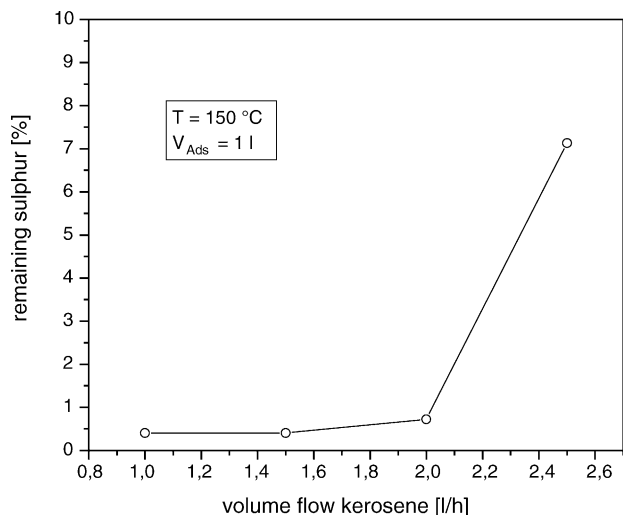


Fig. 13. Adsorptive desulphurisation, Exxon Jet A-1.

fraction oscillates much more than during the reforming of hydrocarbons with low sulphur content.

After switching back to desulphurised Jet fuel, the reforming process regenerates again but does not reach the initial level, the catalyst is permanently damaged.

Palm et al. [16] found a decrease of hydrocarbon conversion during sulphur containing feed from 94 to 78% with a sulphur content of 11 ppmw and of 93 to 74% with a sulphur content of 30 ppmw at an oxygen to fuel ratio $O/C = 0.43$ and a steam to carbon ratio of $S/C = 2.2$. They also found, that the conversion regenerates during the use of desulphurised hydrocarbons, but is only partly reversible.

2.2. SARS

With an adsorbing volume of 11, a volume flow of 21 h^{-1} kerosene (containing 290 ppmw sulphur) can be desulphurised to 1.2 ppmw (Fig. 13). The ordinate shows the actual sulphur content based on the initial sulphur content of Exxon Jet A-1 fuel in per cent. The analysis was made by an external laboratory with inductively coupled plasma optical emission spectroscopy (ICP-OES) from Jobin Yvon. About the long term loading behaviour of the material, no statements can be made after those measurements.

Because of the high reactivity of the material, regeneration is not possible.

2.3. HDS

Fig. 14 shows the results from the HDS experiments. With a combined HDS and ZnO reactor system at a pressure of 10 bar and a temperature of 300°C , Jet fuel can be desulphurised down to 8.7 ppmw using a hydrogen flow of 6.72 slm. Again, the ordinate shows the actual sulphur content based on the initial sulphur content of Exxon Jet A-1 fuel in per cent.

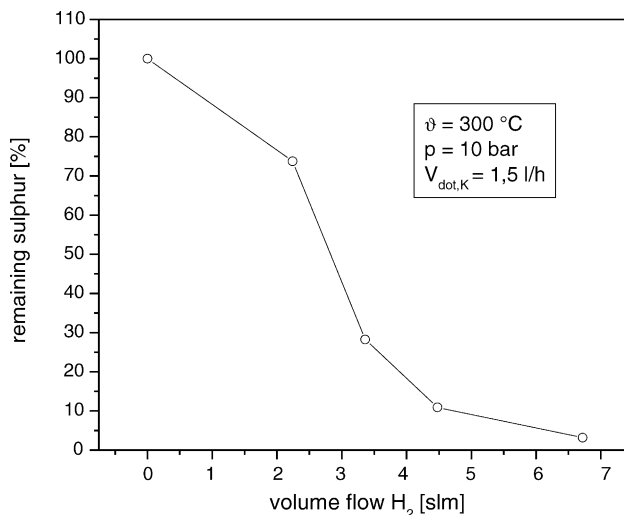


Fig. 14. Hydrodesulphurisation at 10 bar, 300°C , kerosene flow 1.51 h^{-1} , Exxon Jet A-1.

3. Conclusions

The autothermal reforming of desulphurised Jet fuel has been examined on a 15 kW test rig. The best efficiency was achieved with a steam to carbon ration $S/C = 1.5$ and an air to fuel ratio of $\lambda = 0.28$ at a space velocity of $\text{GHSV} = 50,000 \text{ h}^{-1}$. No hydrocarbons larger than C_6 are found in the product stream, but it was found that always the content of alkenes is higher than the content of the corresponding alkanes. The different desulphurisation methods were examined, both worked well. For implementation into an aircraft or other mobile applications the hydrodesulphurisation process seems to be too heavy and large. The SARS process seems to be very promising, although it only works economically at lower sulphur contents.

The investigation of the system behaviour and the detailed study of the heat and mass transfer processes in the reforming reactor will be part of further work.

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