

Novel process to evaporate liquid fuels and its application to the catalytic partial oxidation of diesel

T. Aicher^{a,*}, L. Griesser^b

^a *Fraunhofer-Institut für Solar Energiesysteme (ISE), Heidenhofstrasse 2, D-79110 Freiburg, Germany*

^b *Riedhofstrasse 71, CH-8049 Zürich, Switzerland*

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Abstract

A novel process for evaporation of liquid hydrocarbons, like gasoline, diesel or kerosene, has been developed and tested. It allows to directly transfer a liquid hydrocarbon mixture into the gaseous phase avoiding all problems related to residue and carbon formation due to contact of the fuel with hot heat exchanger surfaces.

This process is especially advantageous when designing combustion or reforming systems for diesel, since this fuel can not be evaporated directly. By transferring diesel into the gaseous phase, thorough mixing of the hydrocarbons with steam and air is possible, reducing the risk of carbon formation in subsequent combustion or reforming processes.

In Part I, this paper describes the evaporation process and presents first experimental results. A map will be given characterizing operating conditions where the evaporation is complete. Furthermore, the composition of the evaporator products were measured providing insight into the processes taking place in the evaporator.

In Part II, the paper explains how the evaporator was connected to a catalytic partial oxidation reformer reactor and presents first experimental results with diesel which showed that at temperatures around 800 °C the thermodynamic equilibrium is reached and no carbon is formed in the evaporator nor in the CPOX reactor.

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1. Introduction

Mixture preparation is a crucial step in both combustion and reforming processes when liquid fuels are used. With the first application good mixing is important to control emissions, with the second to reduce the risk of carbon formation, a problem that many researchers have been battling with and that still is an objective of current research, as reaffirmed recently by Porš et al. [1]. In both processes one of the fundamental steps is to mix liquid and gaseous reaction partners. For instance, in partial oxidation, combustion, or autothermal reforming of liquid fuels, the fuel has to be mixed with air and in latter case also with water.

One way to achieve thorough mixing of all feed streams is to vaporize all liquid reactants prior to mixing with air. The

simplest way to achieve this is the direct evaporation of liquid hydrocarbons. It can be employed for liquid fuels as reported for instance for pure alkanes [2], alcohols, gasoline [3], kerosene [4] or surrogate fuels [2]. In case of diesel, however, problems like the formation of residue, carbon deposits, and fouling on heat exchanger surfaces arise, which make it difficult to evaporate diesel by direct contact with hot heat exchanger surfaces. Therefore, to the authors' knowledge no accounts have been published stating the successful direct evaporation of diesel.

Since the direct evaporation of diesel seems impossible, many concepts have been developed to indirectly transfer diesel into the gas phase. One way is the injection of liquid fuels, e.g. diesel, into the hot combustion air. Even though this concept seems relatively simple, high inlet temperatures and high pressures can yield self-ignition of the fuel and stable flames in parts of the reactor that were designed for pre-mixing only. These problems are more pronounced for heavy hydrocarbons since their boiling points exceed their auto ignition temperatures and their ignition

* Corresponding author. Tel.: +49 761 4588 5194; fax: +49 761 4588 9320.
E-mail address: thomas.aicher@ise.fraunhofer.de (T. Aicher).

delay times are very short under conditions typical for reformers and combustors. This is described in more detail by Krummenacher et al. [5]. Another draw-back of this concept is that the liquid hydrocarbons have to be injected into a hot gas stream by a nozzle, oftentimes a dual-fluid nozzle. This allows the use of a nozzle in a small range of flow rates only, thus reducing the turn-down ratio of the plant.

Numerous papers have been published with more sophisticated approaches to the problem of mixing liquid fuels with combustion and reforming air. Reh and his co-workers suggest in their concept for combustion processes to atomize and evaporate the liquid fuel externally in the absence of air prior to blending it with the combustion air in a static mixer (see for instance Wei et al. [6]). The fuel air mixture is then supplied to a premixed surface burner. They use small amounts of superheated steam (molar steam-to-carbon ratio of about 0.2–0.8) to atomize the diesel fuel before it is mixed with the combustion air. Similar concepts have also been developed by others, for instance by Piwetz et al. [7], Scoles and Perna [8], Steinfeld et al. [9], Mauzey et al. [10], and Perna et al. [11] for mixture preparation upstream of a pre-reforming and autothermal reforming reactors as part of fuel cell systems. All these systems use much higher steam-to-carbon ratios.

Combining the simple direct injection concept with a steam buffer concept to avoid high local heat release yields a process, where diesel is directly injected into a hot steam/air mixture, as described by Aicher and co-workers for the autothermal reforming of diesel [12]. They use a proprietary reactor design to provide good mixing of the diesel spray with hot air/steam mixture and demonstrate that it is possible to control such a system over a wide range of operating conditions (40–100% turn-down operation) without formation of any soot. The same concept of injecting liquid fuels directly into the reaction chamber is also reported by Docter et al. for diesel and gasoline [13] and Roychoudhury et al. for kerosene and diesel [14].

Another, very common approach to mixture preparation with diesel that avoids problems associated with direct evaporation of diesel is to first mix diesel and water and then evaporate the two-phase mixture, as described by Pereira et al. [15], Palm et al. [16], Cheekatamarla and Lane [17], and Sgroi et al. [18] for autothermal reforming of diesel.

Another concept, that completely avoids the problem of blending liquid diesel with gaseous reactants, is the reforming of diesel in supercritical water as reported for instance by Taylor et al. [19] and Pinkwart et al. [20]. While the first team reports difficulties when supplying diesel to their reactor, the latter states that their process does not show any coke formation attributed to low temperatures.

All these aforementioned concepts have in common that they rely on water as one of the reactants. However, in case of partial oxidation for fuel cell applications and ICE exhaust gas treatment or total oxidation for combustion applications, no water is available. For those applications, different methods have been published. A widely investigated way of mixture preparation is the “cool flame” concept reviewed in a very comprehensive paper by Naidja and his co-workers [21]. The cool flame concept uses the effect that sub-stoichiometric mixtures of fuels and air

react chemically producing weak lightly blue flames, so called “cool flames” which release very little heat. The energy released through the partial oxidation of the fuel causes the molecules to break down and recombine producing a variety of stable organic compounds, including alcohols, peroxides, aldehydes and carbon monoxide. The most recent developments by Hartmann et al. [22] applied this concept for mixture preparation to reforming processes, e.g. partial oxidation [23].

Part I of this paper describes a novel, proprietary, very simple process for direct evaporation of diesel by radiative heat exchange with the energy being provided by partial oxidation of the diesel. First experimental results are presented including a map with operating regimes for various diesel and air flow rates. The process can be designed for pure evaporation with and without the presence of water. Thus, it can be applied for mixture preparation in combustors, reforming systems for fuel cell applications and for ICE exhaust gas after-treatment for emission control. A patent for this process has been granted [24].

In Part II, this paper describes how a partial oxidation catalyst (CPOX) was integrated into the evaporator along with static mixing devices to enhance mixture preparation. First experimental results with varying the air flow to the CPOX reactor were obtained and interpreted. No carbon formation was observed, neither in the evaporator nor in the CPOX reactor.

2. Part I

2.1. Process description of the evaporator

This novel evaporation process does not rely on external heat sources because it uses heat generated by partial oxidation of a portion of the fuel with sub-stoichiometric amounts of air. The air flow rate is characterized by the air number λ being in the range of 0.1–0.2. The air number λ is defined as the ratio of air flow supplied to the reactor divided by the air flow required for complete (total) combustion. The partial oxidation takes place on an alumina wash coat with noble metal used as a catalyst supported on a cylindrical metal mesh consisting of stainless steel. The evaporator comprises an inclined tube with the internal cylindrical catalyst. The fuel is supplied to the upper end of the evaporator, flowing down on the lower part of the tube wall as shown in Fig. 1(a). Due to heat transfer by radiation and convection from the hot catalyst support to the thin fuel film, the more volatile components of the fuel evaporate immediately and react on the catalyst surface with the air supplied to the tube, as depicted in Fig. 1(b). By this way, the fuel on the wall is completely evaporated along its way down the tube without forming any residues or coking, since there are no walls heated from outside, which can lead to the formation of crusts and coking. The catalyst is not in direct contact with liquid fuels at any time, because this would result in an accelerated deterioration of the catalyst.

For start-up the evaporator wall or the catalyst support must be preheated, either electrically or by process heat from other sources. After the appropriate start-up temperature of the catalyst (e.g. about 500 °C for diesel) has been reached, the fuel is

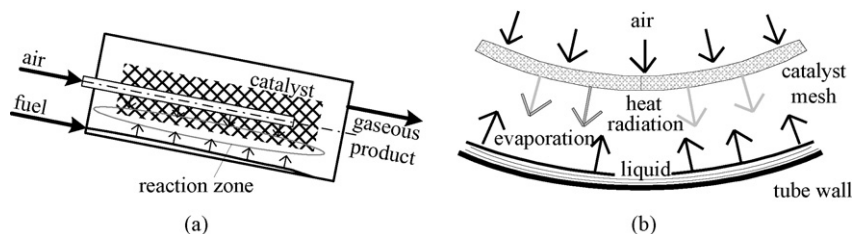


Fig. 1. Longitudinal (a) and cross (b) section of the evaporator. The diameter of the evaporator tube is 30 mm and the length 215 mm. The dimensions of the catalyst are 20 mm in diameter and 150 mm in length.

supplied to the evaporator and the external energy source can be switched off.

2.2. Experimental

The evaporator used for the investigations consists of a stainless steel tube of 30 mm inner diameter and 215 mm length inclined about 15–45° from the horizontal. On both ends stainless steel disks with the inlet and outlet ports are connected via flanges. Sulfur-free diesel (16 ppm total sulfur) is used as a fuel and supplied at the upper end of the tube right onto the bottom of the tube, with the tip of the supply tube pointed perpendicularly to the evaporator tube wall. The diesel flows downwards evaporating completely along the way.

At the center of the evaporator, connected to the upper disk, a thin tube with 6.5 mm inner diameter is located with several holes for air supply along its axis. The openings face downwards to provide air to the reaction zone where partial combustion takes place. The air inlet tube also serves to support the catalyst metal mesh.

The catalyst is finely dispersed Pt supported on a cylindrical stainless steel mesh of 20 mm inner diameter and 150 mm length with an γ -alumina wash coat.

Two possibilities to start up the evaporator have been utilized: (1) by running an electric current through the catalyst metal mesh, or (2) by heating the evaporator tube from the outside with an electric heating coil. Both methods work well with the second being the simpler, since preheating of the evaporator tube from the outside is necessary for start-up in any case.

The diesel flow to the evaporator and the air flow can be started, as soon as the catalyst has reached about 500 °C. The catalyst temperature is measured by a thermocouple attached to the catalyst mesh in the lower half somewhere in the middle between upper and lower end. During start-up the diesel flow is also pre-heated by an electrically heated supply hose. As

soon as the temperature of the catalyst has stabilized all external heat sources can be switched off. During operation the catalyst temperature can be controlled by the air flow to the evaporator.

For analysis the gaseous product leaving the evaporator is cooled down in an air cooler and the condensate is separated from the gas stream in a flash drum (please refer to Fig. 2). Since the gas downstream of the flash drum still contains aerosol-like droplets that look like a mist it became necessary to supply the gas stream to a toluene filled bubble column to retain the fine droplets for analysis. Both, the condensate after the air cooler and the loaded toluene were weighed and, after completion of the test run, analyzed by an external lab. From the clean gas leaving the bubble column a small stream was supplied to a gas chromatograph for analysis of its composition.

2.3. Results and discussion

In a first set of experiments the operating conditions for complete evaporation of the diesel feed were determined varying the diesel mass flow and the air mass flow to the evaporator. To determine whether the evaporation was complete a transparent Teflon tube was connected to the lower end of the evaporator to detect any liquids leaving the apparatus. The operating conditions for complete evaporation can be related to the catalyst temperature. For operating conditions where the catalyst temperature is above a critical temperature of about 460 °C no liquid flow from the evaporator was detected at any time. Fig. 3 shows the catalyst temperature plotted against the diesel feed to the apparatus with the air number being the parameter for the curves. The diesel flow rate to the evaporator is expressed as the energy flow rate based on the LHV of diesel (i.e. 43,000 kJ kg⁻¹), the air flow rate is characterized by the air number λ . With increasing air number the catalyst temperature increases since more diesel is burned causing a higher heat release. Because heat losses from the system depend on the surfaces temperature of the insulated

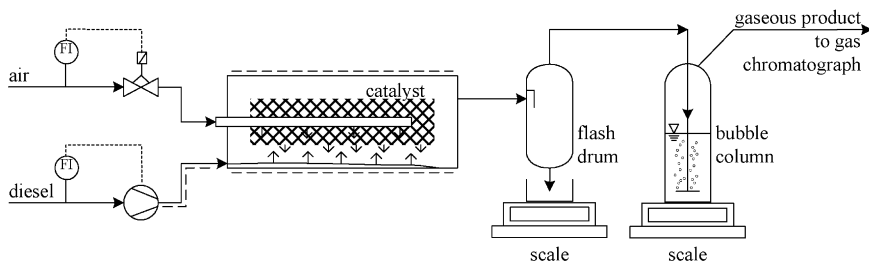


Fig. 2. Process flow diagram of evaporator test rig.

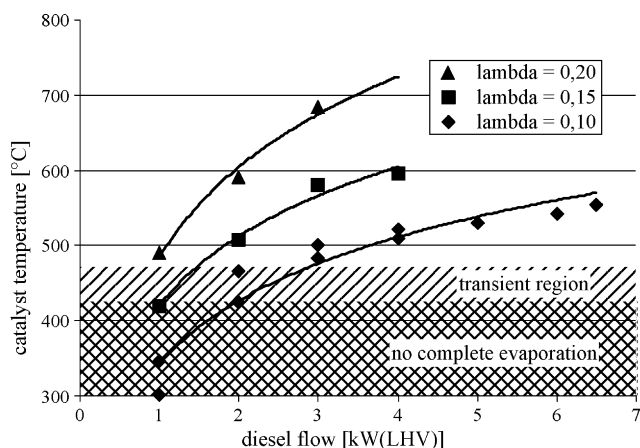


Fig. 3. Operating regimes for diesel: at catalyst temperatures of above 460 °C completed evaporation is observed at all times, below 420 °C the evaporation is not complete.

tube and, therefore, are almost constant, the catalyst temperature rises. An increase in diesel flow rate also increases the heat release inside the evaporator and yields an increase in catalyst temperature.

In the transient region of catalyst temperatures between 420 and 460 °C complete as well as incomplete evaporation has been observed. The reason for this has not been clarified yet. Below catalyst temperatures of 420 °C the diesel supplied to the evaporator is not transferred into the gaseous phase completely. The final boiling point (FBP) of the diesel used for the investigations was 390 °C.

The air flow to the evaporator should be controlled such that the catalyst temperature is between 500 and 800 °C at all times. The actual supplied air flow depends on the diesel flow rate and the insulation of the system. The catalyst temperature has to stay above the lower critical temperature to ensure proper operation of the evaporator with no liquids leaving the apparatus, and below an upper critical temperature of about 800–900 °C to prevent catalyst deterioration.

In a subsequent set of experiments the influence of load changes on the evaporator performance was investigated. In Fig. 4 the catalyst temperature is plotted versus time for a run where the diesel (energy) flow rate was changed from 6.5 down

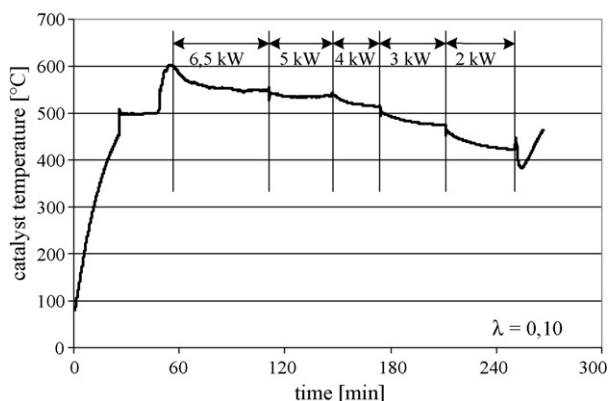


Fig. 4. Catalyst temperature over time for various diesel flow rates (expressed as energy flow rates) at a constant air number λ of 0.10.

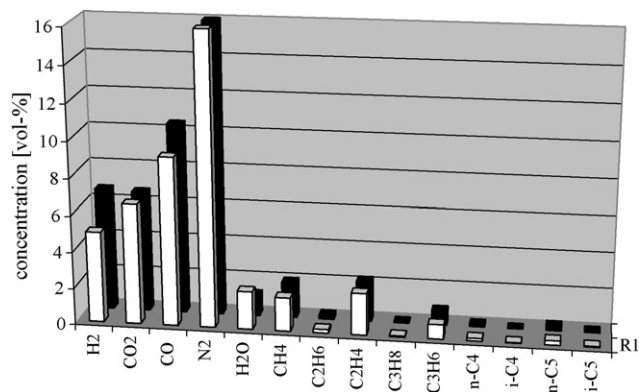


Fig. 5. Product gas composition for operation of the evaporator at 6 kW and λ of 0.14 determined by gas chromatography. The higher hydrocarbons were analyzed separately. The concentrations of nitrogen (N₂), cut off at 16 vol.%, are in the range of 66 vol.%.

to 2.0 kW while the air number λ was held constant at 0.10. After an initial start-up phase of about 60 min the diesel feed to the evaporator was started and all external heat sources were switched off. Then, the diesel feed to the evaporator was reduced in small steps. It takes about 30 min to reach a new steady state. At all times, even during the transition to a new steady state, the evaporation was complete, i.e. no liquids were leaving the apparatus.

As a next step the composition of the product gas leaving the evaporator was analyzed for conditions where the evaporation was complete. Fig. 5 shows the gas composition for a test run at 6.0 kW diesel feed and an air number λ of 0.14, measured during one test run in intervals of about one hour. The dominant component is nitrogen with a concentration of about 66 vol.%, cut off in Fig. 5 at 16 vol.%. The nitrogen is carried in by the partial oxidation air. Significant amounts of hydrogen, carbon monoxide and carbon dioxide in the order of 5–10 vol.% have been observed. These components indicate that partial oxidation of higher hydrocarbons occurs under heat release. The energy released initiates thermal cracking of the hydrocarbon chains, which yields methane and alkenes, like ethene and propene as reaction products. These three components can be found to smaller extent (between 1 and 2.5 vol.%) in the gaseous reaction products. Both, partial oxidation and thermal cracking lead to a reduction in mass flow rate of the liquid reaction products and shift its composition towards lighter hydrocarbons. The first conclusion is confirmed by a mass balance, which reveals that about one quarter of the mass of the diesel feed is converted into gaseous reaction products. The composition of the remaining liquid products was analyzed, the result of which is shown in Fig. 6.

In Fig. 6 the compositions of diesel and the “liquid” evaporator product, i.e. the mixture of the flash drum and the bubble column liquid products, are plotted as molar concentrations for various hydrocarbon fractions between C7 and C33. The toluene has been subtracted from the analysis results. The curve for the reaction products confirms that the “liquid” evaporator product is lighter than the diesel feed, i.e. the curve is shifted towards lighter hydrocarbons while the content of hydrocarbons of C11–C13

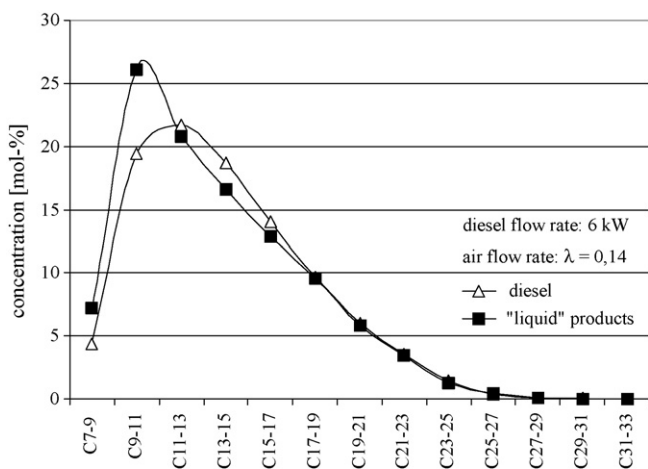


Fig. 6. Analysis of the "condensable" reaction products.

and heavier has decreased. The hydrocarbons above C17–C19 do not change significantly.

3. Part II

3.1. Description of the evaporator-cum-partial oxidation reactor and experimental procedure

After having obtained a good understanding of the processes inside the evaporator and after having determined the range of operating parameters required for a complete evaporation, a partial oxidation catalyst was installed right downstream of the evaporator by adding a second tube, a little shorter than the evaporator tube, as depicted in Fig. 7. In addition, static mixing devices were added to enhance mixture preparation. This step proved to be important to avoid carbon formation and to achieve complete fuel conversion (patent pending).

The catalyst used for the tests was a platinum based CPOX catalyst wash-coated on a ceramic monolith. The monolith was 35 mm in length and 28 mm in diameter. In the test-rig, the reformer air to the CPOX catalyst is provided through a tube that penetrates the core of the catalyst monolith, as shown in Fig. 7. Other ways of supplying the air are also feasible, e.g. feeding the air from the side or from an extension of the evaporator air tube.

For start-up the entire evaporator-cum-reformer was heated from the outside by two electrical heating coils, one for each

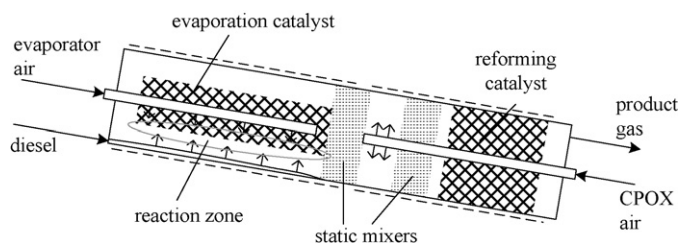


Fig. 7. Sketch of the evaporator-cum-CPOX reformer. Air is supplied separately to the evaporator and the CPOX reformer. The diameter of the evaporator-cum-reformer tube is 30 mm and the length 365 mm. The dimensions of the reformer catalyst are 28 mm in diameter and 35 mm in length.

section. Depending on the heat input of the heating coils, start-up times between 10 and 30 min could be achieved. As soon as the evaporator catalyst reached 400 °C and the CPOX catalyst 500 °C, the diesel flow and both air flows were started. Within seconds after switching on the feed streams the temperature of the evaporator catalyst increased to about 500–550 °C and the temperature in the CPOX catalyst to about 700–900 °C depending on the air flow rates. Both air flows were controlled independently from each other by mass flow controllers.

To minimize heat losses and to increase system efficiency at the same time, the entire evaporator-cum-reformer was insulated with mineral wool.

3.2. Results and discussion

In a first set of experiments the performance of the evaporator-cum-reformer was investigated by measuring the product gas composition and the catalyst temperatures and by inspecting the inside of the apparatus for carbon formation. The diesel feed was kept constant at 1.7 kW (LHV diesel) for this set of experiments. The total air flow was in the range of $\lambda = 0.37$ –0.48, with about 35–50% being supplied to the evaporator, and the balance to the reformer. The reformer gas composition of the product gas was analyzed by gas chromatograph, with mass balances closing to within 3%.

The minimum air flow for a partial oxidation process must exceed an air number of about 0.33 (i.e. $O_2/C = 0.5$), following a consideration by Lutz et al. [25] when just the carbon of the fuel is oxidized and the hydrogen is released. This minimum air number was determined by using $C_{13}H_{26}$ as a diesel surrogate fuel, because it was available in the process simulator's data base (ChemCad[®]) and seems close to the average diesel composition of $C_{12.95}H_{24.38}$, as reported by Amphlett et al. [26].

At the end of each test run the evaporator-cum-reformer was disassembled and the inside was checked for carbon deposits and tar-like residues. However, the inner walls and all the internals proved to be clean at all times.

Fig. 8 shows the product gas composition for constant diesel flow and various air flows. It turned out that the air number for the given diesel flow rate (1.7 kW) has to be at least 0.4 to achieve complete diesel conversion. This was deduced from the fact that at air numbers below 0.4 the product gas was still a slightly aerosol like gas, similar to the product gas from the evaporator, as stated in Part I.

In order to present all gas components in one diagram, the concentrations are plotted using a logarithmic axis. The concentrations shown are wet concentrations including water. In order to obtain accurate values for water all the connection tubes were heated to above 150 °C. As can be seen in Fig. 8, the air flow does not have a significant influence on the concentration of major components, like nitrogen, hydrogen, carbon monoxide and water.

The concentrations of hydrocarbons up to pentane were determined as well. While no pentanes were measured in the product gas, trace amounts of C1, C2's, C3's, and C4's were found. Especially, ethylene and propylene concentrations are in the order of tenths of a percent, which indicates that some cracking is still

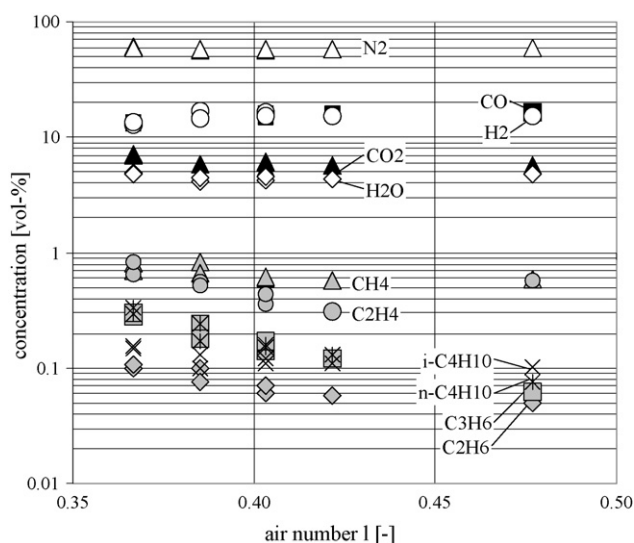


Fig. 8. Product gas composition for operation of the evaporator-cum-CPOX reformer at 1.7 kW and λ of 0.37–0.48, determined by gas chromatography.

occurring inside the catalyst or that some of the unsaturated light hydrocarbons formed in the evaporator were not converted in the CPOX reactor. On the other hand, it can be deduced from Fig. 8 that a higher air number, which corresponds with a higher catalyst temperature, decreases the amount of hydrocarbons in the reformer product gas flow.

At the high air number λ of 0.48, the reformer catalyst temperature measured by thermocouples was 790 °C and the evaporator catalyst temperature was 520 °C. At $\lambda = 0.37$ the catalyst temperature only was 720 °C. Both, evaporator and reformer catalyst temperatures were controlled independently from each other by the air flow rate to the respective section of the apparatus.

For comparison a commercial process simulation software (ChemCad[®]) was used to calculate the thermodynamic equilibrium for a temperature of 790 °C. As a reference point the measurements obtained at a diesel feed corresponding to 1.7 kW (LHV diesel) and an air number λ of 0.48 were utilized. In Table 1 measured and calculated gas concentrations are com-

Table 1
Comparison of measured (gas chromatograph) and calculated (ChemCad[®]) evaporator-cum-reformer gas composition for 1.7 kW (LHV) diesel feed and 0.48 air number

Component name	Measured concentration (vol.%)	Calculated concentration (vol.%)
CO	16.1	16.3
CO ₂	5.5	4.6
H ₂ O	4.7	4.6
H ₂	14.7	17.9
N ₂	57.6	56.6
CH ₄	0.57	0.02
C ₂ H ₄	0.56	0
C ₂ H ₆	0.05	0
C ₃ H ₆	0.06	0
C ₃ H ₈	0.01	0
<i>i</i> -C ₄ H ₁₀	0.10	0
<i>n</i> -C ₄ H ₁₀	0.07	0

The catalyst temperature measured was 790 °C.

pared with each other. The agreement for the major components with the exception of CO₂, H₂ and CH₄, is better than 2%. The trace amounts of methane and the hydrocarbons up to butane are not predicted properly. The significant amounts of methane and ethylene indicate that carbon formation within the catalyst might be occurring. This will be objective of future investigations, including Temperature Programmed Oxidation (TPO) to determine the amount of carbon deposited on the CPOX catalyst.

The problem of carbon formation in the reactor seems to be even more pronounced at lower temperatures and lower air numbers, respectively. For an air number of 0.37, the simulated gas composition deviates quite significantly from the measured one. In fact, the simulation shows that at this temperature carbon is predicted to be present among the reaction products. This is in agreement with the observation stated earlier, that at air numbers below 0.40 the gaseous product stream from the reformer was no longer clear and resembled the aerosol-like evaporator product.

For the operating point at 1.7 kW (LHV diesel) and $\lambda = 0.48$, the conversion efficiency (lower heating value of product gas divided by lower heating value of diesel feed) was calculated to be 78%. This value can be even further increased by using the hot reactor product gas to preheat the feed streams (heat integration) and by a better heat insulation.

4. Conclusions

A novel, very simple evaporation process, which promises significant advantages for combustion and reforming processes, has been investigated. Both combustion and reforming have in common that mixture preparation is the crucial process step, which can be facilitated if the fuel is evaporated prior to being mixed with the air and steam, the latter if present. In case of diesel, evaporation has not been an option until recently, because diesel forms residues and carbon deposits when evaporated over hot heat exchanger surfaces.

The novel process, presented in this paper, transfers hydrocarbon fuels, including diesel, into the gaseous phase by radiative heat transfer from a hot catalyst supported on a metal mesh, thus avoiding contact of the diesel with a hot metal surface. The generated diesel vapor can then be supplied to a combustion chamber or – as described in the second part of this paper – to a reforming reactor.

The performance of the evaporator has been investigated and a map derived that shows operating conditions where the fuel is completely transferred into the gaseous phase. The catalyst temperature is a very simple indicator for proper operation of the evaporator: as long as it is above 460 °C (in case of diesel), the evaporation is complete. The evaporator can be started very easily by electrical preheating. The enthalpy of fuel vaporization is provided by the process itself since about 10% of the fuel are partially oxidized.

In a second step, a catalyst for partial oxidation was added downstream of the evaporator and successfully operated with the evaporator. The evaporator-cum-reformer can be started up by electrical heating from the outside, like the evaporator. In a first set of experiments, it was operated at a constant diesel flow rate with varying air flows. No carbon formation was observed

inside the reactor at any time, however the small amounts of unsaturated hydrocarbons (in the order of up to 0.6 vol.%) in the product gas indicate that some carbon formation occurs within the catalyst for low air numbers and low reforming catalyst temperatures. To further back up this observation and to evaluate how severe this problem is, it is planned to determine the amount of carbon deposited on the catalyst surface by temperature programmed oxidation (TPO). Further objectives of future investigations are the influence of sulfur on the performance and tests with kerosene and gasoline.

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