

Glide-Arc Stabilization of Lean Methane-Air Combustion

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Abstract—Technical application of combustion processes often requires stability and low emissions of noxious compounds in a certain range of fuel-air stoichiometry. An increase of the combustion stability especially near the lean blow-off limit is desirable because this enables to run the combustion process with very low NO_x emissions. Thus we investigated plasma stabilization of premixed methane-air combustion using a miniaturized glide-arc reactor integrated into a Bunsen type burner for plasma-activation of fuel-steam mixtures: The plasma reactor consisted of a concentric electrode system forming an annular discharge gap, which was varied from 0.7 mm to 1.4 mm. A DC power supply (0-6 kV, 0-300 mA) was connected to the electrode system with a current limiting resistor in series. Feed velocities of the preheated methane-steam mixture ranged from 2.0 m/s to 15.5 m/s. Plasma stabilization performed with a discharge gap of 1.4 mm resulted in an increase of the lean blow-off air number from 1.8 to 2.4. Due to the more lean combustion the NO_x-emission decreased from 3 ppm to 1 ppm.

Keywords—Glide-arc, methane, steam reforming, lean combustion.

I. INTRODUCTION

Technical application of combustion processes often requires stability and low emissions of noxious compounds in a certain range of fuel-air stoichiometry. However, normally technical combustors are optimized for a well specified fuel and a certain range of thermal loads. Emissions increase and combustion stability decreases if they run outside of the load limits for which they are designed or are operated with a fuel deviating from the specified one in properties such as the flammability limit, the Wobbe index, or the laminar flame velocity as a function of air number [1]. Fuel flexibility, however, is highly desirable in order to enable utilization of fuels generated from biomass in combustors being designed for fossil fuels and vice versa. If in a grid for electrical energy distribution the average fraction fed by renewable sources such as wind or solar energy exceeds some 10 percent, load flexibility of the conventional, thermal power plants is an important task: Here an increase of the combustion stability of combined cycle power plants especially near the lean blow-off limit is desirable. This does not only help to increase the lean blow off air number but also enables – if done in the right way – to run the combustion process with very low NO_x emissions. State of the art is stabilization of premix combustion of natural gas with air by means of a non-premix pilot flame. The high temperature of the pilot flame causes NO_x-emissions which are unavoidable due to the Zel'dovich mechanism [1]. One way of combustion stabilization demonstrated recently is

secondary fuel injection [2]. Another way to achieve combustion stabilization avoiding the drawbacks of a pilot flame or secondary fuel injection could be electric field control of the combustion process [3], [4], [5]. This process has recently been investigated in more detail, especially regarding scalability towards technical applications, where increased pressures and feed velocities of fuel and air are important [6]. The main advantage of electric field control of combustion processes is the very low power consumption being substantially below 0.1 % of the thermal power of the combustor. Nevertheless, in order to generate the electric field in regions being relevant for combustion stabilization an electrode needs to be implemented in the combustion chamber. Further, as already pointed out in [3], the hydrodynamic effects of an electric field stabilizing the flame are limited if formation of a self sustained gas discharge shall be avoided. However, the stability limit and the emissions of a combustor can much stronger be influenced by means of plasma chemical processes activating at least one of the feed gases of the combustor [7], [8]. Several process schemes been reported in the literature such as dielectric barrier discharge (DBD) activation of the fuel [9], DBD assisted diffusion combustion [10], plasma assisted premix combustion applying ultra-short pulse repetitive discharges inside of a combustion chamber [11], or rotating discharge assisted partial oxidation of fuel-air mixtures [12]. However, in the case of plasma activation of pure fuel the challenge is to avoid soot-formation, and in the case of plasma activation of fuel-air mixtures we observed an increase of NO_x-emissions due to plasma induced NH₃-formation in the fuel-rich gas mixture [13]. In [11] plasma induced formation of NO_x inside of the combustion chamber was observed.

Thus we investigated plasma stabilization of premixed methane-air combustion using a miniaturized

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glide-arc reactor integrated into a Bunsen type burner for plasma-activation of fuel-steam mixtures.

II. EXPERIMENTAL

A General procedure

Two series of experiments were performed: In the first set of experiments the miniaturized glide-arc reactor was characterized. The electrical characteristics and the plasma-chemical conversion of methane-steam mixtures were evaluated for different combinations of experimental parameters such as total gas flow rate, gas mixture (H_2O/CH_4 -ratio), discharge gap of the plasma reactor, and plasma input power. Optimum operation parameters were identified. In the second set of experiments the plasma reactor was integrated into the burner and plasma stabilization of the flame was investigated. Here the plasma reactor was operated using the optimized discharge gap, and parameters such as the total fuel-air ratio, the H_2O/CH_4 -ratio of the plasma reactor, and the plasma input power were varied. In order to evaluate the influence of this large number of factors on combustion stability performing a limited number of experiments, Gen'ichi Taguchi's "Design of experiment" (DoE) method was applied [14]: Independent control parameters (factors) were identified and varied in such a way (steps) that a minimum set of well balanced experimental situations being characterized by an orthogonal array was obtained (see Table I below). For details about this method the reader is referred to text books or professional software guiding the user step by step through the procedure of experiment planning and evaluation.

B Experimental set-up

The experimental set-up (Fig. 1) consisted of a gas supply system, a heated reactor vessel being equipped with the plasma reactor integrated into the burner, a gas probe for extraction of product gas samples, a condenser for de-humidification of the sampled gas, and an Fourier transform infrared absorption spectrometer (FTIR, Perkin Elmer System 2000 equipped with a 1 m White cell heated to 180 °C) and a gas chromatograph (GC, Shimadzu model 14B equipped with columns packed with the following materials: HayeSep R, HayeSep R/Q, and molecular sieve 13x).

The gas mixtures were supplied to the experiment from a centralized gas supply for N_2 and O_2 and pressurized cylinders (CH_4) by means of a gas mixing manifold equipped with MKS mass flow controllers. A constant flow of steam was generated from de-ionized water using a pump pressurizing the water to about 300 kPa, a Bronkhorst mass flow controller for liquids, and a self-manufactured evaporation module being heated to $T > 150$ °C. For good mixing of methane and steam and improved evaporation the methane required in the plasma

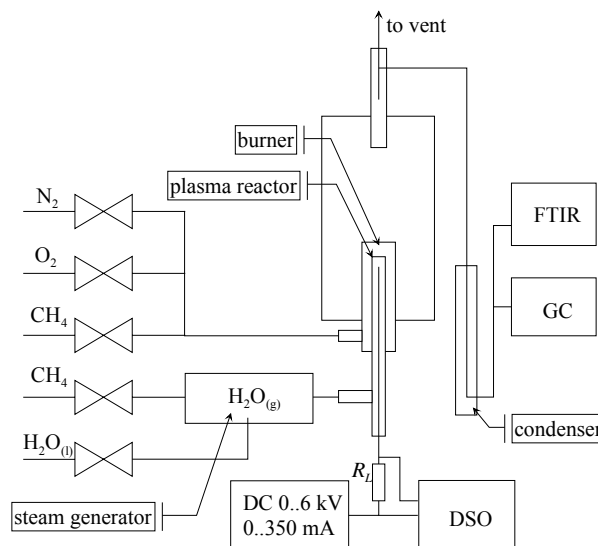


Fig. 1. Experimental set-up

reactor was also fed to the evaporation module. Flow-down of this module all lines and the reactor vessel were heated to 150 °C.

The de-humidified product gases were analyzed quantitatively for CH_4 , CO , CO_2 , NO , NO_2 , and NH_3 using the FTIR, and for N_2 , O_2 , H_2 , CH_4 , and C_2 -hydrocarbons using the GC. Under some conditions an electrochemical sensor system (IMR 3000 P with exhaust gas dryer IMR 500 P) was applied for convenient measurement of NO -, NO_2 -, CO -, and O_2 -concentrations. The feed-gas of the plasma-reactor was analyzed using the FTIR without the condenser.

C Plasma Reactor

The plasma reactor consisted of a concentric electrode system forming an annular discharge gap, which was varied from 0.7 mm to 1.4 mm by exchanging the cathode insert (Fig. 2). A DC power supply (Elektro-Automatik EA-HV 9000-6K-2000, 0-6 kV, 0-300 mA) was connected to the electrode system applying a current limiting resistor R_L of 14 kOhms in series.

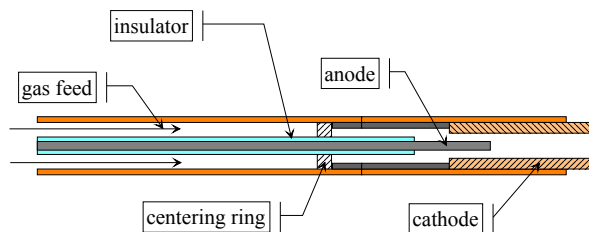


Fig. 2. Plasma reactor for treatment of methane-steam mixtures by means of gliding arc discharges.

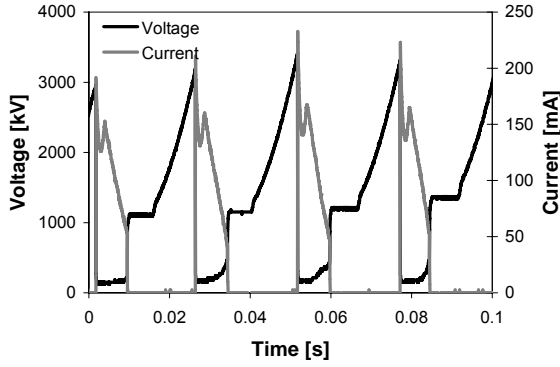


Fig. 3. Reactor voltage and current. Operation parameters of the plasma reactor: Discharge gap 1.1 mm, feed flow rate 2 slm, molar $\text{H}_2\text{O}/\text{CH}_4$ -ratio 2.0, average current limited by the power supply to 35 mA.

The reactor voltage U_R and the voltage drop

$$U_L = U_0 - U_R \quad (1)$$

at R_L (Fig. 3) were recorded on a digital storage oscilloscope (DSO; Lecroy Waverunner 9304) using high voltage probes (Lecroy PPE 20kV). The reactor current was evaluated according to

$$I_R = U_L / R_L \quad (2)$$

The pulsation of the gas discharge seen from the electrical signals in Fig. 3 fits well to visual observations of a rapidly moving arc-like discharge. The frequency of pulsation ranging from 24 Hz to 50 Hz is influenced by various factors such as the gas flow velocity of the methane-steam mixture in the discharge gap ranging from 2.0 m/s to 15.5 m/s.

For the evaluation of energy related quantities such as energy costs of plasma-chemical hydrogen generation an average plasma input power was evaluated according to

$$P_{average} = \frac{1}{T} \cdot \int_0^T U_R(t) \cdot I_R(t) \cdot dt \quad (3)$$

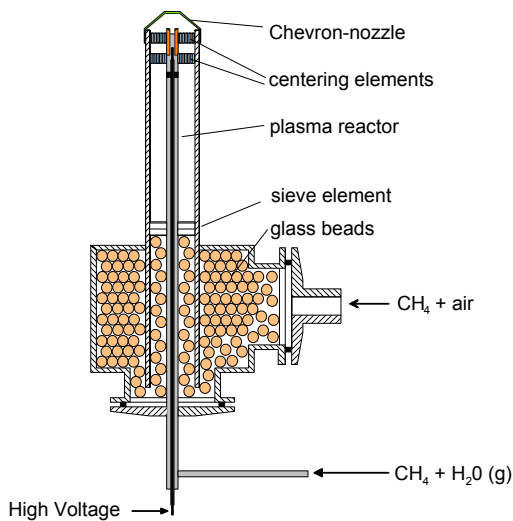


Fig. 4. Set-up of the burner with the integrated plasma reactor.

where T is the period of discharge pulsation. This average plasma input power was varied between 5 W and 15 W. The most convenient way of doing this was by setting the current limitation of the power supply to a certain value corresponding roughly to an average reactor current $I_{average}$.

D Plasma stabilized burner

The burner (Fig. 4) consisted of a cylindrical tube to which a lean methane-air mixture was fed from the bottom through a bed of glass beads. On top of the burner tube a Chevron nozzle was mounted in order to facilitate mixing of the product gas of the plasma reactor centered inside of the burner tube with the lean methane-air mixture.

III. EXPERIMENTAL RESULTS

A. Characterization of the plasma reactor

In order to characterize the plasma reactor, the total gas flow rate $Q_{Vplasma}$ fed to the plasma reactor, the molar ratio of $\text{H}_2\text{O}/\text{CH}_4$ γ_{steam} , and the average reactor current I_{av} were set to the values given in Table I (factors of influence and steps).

Discharge gaps d_{gap} of 1.4 mm, 1.1 mm, and 0.7 mm were investigated by selecting a cathode insert with an inner diameter of 4.3 mm, 3.6 mm, and 2.9 mm, respectively. The diameter of the anode wire was kept constant at 1.5 mm. The anode wire overlapped with the cathode insert for about 7 mm, the length from the wire tip to the outlet of the plasma reactor was about 18 mm. It turned out very soon that stable operation of the plasma reactor was not possible at a discharge gap of 0.7 mm. The 1.1 mm discharge gap showed slightly better results than the 1.4 mm gap. However, because of better long term stability of the reactor the 1.4 mm gap was the preferred one for plasma assisted combustion experiments. Thus in the following only results obtained for a discharge gap of 1.4 mm are shown, where the flow velocity in the discharge gap ranged from 2.0 m/s to 5.9 m/s..

TABLE I
FACTORS INFLUENCING PLASMA-CHEMICAL CONVERSION OF METHANE-STEAM MIXTURES AND VALUES (STEPS) USED IN EXPERIMENTAL INVESTIGATION.

Factor/Step	1	2	3
I_{av} [mA]	35	43	50
γ_{steam} [-]	1.5	2.0	2.5
$Q_{Vplasma}$ [slm]	1.0	2.0	2.9

Variables of interest depending on the factors $Q_{Vplasma}$, γ_{steam} , and I_{av} were the CH_4 -conversion X_{CH_4} , the H_2O -conversion $X_{\text{H}_2\text{O}}$, the yields hydrogen, CO, CO_2 ,

C₂H₆, C₂H₄, and C₂H₂ (Y_{H_2} , Y_{CO} , Y_{CO_2} , $Y_{C_2H_6}$, $Y_{C_2H_4}$, $Y_{C_2H_2}$) the average plasma input power P_{av} , and derived quantities such as specific input energy density (ratio of P_{av} and $Q_{vplasma}$) and the energy costs for H₂-generation.

An example of the results at intermediate average current and flow rate is given in Fig. 5. Under these conditions the concentrations of C₂-hydrocarbons are fairly low ($[C_2H_6] = 777$ ppm, $[C_2H_2] = 105$ ppm) compared to those of CO and CO₂.

The traditional analysis of the plasma chemical conversion of CH₄ to H₂ as a function e.g. of specific input energy density does not give a clear indication of the parameter dependencies (Fig. 6). In order to evaluate the influence of factors, the results of 18 experiments covering 9 different combinations of the three factors $Q_{vplasma}$, γ_{steam} , and I_{av} were analyzed using Taguchi's DoE method. For the hydrogen flow rate the results are given in Fig. 7. The influence of the factors $Q_{vplasma}$ and I_{av} is statistically significant (confidence levels > 95 %), whereas for the factor γ_{steam} the confidence level is slightly below 95 %. In the same way we found that the energy costs of H₂-generation are significantly influenced by γ_{steam} and $Q_{vplasma}$ (Fig. 8).

The energy costs of H₂-generation are in good agreement with those ones obtained with a repetitive spark plasma reactor or with plasma-catalytic hybrid reforming using a commercial Ni-catalyst [15].

B. Plasma assisted combustion

In order to evaluate the influence of the plasma control on the combustion stability and emissions, the operation parameters of the plasma reactor (γ_{steam} , and I_{av}) and the methane flow rate of the burner $Q_{vburner}(CH_4)$ were fixed, and the air flow rate of the burner $Q_{vburner}(air)$ was varied. From these parameters the total air number of the plasma assisted burner

$$\lambda = \frac{0.21}{2} \cdot \frac{Q_{vburner}^{air}}{Q_{vburner}^{CH_4} + Q_{vplasma} / (1 + \gamma_{steam})} \quad (4)$$

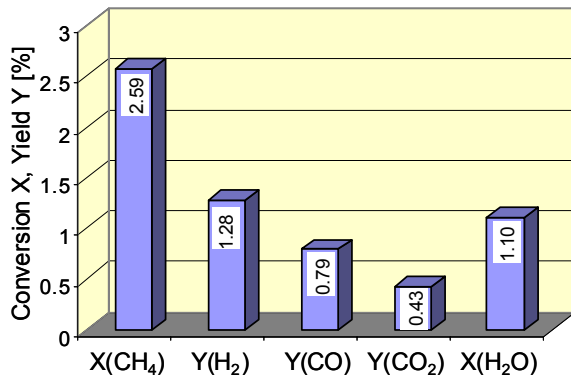


Fig. 5. Conversion of CH₄ and H₂O and yields of H₂, CO, and CO₂. Experimental conditions: $d_{gap} = 1.4$ mm, $Q_{vplasma} = 2$ slm, $\gamma_{steam} = 1.5$, and $I_{av} = 43$ mA

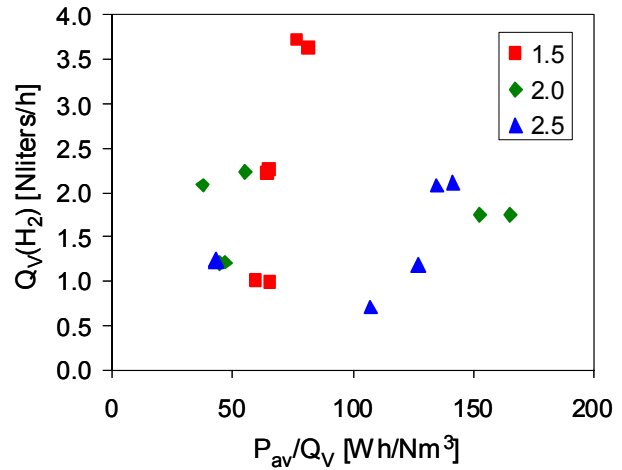


Fig. 6. H₂-flow rate due to plasma-treatment of H₂O/CH₄-mixtures as a function of the specific input energy density for different H₂O/CH₄-ratios ($d_{gap} = 1.4$ mm).

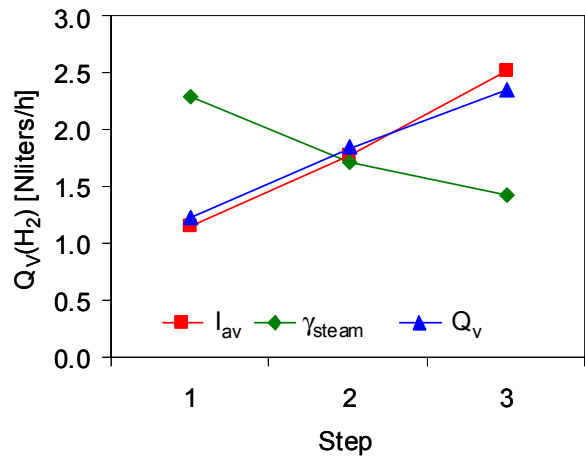


Fig. 7. Influence of factors (steps see 0) on the H₂-flow rate evaluated from 18 experiments using Taguchi's DoE method (results of real value analysis), $d_{gap} = 1.4$ mm.

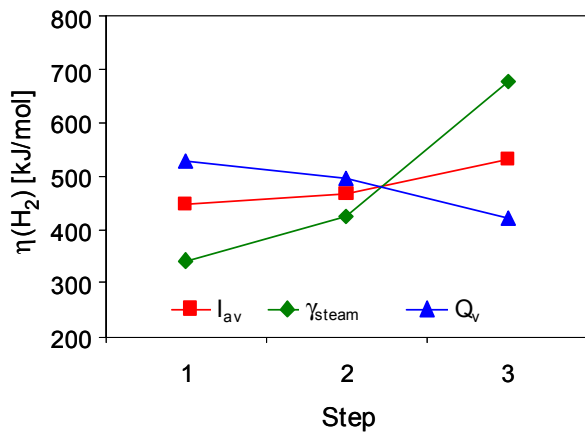


Fig. 8. Influence of factors (steps see 0) on the energy costs of H₂ generation evaluated from 18 experiments using Taguchi's DoE method (results of real value analysis), $d_{gap} = 1.4$ mm.

was evaluated being used for the characterization of the results given in Fig. 9 and Fig. 10. The total gas flow rate of the plasma reactor $Q_{Vplasma}$ and the discharge gap were kept constant at 1.0 slm and 1.4 mm respectively. As a reference the burner was characterized (a) with the H_2O/CH_4 -mixture flowing through the plasma reactor without plasma application (0 mA) and (b) without a gas flow through the plasma reactor. In case (b) the methane flow rate of the burner was increased for the methane flow rate of the plasma reactor in order to have identical thermal power, and a nitrogen volume flow rate being equal to the steam flow rate of the plasma reactor was added to the feed flow of the burner in order to have identical total flow rates. Detailed information about the experimental conditions is given in Table II.

TABLE II
CONDITIONS INVESTIGATED
FOR PLASMA ASSISTED COMBUSTION

Conditions	1	2	reference	
d_{gap} [mm]	1.4			
I_{av} [mA]	0, 35, 50		0	0
γ_{steam} [-]	1.5	2.5	1.5	2.5
Plasma reactor flow rates				
$Q_{Vplasma}$ [slm]	1.00	1.00	0	0
$Q_{Vplasma}^{CH_4}$ [slm]	0.40	0.29	0	0
$Q_{Vplasma}^{H_2O}$ [slm]	0.60	0.71	0	0
Burner flow rates				
$Q_{Vburner}$ [slm]	9.57	8.31	9.97	8.59
min / max	19.11	16.54	20.11	17.54
$Q_{Vburner}^{CH_4}$ [slm]	0.40	0.40	0.80	0.69
$Q_{Vburner}^{air}$ [slm]	9.17	7.91	9.17	7.91
min / max	18.71	16.14	18.71	16.14
$Q_{Vburner}^{N_2}$ [slm]	0	0	0.60	0.71
v_{nozzle} [m/s] ^a	0.9 to 2.1			

a) The feed flow rate was estimated at the cross section of the Chevron nozzle. The following temperatures were assumed: Plasma gas flow 550 K, burner feed flow 330 K.

First of all it can be seen that the stability range of the burner is very sensitive to the total CH_4 flow rate. It needs to be noted that in the case of $\gamma_{steam} = 1.5$ for all the experiments the total CH_4 flow rate

$$Q_{Vtotal}^{CH_4} = Q_{Vburner}^{CH_4} + Q_{Vplasma} / (1 + \gamma_{steam}) \quad (5)$$

was slightly higher (0.8 slm) than in the case of $\gamma_{steam} = 2.5$, where the total CH_4 flow rate was 0.69 slm. This resulted in a substantial larger lean blow-off air number of the burner without plasma assistance (see curves marked with "0 mA, 0 slm"). In this case the burner emits small CO-concentrations for $\lambda < 1.5$. For $\lambda > 1.5$ the CO-emission increases strongly with λ . This is due to the decrease of the adiabatic flame temperature resulting in less complete oxidation of intermediate combustion products.

Addition of steam to the feed gas resulted also in an increase of the CO-emission for the same reason: The steam acts as thermal ballast reducing the adiabatic flame temperature at constant λ . Surprisingly, the addition of steam does not result in a substantial decrease of the lean blow-off air number λ_{lbo} as compared to the burner operation in the corresponding reference case.

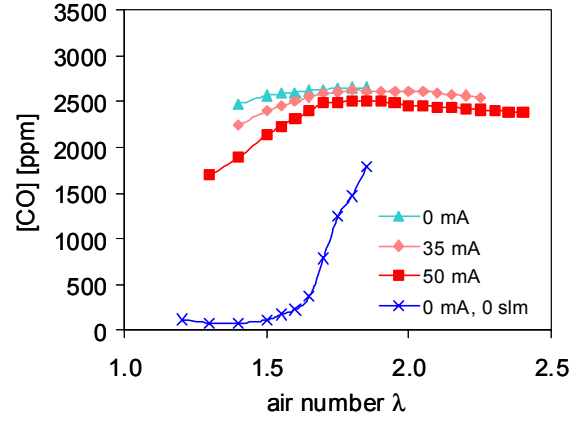


Fig. 9. CO-emissions as a function of total air number. Experimental conditions: 0 mA, 35 mA, 50 mA – gas flow through plasma reactor with $\gamma_{steam} = 1.5$; 0 mA, 0 slm – no gas flow through plasma reactor.

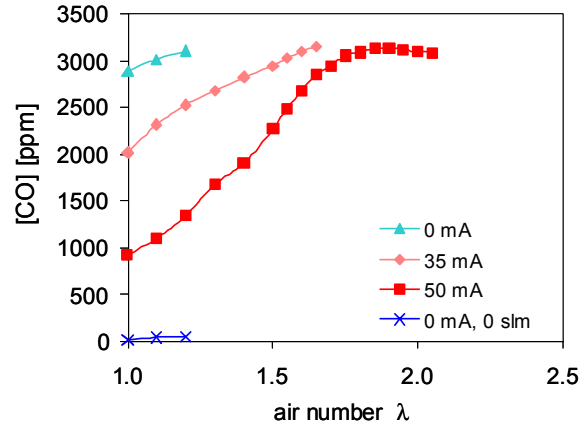


Fig. 10. CO-emissions as a function of total air number. Experimental conditions: 0 mA, 35 mA, 50 mA – gas flow through plasma reactor with $\gamma_{steam} = 2.5$; 0 mA, 0 slm – no gas flow through plasma reactor.

When the plasma is switched on, a decrease of CO-emission with increasing current I_{av} is observed for both γ_{steam} -values. Thus it can be concluded that the CO generated in the plasma reactor is oxidized in the flame. This may be caused by a fast reaction of the CO-molecules generated in the plasma reactor with OH-radicals, which are generated in the flame front at high rate:



Further plasma stabilization resulted in an increase of the lean blow-off air number with increasing I_{av} . This is in good agreement with the idea of lean combustion stabilization using hydrogen produced from a hydrocarbon fuel by plasma reforming published in [16]. In the case of $\gamma_{steam} = 1.5$ an average current of 50 mA results in an increase of λ_{lbo} from 1.8 to 2.4. Due to the more lean combustion the NO_x -emission decreased from 3 ppm to 1 ppm.

In the case of $\gamma_{steam} = 2.5$ an average current of 50 mA results in an increase of λ_{lbo} from 1.2 to 2.1. Due to the more lean combustion the NO_x -emission decreased

from 6 ppm to 2 ppm. This increase being strong as compared to the case $\gamma_{steam} = 1.5$ is surprising because an increase of γ_{steam} under otherwise identical conditions results in a decrease of the H₂ flow rate caused by plasma application (see Fig. 7).

It is interesting to note, that the average plasma input power was small compared to the thermal power of the combustion process. In the case of $\gamma_{steam} = 1.5$ the average plasma input powers were 12.1 W at $I_{av} = 35$ mA and 15.2 W at $I_{av} = 50$ mA, respectively, which corresponds to 2.4 % and 3 % of the thermal power of 529 W.

IV. CONCLUSIONS

A miniaturized glide-arc reactor for plasma-chemical conversion of steam-methane-mixtures was constructed and characterized using Taguchi's "Design of Experiment" method. Stable operation was achieved using a discharge gap of 1.4 mm resulting in average feed gas flow velocities between 1.8 m/s and 5.3 m/s. Depending on the average current varied between 35 mA to 50 mA, the total gas flow rate varied between 1 slm and 2.9 slm, and the molar H₂O/CH₄-ratio varied between 1.5 and 2.5, the average plasma powers ranged from 5 W to 15 W. The H₂-yields ranged from 0.8 % to 2.4 % under these conditions.

For investigation of plasma assisted combustion the miniaturized glide-arc reactor was integrated into a tubular burner being operated at a thermal power around 500 W. Plasma stabilization of the flame resulted in an increase of the lean blow-off air number of the burner from 1.8 to 2.4.

However, the steam fed to the burner by means of the glide-arc reactor resulted in a strong increase of the CO-emission. Since CO generated by plasma-chemical conversion of methane and steam was oxidized in the flame, the stability of flames could be increased without increasing the CO-emission when the fraction of gases supplied to the burner from the plasma reactor could strongly be reduced. Unfortunately due to restrictions of our laboratory scale test bench the maximum thermal power, which could be investigated, was limited to less than 1 kW. Thus we had to work with small gas flow rates. Since on the other hand a large gas flow velocity is required inside of the plasma reactor in order to get a gliding arc discharge, the fraction of gas supplied to the burner by the plasma reactor could not be reduced substantially.

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