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Plasma- and electric field control of natural gas combustion

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Abstract

The influence of electric fields on the stability and emissions of natural gas combustion was investigated at a seven-hole Bunsen burner at pressures p between 0.1 MPa and 1 MPa being representative for combustion processes in gas turbines. For the E-field generation a ring shaped high voltage electrode placed above the burner head was used. Self-sustained gas discharge formation was avoided by keeping the spatial average of the reduced electric field below 40 Td (1 Td $= 10^{-21}$ Vm^2). In this case we found that the reduced electrical current I/p is well described by a power law of the reduced voltage U/p with power 1.5. Due to the applied electric field the air number, at which lean blow off occurred, could be increased from 1.14 to 1.22 resulting in a decrease of nitric oxide emissions of more than 20%. At the same time the carbon monoxide emissions decreased by more than 90%. The electric power consumption for this effect was $< 0.02\%$ of the thermal power of the burner. Plasma combustion control was tested at a small scale atmospheric pressure laboratory set-up consisting of a single flame Bunsen type burner and an integrated gliding arc reactor centered in the cylindrical burner tube. Half of the methane fuel was subjected to plasma induced partial oxidation resulting in formation of hydrogen, carbon monoxide, and small concentrations of by-products, whereas a very lean mixture of the remaining fuel with air was directly fed to the burner tube. An increase of the lean blow off air number from 1.6 to 2.0 was achieved. Further the fuel conversion was improved from 90% to 95%.

Keywords: Combustion control; Electric field; Electro-hydrodynamics; Glide-arc; Plasma; Natural gas; Gas turbine; Lean blow off limit; Combustion stability; Emissions; Nitric oxide; Carbon monoxide

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1 Introduction

Electric power generation sets growing requirements for flexible load, high efficiency, and low emission combustion of liquid and gaseous fuels. Fuel flexibility of combustors is also of growing importance. In order to fulfill these requirements, methods allowing fast control of a combustion process are desirable. Since electric fields with sub-millisecond rise-time can easily be applied to a flame and plasma generation can be accomplished on the same time scale, both electrostatic field combustion control and plasma combustion control attracted some attention.

That electric fields can be utilized to influence the shape and emissions of a flame was discovered more than a century ago and soon related to the electrical properties of the flame [1], which were investigated in more detail in the 1920's. Investigations of electric field combustion control were intensified in the 1950's resulting in a more detailed understanding of the influence of electric fields on the gas flows in combustion processes [2], [3]. Just recently experimental investigations of the influence of electric fields on combustion under micro-gravity conditions were presented [4], and electric fields were utilized to control the combustion synthesis of nano-powders [5]. A proposal for the application of a plasma jet for combustion control can be found in [6]. Recently a comprehensive overview of plasma assisted combustion was given on an international workshop held at the Los Alamos National Laboratory [7]. Nevertheless, due to the complexity of the subject until now there is no agreement about the mechanism, neither of electric field combustion control nor of plasma combustion control. Further very few experiments covering the parameter range of technical application were published.

2 Experiments

Since stationary gas turbines for power generation run at pressures of typically 1.6 MPa we performed an experimental investigation of the potential of electric field combustion control (EFCC) for stabilization and emission reduction of methane premix combustion at pressures ranging from 0.1 to 1 MPa using a 7-hole Bunsen type burner. In order to get a more detailed understanding of EFCC, low pressure premix combustion experiments were performed at a McKenna flat flame burner [9] for 6 kPa $\leq p \leq$ 20 kPa, where the influence of the electric field onto the position of the (thermal) flame front was observed spectroscopically. Both in the high- and in the low-pressure experiments plasma-chemical effects on combustion were avoided by keeping the reduced electric field values below the limit for the formation of self-sustained gas discharges.

Plasma-chemical stabilization of lean methane combustion was investigated in atmospheric pressure experiments, where certain fractions of air and fuel were fed to a miniaturized glide-arc reactor integrated into a tubular burner.

2.1 Electric field combustion control at high pressures (0.1–1.0 MPa)

The experimental set-up consisted of a high pressure combustion chamber equipped with a 7-hole Bunsen burner, which was fed by a premixed methane-air gas flow. A ring shaped high voltage electrode was installed at a height d above the burner head connected to ground (Fig. 1), which was connected to a DC power supply (0..60 kV, 0..10 mA) by means of a current limiting resistor (12 MΩ). Current I and voltage drop at the combustion chamber U were determined from the voltages measured at the HV-electrode and at the resistor relative to ground using high voltage probes (Lecroy, 1:1000). Time resolved measurements were applied in order to detect corona discharge activity. Emissions of noxious compounds (NO_x, CO) were measured in the diluted flue gas using electrochemical sensors (Testo 360-1).

Our investigations covered pressures p from 0.12 MPa to 1 MPa at a feed gas temperature of typically 290 K. Since the pressure in gas turbines is generated by nearly adiabatic compression of air, the maximum gas density investigated in our experiments was even higher than that in gas turbines. In all experiments the air number λ, which gives the actual air to fuel ratio relative to that of stoichiometric combustion, was around 1.2 (slightly lean combustion), and the feed flow velocity was 5 m/s.

In order to get an idea of pressure scaling of the electrical characteristics, the measured voltage- and current values U and I were divided by the pressure p and drawn in a diagram (Fig. 2). The fit shows

$$I/p \sim (U/p)^{3/2} \tag{1}$$

which was derived from a simple 1-dimensional theoretical model [8] describing E-field induced drift of the electrons and ions generated in the reaction zone of the flame due to chemo-ionization. For the solution of the 1-dimensional Poisson equation this model takes into account space charge formation caused by charge separation. Deviations between the fit and experimental values can be attributed to the fact that the reaction zone in the 7-hole Bunsen burner is far from being planar. Thus depending on the distance between reaction zone and burner surface the space charge field develops different. With the exception of two measurement points at 0.12 MPa no indication of corona discharges could be found.

For all pressures a reduction of the CO-emissions for more than 90% caused by more complete combustion of the fuel was achieved. The electric power consumption for this effect was $< 0.02\%$ of the thermal power of the combustion

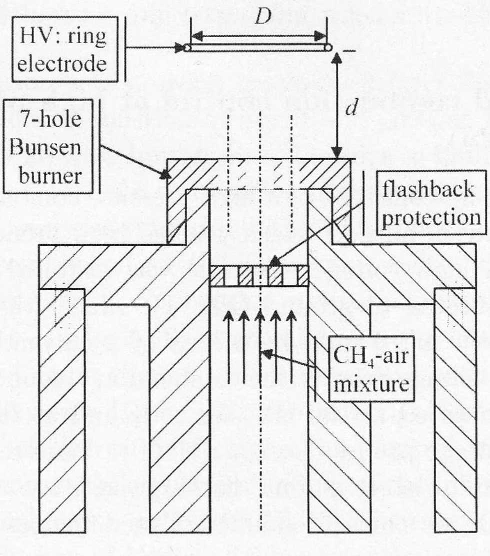

Figure 1. Cross sectional drawing of the burner – electrode set-up inside of the high-p combustion chamber.

process for optimized electrode position and diameter. By application of a reduced voltage U/p of 40 kV/MPa the lean blow-off limit of this combustor could be improved for up to 8%, which finally resulted in a decrease of both CO- and NO_x-emission (Fig. 3): Normally the CO emission of methane combustion increases due to incomplete combustion, when for a certain burner the lean blow-off limit is approached. At the 7-hole Bunsen burner a lift-off of the flames starting at the outer burner rims can be observed. This lift-off causes formation of zones where the fuel is oxidized incompletely. Application of an electric field reduces this lift-off and, at sufficiently large values the flames attach, completely to the burner rims. Thus nearly complete combustion with very low CO emissions is achieved. Since more complete combustion at a fixed air number λ increases the heat release, on application of an electric field an increase of NO_x-emissions is observed. For $\lambda = 1.14$, e.g. without electric field, a CO-concentration of 226 ppm and a NO_x-concentration of 23 ppm were measured respectively. Application of a voltage of 5 kV (10 kV, 15 kV) to the ring electrode resulted in a decrease of CO-concentration to 58 ppm (19 ppm, 3.4 ppm) and in increase of NO_x-concentration to 24 ppm (26.5 ppm, 27.7 ppm). Increase of the air number resulted in a decrease of the adiabatic flame temperature reducing the NO_x-concentrations and increasing the CO-concentrations.

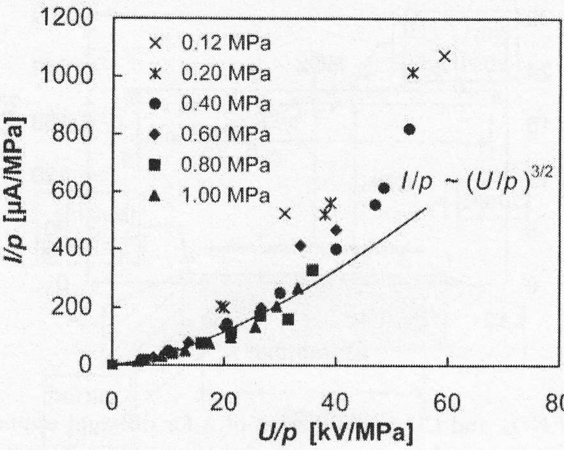

Figure 2. Voltage-current characteristics of the flame (D = 70 mm, d = 40 mm) at different pressures.

2.2 Low pressure EFCC of a McKenna flat flame burner

In low pressure premix combustion experiments (6 kPa $\leq p \leq$ 20 kPa) performed at the McKenna flat flame burner (Fig. 4) the electrical characteristics of the flame could be measured for a much wider range of reduced electric fields than in the high-pressure experiments (Fig. 5).

Since the planar reaction zone has a well defined cross sectional area being equal to the flow cross section provided by the porous bronze body stabilizing the flame, a current density j can be given as a function of the spatial average value of the reduced electric field E_{av}/p. Four different regions can be identified in this diagram:

I. Charge separation and drift – a fraction of the electron-ion pairs generated in the reaction zone recombines

II. Saturation – complete charge separation

III. Non-self sustained gas discharge – electron collision ionization occurs in space charge regions

IV. Reverse polarity – electron drift to the burner head; negative ion formation due to electron attachment in the cold feed gas of the flame.

In the case **IV** a strongly reduced current density results due to mobility of O^--ions being about 200 times smaller than that of electrons. Further, due to the low temperature between the burner head and reaction zone the gas density is increased in this region.

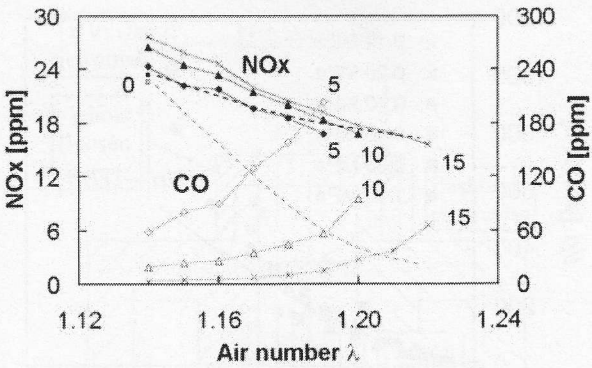

Figure 3. Emissions of NO_x and CO as a function of λ for different applied electrode voltages in kV given by the numbers at the end of the lines ($D = 20$ mm, $d = 20$ mm; $p = 0.4$ MPa). Lines end at lean blow-off limit. The long-dashed lines give the potential for simultaneous NO_x- and CO-emission reduction (black/gray).

Figure 6 gives an idea of the reduced electric field and potential distribution for a 1-dimensional flat flame located 8 mm above burner head at current densities being substantially below saturation: Application of a positive voltage applied to the ring electrode causes formation of a positive space charge between the burner head and reaction zone (0 mm $< z <$ 8 mm) and formation of a negative one between reaction zone and ring electrode (8 mm $< z <$ 60 mm). Since in the experiments the burner head was cooled to about 360 K, the temperature between burner head and reaction zone was about 400 K, and behind the reaction zone temperatures around 2200 K were measured [10]. These temperatures were taken into account for the evaluation of the mobilities of charge carriers. At low temperature electrons can easily form stable O_2^-–ions in collisions with molecular oxygen. Because at high temperature these molecular ions are not stable, behind the reaction zone electrons are the major negative charge carriers. For this reason in the case of a negative voltage applied to the ring electrode the resistance of the flame is more than a factor 10 higher than in the case of positive polarity (see current densities and reduced potential values in Fig. 6).

A more detailed evaluation of the $U - I$ characteristics is given in Fig. 7. Regions I and III can be reproduced qualitatively by simple models. According to Eq. (1) (see [8]) $I \sim U^{3/2}$. Under the assumption, that no secondary electrons are generated at the burner head, the current in region III can be reproduced by a simple avalanche model: $I \sim \exp\{\alpha_{eff}\, d\}$, where α_{eff} is the effective ionization coefficient of the gas mixture behind the reaction zone (assumed to consist of N_2, H_2O, CO_2, and O_2) at a temperature of 2200 K evaluated at a spatial average value of the reduced electric field. From the saturation current density

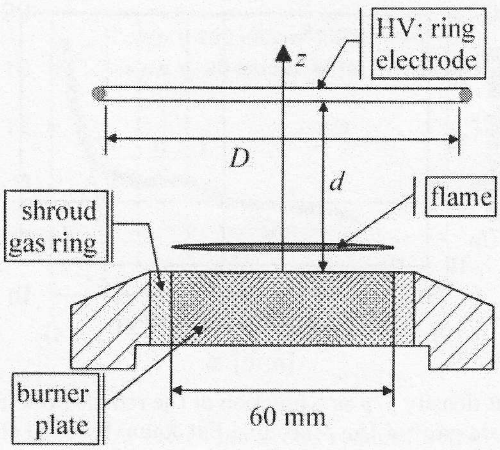

Figure 4. Scheme of the McKenna flat flame burner. The burner plate and the shroud gas ring consist of porous bronze distributing the gas flow very uniformly over the complete cross section. For water cooling a copper line is embedded into the burner plate. This allows precise control of the position z of the planar flame above burner plate.

(region II) and the recombination rate coefficient the chemo-ionization rate r_{ion} can be evaluated according to [2].

In the case of applied voltages leading to a non self-sustained glow-discharge (region III) the spatial profile of near UV radiation emitted from electronically excited molecular nitrogen indicated regions of high electric fields, e.g. around the ring electrode (Fig. 8), where electron collision ionization can be expected to take place. A shift of the flame front being proportional to the applied electrode voltage could be demonstrated for a wide range of voltages covering regions I–III in Fig. 5. However, because near the reaction zone of the flame the electric field was low we could not find any direct influence of electron collision induced effects on combustion.

Both high and low pressure experiments reinforce the sketch of electric field combustion control model given by Weinberg [2]:

(a) continuity equations for electrons and ions with chemo-ionization and recombination as source and loss terms;

(b) introduction of an electrostatic force term into the momentum balance equation;

(c) Ohm's law for the relation between electric field and current density;

(d) Poisson equation for the calculation of the electric field.

Nevertheless, a comprehensive numerical simulation can be difficult because this

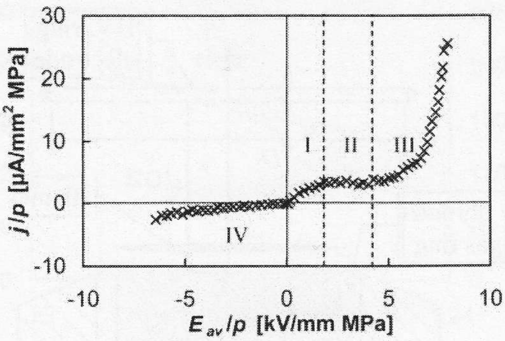

Figure 5. Reduced current density j/p as a function of the reduced electric field E_{av}/p (averaged over the electrode gap) of the McKenna flat flame burner ($D = 90$ mm, $d = 60$ mm; $p = 6$ kPa).

requires coupling of computational fluid dynamics, chemical kinetics of combustion and chemo-ionization, and electrostatic field simulation.

2.3 Plasma-chemical combustion control

Plasma assisted combustion was investigated in a configuration consisting of a miniaturized glide-arc reactor inserted into a tubular burner head (Fig. 9). A fuel rich methane-air mixture (air number $0.4 < \lambda < 0.6$) was fed to the glide-arc reactor, whereas a lean methane-air mixture was fed to the tubular burner head. The mass flows and the dimensions of the glide-arc reactor and of the tubular burner head were chosen such that the stoichiometry of the combined gas flows were lean ($1.2 < \lambda < 2.0$), the feed flow velocities in the glide-arc reactor were 15-20 m/s, and the flow velocity of the combined gas flows in the tubular burner were 0.6-0.9 m/s. In order to enhance mixing of the fuel rich gas flow exiting the glide-arc reactor with the surrounding, lean gas flow, a baffle plate was mounted at the exit of the plasma reactor. The combustion was stabilized by a grid mounted about 5 cm flow-down of the baffle plate.

The influence of feed gas composition, flow velocity, and plasma input power on the plasma conversion of fuel was investigated by means of FTIR-absorption spectroscopy and gas chromatography. Hydrogen yields

$$y_{H_2} = \frac{\dot{\nu}_{H_2}}{2 \cdot \dot{\nu}_{CH_{4ed}}} \tag{2}$$

of up to 12% and CO-yields

$$y_{CO} = \frac{\dot{\nu}_{CO}}{\dot{\nu}_{CH_{4ed}}} \tag{3}$$

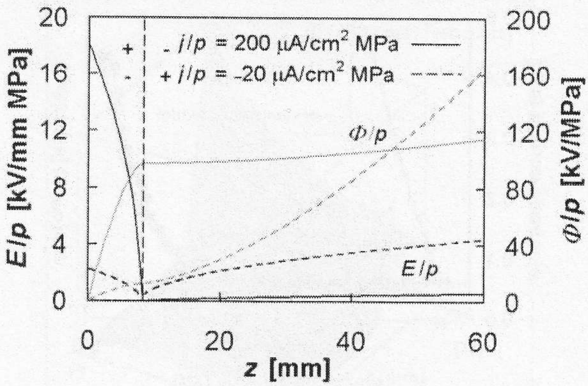

Figure 6. Reduced electric field E/p (black lines) and reduced potential F/p (gray lines) evaluated from a two-zone drift model. The vertical dashed line gives the position of the flame front. Full lines – positive voltage (~2 kV) applied to the ring electrode resulting in a positive space charge (+) flow-up and a negative space charge (–) flow-down of the flame front, and in a reduced current density of 200 mA/cm^2 MPa; dashed lines – negative voltage (~–16 kV) applied to the ring electrode resulting in a negative space charge (–) flow-up and a positive space charge (+) flow-down of the flame front, and in a reduced current density of – 20 mA/cm^2 MPa. In the region flow-up of the flame front $T = 400$ K was assumed, in the flow-down of the flame front $T = 2200$ K.

of more than 25% were achieved. Here $\dot{\nu}_{H_2}$, $\dot{\nu}_{CO}$, and $\dot{\nu}_{CH_{4ed}}$ are the molar flow rates of H_2 and CO in the product gas and of CH_4 in the feed gas, respectively.

In spite of the baffle plate, which obviously quenches chemically active radicals generated in the glide-arc plasma, due to plasma pre-treatment of a fraction of the fuel the lean combustion limit could be extended from $\lambda = 1.6$ to $\lambda = 2.0$. For lower air numbers between 1.2 and 1.6 more complete combustion of the fuel was achieved. Without plasma pre-treatment the residual methane concentration measured in the combustion flue gas was between 0.6 vol% and 0.45 vol%, whereas with plasma pre treatment CH_4-concentrations around 0.3% were evaluated. Thus the conversion of fuel was improved from 90 % to 95%. Due to the burner design this improvement can only be attributed to the generation of species having lifetimes exceeding 50 ms such as H_2 or higher hydrocarbons (C_2H_6, C_2H_4, C_2H_2). However, the latter ones were found in low concentrations (in total < 0.5 %), only. Thus hydrogen can be expected to dominate the combustion control. Indeed there have been several proposals and investigations published in the literature stating improved efficiency and reduced emissions of internal combustion engines due to conversion of a fraction of fuel to hydrogen [11, 12].

Nevertheless, a configuration allowing utilization of reactive radicals will have the potential for higher efficiency plasma assisted combustion [13].

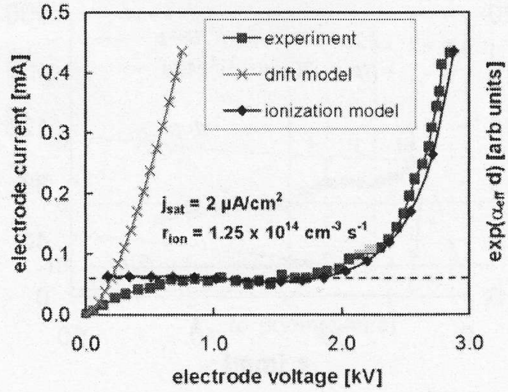

Figure 7. Voltage-current characteristics of the McKenna flat flame burner operated at p = 6 kPa: experiment – measured data; model – result of a 1-dimensional drift model [8]; ionization model (right scale) – electron avalanche simulated using the effective ionization coefficient evaluated at a spatial average value of the reduced electric field; dashed line – saturation current.

3 Summary and conclusions

Electric field control of premix methane combustion applying a 7-hole Bunsen burner could be demonstrated in a wide range of pressures relevant for practical application: First the lean blow-off limit could be increased by 8% resulting in a NO_x emission reduction of by nearly 30%. More than 90% reduction of the CO emission was achieved applying electric powers of less than 0.02 % of the thermal power. The CO emission reduction can be attributed to more complete combustion due to better attachement of the flames to the burner rims. From the measured electrical characteristics of the burner we have the evidence, that gas discharge effects do not play a role in these high pressure experiments. This is supported by the agreement of the measured $U-I$ characteristics and its pressure scaling with that evaluated from a simple 1-dimensional model. Thus we conclude that electro-hydrodynamic (EHD) effects are responsible for the attachment of the flame to the burner rims. Because for the electrical force density f_{el} the model predicts

$$f_{el}/p = j/\left(n\,\mu_{ion}\,k_b\,T\right) \tag{4}$$

where j is the current density, $n\,\mu_{ion}$ is the reduced ion mobility, k_b is the Boltzmann constant, and T is the gas temperature, high pressures should be no limitation for the utilization of EHD effects.

Plasma-chemical stabilization of combustion was investigated at atmospheric pressure. Here the lean combustion limit of a tubular burner could be extended from $\lambda = 1.6$ to $\lambda = 2.0$. At the same time better utilization of the fuel was

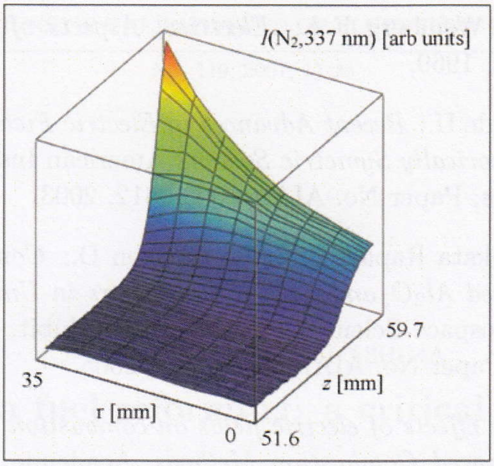

Figure 8. Emission intensity of the N_2 $C^3\Pi_u$-$B^3\Pi_g(0,0)$ band at 337 nm integrated along line of sight (side on measurement using a gating spectrograph and an intensified CCD-camera [10]; data not Abel inverted).

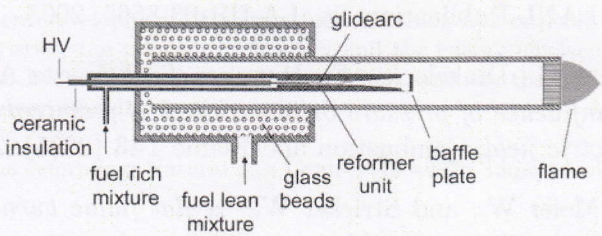

Figure 9. Set-up of the plasma-chemical combustion control experiment.

observed. Due to the specific reactor design this effect is most likely caused by plasma induced partial oxidation of a fraction of the fuel to H_2 and CO. Thus the energy requirements were rather large (plasma powers of the order of several percent of the thermal power).

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