# Numerical and experimental investigation of streamer discharges leading to the ignition of hydrogen/air mixtures

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## 1 Introduction

The safety-related assessment of electrical discharges for usage in hazardous areas, which is done to avoid the ignition of explosive atmospheres, has to consider all types of electrical discharges. Using converter fed drives, fast switching processes can lead to significant overvoltages resulting in streamer discharges. A single streamer discharge leads only to a low temperature rise inside the discharge volume. However, repetitive streamer discharges using high frequency alternating current can ignite hydrogen/air mixtures due to the accumulation of the discharge energy [1].
Streamer discharges are a typical example of non-equilibrium plasma and occur during the formative phase of a spark discharge. A conductive channel is produced with a significant energy deposit due to the interaction of charge carriers with the electric field and with gas molecules. In the case of an alternating current with high frequency, it persists only for several nanoseconds and, depending on the peak voltage and gap distance, a flashover can be avoided. However, not only thermal heating inside the discharge volume has to be taken into account concerning streamer discharges. The electron energy of a streamer which is much higher than the electron energy of a spark discharge is in the range of the energy necessary for ionization or dissociation, for instance, of oxygen and nitrogen [2, 3].
This paper presents temperature measurements of repetitive streamer discharges using optical emission spectroscopy. The results demonstrate that ignition of hydrogen/air mixtures takes place at comparatively low temperatures. Numerical simulations of the ignition process using detailed chemistry combined with an analysis of the chemical reaction pathways confirm the experimental results and show that electron impact reactions during a discharge produce significant amounts of radicals, which drastically influence the ignition process.

## 2 Experimental setup

A detailed description of the experimental setup used for repetitive streamer discharges has been given elsewhere [1]. Streamer discharges between a rod/plane electrode configuration with a gap distance of 40 mm were created by a power supply unit, consisting of a power amplifier, a frequency generator and a resonating air-core coil. This power supply unit delivers peak voltages up to 20 kV with a frequency between 600 and 750 kHz. The frequency generator was used to define the number of voltage cycles and the voltage amplitude. Figure 1 shows the rod/plane configuration inside an

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optically accessible combustion chamber. The chamber can be filled with hydrogen/air mixtures or air using Bronkhorst mass flow meters.
The emitted light from the streamer discharges was focused onto the spectrograph (Acton Research SpectraPro-300i) with a UV lens (B. Halle Nachfl., *f*/2, focal length 100 mm). The spectrograph was coupled with an image intensifier (LaVision IRO) and an ICCD camera (LaVision ImagerProPlus 2M). The system was calibrated concerning the wavelength by means of a mercury-vapour lamp at a wavelength of 253.5 nm and concerning the spectral sensitivity at different wavelengths by means of a tungsten halogen lamp.

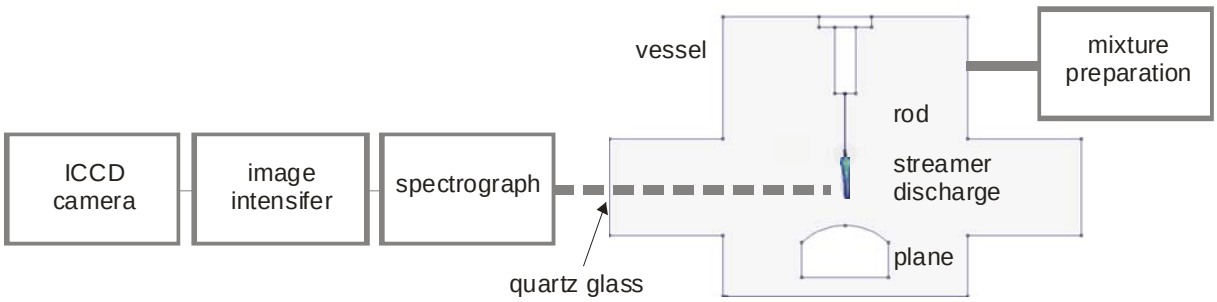


Figure 1. Experimental setup and combustion chamber.

# 3 Kinetic model

The reaction mechanism of Bowman et al. [4] contains low temperature $H_2$/air chemistry and non-equilibrium plasma discharge processes. The mechanism consists of 168 equations and 29 neutral and excited species. Ions are not accounted for in this work as they have no significant influence on major species chemistry [5]. The rate coefficients of the electron impact reactions for excitation and dissociation due to electron impact are calculated for reduced electrical fields between 1 and 1000 Td by means of an electron energy density function (EEDF) which is solved using the software BOLSIG+ [6].
Having a frequency of 740 kHz during one single voltage cycle of 1350 ns, one or more streamer discharges occur within approximately 100 ns during the positive voltage peak [1]. The streamer formation at the negative voltage peak can be neglected. Therefore, the reduced electrical field is considered as a square pulse with a duration of 100 ns. Only during this pulse are electron impact reactions considered. During the remaining 1250 ns, quenching of excited species takes place before the next square pulse of the reduced electrical field occurs. More details are given in [7]. In the model the formation of electrons is not considered. Hence, for each single pulse of the reduced electrical field a defined electron density in the range of $10^{11}$ $cm^{-3}$ to $10^{12}$ $cm^{-3}$ is assumed as a start value [3]. With increasing temperature a decreasing electron density is necessary for streamer formation which is taken into account [8].
A cylindrical configuration of the streamer with a radius of 230 µm is assumed, based on experimental results [1], which is represented using a one-dimensional model [9]. Perpendicular to the symmetrical axis of the cylindrical configuration, the well-known mass, species and energy equations are solved as a function of time. Having a constant outer radius, zero gradients for species and temperature at the inner and outer boundary are assumed. During each pulse of the reduced electrical field a source term is considered leading to a temperature rise of several Kelvin during one voltage cycle. The artificial production of electrons using a defined electron density at the beginning of a voltage cycle has to be taken into account as an additional energy source term which represents the electron energy deposition due to inelastic collisions between electrons and molecules leading to the excitation and dissociation of molecules [7].

# 4 Results and discussion

Averaged experimentally measured spectra of the emission of the second positive system of $N_2$ C-B in the spectral range between 365 and 381 nm were compared with 275 simulated spectra (using different rotational and vibrational temperatures) calculated with the Specair software [10]. The database of simulated spectra covers rotational temperatures $T_{rot}$ between 500 and 1500 K with a step size of 50 K and vibrational temperatures $T_{vib}$ between 2000 and 5000 K with a step size of 250 K. The rotational temperature of an experiment was yielded from the best fit Specair spectrum whereas the vibrational temperature of the $N_2$ C state was determined from the intensity ratio of the different vibrational bands in the given spectral range [11]. Assuming a conservation of the rotational quantum number during the electron impact processes, $T_{rot}$ is a good approximation of the gas temperature [12].

Figure 2a shows rotational and vibrational temperatures of $N_2$ as a function of the number of voltage cycles producing repetitive streamer discharges in air with a voltage amplitude of 11.3 kV. An increasing number of voltage cycles leads to an increase of the streamer channel length [1]. Therefore, the energy deposited during one streamer discharge is given to a larger volume with an increasing number of voltage cycles decreasing the energy density per streamer discharge. Hence, starting with a rotational temperature of (900±50 K) using 30 voltage cycles, $T_{rot}$ increases only slightly with an increasing number of voltage cycles. The vibrational temperature of the $N_2$ C state is nearly independent of the number of voltage cycles as can be seen in Figure 2a.
Using a fixed number of voltage cycles, the increase of the voltage amplitude results in an increase of rotational temperatures as shown in Figure 2b. Starting with a rotational temperature of (750±50) K at a peak voltage of 8.6 kV, $T_{rot}$ increases linearly up to (1500±50) K. The same dependency can be observed in Figure 2b for the vibrational temperature of the $N_2$ C state. The measured vibrational temperatures are always significantly higher than the rotational temperatures. This confirms the assumption of a non-equilibrium plasma, even though the vibrational population of the $N_2$ C state does not have to be identical to the vibrational population of the ground level.

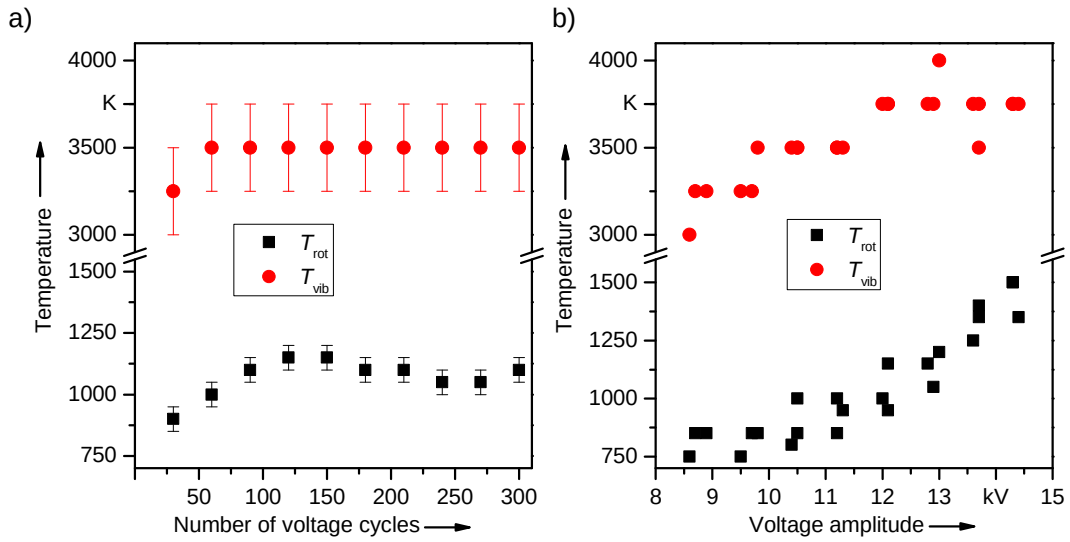


Figure 2. Dependency of rotational and vibrational temperatures of $N_2$ from a) the number of voltage cycles (11.3 kV voltage amplitude) and from b) the voltage amplitude (30 voltage cycles).

Repetitive streamer discharges produced from 30 voltage cycles with a voltage amplitude of 11.3 kV can ignite a 25 vol.% $H_2$ in air mixture [1]. Based on the results shown in Figure 2, temperatures around 1000 K can be expected before ignition takes place.

Figure 3a shows the maximum temperature inside the computational domain using the minimum energy necessary for the ignition of a 25 vol.% $H_2$ in air mixture using 30 voltages cycles with a duration of 40.5 µs both for a reduced electrical field of 0 Td and of 300 Td. Regarding a reduced electrical field of 0 Td, electron impact reactions are neglected and only thermal heating by streamer discharges is considered. As can be seen in Figure 3a the temperature rises up to 1431 K at the end of the energy deposition and subsequently cools down mainly due to heat conduction. After 78 µs a strong temperature rise is visible, the mixture has been ignited. Having a reduced electrical field of 300 Td, a significant amount of the total discharge energy is used for the population of excited electronic states and for molecular dissociation. Therefore, only a temperature rise of up to 1178 K is visible in Figure 3a for the non-equilibrium plasma. However, the mixture ignites in this case after 125 µs.

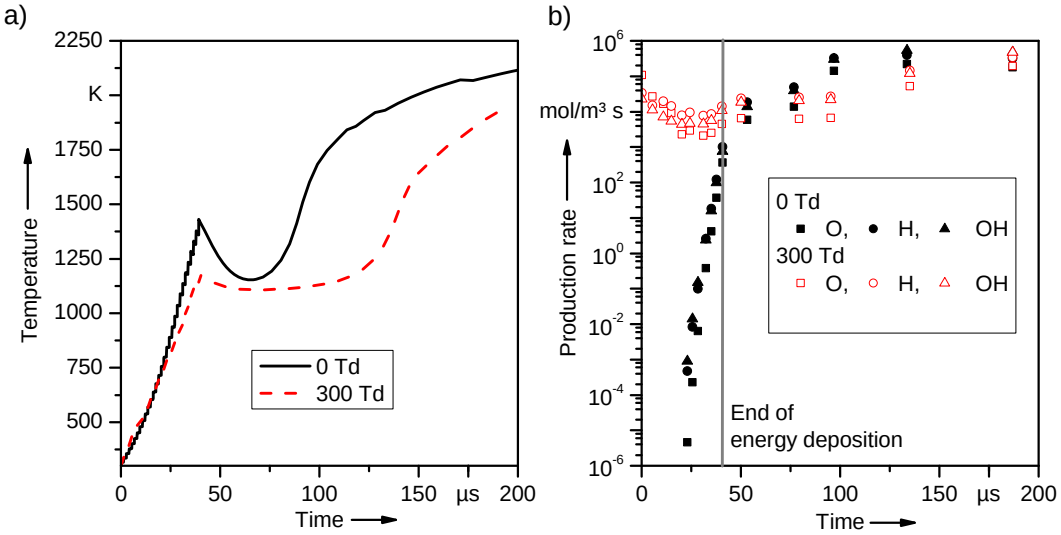


Figure 3. a) Calculated maximum temperature igniting a 25 vol.% $H_2$ in air mixture by a thermal plasma (0 Td) and a non-equilibrium plasma (300 Td), b) production rate of O, H and OH during and after energy deposition.

Therefore, in case of a non-equilibrium plasma, the ignition can take place at lower temperatures. As can be seen in Figure 3b having a reduced electrical field of 300 Td, a significant production rate of O, H and OH is visible starting with the beginning of the energy deposition due to the electron impact reactions. In case of a reduced electrical field of 0 Td, the production rate of the radicals increases more slowly due to the strong temperature dependence of the chain branching reactions.

Reaction flow analyses have been performed at selected time steps during the ignition with a reduced electrical field of 300 Td in order to identify the important reaction paths in the non-equilibrium plasma. Figure 4a shows the results of an analysis during the discharge of the fifth voltage cycle after 5.4 µs. The maximum gas temperature $T_{max}$ in the streamer channel has increased only up to 411 K. As can be seen from Figure 4a, 91 % of atomic oxygen originates from the electron impact dissociation of $O_2$ into O+O and O+O($^1$D). After the end of the discharge during the fifth voltage cycle at a maximum temperature of 431 K, as shown in Figure 4b, the formation of radicals is dominated by reactions including electronically excited species. Atomic oxygen, for example, is mainly produced due to the interaction of $O_2$ with vibrationally excited $N_2(A^3\sum_u^+)$.

Therefore, at low temperatures and during a streamer discharge the reactions describing the non-equilibrium plasma processes are responsible for the dominant pathway of radical production. At the end of the burst of streamer discharges the streamer channel has heated up significantly - as shown in Figure 3a - and the well-known chain branching reactions are important for the formation of O, H and OH.

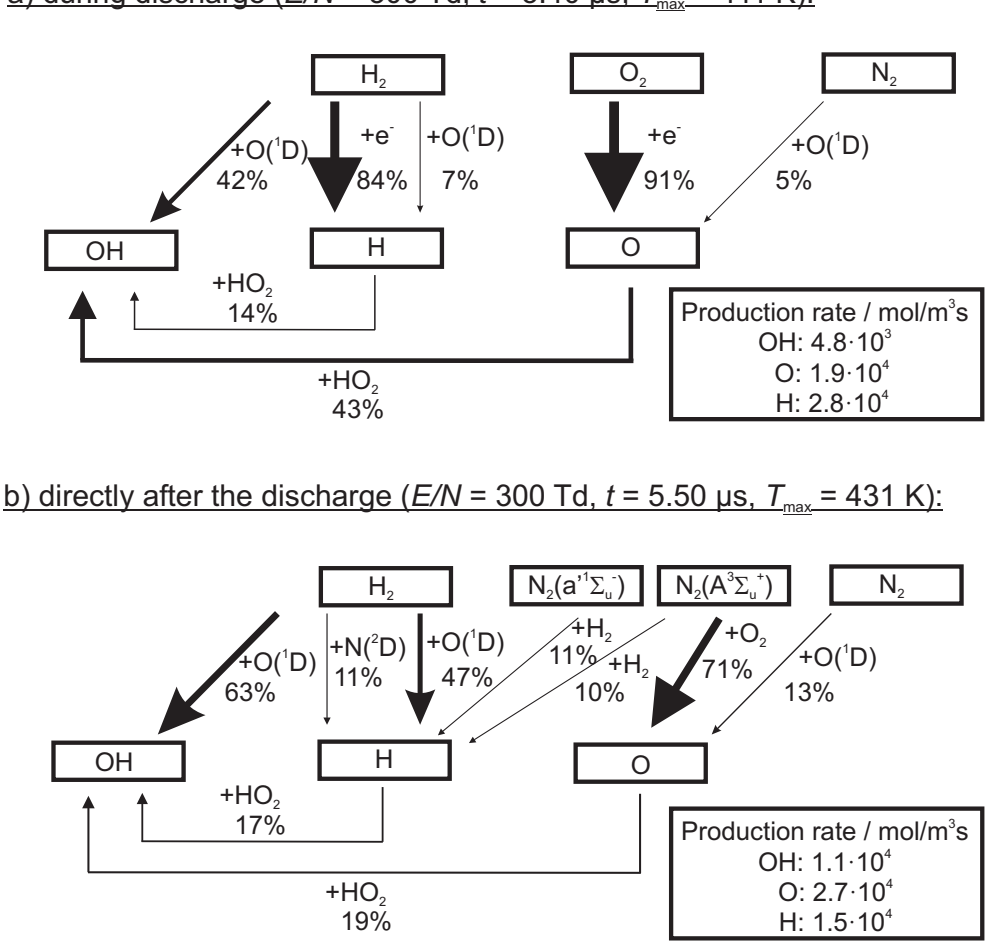


Figure 4. Reaction flow analysis concerning the production of O, H and OH a) during a discharge in the fifth voltage cycle, b) directly after this discharge.

# 5 Conclusions

The experimental investigation of repetitive streamer discharges in air using optical emission spectroscopy shows that the temperature rise depends only weakly on the number of voltage cycles, but increases linearly with the voltage amplitude. The final temperatures in experiments leading to the ignition of hydrogen/air mixtures are in the range of 900 to 1100 K, which is in excellent agreement with numerical simulations. Even though these temperatures are significantly lower compared to spark ignition experiments, high production rates of O, H and OH can be observed due to the non-equilibrium plasma processes.

The reaction flow analysis of the ignition by repetitive streamer discharges shows that the electron impact reactions leading to the dissociation of hydrogen and oxygen during a discharge are the most important reactions concerning the formation of O, H and OH at low and intermediate temperatures. After the discharge the reactions of hydrogen and oxygen with excited species have to be considered in this temperature range, too. With increasing temperature, however, the well-known chain branching reactions become more and more important.

# 6 Acknowledgments

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# References

[1] Langer T et al. (2010). Streamer discharges caused by high-frequency voltage leading to ignition of hydrogen/air mixtures. Comb. Sci. Technol. 182: 1718.

[2] Liu J et al. (2004). Effect of discharge energy and cavity geometry on flame ignition by transient plasma. 42nd Aerospace Sciences Meeting, 6th Weakly Ionized Gases Workshop, Reno, AIAA-2004-1011.

[3] Starikovskaia S. (2006). Plasma assisted ignition and combustion. J. Phys. D.: Appl. Phys. 39: R265.

[4] Bowman S et al. (2010). Kinetics of low-temperature hydrogen oxidation and ignition by repetitively pulsed nonequilibrium plasmas. 48th Aerospace Sciences Meeting and Exhibit, Orlando, AIAA-2010-1590.

[5] Mintsunov E et al. (2008). Mechanism of plasma assisted oxidation and ignition of ethylene-air flows by repetitively pulsed nanosecond discharges. 46th Aerospace Sciences Meeting and Exhibit, Reno, AIAA-2008-1106.

[6] Hagelaar GJM, Pitchford LC. (2005). Solving the Boltzmann equation to obtain electron transport coefficients and rate coefficients for fluid models. Plasma Sources Sci. Technol. 14: 722.

[7] Langer T, Markus D, Maas U. (2010). Influence of non-equilibrium plasma on the ignition process due to repetitive streamer discharges. 44th AIAA Plasmadynamics and Laser Conference, Honolulu, AIAA-2011-3450.

[8] Luque A, Ebert U. (2012). Density models for streamer discharges: beyond cylindrical symmetry and homogeneous media. J. Comput. Phys. 231: 904.

[9] Maas U, Warnatz, J. (1988). Ignition processes in hydrogen-oxygen mixtures. Combust. Flame, 74: 53.

[10] Specair: http://www.specair-radiation.net (accessed 20-November-2012) (online).

[11] Laux CO et al. (2003). Optical diagnostics of atmospheric pressure air plasmas. Plasma Sources Sci. Technol. 12: 125.

[12] Fantz U. (2006). Basics of plasma spectroscopy. Plasma Sources Sci. Technol. 15: 137.