Abstract

Ignition delay time, time-resolved temperature, and time-resolved absolute OH concentration are measured in mildly preheated H₂–air mixtures (T = 473–500 K), excited by a repetitive nanosecond pulse discharge in a plasma flow reactor. The measurements are done in decaying plasma after the discharge pulse burst (pulse repetition rate 10–40 kHz). Ignition delay increases steeply as the number of pulses in the burst is reduced, exhibiting threshold-like behavior. Temperature and OH concentration during the ignition process are measured by two-line OH LIF, as functions of time delay after the discharge burst. Absolute calibration is done using a near-adiabatic flame. The results show that the temperature at the end of the discharge burst is 700–750 K, rising to 1500–1600 K within a few ms after the burst, indicative of ignition. During ignition, OH concentration increases by a factor of 20–50, from \((0.5–1.1) \times 10^{14} \text{ cm}^{-3}\) at the end of the burst to peak value of \((2.3–2.6) \times 10^{15} \text{ cm}^{-3}\). Kinetic modeling calculations show good agreement with the experimental results, demonstrating quantitative insight into kinetics of plasma-assisted ignition. Modeling calculations demonstrate that ignition is induced primarily by accumulation of H atoms and gradual temperature rise in the discharge, at temperatures significantly lower than autoignition temperature, by up to 200 K.

1. Introduction

Recently, nanosecond pulse discharges have been demonstrated to be effective for ignition and flameholding [1–4], ignition delay reduction [5,6], and flame stabilization [7,8]. One key advantage of these discharges is their operation at high reduced electric fields, \(E/N \sim 10^5–10^6 \text{Td} \) (1 Td = \(10^{-17} \text{ V cm}^2\)), when a significant fraction of input power is spent on electronic excitation and molecular dissociation, resulting in efficient generation of radical species in fuel–air mixtures. Insight into kinetics of radical generation in nanosecond pulse discharges, as well as into low-temperature fuel oxidation and ignition, requires time-resolved
measurements of radical species concentrations and temperature. Data obtained above room temperature and below ignition temperature remain scarce. Previously, time-resolved NO and O atom concentrations have been measured by LIF and two-photon absorption LIF (TALIF) in single-pulse discharges in air, methane-air, and ethylene-air [9,10]. Time-resolved temperature in repetitively pulsed discharges in air, ethylene-air, and hydrogen-air has been measured by pure rotational CARS [11,12]. Time-resolved absolute OH concentration has been measured by LIF in single-pulse and repetitively pulsed nanosecond discharges in hydrogen-air [13].

Our previous measurements of OH concentration and ignition in H2–air [13,14] have shown that ignition time dependence on the details of kinetic mechanism used is fairly weak. This is primarily due to maintaining the nanosecond discharge, pulsed at a high repetition rate, through the ignition process. Although plasma-assisted ignition has strong, threshold-like dependence on radical species concentrations and temperature [14], continuous generation of radicals during the discharge, overlapping with ignition, “masks” this dependence to a large extent. The present paper is focused on simultaneous OH concentration and temperature measurements by two-line OH LIF thermometry after the discharge pulse burst is terminated. At these conditions, electronically excited molecules and atoms generated during the discharge decay rapidly, such that ignition is controlled by “conventional” chemical reactions of radicals generated during the discharge (primarily H, O, and OH). The experiments are conducted in a mildly preheated plasma flow reactor, with initial gas temperature, \( T_0 = 473–500 \text{ K}, \) kept significantly below auto-ignition temperature \( (T \approx 900 \text{ K} \text{ at } P = 100 \text{ torr}) [14]. \) The nanosecond pulse discharge hydrogen-air plasma generated in this reactor is diffuse and uniform up to \( P = 150 \text{ torr} [14], \) resulting in controlled and reproducible ignition, and lending it suitable for optical diagnostics using repetitively pulsed lasers.

2. Experimental

The experimental setup used in the present work (see Fig. 1) is similar to the one used in our previous studies [13,14]. The discharge cell/plasma flow reactor consists of a 280 mm long, \( 22 \times 10 \text{ mm} \) rectangular cross section quartz channel with wall thickness of 1.75 mm. Two plane quartz windows are fused to the ends of the channel, providing optical access in the axial direction. A 60 mm long, right angle fused silica prism is placed along the channel to provide optical access from the side (see Fig. 1). The entire assembly is heated in a tube furnace (Thermcraft, Ltd., with 6 in. diameter, 12 in. long heated section), to improve plasma stability. A 1 m long quartz tube coil inlet (see Fig. 1) preheats the fuel-air flow to the furnace temperature, which was verified by thermocouple measurements. The furnace, mounted on rails, can be moved aside and replaced by an adiabatic Hencken burner used for absolute calibration of LIF diagnostics. Two \( \frac{1}{4} \text{ in.} \) diameter quartz-to-stainless-steel adaptors connect the reactor to the gas delivery system. Fuel and air flow rates through the reactor are controlled by MKS mass flow controllers. Before entering the cell, fuel and air flows are premixed. Pressure gages are placed in the delivery lines upstream and downstream of the reactor.

Two \( 14 \times 60 \text{ mm} \) rectangular plate copper electrodes, rounded at the edges, are placed on the top and bottom of the quartz channel (see Fig. 1) and held in place by ceramic clamps. A 1/16-in. thick high-temperature perfluoroelastomer sheet (Kalrez, DuPont) is placed between each electrode and the channel wall, to reduce air gaps and prevent corona discharge outside the cell. The electrodes are connected to a Chemical Physics Technologies (CPT) high voltage pulsed power supply, which produces 25 kV peak voltage, 25 ns duration pulses, with repetition rate up to 50 kHz, or to an FID GmbH FPG 60-100MC4 pulse generator (peak voltage up to 30 kV, pulse duration 5 ns, repetition rate up to 100 kHz). Both pulsers are operated in repetitive burst mode, producing bursts of 100–200 pulses at a pulse repetition rate of 10–40 kHz and burst repetition rate of 2 Hz. During operation using the CPT pulser, the cell is irradiated by a UV emission source (Hg–Ar lamp), to produce breakdown by the first discharge pulse. No UV pre-ionization is necessary when the FID pulser is used. Ignition is detected by monitoring OH* emission through the side wall of the cell, using a narrow bandpass filter (centered at 310 ± 2 nm, bandpass 10 ± 2 nm FWHM), a photomultiplier tube (PMT) (Hamamatsu, Model R106), and a digital oscilloscope. The PMT response time, approximately 10 μs, is controlled by a 50 kΩ terminator between the PMT and the oscilloscope.

The experiments are conducted in premixed, lean hydrogen-air flows at an initial temperature of \( T_0 = 473–500 \text{ K}, \) \( P = 80–100 \text{ torr}, \) and equivalence ratio of \( \phi = 0.4–0.5, \) at flow velocity through the reactor of \( u = 40 \text{ cm/s}. \) Low discharge burst repetition rate, 2 Hz, is chosen to keep the delay between the bursts longer than the residence time of the flow in the plasma, \( \sim 0.1 \text{ s}, \) such that the flow in the reactor would experience only one burst. Low equivalence ratio reduces pressure overshoot during ignition, such that the flow is restored to baseline conditions before the next burst, and prevents water vapor condensation in the exhaust line. Operation at a higher burst repetition rate (5 Hz) results in poor ignition reproducibility burst-to-burst. Slow flow velocity
improves flow preheating in the inlet coil and reduces pressure drop across the reactor.

A separate low-temperature discharge cell of the same geometry, but without Kalrez sheets between the electrodes and the quartz channel, is used for measurements of energy coupled to the plasma by a nanosecond pulse discharge. In this cell, the electrodes are attached to the channel by a thin layer of silicone adhesive, which also covers the entire electrode surface to preclude corona discharge and the resulting energy loss. Energy coupled to the plasma is determined by simultaneous measurements of pulse voltage and current using custom designed, short response time capacitive voltage probes and shunt current probes [15].

The OH LIF apparatus is similar to the one used in our previous work [13]. Briefly, a 532 nm second harmonic output of an Nd:YAG laser (Continuum, Model Powerlite 8010) pumps a tunable dye laser (Laser Analytical Systems, Model LDL 20505), generating output at 566 nm with \( \sim 0.25 \text{ cm}^{-1} \) linewidth, measured using a Fizeau wavemeter (High Finesse, Model Angstrom WS/6). While the flash lamp is operated at 10 Hz, Q-switching controlling the Nd:YAG laser output is maintained at 2 Hz, to match the burst repetition rate of the pulser. The dye laser output is frequency doubled by a BBO crystal, and the 283 nm beam is directed along the centerline of the quartz test cell (see Fig. 1). A half wave plate and thin film polarizer pair is used to control the UV beam energy for LIF operation in the linear regime (below 5 \( \mu \text{J/pulse} \)). The LIF signal is collected from approximately 1/4 of the 6 cm long discharge region, using the prism shown in Fig. 1, fused silica lenses, and a PMT (same as used for ignition delay measurements but terminated at 50 \( \Omega \)) in combination with a monochromator (Horiba, Model H-10). The monochromator, centered at 310 nm, has a band pass of approximately 20 nm to incorporate the entire (1,1) and (0,0) OH fluorescence bands and avoid spectral bias. A neutral density filter is used at high signal incidences to avoid PMT saturation. The LIF signal is integrated over a 100 ns gate and analyzed in real time using a programmable digital oscilloscope with 1 GHz bandwidth (LeCroy, Model WAVE-PRO 7100A).

For two-line OH LIF measurements, the laser system is tuned to two absorption transitions in the OH A-X (1,0) band, \( Q_1(5) \) at 282.667 nm and \( Q_2(11) \) at 285.073 nm, chosen for optimum sensitivity in preheated mixtures during ignition, i.e. at \( T = 500–2000 \text{ K} \). The excitation spectrum is obtained by scanning the laser system across each absorption line, at the tuning speed of 1.25 pm per 60 laser shots. The spectrum is fitted to a Voigt profile to remove the satellite lines (such as \( Q_21(5) \)), and to integrate the signal over the same wavelength range for both pump transitions. Fitting residuals are used to estimate statistical uncertainties in inferred temperature and OH concentration. Relative OH concentrations were put on an absolute scale using calibration in an atmospheric pressure near-adiabatic ethylene–air...
flat flame generated by a Hencken burner (Technologies for Research, Model RD5X5). The burner consists of a 12.7-mm square hastalloy honeycomb burner surface surrounded by a 6.5-mm-wide nitrogen coflow, used to isolate the flame from the surrounding air and to improve flame stability [16]. Air flow rate through the burner was 4000 SCCM, with ethylene flow rate varied from 140 to 350 SCCM (equivalence ratio $\phi = 0.5–1.5$). At these conditions, the flame is nearly adiabatic and temperature and OH concentration are close to thermochemical equilibrium [17]. During calibration, the burner was positioned at the same location as the discharge cell to maintain the same signal collection efficiency. For LIF measurements in the flame, OH($A^2\Pi$) quenching rate is calculated based on the cross sections and temperature scaling relations [18]. Vibrational energy transfer (VET) rate coefficients for OH($A^2\Pi$, $v' = 1$) are taken from Ref. [19]. In the plasma flow reactor, the quenching rate was determined directly from the fluorescence signal decay rate.

Statistical uncertainty of temperature inference using the line area ratio, controlled primarily by the energy separation of the two pump transitions, is estimated to be $\pm 2.5\%$ at relatively low temperatures at the end of the discharge burst (see Section 4), increasing to $\pm 5\%$ at near peak temperature. At $T = 1000–2000$ K, the systematic error is likely to be small, since in this range temperature inferred from LIF data is validated using measurements in the flame. At $T < 1000$ K, the inferred temperature uncertainty may increase since LIF signal from Q$_{\delta}(11)$ transition at these conditions becomes rather weak. The estimated uncertainty in inferred OH concentration caused by the uncertainty in temperature is $\pm 15\%$ at the end of the burst and $\pm 10\%$ at near peak temperature. The uncertainty in OH concentration due to the uncertainty in the VET rate is estimated to be approximately $25\%$. Intrinsic uncertainties of the calibration process, such as the difference in the optical efficiency of signal collection from the flame and from the plasma may also affect the overall systematic error in the absolute OH concentration.

3. Plasma chemistry/pulsed discharge model

To obtain insight into kinetics of plasma/chemical fuel oxidation and ignition, we used a kinetic model developed in our previous work [10,12–14]. Briefly, the model incorporates non-equilibrium air plasma chemistry [20], expanded to include hydrogen dissociation processes in the plasma [14] and hydrogen–oxygen chemistry model developed by Popov [21]. The list of air plasma chemistry processes and hydrogen–oxygen chemical reactions incorporated in the present model is given in Refs. [20] and [21], respectively.

The dominant radical species generation processes in H$_2$–air plasma, as well as principal hydrogen oxidation reactions are listed in Ref. [14]. The species concentration equations are coupled with the two-term expansion Boltzmann equation for plasma electrons. The model incorporates energy equation for the temperature on the discharge centerline [14], with heat transfer to the walls being the dominant energy loss mechanism, as well as quasi-one-dimensional flow equations. The model is validated using TALIF O atom concentration measurements [9,10], rotational CARS temperature measurements [12], OH LIF and ignition time measurements [13,14] in nanosecond discharges in air and in hydrogen–air, showing good agreement.

Key parameters controlling plasma chemistry in the nanosecond pulse discharge are the reduced electric field, $E/N$, and coupled pulse energy. In the present work, energy coupled to the plasma by CPT and FID pulse generators was measured in air in the low-temperature cell (see Section 1), showing that coupled pulse energy is nearly independent on pulse repetition rate (at $v = 0.1–100$ kHz), remains nearly constant during the pulse burst, and is proportional to discharge pressure at $P = 13–100$ torr, i.e. energy coupled per molecule remains constant. Figure 2 plots pulse voltage and coupled energy in air at $P = 40$ torr and room temperature, for CPT and FID pulsers (0.8 mJ/pulse and 2.0 mJ/pulse, see Fig. 2). Higher energy coupled by the FID pulser is mainly due to steeper pulse voltage rise (~5 kV/ns vs. ~1 kV/ns for the CPT pulser).

These results are well reproduced by the analytic model of energy coupling in a nanosecond pulse discharge [22], which incorporates key effects of pulsed breakdown and sheath development on nanosecond time scale. The model predicts pulse energy coupled to the plasma vs. pulse voltage waveform, discharge geometry, pressure, and temperature (1.0 mJ/pulse and 1.6 mJ/pulse for CPT and FID pulsers, respectively, at the conditions of Fig. 2). In the present work, the model of Ref. [22] is used to calculate coupled pulse energy, as well as $E/N$ (approximated as a Gaussian pulse with the same time constant as the experimental voltage waveform) and electron density in hydrogen–air plasma in the preheated flow reactor.

4. Results and discussion

Ignition delay time was measured using the CPT pulser, which provides better shot-to-shot voltage waveform reproducibility. Figure 3 plots three UV emission traces taken in a stoichiometric H$_2$–air flow at $T_0 = 473$ K and $P = 80$ torr, excited by a burst of pulses at $v = 40$ kHz, for different burst durations. All traces are plotted on the same
Emission during the discharge burst is primarily from nitrogen second positive band system \[ [14] \]. \( \text{N}_2 \) emission decays rapidly after each discharge pulse (see inset in Fig. 3) and disappears after the burst. Self-sustained OH (\( \text{A} \rightarrow \text{X} \)) emission, indicative of ignition, appears after the burst and lasts for several milliseconds. As can be seen from Fig. 3, both delay time until \( \text{OH}^* \) emission rise (ignition delay) and peak \( \text{OH}^* \) emission intensity are sensitive to the number of pulses in the burst. The results exhibit well pronounced and well reproduced threshold behavior, such that when burst duration is reduced by a single pulse (from 115 to 114 in Fig. 3), ignition is no longer detected.

Ignition delay time is plotted in Fig. 4(a) vs. the number of pulses in the burst, for \( P = 80 \text{ torr} \).
and 100 torr. Figure 4(a) also plots ignition delay time and temperature at the end of the discharge burst, predicted by the kinetic model. Ignition delay increases steeply as the number of pulses is reduced and approaches the threshold value, a trend well reproduced by the model. The calculations predict that every discharge pulse increases the plasma temperature by ≈2–3 K. It can be seen that ignition after the burst occurs when the temperature predicted at the end of the burst exceeds $T_f = 715$ K (at $P = 80$ torr) or $T_f = 725$ K (at $P = 100$ torr). This is approximately 200 K lower than auto-ignition temperature at these conditions [14].

Figure 4(b) illustrates the effect of discharge burst duration, predicted by the model, in greater

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Fig. 4. (a) Ignition delay and temperature at the end of the burst; (b) radical number densities at the end of the burst at the conditions of Fig. 3.
It can be seen that after the first 50 pulses, the number densities of O atoms and OH generated in the discharge have leveled off and are actually decreasing with the burst duration (primarily by reaction \( \text{O} + \text{OH} \rightarrow \text{H} + \text{O}_2 \)). While the number density of H atoms keeps increasing. As the temperature during the burst continues to rise, due to both Joule heating and gradual hydrogen oxidation, the rates of chain branching reactions, \( \text{H} + \text{O}_2 \rightarrow \text{O} + \text{OH} \) (activation energy \( E_a = 7470 \) K) and \( \text{O} + \text{H}_2 \rightarrow \text{H} + \text{OH} \) (\( E_a = 3160 \) K) increase rapidly. This initiates a well-known chain reaction mechanism, resulting in a non-linear increase of radical concentrations and temperature (see Fig. 4(b)). These results illustrate that both radical concentrations (primarily H atoms) and temperature rise are key for ignition.

Figure 4(a) presents results of single-burst measurements of ignition delay in the near-threshold region, when a small increase in temperature and radical concentrations (varying burst duration by only 1–2 pulses) results in a significant ignition delay change. However, operating near ignition threshold in repetitive burst mode, for temperature and species concentrations measurements by LIF, is difficult for the exact same reason. In this case, variations of flow parameters caused by ignition during a previous burst adversely affect reproducibility. The present work is focused on time-resolved temperature and OH concentration measurements after the discharge burst, for burst durations above the threshold.

All present OH LIF measurements have been conducted using the FID pulser, which produces higher output pulse energy (see Fig. 2). Figure 5 shows broadband images of the plasma and the flame taken with a PI-MAX ICCD camera with a UV lens (Nikon Nikkor 105 mm f/4.5), using the prism placed next to the discharge cell (see Fig. 1). The images are taken in a hydrogen–air mixture at \( \phi = 0.5 \), \( T_0 = 500 \) K, and \( P = 80 \) torr, excited by a 125-pulse burst at \( v = 20 \) kHz. Emission from the plasma (mainly nitrogen second positive bands) is taken using a 2 s camera gate “wrapped around” a single discharge pulse (see inset in Fig. 3). Emission from the flame (mainly OH(A \( \rightarrow X \) bands) is taken after the burst using a 30 \( \mu \)s gate, to improve signal-to-noise and discriminate against strong plasma emission. Flame images in Fig. 5 are labeled according to delay time after the burst.

From Fig. 5, it can be seen that the plasma appears diffuse and quite uniform, although brighter regions can be identified near the electrode edges. Ignition is first initiated in one of these regions, and is followed rapidly by ignition in the rest of the plasma (time delay between 0.63 ms and 0.75 ms), without clearly defined propagating flame front, such as detected in our previous experiments in plasma assisted ignition of ethylene–air [14]. This suggests that ignition in the bulk of the plasma is not caused by earlier ignition near the edge. Large-volume ignition in the entire field of view provides a fairly uniform region for LIF diagnostics. At burst repetition rate of 2 Hz, both ignition delay time and peak OH* emission intensity are reproduced very well burst-to-burst.

Absolute OH concentration and temperature in H\(_2\)–air mixtures excited by a discharge pulse burst \((T_0 = 500 \text{ K, } P = 80 \text{ torr, } v = 10 \text{ kHz})\), inferred from LIF data, are plotted vs. time delay after the burst in Fig. 6. The data in Fig. 6 are shown for three cases, (a) \( \phi = 0.5 \), 115 pulses, (b) \( \phi = 0.4 \), 115 pulses, and (c) \( \phi = 0.5 \), 105 pulses. Figure 6 also plots OH(A \( \rightarrow X \)) emission traces (in arbitrary units) taken simultaneously with the LIF data. It can be seen that temperature immediately after the discharge burst is \( T_f \approx 700–750 \text{ K, i.e. } 200–250 \text{ K higher than the initial temperature of the mixture and significantly lower than autoignition temperature at these conditions, } \approx 900 \text{ K} \) [14]. This shows that flow temperature rise in the discharge is fairly modest, approximately 2 K/pulse. OH concentrations accumulated by the end of the burst are \( 0.86 \times 10^{14} \text{ cm}^{-3} \), \( 1.1 \times 10^{14} \text{ cm}^{-3} \), and \( 0.52 \times 10^{14} \text{ cm}^{-3} \) for the three cases shown, respectively.

The temperature increases up to \( T_{\text{peak}} \approx 1500–1600 \text{ K} \) within 1–5 msec from the end of the discharge burst (see Fig. 6). This is accompanied by an OH concentration jump by a factor of 20–50 compared to the value measured at the end of the burst, up to \( (2.3–2.6) \times 10^{15} \text{ cm}^{-3} \). For the shorter burst duration, OH concentration actually decreases before the rise, by approximately a factor of three (see Fig. 6(c)). The rise of temperature and OH concentration is indicative of ignition, which is also detected from OH* emission traces and flame images (see Fig. 5). In all cases shown in Fig. 6, OH* emission peaks approximately at the same time when ground state OH concentration reaches maximum. After that, temperature and OH concentration reduction are due to heat transfer to the reactor walls and convective cooling of the flow. As expected, ignition delay time increases as the burst duration is reduced, from...
1.0 ms for 115 pulses (case a) to 2.5 ms for 105 pulses (case c). Reducing the equivalence ratio from $\phi = 0.5$ to $\phi = 0.4$, while keeping burst duration the same, increases ignition delay time to 1.5 ms (case b).

Figure 6 also plots temperatures and OH number densities predicted by the kinetic model. Temperatures predicted at the end of the burst, $T_f \approx 750–800$ K, are close to the experimental values. The model predicts that at the present conditions, threshold temperature at the end of the burst necessary to produce ignition is approximately $T_f = 750$ K. Predicted peak temperature after the burst, $T_{\text{peak}} = 1300$ K, is somewhat lower than in the experiment, $T_{\text{peak}} \approx 1500–1600$ K. Underpredicting peak temperature may be due to (i) simplified transient conduction heat transfer model [14], which is valid only for Fourier numbers $Fo > 0.1$ and does not resolve temperature changes on a short time scale, $t \sim 1$ ms, and (ii) pressure overshoot in the flow reactor during ignition. The estimated time for pressure equilibration after ignition is several ms, comparable to experimental temperature rise time (see Fig. 6). This may cause nearly constant volume conditions (i.e. pressure overshoot) in the reactor during ignition, rather than nearly constant pressure conditions before and long after ignition.

OH number densities at the end of the burst predicted by the model for the three cases shown in Fig. 6, $0.97 \times 10^{14}$ cm$^{-3}$, $0.72 \times 10^{14}$ cm$^{-3}$, and $0.53 \times 10^{14}$ cm$^{-3}$, respectively, are close to the experimental values. Predicted peak OH concentration, as well as OH rise time and decay time are also close to the experimental results. However, OH concentration “dip” before ignition for the 105-pulse burst (case c) is not reproduced by the model. In general, the agreement between the model and the experiment is better for 115-pulse bursts (cases a and b). Note that reducing burst duration to 105 pulses brings the excited mixture closer to ignition threshold (illustrated in Fig. 3 for the CPT pulser), when burst-to-burst data reproducibility is not as good.

As discussed in our previous work [13,14], dominant processes of O and H atom generation in nanosecond pulse discharge include electron impact dissociation, $O_2 + e^{-} \rightarrow O + O + e^{-}$, $H_2 + e^{-} \rightarrow H + H + e^{-}$, and dissociation by electronically excited nitrogen molecules, $N_2(A^3 \Sigma) + O_2 \rightarrow N_2 + O + O$, $N_2(a^1 \Sigma, A^3 \Sigma) + H_2 \rightarrow N_2 + H + H$. Key processes of OH production and decay (a reduced 9-reaction mechanism) are identified in Ref. [13]. As discussed above, H atom accumulation during the discharge burst occurs primarily by reaction $O + OH \rightarrow H + O_2$. Analysis of the experimental data and modeling calculations shown in Figs. 4 and 6 demonstrates that ignition of $H_2$–air mixtures by a repetitive nanosecond pulse discharge occurs due to accumulation of H atoms, triggering a chain mechanism, $H + O_2 \rightarrow O + OH$ and $O + H_2 \rightarrow H + OH$, below autoignition temperature (by up to 200 K at the present conditions). The present results suggest that future studies of
plasma assisted ignition of hydrogen–air mixtures should focus on time-resolved measurements of H atoms (the most abundant radical generated in the plasma). These data would provide further validation of the present kinetic mechanism and test its predictive capability.

5. Summary

Ignition delay, time-resolved temperature, and time-resolved absolute OH concentration are measured in mildly preheated H2–air mixtures excited by a repetitive nanosecond pulse discharge in a plasma flow reactor. The measurements are conducted in decaying plasma after the discharge burst, to reduce the effect of electronically excited molecules and atoms on ignition kinetics. OH LIF measurements are obtained with the discharge operating in burst mode at a low burst repetition rate, 2 Hz, to improve data reproducibility.

Ignition delay in single discharge burst experiments is measured by monitoring time-resolved OH(A → X) emission near 310 nm. The results demonstrate that ignition delay time increases steeply as the number of pulses in the burst is reduced, exhibiting threshold-like behavior. Near ignition threshold, varying burst duration by a single pulse results in a significant, reproducible change in ignition delay, in good agreement with kinetic modeling. The calculations show that ignition occurs due to (i) accumulation of radicals generated in the discharge, primarily H atoms, and (ii) temperature rise in the discharge, due to Joule heating and energy release in low-temperature hydrogen oxidation reactions. When temperature at the end of the burst becomes sufficient to initiate a chain reaction mechanism in the presence of significant amounts of H atoms (up to ~0.1%), ignition occurs. At the present conditions, this effect results in ignition temperature reduction by up to 200 K, compared to thermal ignition.

Temperature and absolute OH concentration are measured during the ignition process, as functions of time delay after the discharge burst, using two-line OH LIF and absolute calibration in a near-adiabatic, atmospheric pressure C2H4–air flat flame. The results show that the temperature at the end of the discharge burst increases to \( T_f \approx 700–750 \) K, 200–250 K higher compared to the initial flow temperature of 500 K. After a delay of ~1–5 ms after the burst, the temperature rises to 1500–1600 K, indicative of ignition. These results confirm that ignition temperature is reduced by about 200 K compared to autoignition temperature, approximately 900 K at the present conditions [14]. During the ignition process, the OH concentration increases by a factor of 20–50 from the value measured at the end of the discharge burst. A significant delay in the OH concentration buildup, and longer ignition delay time are detected at shorter burst durations. At all conditions tested, OH(A → X) emission peaks at the same time when ground state OH concentration reach maximum. Kinetic modeling calculations show good agreement with the experimental results, demonstrating quantitative insight into kinetics of plasma-assisted ignition of H2–air.

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References

