

Calorimetry and Imaging of Plasma Produced by a Pulsed Nanosecond Discharge Igniter in EGR Gases at Engine-Relevant Densities

Benjamin Matthew Wolk and Isaac Ekoto
 Sandia National Laboratories

ABSTRACT

Pulsed nanosecond discharges (PND) can achieve ignition in internal combustion engines through enhanced reaction kinetics as a result of elevated electron energies without the associated increases in translational gas temperature that cause electrode erosion. Atomic oxygen (O), including its electronically excited states, is thought to be a key species in promoting low-temperature ignition. In this paper, high-voltage (17-24 kV peak) PND are examined in oxygen/nitrogen/carbon dioxide/water mixtures at engine-relevant densities (up to 9.1 kg/m³) through pressure-rise calorimetry and direct imaging of excited-state O-atom and molecular nitrogen (N₂) in an optically accessible spark calorimeter, with the anode/cathode gap distance set to 5 mm or with an anode-only configuration (DC corona). The conversion efficiency of pulse electrical energy into thermal energy was measured for PND with secondary streamer breakdown (SSB) and similar low-temperature plasmas (LTP) without. The calorimetry measurements confirm that, similar to inductive spark discharges, SSB discharges promote ignition by increasing the local gas temperature. LTP discharges, on the other hand, had very little local gas heating, with electrical-to-thermal energy conversion efficiencies of ~1% at 9 bar. Instead, LTP discharges were found to generate substantial electronically-excited O-atom populations at lower pressures, but the observed image intensity decreased rapidly as the initial pressure was increased. The observed O-atom emission peaked ~20 ns after the start of the pulse and was concentrated near the anode and cathode tips, indicating that the presence of the cathode was beneficial for increasing radical production (although the likelihood of SSB increased). Decreasing oxygen and increasing carbon dioxide concentrations were found to reduce the observed image intensity, but had minimal impact on SSB probability and electrical-to-thermal conversion efficiency. The impact of changes in collisional quenching and the electron energy distribution on image intensity were evaluated.

CITATION: Wolk, B. and Ekoto, I., "Calorimetry and Imaging of Plasma Produced by a Pulsed Nanosecond Discharge Igniter in EGR Gases at Engine-Relevant Densities," *SAE Int. J. Engines* 10(3):2017, doi:10.4271/2017-01-0674.

INTRODUCTION

Plasmas - defined here as globally neutral gaseous mixtures of positively- and negatively-charged particles that interact by electric forces in a collective manner - have long been used to control ignition and combustion processes. Conventional inductive coil sparks that feature thermal plasmas with electron energies in equilibrium with the bulk gas energy have fallen out of favor for next-generation automotive engines due to excessive electrode erosion rates from high-temperature arc discharges and poor tolerance to dilution by air or exhaust-gas recirculation (EGR) [1]. Conversely, ignition induced by non-equilibrium low-temperature plasma (LTP), where an imbalance exists between ion and electron temperatures, leads to enhanced reaction kinetics as a result of elevated electron energies without the associated translational gas temperature increases that cause electrode erosion. Automotive low-temperature plasma-assisted ignition (PAI) systems extend dilution tolerance limits through the formation of active radicals that shorten ignition delays [2] and rapid heating via electron energy transfer processes [3, 4, 5]. Ignition kernel expansion speeds may also be influenced by hydrodynamic instabilities and ionic winds [6, 7]. LTP generated atomic oxygen (O-atom) in particular, including its electronically-excited states, has been identified as an important radical species that accelerates

hydrogen abstraction from the fuel, which in turn increases overall mixture reactivity [8]. For automotive engine combustion, these attributes reduce ignition delay times, suppress knock inducing end-gas auto-ignition, alter fuel reactivity and diffusivity characteristics, and improve noxious emission oxidation [9].

LTP are generally classified by the reduced electric field strength, or the electric field strength (E) normalized by the gas number density (N), in units of Townsend (Td). Broadly speaking, PAI systems sorted by increasing E/N include: microwaves (10 - 50 Td) [3, 10, 11, 12, 13], high-frequency resonant discharges (50 - 200 Td) [7, 13, 14, 15, 16, 17], and pulsed nanosecond discharges (PND) (100 - 1000 Td) [2, 4, 13, 18, 19, 20, 21]. PND igniters typically have electron temperatures above 10 electron volts (eV) using only modest pulse energies (10s of mJ) due to the truncated discharge [22, 23, 24, 25]. PND are characterized by higher electron energy, higher peak current, and lower gas temperature compared to both microwave and high-frequency resonant discharges [13]. Since LTP-related ionization and dissociation rates increase with higher E/N due to limited collisional electron energy transfer effects [26], several recent PAI experiments have focused on PND systems [22-25, 27].

Cathode-directed PND, such as those used in this work, have two discharge phases: the primary streamer and the secondary streamer. The primary streamer propagates as a “plasma bullet” across the electrode gap ($O(1\text{ ns})$), with intense E/N at the primary streamer head. After the primary streamer reaches the cathode, a slower secondary streamer with lower E/N begins to propagate from the anode. If the secondary streamer reaches the cathode ($O(10\text{ ns})$), breakdown occurs; this will be referred to as secondary streamer breakdown (SSB) throughout this paper. The likelihood of SSB increases for decreased gas resistance; i.e., from low pressures, high temperatures, short electrode gap distances, or gases with low ionization potential [28, 29]. A SSB discharge more closely resembles a thermal plasma spark than a LTP discharge with 2 key differences: (1) the electrical-to-thermal energy transfer is generally more efficient [1, 30], and (2) electrode wear greatly increases due to the elevated discharge current. The electrical-to-thermal energy conversion efficiency also increases with increasing pressure, increased gap size, and reduced pulse energy [1, 30]. Due to the elevated electrode wear from SSB, PND igniters should be designed to avoid SSB.

While SSB-related electrode erosion issues can be mitigated through the use of dielectric electrode insulators that repel the discharge current [31, 32], the electric field strength becomes compromised with an associated decrease in radical formation rates. Instead it may be preferable to generate LTP without the use of dielectrics, with SSB instead inhibited through the selection of appropriate electrode geometry and pulse discharge characteristics. It is important to note that at engine-relevant pressures, LTP streamer propagation deviates from the well-established self-similar laws (i.e., increased streamer branching, thinner leader structures, and slower than expected streamer velocities) with unknown impact on discharge kinetics [33, 34, 35]. Potential causes for the scaling deviation at high-pressure include a change in electron recombination physics [33], less effective streamer cooling by thermal conductivity [36], and photoionization by excited-state nitrogen relaxation [34].

Direct imaging of single-pulse PND discharges using intensified cameras with short gate times (e.g. 5 ns) has provided insight into primary and secondary streamer propagation and structure [29, 37, 38, 39], specifically the impact of oxygen (O_2) and water concentration on streamer branching at atmospheric or sub-atmospheric pressure. Ono et al. [37] identified specific species produced during each streamer phase: the excited state transition of molecular nitrogen $N_2(C^3\Pi_u \rightarrow B^3\Pi_g)$ was observed in both the primary and secondary streamers, while atomic oxygen was inferred from ozone measurements to only be created in the secondary streamer. In this work, the $N_2(C^3\Pi_u \rightarrow B^3\Pi_g)$ transition was directly-imaged in order to visualize primary and secondary streamer structure, while O-atom was visualized using the $O(3p^3P \rightarrow 3s^3S)$ transition.

Previous engine research studies have investigated the use of multi-pulse PND operation, where close-coupled PND with pulse dwell times of $\sim 100\text{ }\mu\text{s}$ were used to continuously augment radical production while ideally avoiding SSB [22, 23]. However, direct discharge imaging during stable engine operation revealed that SSB almost always occurred at some point during the pulse burst; an example image of the luminous SSB event from a 10-pulse burst

from Sjöberg et al. [23] is shown in Figure 1. Moreover, once SSB occurred during the burst, each successive discharge in the pulse-burst exhibited SSB biased towards the same cathode site as the first SSB event. Wolk and Ekoto [40] identified fast-gas heating along streamer channels as a likely mechanism that leads to this behavior for PND in air. The impact of gas composition, specifically EGR constituents and fuel components, on SSB propensity in high-voltage PND has not yet been evaluated.

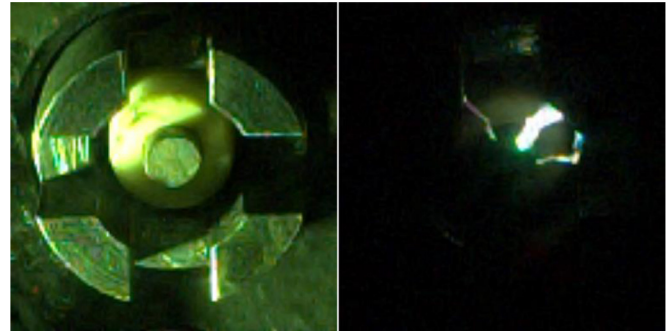


Figure 1. Image of a 4-prong PND plug in a single-cylinder research engine along with imaging for 10-pulse PND bursts with a $100\text{ }\mu\text{s}$ dwell between successive pulses. Intense luminosity between the central anode and one of four separated cathode sites is indicative of SSB.

The current paper describes experiments performed in a custom-built, optically accessible spark calorimeter used to evaluate high-voltage PND at engine-relevant densities, specifically in EGR-like gases at elevated pressure. For all experiments, the test gas mixture was a combination of ultra-air, N_2 , carbon dioxide (CO_2), and water (H_2O) with a point-to-point electrode configuration selected so that the local E/N , and hence active radical formation, was maximized near the anode and cathode. Pressure-rise calorimetry was used to measure the electrical-to-thermal energy conversion efficiency for single-pulse PND discharges as a function of ambient pressure and gas composition. High-speed photodiode measurements were used to determine the SSB probability for each pressure and gas composition; the pulse electrical energy and electrical-to-thermal energy conversion efficiency were conditionally-averaged based on whether the PND remained an LTP or transitioned to a SSB discharge. Simultaneous direct imaging of excited-state O-atom and molecular nitrogen emission was performed for select LTP discharges at a range of initial pressures (P_{init}) and gas compositions to evaluate the impact on O-atom production and streamer structure.

APPROACH

All experiments were performed in a custom-built, optically accessible, spark calorimeter illustrated in Figure 2. The calorimeter body is constructed of 316 stainless steel, with a cylindrical 29 cm^3 internal chamber volume (v_{cal}). An inlet port was used to fill the chamber to the desired pressure (up to 9 bar absolute) with a premixed ultra-air/ N_2 / CO_2 / H_2O mixture. The gas mixture was prepared using MKS GE50A flow controllers and a syringe pump (Valcor SV690) and was stored in a 1-liter reservoir with a metered bypass valve. The calorimeter and reservoir were either unheated or heated to 70°C using resistive heating tape with the temperature

monitored using embedded type-K thermocouples. The calorimeter thermocouple was installed flush with the wall of the calorimeter internal volume.

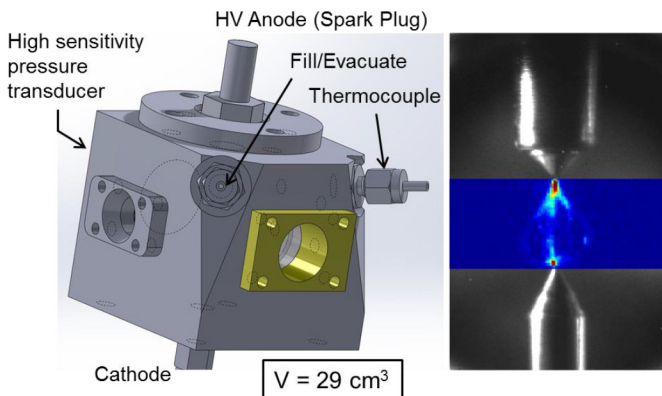


Figure 2. Illustration of the optically accessible spark calorimeter (left) and an image of the spark gap as visualized through the optical viewing port (right). An example image of filtered natural luminosity from the discharge has been superimposed between the electrodes.

The calorimeter body also features viewing access of the discharge event through an optical port on the front side wall along with input and output laser access ports that facilitate acquisition of laser-based spectroscopy measurements (not employed in this work). The 20 mm clear aperture viewing window enabled direct imaging of the discharge. The 16 mm diameter entrance and outlet laser windows had a 12.9 mm clear aperture and were installed orthogonal to the viewing window in opposing calorimeter sidewall ports. Calorimeter window ports included a 45° beveled seat and copper gasket that mated with a complementary bevel on the front window surface to provide a primary seal and minimize mechanical stresses that could induce window fracture when the calorimeter was pressurized. A metal retainer was used to secure the windows into the calorimeter via 4 screw holes that mated with tapped holes around the window ports; another copper gasket was placed between the retainer and the window back surface to minimize mechanical stresses. A Kalrez O-ring was placed into a machined O-ring groove in each window port to further prevent leakage around the windows when the calorimeter was pressurized.

To generate the PND within the spark calorimeter, a Transient Plasma Systems Inc. SSPG-101-HF high-voltage (28 kV peak) pulse generator with a 12 ns full width at half max (FWHM) pulse width and a 5 ns rise time was used. A low-impedance inline attenuator was used to monitor pulse voltage and current for each discharge event. The high-voltage anode was a modified NGK DP7EA-9 size M12 non-resistive sparkplug that was centrally positioned on the calorimeter top. The plug J-hook and the top 1 cm of material from the outer body ground was removed, with the anode tip machined to a rounded point (~125 μm radius of curvature) so that local electric field strengths were maximized, while maintaining relatively repeatable discharge characteristics. The cathode was replaced by a sharpened 3.18 mm diameter steel rod installed from the calorimeter base and secured in place by a Swagelok fitting. The inter-electrode distance between the anode and cathode was set to 5.0 mm. Alternatively, an “Anode-only” configuration was studied where the

cathode was replaced by a stainless steel rod flush with the calorimeter interior bottom surface (19.6 mm from anode tip). Key calorimeter specifications and operating conditions are summarized in Table 1.

Table 1. Calorimeter specifications and operating conditions.

Calorimeter Internal Volume [cm^3]	29.0
Purge and fill cycle time [s]	105
Initial Temperature [$^{\circ}\text{C}$]	70 ± 1.0 , 21 ± 1.0
Initial Pressure [bar]	$1.07 - 9.0$
Gas Density [kg/m^3]	$1.08 - 9.12$
Inter-Electrode Distance [mm]	5.0, Anode Only
Electrode Tip Radius of Curv. [μm]	~125
Gas Mixtures [mole-%] ^{†‡}	Ultra-air: 21.9% O_2 , 78.1% N_2 ($\gamma = 1.397$) 15.9% O_2 , 84.1% N_2 ($\gamma = 1.397$) 15.9% O_2 , 78.8% N_2 , 5.3% CO_2 ($\gamma = 1.387$) 15.9% O_2 , 77.0% N_2 , 3.7% CO_2 , 3.4% H_2O ($\gamma = 1.387$)

[†]Ratio of specific heats (γ) calculated at 70 $^{\circ}\text{C}$.

[‡]Additional minor species in ultra-air neglected.

Four gas compositions were studied: (1) ultra-zero air (Matheson) with 21.9 mole-% O_2 , (2) ultra-zero air mixed with additional nitrogen to achieve 15.9 mole-% O_2 , (3) ultra-zero air mixed with nitrogen and CO_2 to achieve 15.9 mole-% O_2 and 5.3 mole-% CO_2 , and (4) ultra-zero air mixed with nitrogen, CO_2 , and H_2O to achieve 15.9 mole-% O_2 , 3.7 mole-% CO_2 , and 3.4 mole-% H_2O . The concentrations of O_2 and CO_2 in the tested gas mixtures were verified using a CAI 600 Series multi-gas analyzer. The O_2 concentration of 15.9% was adopted from the intake composition estimated from experimental engine data from Argonne National Laboratory using a similar PND system [22]: 15.9% O_2 , 3.7% CO_2 , 3.4% H_2O , 77.0% N_2 (excluding hydrocarbons). In [22], the 12.1:1 compression ratio engine was operated at 1500 rpm, 5.6 bar indicated mean effective pressure (IMEP), and 23 mole-% EGR. The CO_2 concentration of 5.3% in the ultra-air/ N_2 / CO_2 mixture was chosen to achieve the same heat capacity as the intake composition in [22] in order to isolate the impact of H_2O from changes in heat capacity. Although not included in this work, the presence of hydrocarbons may have an important impact on discharge behavior and will be evaluated in a future study.

Pressure-Rise Calorimetry

For the calorimetry experiments, P_{init} was varied between 1.07 bar and 9 bar absolute with initial temperatures fixed at 70 ± 1.0 $^{\circ}\text{C}$. For these conditions, the chamber gas densities were similar to the densities encountered in the engine in [22]. This is illustrated in the upper plot of Figure 3 where the in-cylinder density from the fired engine in [22] is plotted versus crank angle, with the densities tested in the calorimeter indicated for the range of P_{init} . The spark timing from the engine experiments is indicated in Figure 3 by the vertical dashed line - the in-cylinder density at the time of spark was equivalent to $P_{init} = 4.75$ bar in the calorimeter. The in-cylinder pressure and bulk-averaged temperature from [22] are also plotted versus crank angle in the bottom of Figure 3 for reference.

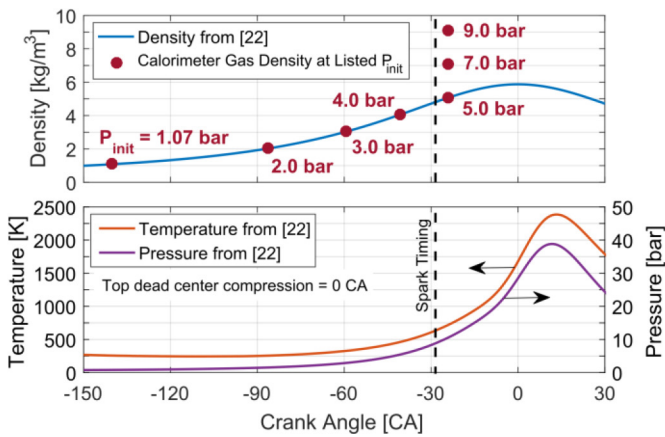


Figure 3. In-cylinder density (top), temperature (bottom), and pressure (bottom) as a function of crank angle for a fired engine experiment in [22]. The densities tested in the calorimeter are indicated for the range of P_{init} .

The calorimeter gas densities for $P_{init} = 7.0$ bar and $P_{init} = 9.0$ bar were greater than the top dead center density from the engine experiments. However, note that the intake was slightly throttled in [22] such that the intake pressure was ~ 71 kPa (absolute). If the engine in [22] was operated at an intake pressure of ~ 105 kPa, the gas density at the time of spark would be equivalent to the calorimeter gas density for $P_{init} = 7.0$ bar. Similarly, the calorimeter gas density at $P_{init} = 9.0$ bar would be representative of the engine operated at an intake pressure of ~ 135 kPa.

For each gas composition, initial pressure, and electrode configuration studied, 30 calorimetry runs were performed. To ensure that residual discharge products did not persist between runs and possibly influence the subsequent discharge, chamber contents after each run were cycle-purged twice using fill and vacuum solenoid valves connected to the gas supply and an available vacuum turbo-pump, respectively. Chamber fill pressure was accurately controlled using a Proportion-Air QPV pressure control valve. Differential chamber pressures were measured using a PCB 106B51 high-sensitivity pressure transducer ($145 \mu\text{V}/\text{Pa}$). The transducer was secured to the calorimeter using an acetal mounting adapter for electrical isolation with the signal amplified by a Kistler 5134B coupler. All data were recorded using a LeCroy HDO 6054 500 MHz high-definition oscilloscope. Recorded pressures were filtered by a 1 kHz low-pass filter during post-processing to remove noise from the discharge event. The SSB probability at each condition was determined from light emission recorded by a Thorlabs DET210 high-speed photodiode placed against an available window port.

Electrical-to-thermal energy conversion efficiency - defined here as the thermal energy deposited into the gas (E_{therm}) normalized by the pulse electrical energy delivered (E_{elec}) to the igniter - was determined for each discharge. Pulse electrical energy was obtained by direct integration of the instantaneous electrical power (W), calculated from the product of the inline attenuator voltage (V) and current (I) measurements over the length of each pulse (t_p):

$$E_{elec} = \int_0^{t_p} W(t)dt = \int_0^{t_p} V(t)I(t)dt \quad (1)$$

Gas thermal energy deposition was determined from the volumetric integration of the change in internal energy (δu):

$$E_{therm} = \int \delta u dV = \int \rho c_v \Delta T dV = \left(\frac{c_v}{R}\right) V_{cal} \Delta P = \frac{V_{cal} \Delta P}{\gamma - 1} \quad (2)$$

where ρ , R , c_v , and γ respectively are the density, gas constant, volumetric heat capacity, and specific heat ratio of the gas mixture. The pressure-rise (ΔP) was the highest recorded differential pressure measurement from a calorimetry experiment. Due to the small pressure-rises encountered in the experiments, a constant γ based on the gas composition and initial temperature was assumed.

Direct Imaging of Discharge

For the direct imaging experiments, P_{init} was varied between 1.7 bar and 2.0 bar absolute with initial temperatures fixed at 21 ± 1.0 °C. A lower temperature was used for the imaging studies compared to the calorimetry because no image signal was observed for LTP-only conditions at 70°C. The limited pressure range studied at 21°C was a result of SSB occurrence preventing imaging below $P_{init} = 1.7$ bar and an absence of signal for $P_{init} > 2.0$ bar.

In order to qualitatively evaluate the production and distribution of atomic oxygen, direct imaging was performed using the $\text{O}(3p^3P \rightarrow 3s^3S)$ transition at 844.9 nm. Excitation of atomic oxygen to the $\text{O}(3p^3P)$ state occurred via electron impact excitation during the discharge and emission at 844.9 nm was isolated using a narrowband interference filter centered at 840 nm with 10 nm bandwidth (Andover 840FS10-50; 57% transmission at 844.9 nm). Light emitted from the discharge through the viewing window was separated using a dichoric beam splitter at 45° angle of incidence with high transmissivity ($>98\%$) at 844.9 nm and high reflectivity ($>95\%$) from 310 nm to 360 nm (Lattice Electro Optics LWP-45-R337.1-T844.9-UF-2025), as shown in Figure 4. To determine the streamer structure, direct imaging of excited state N_2 transitions near the $\text{N}_2(\text{C}^3\Pi_u \rightarrow \text{B}^3\Pi_g)$ transition at 337.1 nm was performed simultaneously with the $\text{O}(3p^3P \rightarrow 3s^3S)$ imaging by using the unfiltered reflection from the dichoric beam splitter.

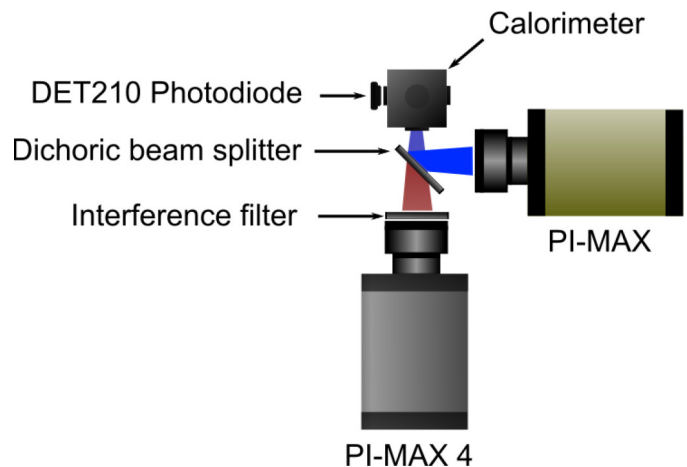


Figure 4. Diagram of the imaging setup used to simultaneously image excited state transitions of O-atom (PI-MAX 4; 844.9 nm) and N_2 (PI-MAX 4; 337.1 nm) transitions near 337.1 nm).

O-atom emission images were recorded using a Princeton Instruments PI-MAX 4: 1024 EM intensified camera with a 1024×1024 pixel imaging array, 2x2 hardware binning, an 85 mm *f*/1.8 Nikkor lens, and a 56 mm extension tube. The PI-MAX 4 was operated at a gain (*G*) setting between *G* = 400 and *G* = 10,000 (maximum) with gate durations of 5 ns or 500 ns. The gate duration of 5 ns was used to capture time-resolved excited-state O-atom images, while the 500 ns gate duration was used to image the entire discharge event. Similarly, N₂ emission images were recorded using a Princeton Instruments PI-MAX intensified camera with a 512×512 pixel imaging array, an 85 mm *f*/1.8 Nikkor lens, and a 56 mm extension tube. The PI-MAX was operated at maximum gain (*G* = 255) with 500 ns gate duration only. To prevent intensifier damage for both cameras, only consistent LTP conditions (i.e. SSB probability equals zero) were imaged.

In order to increase the data acquisition rate for the direct imaging experiments, the pressure transducer was removed and replaced with a metal plug containing a threaded outlet port. The desired gas mixture flowed continuously through the calorimeter with discharges at 10-second intervals (>2 calorimeter flow through times), for a total of 30 discharges using the 500 ns gate duration. For the 500 ns gate duration imaging, the intensifier gate delays were adjusted to achieve coincidence of the intensifier gate timing between the two cameras, verified using a high-speed oscilloscope. For the 5 ns gate duration, 100 discharges were recorded at each tested operating condition. Only one image could be recorded per discharge; however, normally-distributed jitter in the start of discharge time ($\sigma = 60$ ns) led the 5 ns gate duration images to span a range of times after the discharge. The image time relative to the start of the discharge was determined in post-processing using the intensifier monitor signal from the PI-MAX 4 and the rising edge of the discharge voltage trace.

Images from each camera were separately background subtracted and flat-field corrected. The intensity of excited-state O-atom images collected using *G* < 10,000 were corrected up to *G* = 10,000 on a pixel-by-pixel basis using the ratio of the flat-field images at the *G* used (e.g. *G* = 400) and *G* = 10,000. As a result, unless otherwise noted, the intensity of all images for each gate duration can be directly compared. Average images for the 500 ns gate duration were constructed by median-averaging the image sets on a pixel-by-pixel basis. The 5 ns gate duration images were spatially median-filtered using a 5×5 pixel window.

RESULTS AND DISCUSSION

Pressure-Rise Calorimetry

SSB Propensity

Fundamental knowledge of the SSB propensity of PND in air and EGR-like gas mixtures will facilitate the development of strategies to avoid SSB in automotive engines. To this end, the SSB occurrence probability was determined for the gas mixtures listed in Table 1 as a function of P_{init} from 1.07 bar to 9.0 bar at 70°C using the 5.0 mm gap, with the results plotted in Figure 5. The Anode-only configuration was also evaluated for the 21.9% O₂ mixture. The filled

symbols in Figure 5 indicate experiments that were conducted using a different bottle of ultra-air than the open symbols - the impact of the change in ultra-air bottle will be discussed later in this section.

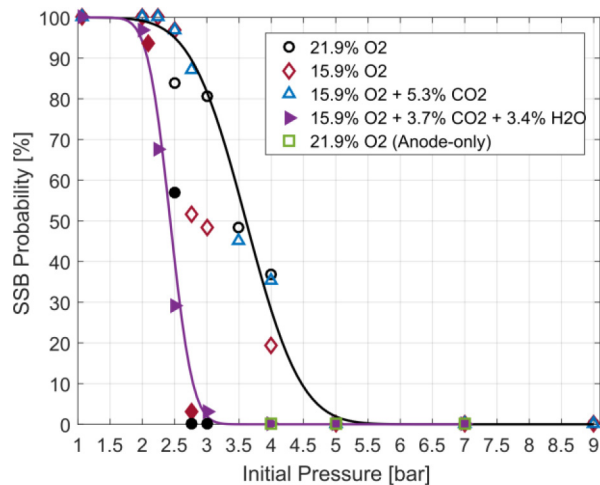


Figure 5. SSB occurrence probability at 70°C as a function of initial pressure and gas composition for a single ~24 kV_{peak} discharge using the 5.0 mm gap. 30 experiments were conducted at each condition. The solid lines are inverse error function curve fits. Filled symbols indicate data points that were taken using a different ultra-air bottle compared to the open symbols.

Looking first at the open symbols in Figure 5, the SSB occurrence probability did not depend appreciably on the O₂ or CO₂ mole fractions at the tested conditions. For all tested gas mixtures, the discharge exhibited 100% SSB occurrence at $P_{init} \leq 2.0$ bar. For $P_{init} > 2.0$ bar, the SSB occurrence probabilities decreased with increasing pressure. For the 5 mm gap at $P_{init} \geq 5.0$ bar, only LTP discharges were observed.

The Anode-only configuration was run for $P_{init} \geq 4.0$ bar as arcing upward towards the calorimeter top was observed for $P_{init} < 4.0$ bar, potentially along the ceramic insulator of the spark plug. The Anode-only configuration operated as a LTP discharge at $P_{init} = 4.0$ bar, where the SSB occurrence probability was near 30% for the 5 mm gap. This illustrates the main advantage of the Anode-only configuration: to reduce the occurrence of SSB.

The SSB occurrence probability curve shifted to lower pressures with the change in ultra-air bottle (filled symbols in Figure 5), so repeat conditions for the 21.9% O₂ and 15.9% O₂ mixtures were run to compare to the 15.9% O₂ + 3.7% CO₂ + 3.4% H₂O gas mixture. From the filled symbols in Figure 5, it is evident that the SSB occurrence probability did not depend appreciably on the H₂O or CO₂ concentrations examined. However, it can be inferred that some change in the ultra-air composition decreased the SSB occurrence probability for pressures between 2 bar and 5 bar. The authors speculate that the argon concentration may have decreased between the first and second ultra-air bottles, although argon concentration information was not available for the ultra-air used in this study.