



# Stability enhancement of ozone-assisted laminar premixed Bunsen flames in nitrogen co-flow



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## ABSTRACT

Ozone (O<sub>3</sub>) is known as one of the strongest oxidizers and therefore is widely used in many applications. Typically in the combustion field, a combination of non-thermal plasma and combustion systems have been studied focusing on the effects of ozone on flame propagation speeds and ignition characteristics. Here, we experimentally investigated the effects of ozone on blowoff of premixed methane/air and propane/air flames over a full range of equivalence ratios at room temperature and atmospheric pressure by using a co-flow burner and a dielectric barrier discharge. The results with ozone showed that a nozzle exit jet velocity at the moment of flame blowoff (blowoff velocity) significantly increased, and flammability limits for both fuel-lean and rich mixtures were also extended. Ozone had stronger effects of percent enhancement in the blowoff velocity for off-stoichiometric mixtures, while minimum enhancements could be observed around stoichiometric conditions for both fuels showing linear positive dependence on a tested range of ozone concentration up to 3810 ppm. Through chemical kinetic simulations, the experimentally observed trends of the enhancement in blowoff velocity were identified as a result of the modification of the laminar burning velocity. Two ozone decomposition pathways of O<sub>3</sub> + N<sub>2</sub> → O + O<sub>2</sub> + N<sub>2</sub> and O<sub>3</sub> + H → O<sub>2</sub> + OH were identified as the most controlling steps. These reactions, coupled with fuel consumption characteristics of each fuel determined the degree of promotion in laminar burning velocities, supporting experimental observations on blowoff velocities with ozone addition.

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## 1. Introduction

Plasma-assisted combustion has been extensively studied over the last few decades and has shown promising effects on controlling or enhancing fundamental combustion phenomena, such as reducing ignition delay time [1–4], increasing flame propagation speed [5], enhancing flame stabilization [6–9], and reducing emissions [10,11]. Many feasibility tests of plasma-assisted combustion have been applied to systems using various types of plasmas ranging from thermal to non-thermal discharges, with care taken to consider how the discharge is coupled to the reactive system. Nevertheless, the coupling process can be complicated enough that it is difficult to quantify the enhancement mechanisms. These enhancement mechanisms can be summarized in three categories [3,12–17]; (1) thermal enhancement by joule heating, (2) chemical kinetic enhancement by active plasma species, and (3) manipulation of transport (diffusion and convection) properties through ionic wind effects.

Although thermal plasmas have shown the instantaneous impacts on enhancing ignition [18–20] and flame stabilization [21], more advanced semi-volumetric applications have been limited because of the inefficiency of plasma energy deposition caused by its intrinsic nature to be a localized discharge. In this regard, recent studies have devoted extensive efforts towards revealing the chemical kinetic enhancement mechanisms by implementing non-thermal plasmas, targeting its unique characteristics in generating active plasma species [9,14,22]. Non-thermal plasma has been known to be more effective than thermal plasma in producing active plasma species, such as active radicals, excited species, and ions/electrons, thus promoting the rate of chemical energy conversion, which consequently can significantly enhance combustion performance [16]. Furthermore, high electrical potentials of the order of kilovolts have been applied to combustion systems and it has been found that electric fields can also alter important transport properties in combustion processes. The changes of transport properties led to enhanced stability of non-premixed jet flames [17,23,24] and premixed flames [15,25], as well as increased flame propagation speeds for tribrachial flames [17,26] and outwardly propagating spherical premixed flames [27].

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In terms of the methodology to couple the plasma energy into a reactive system non-thermally, there are three different strategic approaches; (1) pretreatment of reactants to reform/crack fuel molecules [28,29], or to supply plasma-generated chemically active species, (2) direct in situ plasma discharges on the reaction zone to activate electron induced chemistry (plasma chemistry) in a flame [11], and (3) after-treatment of combustion emission such as soot and  $\text{NO}_x$  [30]. Among the aforementioned approaches, the pretreatment of the oxidizer has been found to be very efficient in generating a more vigorous oxidizer, such as electronically excited oxygen, atomic oxygen, and ozone [12,13,31–33]. Furthermore, these oxidizer species can be selectively produced and/or isolated, thus enabling quantitative identification of the enhancement mechanisms [12,32,33].

By supplying oxygen to a non-thermal electrical discharge, oxygen atoms (O) can be released from  $\text{O}_2$  by electron impact dissociation even at room temperature conditions, upon which they readily combined with molecular oxygen ( $\text{O}_2$ ) to produce the stable species of ozone ( $\text{O}_3$ ). Ozone is known as a powerful oxidant and has been widely investigated to increase the performance of combustion and to reduce emissions [31,34–36]. However, throughout the electrical discharge processes, other byproducts such as  $\text{O}_2(v)$ ,  $\text{O}(^1\text{D})$ ,  $\text{O}(^1\text{S})$ ,  $\text{O}_2(a^1\Delta_g)$ , and  $\text{O}_2(b^1\Sigma_g)$ , have to be inevitably produced as well as other excited species, ions, and electrons [12,13]. Recently, Ombrello et al. [12,13] demonstrated that the effects of ozone, which has a relatively long lifetime compared to other plasma-produced species, could enhance the laminar burning velocity. The enhancement by ozone has been attributed to the fact that the decomposition of ozone early in the flame front provides O atoms, thus accelerating the fuel oxidation rate. Consequently, by adding several thousand parts per million (ppm) of ozone, it was found that the laminar burning velocities of hydrocarbon flames could be increased by a few percent [5,12,32,33].

Although the effects of ozone on laminar burning velocity of premixed flames have been reported, the effects of ozone on other flame phenomena, such as blowoff velocity and flammability limits, where the ozone impact might be magnified through the coupling between kinetics and hydrodynamics [12], remain unknown. In this regard, the objective of this study was to investigate the effect of ozone on the flame stability of premixed Bunsen flames. The blowoff velocities and flammability limits for fuel/air pre-mixtures were investigated in terms of ozone addition at atmospheric pressure and room temperature conditions. Two gaseous fuels, methane and propane, which have very different fuel fragmentation processes, were used to clarify the difference in chemical kinetic pathways by ozone addition. Detailed features of the enhancement of blowoff and the extension of flammability are discussed and supported by numerical simulations taking into account chemical kinetic pathways involved.

## 2. Experiment

The experimental apparatus consisted of a co-flow burner, flow controllers, an ozone generator, an ozone monitor, and a laser-induced fluorescence system for OH radicals, as schematically shown in Fig. 1. The co-flow burner had a central nozzle made of stainless steel with its inner and outer diameters of 7.53 mm and 9.54 mm, respectively. To ensure a fully developed parabolic velocity profile at the nozzle exit up to the maximum velocity of the present experiment, well known laminar flow correlations for an entrance length for the fully developed velocity profile, (length of nozzle)/(inner diameter of nozzle)  $\sim 0.06 \text{ Re}$  [37], was considered. By using a 470 mm-length tube, 60 times the nozzle diameter, a fully developed parabolic velocity profiles at the nozzle exit was assured for all flow conditions (Reynolds number  $< 1000$ , thus laminar

regime) in this study. To obtain a uniform flow distribution throughout the co-flow section, a layer of glass beads was packed under a ceramic honeycomb. A nitrogen ( $\text{N}_2$ ) co-flow was used to isolate premixed flames from secondary diffusion flames formed with ambient air at fuel rich conditions, and its flow velocity was fixed at 10.2 cm/s for all test conditions. Mass flow controllers (Brooks Instrument, 5850E) calibrated by a dry-test gas meter (Bios, Definer 220M) were used to control the flow rates of the gases. Methane ( $\text{CH}_4$ , 99.95%) and propane ( $\text{C}_3\text{H}_8$ , 99.5%) were selected as the fuels, and oxygen ( $\text{O}_2$ , 99.9995%) and nitrogen ( $\text{N}_2$ , 99.9999%) were separately supplied keeping their composition as that of air.

A coaxial type dielectric barrier discharge (DBD) reactor was employed as the ozone generator. A quartz tube was used as a dielectric material having an outer diameter of 30 mm, an inner diameter of 27 mm, and a length of 300 mm. Stainless steel mesh wrapped around the quartz tube with 200 mm in length served as a ground electrode. At the center of the quartz tube, a stainless steel rod with an outer diameter of 25 mm was used as a high voltage electrode; creating a gap between the center electrode and the quartz tube of 1 mm. A power amplifier (Trek, 40/15-H-CE), which could supply voltage and current up to 40 kV and 15 mA, respectively, in conjunction with a function generator (Tektronix, AFG3021B) was used to generate high voltage AC with an arbitrary frequency and energize the ozone generator. The operating frequency and the applied voltage were changed to adjust ozone concentration in the oxidizer stream. It was found that the conditions of  $V = 7.28 \text{ kV}$  in rms value and  $f = 700 \text{ Hz}$  provided a maximum concentration of ozone of 3810 ppm. High-purity oxygen was flowed into the ozone generator to achieve higher yields of ozone without undesired plasma byproducts, such as  $\text{NO}_x$  [12]. Then, as shown in Fig. 1, the ozone containing oxygen stream was mixed with nitrogen to match the air composition (21%  $\text{O}_2$  and 79%  $\text{N}_2$  by volume). The ozone concentration in the  $\text{O}_2$ - $\text{N}_2$  mixture (synthetic air) was measured with a UV ozone monitor (Ebara Jitsugyo, EG-3000). Finally, the ozone containing synthetic air stream was mixed with fuel to form the target equivalence ratio of the fuel/air mixture.

In order to ensure that the ozone measured in the UV ozone monitor was in fact what was entering the flame, a parametric study was performed. The DBD discharge produces many other species besides ozone, such as O,  $\text{O}(^1\text{D})$ , and  $\text{O}_2(a^1\Delta_g)$ . However, since the lifetime of these metastable oxygen species is extremely short at atmospheric pressure [12], the only long lived species is ozone. Therefore, to confirm experimentally that the ozone concentration was constant, different lengths of tubing were tested to vary the flow residence time for the range of conditions used in the experiments. The lack of change in the measured ozone concentration with the different length tubes verified that ozone was the only plasma-produced species present at the ozone monitor. Furthermore, changing the length of tubing between the fuel mixing location and the burner resulted in no changes to the flame. The result confirmed there was no reaction of ozone with the fuels, which is reasonable because ozone reactions with alkanes are very slow at low temperature (295 K) [12,38].

The most important parameter investigated was the blowoff limit of the premixed flames along with the equivalence ratios,  $\phi$ , of the mixtures. Experimentally, to determine a critical nozzle exit velocity at the moment of flame blowoff, a nozzle exit velocity,  $u_{\text{jet}}$ , needed to be varied while maintaining a given equivalence ratio. Since it was difficult to maintain the same concentration of ozone when  $u_{\text{jet}}$  was varied, a bypass system was designed into the experiment. First, the flow rate of oxygen was fixed at 2 l/min, which could accommodate the highest flow rate condition used in the experiments. By fixing the applied voltage and frequency to the ozone generator, the production rate of ozone could be locked,

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