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# Effect of strain rate on sooting limits in counterflow diffusion flames of gaseous hydrocarbon fuels: Sooting temperature index and sooting sensitivity index

Yu Wang, Suk Ho Chung *

*Clean Combustion Research Center, King Abdullah University of Science and Technology, Thuwal, Saudi Arabia*

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ABSTRACT

The effect of the strain rate on the sooting limits in counterflow diffusion flames was investigated in various gaseous hydrocarbon fuels by varying the nitrogen dilution in the fuel and oxidizer streams. The sooting limit was defined as the critical fuel and oxygen mole fraction at which soot started to appear in the elastic light scattering signal. The sooting region for normal alkane fuels at a specified strain rate, in terms of the fuel and oxygen mole fraction, expanded as the number of carbon atoms increased. The alkene fuels (ethylene, propene) tested had a higher propensity for sooting as compared with alkane fuels with the same carbon numbers (ethane, propane). Branched iso-butane had a higher propensity for sooting than did n-butane. An increase in the strain rate reduced the tendency for sooting in all the fuels tested. The sensitivity of the sooting limit to the strain rate was more pronounced for less sooting fuels. When plotted in terms of calculated flame temperature, the critical oxygen mole fraction exhibited an Arrhenius form under sooting limit conditions, which can be utilized to significantly reduce the effort required to determine sooting limits at different strain rates. We found that the limiting temperatures of soot formation flames are viable sooting metrics for quantitatively rating the sooting tendency of various fuels, based on comparisons with threshold soot index and normalized smoke point data. We also introduce a sooting temperature index and a sooting sensitivity index, two quantitative measures to describe sooting propensity and its dependence on strain rate.

## 1. Introduction

Emissions of particulate matter, especially soot, from incomplete combustion have implications for both human health and the environment. Small soot particles can travel deep into the respiratory system when inhaled and subsequently enter the blood circulation system, causing serious health problems [1]. Emissions of soot have also been identified to be important contributors to global warming [2]. Thus, fundamental understanding and control of soot emissions from practical combustion devices are of crucial importance.

Previous studies have shown that soot formation depends on various factors including fuel structure [3,4], flame temperature [5,6], pressure [7,8] and fuel dilution [9,10]. Among these, the effect of fuel structure is of particular interest because of the chemical complexity and compositional variations of practically relevant hydrocarbon fuels [3,11–14]. In particular, in diffusion flames, the order of decreasing sooting tendency follows the order of aromatics, alkynes, alkenes, and alkanes [14].

Quantitative sooting metrics for a wide range of hydrocarbon fuels have been previously proposed. Calcote and Manos [11] introduced the threshold soot index (TSI) with the scale of 0 (ethane) and 100 (naphthalene) where higher values mean higher sooting propensity. This index is the normalization of smoke point data in laminar diffusion flames, which can be obtained, e.g., through the standard test procedure defined in American Society for Testing and Materials (ASTM) D1322. However, there is a certain level of subjectivity (about ±3 mm ambiguity in determining the smoke point height as described in [15]), despite the well-defined test standard. This can be a source of data scattering in different test campaigns.

For gaseous hydrocarbons, TSI values, determined in co-flow experiments [11], are typically in the range from 0 to 3, which is quite small compared to the defined higher scale of 100. Nonetheless, TSI is widely adopted [16–18] due partly to the simplicity in experimental setup. This sooting propensity data of TSI for liquid fuels could be related with engine-out soot in compression ignition engines [19]. Recently, TSI was also used as one of targets in surrogate fuel development to match with real fuel in gas turbine combustion [16,20,21].

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* Corresponding author.
*E-mail address:* sukho.chung@kaust.edu.sa (S.H. Chung).

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Part of the reason why gaseous fuels have very low TSI values is the inclusion of the molecular weight (MW) of the fuel in the definition of TSI, which is low for gaseous fuels. However, some researchers pointed out that this direct inclusion of MW may not be physics motivated [22,23]. Recently, Li and Sunderland [22] revisited smoke point data and proposed a new sooting metric called the normalized smoke point (NSP). Barrientos et al. [23] proposed the oxygen extended sooting index (OESI) for oxygenated fuels to account for the effect of oxygen atoms in the normalization of smoke point data.

Smoke point flames, on which both TSI and NSP are based, are quite short in length for highly sooting fuels and, as such, their flame heights are difficult to measure accurately [24]. This in turn results in appreciable uncertainties in quantitative indices for such fuels, although a recent study proposed an improved methodology to determine TSI based on image processing techniques [15]. Apart from smoke-point-based methods, McEnally and Pfefferle doped target fuels in a methane coflow diffusion flame and measured the peak soot volume fraction, which was then normalized to determine the yield soot index (YSI) [24–26].

The above-mentioned indices are based on either coflow flames or smoke points in which pure fuel and air were frequently used. Recent focus on oxyfuel/oxygen-enriched combustion, low-temperature combustion, mild combustion, and flue gas recirculation [27] requires information on the effect of dilution of fuel and oxidizer streams on sooting behaviors in diffusion flames. Furthermore, soot formation is a rate-limiting process and is therefore sensitive to the characteristic flow time scale [28,29], which can be represented by the strain rate. In coflow diffusion flames, the strain rate is usually very low (long residence time) [30] and difficult to control, especially in TSI measurements. In addition, in coflow/smoke point flames, soot once formed will be transported toward the high-temperature flame zone where soot oxidation takes place, behaving as a soot formation/oxidation (SFO) flame [31]. As such, the smoke point is essentially a critical condition when soot formation and oxidation are balanced. To quantify the sooting tendency for different fuels, it may also be useful to separate soot formation from oxidation.

Counterflow experiments can provide valuable information regarding dilution and strain effects on soot formation. Research has looked at the effect of the strain rate on soot and PAH formation in such a configuration. Wang et al. [32] reported a reduced soot volume fraction with an increasing strain rate. Decroix and Roberts [33] observed a similar trend in addition to the finding that different fuels have different sensitivities to the strain rate. Yamamoto et al. [34] used gas sampling and GC/MS to study acetylene counterflow flames and found that increasing the strain rate could reduce concentrations of polycyclic aromatic hydrocarbons (PAHs), which are the precursors to soot formation. Huijnen et al. [35] conducted numerical simulations to study the behavior of soot and PAH in benzene and ethylene counterflow flames with varying strain rates and showed that the sensitivity of the soot volume fraction to the strain rate was lower in benzene flames than in ethylene flames. Although these studies provided interesting results, systematic data on critical sooting conditions (i.e., sooting limits) considering dilution and strain rate effects in counterflow diffusion flames remain limited.

In a counterflow, the soot zone structure can be quite different depending on the location of the flame relative to the stagnation plane, classified as soot formation (SF) and soot formation oxidation (SFO) flames [31]. In a SF case, the flame is located on the oxidizer side of the stagnation plane such that the soot formed on the fuel side from the flame will be convected away from the flame and subsequently it will leak out along the stagnation plane without being oxidized. In the SFO case, the flame is on the fuel side of the stagnation plane and the soot migrates toward the flame to be oxidized. These two different behaviors suggest that studies of sooting conditions for both SF and SFO flames are necessary.

An important advantage in adopting a counterflow is that the strain rate effect can be systematically investigated. Additionally in soot formation (SF) counterflow flames, the propensity of the fuel to form soot can be studied without much interference from soot oxidation, as opposed to coflow experiments where soot is oxidized when crossing the flame toward the oxidizer side.

This study aims to identify the effects of dilution and the strain rate on the sooting limit of various gaseous hydrocarbon fuels ($CH_4$, $C_2H_4$, $C_2H_6$, $C_3H_6$, $C_3H_8$, n-$C_4H_{10}$ and iso-$C_4H_{10}$) in counterflow diffusion flames, extending our previous work on ethylene/propane mixtures [36]. A systematic comparison among different fuels is presented followed by analysis of the fuel-dependent effects of the strain rate on the sooting limit. The potential use of the limiting flame temperature as a quantitative sooting metric is proposed as a sooting temperature index (STI) together with a sooting sensitivity index (SSI).

## 2. Experiment

The apparatus consisted of a counterflow burner and a laser light scattering system. The burner had two opposing nozzles (10 mm exit diameter) with a separation distance of 8 mm. The nozzle had diverging, settling, and converging sections with an area contraction ratio of 36 to achieve a near uniform flow at the exit. The upper nozzle was cooled with water to prevent heating by buoyancy. A concentric slit with a 2 mm width was used as a nitrogen flow shield to eliminate secondary flames formed by the ambient air. The fuel stream consisting of fuel and nitrogen was supplied through the lower nozzle and an oxidizer consisting of oxygen and nitrogen was supplied through the upper one. The nozzle exit velocities ($V_0$) for the two nozzles were the same. The initial temperatures of the fuel and oxidizer streams were 298 K. The burner setup was mounted on a two-dimensional translational stage with a spatial resolution of 100 μm.

The fuels used were commercially pure grade (>99.5%) normal alkanes of methane, ethane, propane, and n-butane, alkenes of ethylene and propene, and a branched alkane of iso-butane. The flow rates of the fuel and oxidizer were controlled by mass flow controllers (MFCs), calibrated with piston-driven dry calibrators. In all test conditions, the dynamic range of MFCs was maintained below 25% for accuracy.

A schematic of the light scattering and light extinction (LS/LE) setup is shown in Fig. 1. A cw Ar-ion laser was operated at 514.5 nm and 0.5 W as the light source. A mechanical chopper (1310 Hz) and a lock-in amplifier were used to enhance the signal-to-noise ratio. Two pinholes were used to shape the incident laser beam and a half-wave plate (514.5 nm) was used to rotate the laser beam to vertical polarization. A plano-convex lens ($f = 1$ m) focused the beam into the center axis of the flame. A photodiode (Thorlabs, DET100A) mounted at the exit port of an integrating sphere was used to detect the attenuated beam after passing through a flame for monitoring.

The scattered light was detected at 90° to the incident beam path. An iris was used to define the collection solid angle. A bi-convex lens ($f = 12$ cm) imaged the measurement volume into the photo-multiplier tube (PMT; Hamamatsu) with unity magnification. Scattering signal was calibrated by supplying propane and nitrogen at ambient temperature through the burner. The ratio of the scattered intensities from these two gases was measured to be 13.5, which is consistent with the known values for scattering cross-sections of these gases [37]. This ensures that spurious laser light reflected from other surfaces does not influence the scattering