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Smoke points of fuel-fuel and fuel-inert mixtures

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ABSTRACT

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Keywords: Fire emissions Laminar diffusion flame Soot Sooting propensity Laminar smoke points are important fuel properties for understanding soot processes in fires and flames. Several past studies have measured laminar smoke points of fuel-fuel and fuel-inert mixtures, but scaling laws for these smoke points have been lacking and thus are considered here. To supplement published data from 8 past studies, smoke points are measured for ethylene-inert mixtures. The resulting dataset includes 160 smoke points for fuel-fuel mixtures and 78 for fuel-inert mixtures. For fuel-fuel mixtures, the most successful scaling equates mixture smoke point to the reciprocal of the sum of the constituent mass fractions divided by their smoke points. This scaling is also reasonably successful for fuel-inert mixtures (except those involving Ar) when infinite smoke points are assumed for the inerts. The effectiveness ranking of the inerts to increase smoke points is, by volume: $SO_2 > CO_2 > N_2 > Ar > He$, and by mass: $He > CO_2 \approx SO_2 > N_2 > Ar$. Argon's unusual behavior and low effectiveness is attributed to its low heat capacity.

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1. Introduction

An improved understanding of soot processes in fires is crucial to fire research. Rates of emissions of carbon monoxide and other toxins from fires are proportional to those of soot. Soot radiation often dominates fire spread rates and flashover hazards. Smoke deposition is a leading cause of property damage in fires. Unfortunately, accurate predictions of soot formation and emission are not obtained in most numerical simulations of fires.

Laminar smoke points are the prevailing measure of fuel sooting tendency in diffusion flames and in numerical models of fires. A smoke point is the condition of incipient sooting of a laminar diffusion flame. Smoke points have been reported for hundreds of pure fuels (e.g. [1–4]). These are found to vary widely with fuel type, but to be much less affected by changes in burner diameter, burner material, or co-flow velocity [1–4]. Smoke points of fuel–fuel or fuel–inert mixtures have received less experimental or analytical attention, thus motivating the present work. An improved understanding of smoke points of such mixtures is important for understanding soot processes in fires, and in engines with exhaust gas recirculation or surrogate fuels [3,5].

Laminar smoke points are good indicators of the sooting behavior of turbulent diffusion fires and flames [6-13]. Smoke points are widely used in computational soot models, including the two

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leading fire simulation models, Fire Dynamics Simulator (FDS) and FireFOAM [10–12,14,15].

Smoke points are widely used to characterize fuels [16,17]. For example, most aviation fuels are required to have a smoke point of at least 25 mm [18]. Surrogate fuels, which are mixtures of pure compounds that match various properties of more complex fuels, can be expected to mimic sooting propensity when the laminar smoke point is matched [3,17,19].

Previous studies have done little to relate mixture smoke points to the constituent smoke points. The absence of such scaling laws is an impediment to the understanding and application of smoke points. Thus motivated, the objectives of this work are to collect published data on smoke points of fuel–fuel and fuel–inert mixtures, to measure smoke points of ethylene–inert mixtures, and to seek successful scaling laws of these smoke points.

2. Past mixture smoke points

Table 1 summarizes the 8 studies to date that report smoke points of 8 or more fuel–fuel mixtures. Included are the burner or wick diameter, *d*, the number of smoke points, and the constituent fuels, all of which are hydrocarbons. These studies contain 160 smoke point measurements, these being binary, ternary, and quaternary fuel mixtures (i.e., comprising 2, 3, and 4 constituent fuels).

Most of the tests in Table 1 burned liquid fuels supplied with wicks [2,3,20–22], although Van Treuren [23] burned liquids supplied to a conical pool. For Van Treuren [23], it was assumed here that the partial volumes of liquid fuels are conserved upon

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Table 1	
Summary of past measurements of smol	ke points of fuel-fuel mixtures. ^a

Study	d (mm)	Number of smoke points	Constituent fuels
Aksit and Moss [21]	5.5	12 binary	<i>n</i> -C ₉ H ₁₂ , 1,3,5-C ₉ H ₁₂ , <i>n</i> -C ₁₀ H ₂₂
Gill and Olson [2]	5.5	18 binary; 7 ternary	$n-C_8H_{10}$, C_8H_{12} , $i-C_8H_{18}$, $i-C_9H_{12}$, $C_{10}H_{12}$, $C_{10}H_{18}$, $\alpha-C_{11}H_{10}$
Hunt [20]	6.4	8 binary	sec-C ₁₀ H ₁₄ , α-C ₁₁ H ₁₀ , n-C ₁₂ H ₂₆
Markstein [25]	4.1	15 binary	C ₂ H ₄ , C ₃ H ₆ , 1,3-C ₄ H ₆
Mensch et al. [3]	5.5	42 binary; 3 ternary	C ₇ H ₈ , C ₇ H ₁₄ , m-C ₈ H ₁₀ , i-C ₈ H ₁₈ , 1,3,5-C ₉ H ₁₂ ,
			1,2,4-C ₉ H ₁₂ , C ₁₀ H ₁₈ , α -C ₁₁ H ₁₀ , <i>n</i> -C ₁₂ H ₂₆ , <i>i</i> -C ₁₆ H ₃₄
Schug et al. [1]	10	24 binary	C_2H_2, C_2H_4, C_2H_6
Van Trueren [23]	11.3-62.2	19 binary	C_6H_{12} , $n-C_6H_{14}$, C_7H_8 , $n-C_7H_{16}$, $o-C_8H_{10}$, $m-C_8H_{10}$, $p-C_8H_{10}$
Yang et al. [22]	5.5	12 quaternary	$C_{10}H_8$, $n-C_{10}H_{14}$, $C_{10}H_{18}$, $n-C_{12}H_{26}$

^a All tests involved steady axisymmetric flames in air at 1.01 bar.



Fig. 1. Smoke point lengths plotted with respect to smoke point fuel mass flow rates for the hydrocarbon measurements of Olson et al. [28].

mixing. The smoke points of Gill and Olson [2] and Mensch et al. [3] were reported as threshold sooting indices, TSI [24], requiring conversion here to smoke point lengths.

Two studies that measured gaseous fuel-fuel mixtures appear in Table 1: Schug et al. [1] and Markstein [25]. Markstein [25] reported only smoke point fuel flow rates, which were converted here to approximate smoke point lengths using [4]:

$$L_{SP} = c \dot{m}_{SP}, \tag{1}$$

where L_{SP} is smoke point length, *c* is a constant for all hydrocarbons burning in air at 1.01 bar, and \dot{m}_{SP} is smoke point fuel mass flow rate. Eq. (1) is supported by diffusion flame theory and measurements [4,26,27]. As shown in Fig. 1, a value of *c*=24.2 m-s/g was obtained with low scatter for the 42 hydrocarbon smoke point measurements of Olson et al. [28].

Only two past studies have measured the smoke points of fuelinert mixtures. As summarized in Table 2, Schug et al. [1] measured smoke points of 48 ethylene–inert mixtures. Berry Yelverton and Roberts [29] reported smoke points of 23 ethylene–inert mixtures at 1.01 bar, but did not report the inert mole or mass fractions.

3. Experimental methods

Smoke points of ethylene-inert mixtures were measured to supplement and confirm the measurements of Schug et al. [1]. A

Tabl	e 2		
Sum	imai	v of	m

Inert	MW (g/mol)	$c_p^{\mathbf{b}}$	$\overline{c}_p^{\mathbf{b}}$	α^{c}	Number of smoke points	
	(g/IIIOI)	(J/g-K)	·k) (j/moi-k) (cm/s)	(CIII /3)	Present	Schug et al. [1]
Ar	39.95	0.519	20.73	0.335	7	7
CO_2	44.01	0.815	35.86	0.202	8	12
H_2O	18.02	2.102	37.88	0.345	-	8
He	4.00	5.204	20.82	2.75	7	7
N_2	28.01	1.03	28.85	0.367	8	8
SO_2	64.06	0.607	38.88	0.120	-	6

^a MW is molar mass; c_p is specific heat; \overline{c}_p is molar specific heat; and α is thermal diffusivity.

 $^{\rm b}$ Properties at 1.01 bar, and 0 $^\circ C$ (except H2O, at 100 $^\circ C$) from El-Mahallawy and Habik [30].

^c Properties at 1.01 bar, and 400 K from Glassman [31].

coflow burner was used with upward flow of ethylene-inert mixtures from a 14 mm inside diameter brass port with a wall thickness of 1 mm and a blunt tip. No chimney was used. The fuel discharge was fully developed and laminar, with Reynolds numbers of 40–95. This was surrounded by concentric air coflow from a 100 mm inside diameter port. The air passed through glass beads followed by a ceramic honeycomb (1 mm cell size by 10 mm long) to provide uniform flow. The air flow rate, 0.41-2.41 g/s, was maintained slightly above the minimum required to prevent flame flicker, and thus generally increased with fuel flow rate. The air flow rate was found to have a negligible effect on smoke point length [1,32]. Ethylene (99.5% pure), inert, and filtered shop air flow rates were controlled with needle valves and measured with rotameters, which were calibrated with soap bubble meters. The inert gases, all with purities of 99.998%, were Ar, CO₂, He, and N₂. Ambient conditions were 1.01 bar and 21 °C.

Smoke points in air at atmospheric pressure and temperature depend weakly on the method of soot emission detection. Smoke points were identified here visually using the *equal wings* (i.e., equal ears) criterion of Schug et al. [1]. This is the condition for which the visible boundaries of the soot annulus and the centerline are equally high. This method was found to yield smoke points in agreement with, but with higher precision than, those identified from visible smoke emissions in front of either light or dark backgrounds. For the longest smoke points, asymmetries required smoke points to be identified from conditions where a line between the left and right wings passed through the limit of the centerline intensity.

After each smoke point was established visually, a color image was recorded using a digital color camera operating at f/4 and 67 ms. These images were used to measure smoke point lengths with respect to the top of the fuel port. The luminous flame tips

generally transitioned from bright yellow to dark over an axial distance of 2–5 mm. Luminous lengths were measured to the point where the centerline grayscale intensity changed most rapidly between these yellow and black regions, which minimized the sensitivity of the measured lengths to camera exposures. Uncertainties in the measurements are estimated at \pm 5% for the flow rates of ethylene, inert, and air, and \pm 10% for the smoke point lengths.

4. Scaling laws

Many properties of gas mixtures, ϕ_{mix} , are defined or can be estimated from the properties of the constituent gases by either of two scaling laws:

$$\phi_{\rm mix} = \sum Y_i \phi_i, \text{ or }$$
(2)

$$\phi_{\rm mix} = \sum X_i \phi_i,\tag{3}$$

where X_i , Y_i , and ϕ_i are the constituent mole fraction, mass fraction and property, and the summation is over all species present. For example, Eq. (2) applies when ϕ is specific enthalpy, specific heat capacity, or speed of sound squared, while Eq. (3) applies when ϕ is compressibility factor, density, lean or rich inverse flammability limit (expressed as fuel mole fraction), molar enthalpy, molar mass, or pseudocritical temperature. When ϕ is defined as the reciprocal of smoke point length, Eqs. (2) and (3) become

$$L_{SP, mix} = (\sum Y_i / L_{SP, i})^{-1},$$
(4)

and

$$L_{SP, mix} = (\sum X_i / L_{SP, i})^{-1}$$
(5)

Note that the correlations using $\phi = L_{SP}$ have been found, here and previously, to yield much poorer empirical fits than Eqs. (4) and (5).

Several past studies have correlated laminar smoke point properties of fuel-fuel mixtures according to scaling laws similar or identical to Eqs. (4) and (5). For smoke points of mixtures of liquid fuels, Van Treuren [23] proposed

$$L_{\text{SP. mix}} = (\sum v_i / L_{\text{SP. i}})^{-1}, \tag{6}$$

where v is liquid volume fraction. Eq. (6) is equivalent to Eq. (4) when all the liquid densities are the same, which is a reasonable approximation for the liquid hydrocarbons of Table 1. For smoke points of mixtures of either liquid or gaseous fuels, Gill and Olson [2] proposed

$$TSI_{mix} = \sum X_i TSI_i \tag{7}$$

Note that Eq. (7), combined with the definition of TSI, yields the more simple and direct Eq. (4), and that a recent paper from our laboratory outlines several shortcomings of the TSI approach [4]. Eq. (7) has been invoked by several studies [3,5,19,22,33,34]. Yang et al. [22] replaced X_i in Eq. (7) with Y_i , thus yielding an expression that is equivalent to neither Eqs. (4) nor (5). For smoke points of mixtures of gaseous fuels, Markstein [25] proposed

$$Q_{SP, mix} = (\sum X_i / Q_{SP, i})^{-1},$$
(8)

where Q_{SP} is the smoke point volumetric flow rate. Eq. (8) is equivalent to Eq. (4) when, as expected, Eq. (1) holds.

To summarize, smoke points of fuel-fuel mixtures have been correlated with the constituent fuel smoke points using at least three different scaling laws, but the results have not been compared or tested with a large number of measurements. Smoke points of fuel-inert mixtures have not previously been correlated using any scaling law.

5. Results

5.1. Fuel-fuel mixtures

For binary fuel-fuel mixtures, smoke point lengths vary monotonically as the composition changes from one fuel to the other. This is illustrated in Fig. 2, using axes that have appeared previously [20,23].The curves shown are the mass fraction scaling of Eq. (4), although the mole fraction scaling of Eq. (5) fits the data nearly as well.

The utility of plots like Fig. 2 is limited because they do not admit ternary or quaternary mixtures and each fuel pairing yields a separate curve. These limitations are addressed by using different axes, as shown in Figs. 3a and b for the mass and mole fraction scaling of Eqs. (4) and (5), respectively. This is the first time such plots have been presented. Shown here are the 138 binary fuelfuel mixtures of Table 1 and the linear predictions of Eqs. (4) and (5). The success of the fits indicate that both Eqs. (4) and (5) capture the smoke point behavior of these diverse binary mixtures. While the difference in R^2 between Figs. 3a and b is insignificant, the Eq. (4) prediction becomes superior upon removing mixtures whose constituent fuel molar masses are very close. Scatter in the data generally increases with increasing smoke point length, and is attributed to increased unsteadiness. Motivated by Eqs. (2)–(5), many other correlations were attempted, but were found to be statistically inferior to those of Fig. 3.

The ternary and quaternary smoke points of Table 1 are not shown in Fig. 3 for clarity. Instead, these are shown in Fig. 4 using the mass fraction scaling of Eq. (4). The low scatter in Fig. 4 indicates the success of Eq. (4) for these data (although Eq. (5) yields a slightly higher R^2 of 0.995). Figs. 3 and 4 reveal the success of scaling laws that predict smoke points of fuel-fuel mixtures from the constituent smoke points.

5.2. Fuel-inert mixtures

Representative color images of the smoke point flames observed in this work are shown in Fig. 5, where the inert is nitrogen and its mass fraction increases from left to right. These luminous flame lengths are roughly proportional to fuel flow rate



Fig. 2. Smoke points for selected binary fuel-fuel mixtures plotted with respect to isooctane mass fraction. Measurements are from Gill and Olson [2] and the curves are from Eq. (4).

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Fig. 3. Measured smoke points for the 138 binary fuel-fuel mixtures of Table 1 plotted with respect to the predictions of (a) Eq. (4), and (b) Eq. (5).



Fig. 4. Measured smoke points for the 22 ternary and quaternary fuel-fuel mixtures of Table 1 plotted with respect to the prediction of Eq. (4).

because the addition of inerts at fixed fuel flow rate does not have a significant effect on the stoichiometric [26] or luminous [1] flame length for gas jet diffusion flames. The soot wings are clearly visible, but any soot emissions are minimal and not visible here. The wings become less symmetric for longer flames, which is attributed to burner asymmetries.

Fig. 5 shows that smoke point length increases with nitrogen addition, which is confirmed below for the other inerts. This is because inert addition reduces hydrocarbon concentrations, temperature, and residence time in the soot formation region. In contrast, it does not significantly reduce oxidizer concentrations, temperature, or residence time in the soot oxidation region higher in the flame, where much of the inert has diffused outward and velocities are dominated by buoyancy.

Fig. 6 quantifies the effect of inert addition on ethylene smoke points. This choice of axes follows Schug et al. [1] and Glassman [31]. Within experimental uncertainties, the present measurements confirm those of Schug et al. [1] for the inerts observed here. Schug et al. [1] H_2O data are not shown because these



Fig. 5. Color images of ethylene-nitrogen flames at their smoke points.



Fig. 6. Ethylene volume flow rate plotted with respect to inert flow rate for the 70 ethylene–inert smoke points of Table 2 (excluding H₂O). The solid lines, whose slopes and R^2 are shown in the legend, are the best linear fits for each inert with an intercept of 4.01 cm³/s, corresponding to the present undiluted ethylene smoke points. The dashed lines are for selected values of X_{inert} .

involved heated burner gases and thus a shorter ethylene smoke point. For all inerts, smoke points increase with increased inert addition. If the only effect of inert addition were to dilute the reactants and increase velocities, all the inerts would collapse onto a single line in Fig. 6. Instead, inerts also increase smoke points by acting as heat sinks [1] and by changing the thermal and mass diffusivities [31]. The effectiveness on a volumetric basis to increase smoke points is seen from the fit slopes in Fig. 6 to rank as: $SO_2 > CO_2 > N_2 > Ar > He$. These slopes correlate approximately with inert \overline{c}_p and $1/\alpha$, but not with any other Table 2 property.

Fig. 7 shows the same data as Fig. 6, where volumetric flow rates are replaced with mass flow rates. The effectiveness on a mass basis to increase smoke points is seen from the line slopes in Fig. 7 to rank as: $He > CO_2 \approx SO_2 > N_2 > Ar$. These slopes correlate approximately with inert c_p , but not with any other Table 2 property.

Schug et al. [1] presented a plot that collapsed their Fig. 5 measurements onto a single line. This was not predictive, as the smoke point fuel flow rate appeared in both axes and the abscissa required measured peak temperatures. An alternate approach is considered here. As Figs. 4–6 show, smoke points become infinite as inert mole or mass fraction approaches unity. When L_{SP, inert} is assumed to be infinite, the summation in Eq. (4) reduces to a summation over the fuel(s). This yields Fig. 8 for the fuel-inert smoke points of Table 2. Eq. (5) was similarly tested, but was slightly less successful. Although the abscissa label has been simplified, the axes of Fig. 8 are identical to those of Fig. 3a. The fuel-inert smoke points are reasonably well correlated when Ar mixtures are excluded. Smoke points with Ar dilution fall below the Eq. (4) prediction because Ar has the lowest c_p and \overline{c}_p of the inerts considered. Such low heat capacities limit argon's ability to cool the soot formation region, as is also evident in Figs. 6 and 7. Motivated by Eqs. (2)-(5), many other correlations were attempted, but were found to be statistically inferior to those of Fig. 8.

Although the correlations are not perfect, it is significant that Eq. (4) yields reasonable correlations of binary, ternary and quaternary fuel–fuel mixtures, and of fuel–inert mixtures not involving argon.



Fig. 7. Ethylene mass flow rate plotted with respect to inert flow rate for the 70 ethylene–inert smoke points of Table 2 (excluding H_2O). The solid lines, whose slopes and R^2 are shown in the legend, are the best linear fits for each inert with an intercept of 4.70 mg/s, corresponding to the present undiluted ethylene smoke points. The SO₂ fit is omitted for clarity. The dashed lines are for selected values of Y_{inert} .



Fig. 8. Measured smoke points for the 78 ethylene–inert mixtures of Table 2 (including H₂O) plotted with respect to the Eq. (4) prediction (solid line). The dashed lines are at \pm 15%. The present L_{SP, C_2H_4} is 92 mm. For Schug et al. [1] this is 86 mm for H₂O and 106 mm for the other inerts. The fit has a correlation coefficient of R^2 =0.87 excluding the Ar data or R^2 =0.58 otherwise.

This should yield insight into the sooting behavior of fires, surrogate fuels, fuel–gas recirculation, and other soot phenomena.

6. Conclusions

Smoke points of fuel–fuel and fuel–inert mixtures were examined empirically and experimentally. Scaling laws relating these smoke points to those of the constituent fuels were considered. The reciprocal of the sum of the constituent mass fractions divided by their smoke point lengths correlates the smoke points of diverse mixtures of 2–4 fuels. This is also reasonably successful for fuel–inert smoke points (except those involving Ar) when inert smoke points are taken as infinite. The effectiveness ranking of the inerts to increase smoke points is, by volume: $SO_2 > CO_2 > N_2 > Ar > He$, and by mass: $He > CO_2 \approx SO_2 > N_2 > Ar$. Argon is particularly ineffective at increasing smoke points owing to its low specific and molar heat capacities. These are the most extensive tests to date of the scaling laws of fuel–fuel mixtures and are the first demonstrated scaling laws for fuel–inert smoke points.

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