THE EFFECT OF STRAIN RATE ON NANOPARTICLES AND SOOT IN ETHYLENE COUNTERFLOW FLAMES BLENDED WITH ETHANOL AND OME_3

V. Esposito*, M. Sirignano*
vincenzo.esposito9@unina.it
*Dipartimento di Ingegneria Chimica, dei Materiali e della Produzione Industriale,
Università degli Studi di Napoli Federico II, Napoli, 80125, Italy

Abstract
Nowadays, the levels of carbon dioxide in the atmosphere have reached alarming values. In order to decrease or halt global warming, and thus mitigate climate change, the ongoing challenge is to reduce dependence on fossil fuels. Biofuels, especially ethanol, emerge as valuable substitutes for traditional fossil fuels due to their lower carbon emissions. Additionally, among e-fuels, the oligomers of dimethyl ether (DME) – oxymethylene ethers (OMEs) – stand out as potential alternatives for achieving carbon-neutral combustion in diesel engines. Therefore, this work delves into investigating the impact of strain rate on nanoparticles and soot in counterflow diffusion flames (CDFs) of ethylene, ethylene/ethanol, and ethylene/OME_3 using in-situ spectroscopic diagnostics to advance cleaner combustion technologies.

Introduction
In the 20th century, the urgency of transitioning from fossil fuels to sustainable renewable energy is crucial. Among several eco-friendly alternatives, biofuels and e-fuels are being widely explored. Biofuels, especially ethanol - the most frequently produced - can emit less carbon than traditional fossil fuels when blended with conventional gasoline or diesel fuel in existing engines [1]. In the Power-to-Fuel (PtF) technology, oxymethylene ethers (OMEs) as e-fuels, and particularly OME_3, are promising candidates for achieving carbon-neutral combustion, as additives or substitutes in diesel engines [2,3]. Understanding soot formation in turbulent diffusion flames is critical because of its propensity to enhance combustion efficiency. In the context of turbulent diffusion flames, the CDFs delve into the dependence on the characteristic flow time scale of soot formation. In CDFs, the flow time is represented by the strain rate [4]. While strain rate is generally known to reduce soot, understanding of nanoparticle formation remains limited. The strain rate (up to 206.92 s^{-1}) effect for ethylene is here investigated along with ethanol and for the first time OME_3. This study aims to elucidate the intricate relationship between strain rate, oxygenated fuel, and soot and nanoparticle formation in soot forming (SF) flame configuration, characterized by two distinct zones: a pyrolytic and an oxidative zone. In order to provide the impact of strain rate on particle

formation, a pulsed Nd:YAG laser with an excitation source at 266 nm is used. The resulting insights contribute to developing cleaner combustion technologies, especially in blending oxygenated fuels with traditional ones. Further research is recommended for broader applicability, considering high strain rates and varied dilution conditions.

Materials and Methods

The counter-flow burner system is the same as in previous works [5-7]. It consists of two vertically positioned jet nozzles (I.D. 2.54 cm), separated by a constant distance of 1.5 cm. The oxidizer stream was introduced from the upper nozzle, while the fuel stream was introduced by the lower nozzle. Nozzle velocities were varied from 10 cm/s to 80 cm/s (at STP), corresponding to a global strain rate (K), calculated according to [8], which ranged from 25.86 s^{-1} to 206.92 s^{-1}. Ethanol and OME_3 concentrations (20% of total carbon fed) were introduced into the fuel stream, where they are electrically pre-heated at 150 °C to ensure their complete evaporation. The total carbon flow rate is kept constant through the different fuel blends, providing a direct comparison of the results. In the investigated flames (Table 1), the flame front is always on the oxidizer side of the stagnation plane, indicating the soot forming (SF) counterflow diffusion flames configuration. It is worth to underline that as in the fuel stream as in the oxidizer stream, the fuels and oxidizer molar fractions were balanced with nitrogen (75% and 79%, respectively).

Table 1. Strain rate and stream compositions of the investigated flames.

	Fuel stream					**Oxidizer stream**
	Pure C_2H_4	C_2H_4/EtOH		C_2H_4/OME_3		
K	$y_{C_2H_4}$	$y_{C_2H_4}$	y_{EtOH}	$y_{C_2H_4}$	y_{OME_3}	y_{O_2}
[s^{-1}]	[-]	[-]	[-]	[-]	[-]	[-]
25.86	**0.25**	**0.20**	**0.05**	**0.20**	**0.020**	**0.21**
38.80	**0.25**	**0.20**	**0.05**	**0.20**	**0.020**	**0.21**
51.73	**0.25**	**0.20**	**0.05**	**0.20**	**0.020**	**0.21**
103.46	**0.25**	**0.20**	**0.05**	**0.20**	**0.020**	**0.21**
206.92	**0.25**	**0.20**	**0.05**	**0.20**	**0.020**	**0.21**

The experimental diagnostic is similar to that used in previous works [5-7]. Spatially and spectrally resolved Laser-Induced Emission (LIE) measurements were performed using the fourth harmonic (266 nm) of a pulsed Nd:YAG laser as the excitation source. Laser-Induced Fluorescence (LIF), attributed to nanoparticles, Laser-Induced Incandescence, attributed to soot, and scattering, coupled with the capability of the entire burner assembly to move up or down with respect to the sampling point (spatial resolution of 0-1-0.2 mm), have allowed to well characterize the flames at different locations between the two opposed jet nozzles.

Results and discussions

In Figure 2a it is possible to observe the effect of strain rate in the pure ethylene counterflow flames, plotting scattering, LIF@350 nm, and LII@500nm profiles. As expected, due to the reduced residence time available for nucleation and growth, a general intense decrease is observed in the signals as the strain rate increases. The stagnation plane was hardly individuated to be located between 6.0 and 6.2 mm, because of the difficulty of detecting it above the gas phase for high strain rates.

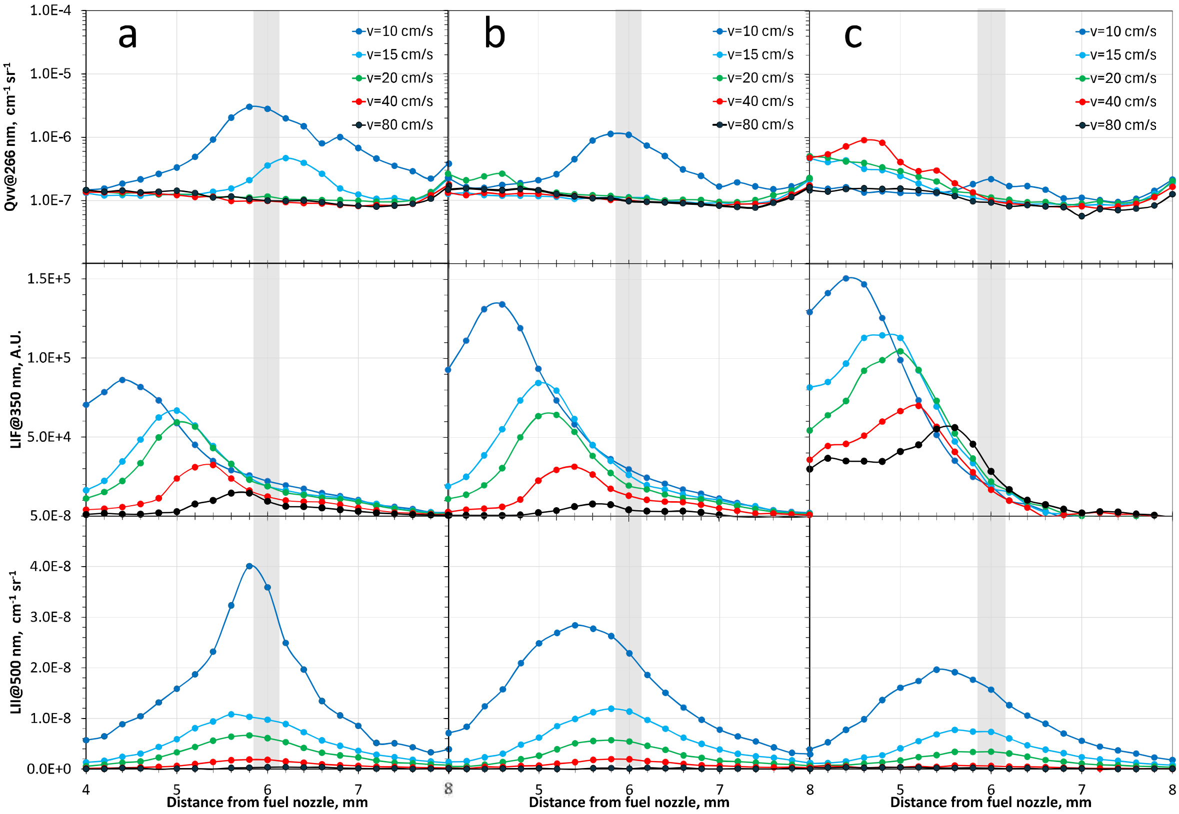

Figure 2 LIE signals measured at different wavelengths in the pure ethylene (a), ethylene/ethanol (b), and ethylene/OME_3 (c) CDFs at different strain rates.

The same trends were observed in the counterflow diffusion flames of ethylene/ethanol and ethylene/OME_3 at different strain rates (Figure 2b-2c).

A direct comparison of scattering, LIF@350nm, and LII@500nm for the three fuel blends with a strain rate of 38.8 s^{-1}, is presented in Figure 3. It is possible to observe the increasing LIF signal in the pyrolytic zone for the two oxygenated fuel blends, with OME_3 showing a more significant reduction in large soot particles detected by LII. On the right side of Figure 4 (left side), LII@500nm and LIF@350nm normalized signals against the strain rate are reported. Full dots and empty squares indicate the normalization of the signals on the maximum in the fuel zone and oxidant zone, respectively. The pure ethylene flame presents different trends in the pyrolytic and oxidant zones, with LII decreasing from K=25.86 s-1 to K=38.8 s-1. OME_3 has a trend similar to pure ethylene, indicating less sensitivity to strain rate but generally results in decreased particle formation due to the absence of C-C bonds.

Moreover, the ethanol blend exhibits a more effective reduction in particle nucleation and growth in the pyrolytic zone.
Finally, soot volume fraction profiles for the three fuel blends at different strain rates have been calculated. Their maximum values were plotted against strain rate such as their normalization in Figure 4 (right side), and once again it is demonstrated that when the strain rate rises, the former consequence is a reduction in soot formation.

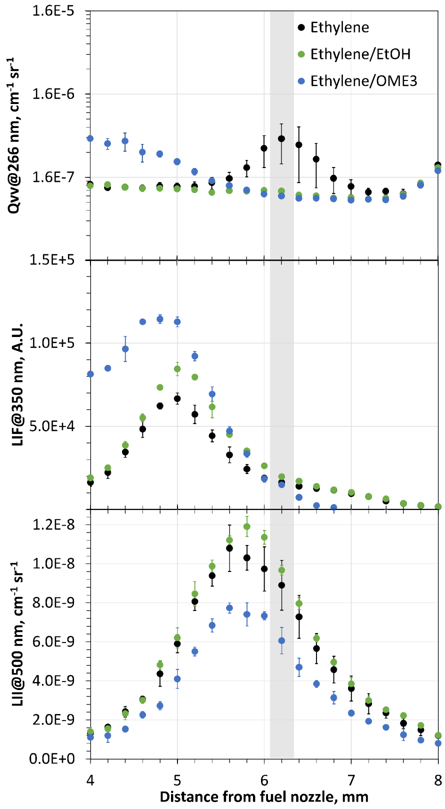

Figure 3 LIE signals measured at different wavelengths in CDFs of pure ethylene, ethylene/ethanol, and ethylene/OME_3 at a strain rate of $K=38.8\ s^{-1}$ (left side).

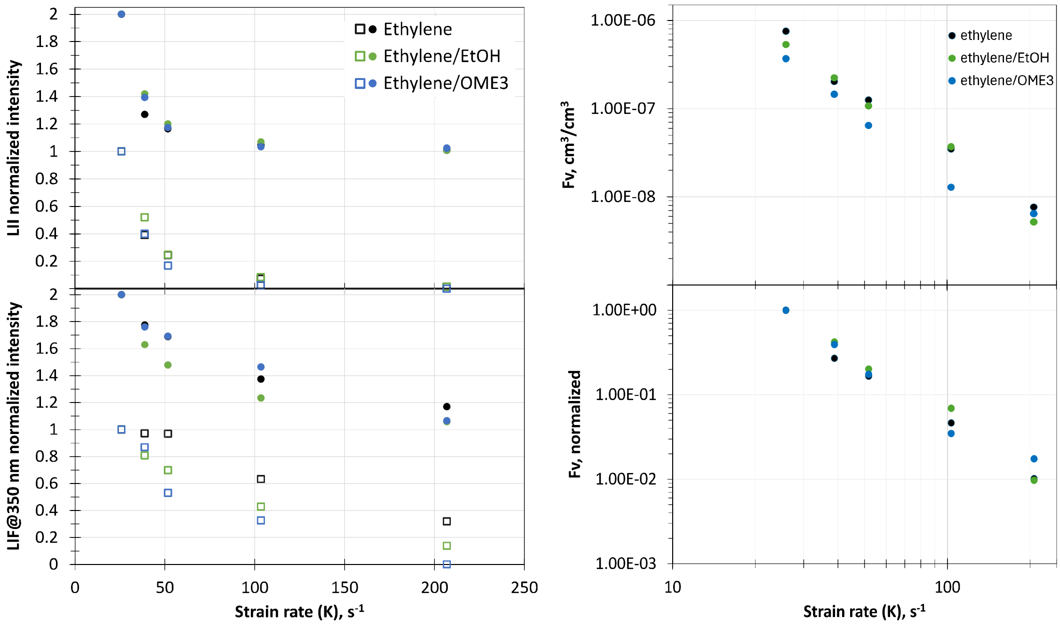

Figure 4 LII@500nm (top) and LIF@350nm (bottom) normalized intensity vs strain rate (left side). Soot volume fraction (top) and normalized soot volume fraction (bottom) vs strain rate for the three fuel blends (right side).

Conclusions

This study has examined the strain rate effect on nanoparticles and soot in counterflow diffusion flames (CDFs) of pure ethylene, ethylene/ethanol, and ethylene/OME_3. Using the fourth harmonic (266nm) of a pulsed Nd:YAG laser it was possible to analyse flame dynamics:

- Particle nucleation and growth are inhibited by decreasing strain rate.
- There is a non-linear strain rate effect on particle formation, and this latter sees the contributions of both pyrolytic (smaller particles due to dominant nucleation and coagulation) and oxidant zones.
- In the pyrolytic zone ethanol and OME3 increase particle formation, effect that goes to diminishes at higher strain rates.
- OME_3 presents a lower sensitivity to strain rate because of its higher reactivity and lack of C-C bonds.

References

[1] Cherwoo, L., Gupta, I., Flora, G., Verma, R., Kapil, M., Arya, S.K., et al. "Biofuels an alternative to traditional fossil fuels: A comprehensive review", *Sustain. Energy Technol. Assessments.* 60: 103503 (2023).

[2] Nemmour, A., Inayat, A., Janajreh, I., Ghenai, C., "Green hydrogen-based E-fuels (E-methane, E-methanol, E-ammonia) to support clean energy transition: A literature review". *Int. J. Hydrogen Energy*. 48:29011-33 (2023).

[3] Lumpp, B., Rothe, D., Pastötter, C., Lämmermann, R., Jacob, E., "OXYMETHYLENE ETHERS AS DIESEL FUEL ADDITIVES OF THE FUTURE", *MTZ world.w..* 72:34-8 (2011).

[4] Chung, S.H., Law, C.K., "An invariant derivation of flame stretch", *Combust Flame.* 55: 123-5 (1984).

[5] Salamanca, M., Sirignano, M., D'Anna, A., "Particulate formation in premixed and counter-flow diffusion ethylene/ethanol flames". *Energy Fuels.* 26(10), 6144-6152 (2012).

[6] Sirignano, M., Collina, A., Commodo, M., Minutolo, P., D'Anna, A., "Detection of aromatic hydrocarbons and incipient particles in an opposed-flow flame of ethylene by spectral and time-resolved laser induced emission spectroscopy". *Combust. Flame.* 159(4), 1663-1669 (2012).

[7] Sirignano, M., Bartos, D., Conturso, M., D'Anna, A., Masri, A.R., "Detection of nanostructures and soot in laminar premixed flames", *Combust. Flame.* 176:299-308 (2017).

[8] Seshadri, K., Williams, F.A., "Laminar flow between parallel plates with injection of a reactant at high Reynolds number.", *Int. J. Heat Mass Transf.* 21:251-3 (1978).