Fuel 186 (2016) 422–429

Contents lists available at ScienceDirect

Fuel

journal homepage: www.elsevier.com/locate/fuel

Full Length Article

Effect of methane doping on nascent soot formation in ethylene-based laminar premixed flames

Can Shao, Bin Guan, Baiyang Lin, Hao Gu, Chen Gu, Zhongzhao Li, He Lin *, Zhen Huang

Key Lab. for Power Machinery and Engineering of M.O.E., Shanghai Jiao Tong University, 200240 Shanghai, PR China

HIGHLIGHTS

- The particle size distribution functions of soot particles in methane-doped ethylene flames behave bimodality.
- The number densities in downstream zone of the methane-doped flames are much higher than pure flame.
- Small ratio of methane doping can promote the volume fractions, while in large ratio cannot.

ARTICLE INFO

Article history:
Received 13 May 2016
Received in revised form 20 July 2016
Accepted 19 August 2016
Available online 1 September 2016

Keywords:
Soot
Particle size distribution function
Premixed flame
Methane-doping

ABSTRACT

The impact of methane doping on sooting behavior of ethylene-base burner stabilized stagnation (BSS) flames was investigated by following the evolution of particle size distribution functions (PSDFs) of nascent soot. The M-series of flames with the equivalence ratio of 2.07 was doped by methane with the mixture ratio of 5%, 10% and 40%. All the methane-doping flames showed obvious bimodality in the PSDFs, and more particles in nucleation stage than the C3 reference flame. The synergistic effect appeared in small ratio (5% and 10%) methane doping cases, leading to the increase of the soot number density and soot mobility volume fractions. While the synergistic effect was not found under large methane doping condition (40%), in which the mobility volume fraction was less than that in C3 flame. By calculating the key species and analyzing the reaction pathways of pyrene formation with KM2-Mech, we found that small ratio of methane doping benefits C3H3 and pyrene (A4) formation, which strengthened the sooting tendency. However too much methane doping reduced the yield of C2H2 that is a critical intermediate of PAHs formation, leading to a lower sooting tendency compared with the pure ethylene flame.

1. Introduction

Soot particle emission from engine combustion contributes significantly to anthropogenic aerosol that does great harm to environment and human health. In the past two decades, a number of studies have been demonstrated to understand the knowledge of fundamental sooting processes in combustion, and significant progress has been achieved in developing the soot models, such as the initial WF-Mech, the much celebrated ABF-Mech and the USC-Mech II [1–12]. Despite the significant progress in soot models, critical gaps remain in many areas of our knowledge [12]. The soot models are still far from being complete without comprehensive understanding of soot nucleation and growth in flame [11], causing remarkable discrepancy between the modeling outcomes and the experimental results. Therefore the mechanism of soot nucleation and soot growth, that reflects the coagulation of the soot particles as well as the particle surface growth, has to be studied thoroughly with both theoretical and experimental methodologies. Over last ten years, probe sampling technique [10,12–29] coupled with mobility sizing was employed to investigate the evolution of particle size distribution function (PSDFs) of nascent soot from particle nucleation to mass growth. The PSDFs of nascent soot combined with soot model prediction enable us to improve the quantitative understanding of soot formation processes [10,12–26]. However, present works mostly focused on the effect of equivalence ratio and temperature on soot particle evolution, little involved in the factor of fuel blending. The impact of fuel blending on soot formation is certainly worthy of attention, due to the fact that most hydrocarbon fuels are mixture containing many kinds of hydrocarbon molecule.

In 2005, Chung et al. [30] measured the soot volume fraction and the number density of ethylene-based opposed flow flame with LII (laser induced incandescence) method, and found that the flame of ethylene doped with methane produced more soot than the pure ethylene flame. They regarded this phenomenon as

---

* Corresponding author.
E-mail address: linhe@sjtu.edu.cn (H. Lin).

http://dx.doi.org/10.1016/j.fuel.2016.08.081
0016-2361/

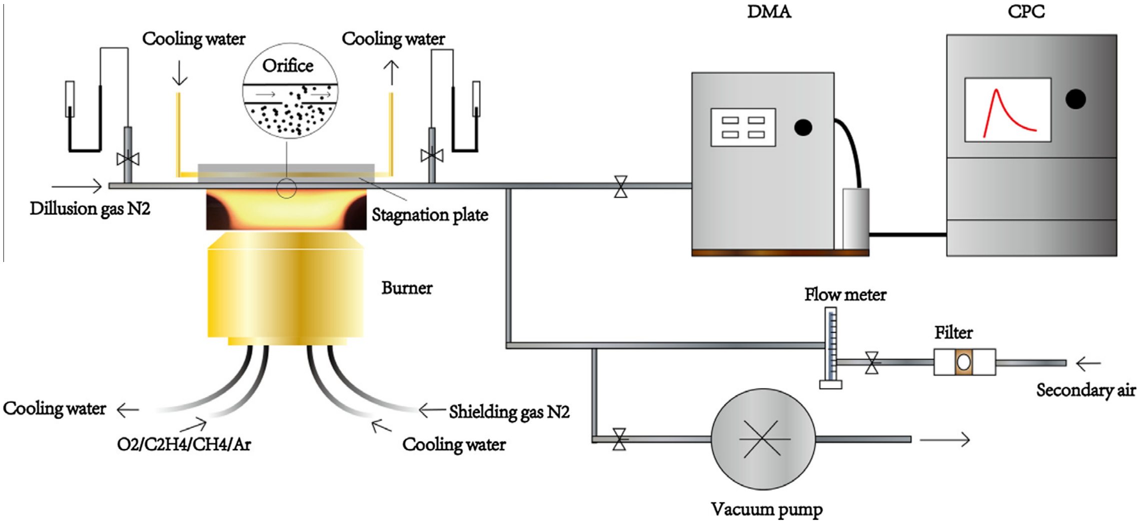

Fig. 1. Schematic of experimental setup.

Table 1
Summary of the C3 and M-series flames studied.

Flame	Mixture ratio	Mole fraction				Φ	Cold gas velocity (cm/s)	$T_{f,max}$ (K)
		CH_4	C_2H_4	O_2	Ar			
C3	0	0	0.1630	0.2370	0.6	2.07	8	1856 ± 79
M1	5%	0.0082	0.1564	0.2354	0.6	2.07	8	1849 ± 82
M2	10%	0.0166	0.1497	0.2337	0.6	2.07	8	1852.5 ± 82.5
M3	40%	0.0708	0.1062	0.2230	0.6	2.07	8	1858 ± 81.5

the synergistic effect, which means that fuel doping may strengthen the sooting tendency, though the doped fuel has lower sooting tendency. This kind of synergistic effect also has been confirmed in DME-based opposed flow flames doping with methane, ethane, propane and ethylene [31]. Afterwards, a renewed soot model, KM2-Mech was developed with taking the effects of fuel blending into account [32].

Through LII method, one can learn about the information of soot volume fraction and number density, but is blind to the evolution of soot particle. In this study, the evolution of soot particle in ethylene-methane premixed BSS (Burner Stabilized Stagnation) flames was studied using probe sampling technique coupled with mobility sizing. Cross comparisons with respect to the PSDFs of soot particle were made between the methane-doped ethylene flames and the pure ethylene flame. The purpose of this study is to give an insight into the impact of methane doping on nucleation and growth of soot particle. To date, the model for precise prediction of soot is unavailable. But existing models are rather sound on predicting the formation of polycyclic aromatic hydrocarbons (PAHs), which are precursors of soot in flame. To better understand the evolution of the PSDFs of methane-doped ethylene flames, the mole fractions of some PAHs were calculated with KM2-Mech in this study. Additionally, analysis of the reaction pathways toward pyrene (A2) formation were made by inspecting the formation of some critical radicals, as such an analysis provides insight into the mechanism of synergistic effect of fuel blending. Furthermore, brute force sensitivity analysis [33] were also performed to highlight the key reactions, and to give a glance to the synergistic effect from the other side.

2. Experimental and computation method

The experimental system using in this study, as shown in Fig. 1, has been described in previous papers [21,22]. In brief, the measurement of PSDF of nascent soot particles in ethylene-methane flames were done using the BSS flames configuration, in which a sampling tube embedding in the stagnation plate was used to take soot samples in the BSS flame. Nascent soot particles were inhaled into the sampling orifice with 130 μm in diameter and immediately diluted by cold nitrogen (30 L/min, STP) to minimize particle coagulation losses in the sampling channels.

The dilution ratio will be optimized by adjusting the second-air flow rate. A small fraction of the diluted sample (1.5 L/min, STP) was analyzed by TSI SMPS system consisting of a 3085 nano-Differential Mobility Analyzer (DMA), a Kr-85 Neutralizer and a 3776 ultrafine Condensation Particle Counter (CPC). The mobility diameter of soot particle was corrected based on the nano-particle transport theory for the reason that particle size smaller than 10 nm was overestimated by Cunningham slip correction [34–36].

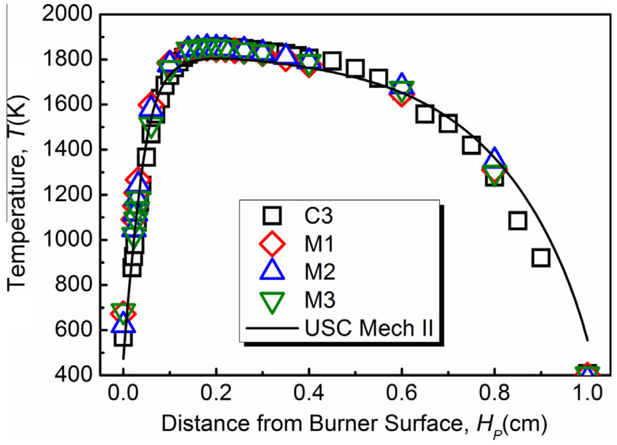

Fig. 2. Comparison of measured temperature of C3 and M-series flame (symbols) and computed temperature profile (line).