Combustion and Flame 191 (2018) 394–407

Contents lists available at ScienceDirect

Combustion and Flame

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

Experimental and numerical investigation on soot volume fractions and number densities in non-smoking laminar n-heptane/n-butanol coflow flames

Liang Qiu, Xiaobei Cheng*, Zhongqiu Li, Hui Wu

State Key Laboratory of Coal Combustion, School of Energy and Power Engineering, Huazhong University of Science and Technology, Wuhan, Hubei 430074, PR China

ARTICLE INFO

Article history:
Received 23 April 2017
Revised 20 July 2017
Accepted 16 January 2018

Keywords:
n-Heptane
n-Butanol
Laminar coflow flame
Soot volume fraction
Soot number density

ABSTRACT

In this work, the soot volume fractions (SVF) and number densities in laminar coflow flames of n-heptane/n-butanol blends were experimentally and numerically investigated. Five blends, B00, B25, B50, B75, and B100, based on the volume fraction of n-butanol in the liquid fuel, were employed to explore the soot behavior when n-butanol is added to the fuel stream. Mass flow rates of the blends were adjusted to keep the mole flow of carbon constant. The fuel stream was heavily diluted with N_2 to carry the fuel vapor. Measured visible heights are 5.7 cm, 5.6 cm, 5.3 cm, 5.1 cm and 4.8 cm for the five target flames, respectively. A thermophoretic probe was used to sample particles at different height of the flames. SVF, soot number density, average primary diameter, and number of primary particles per aggregate were measured and calculated through transmission electron microscopy (TEM) images. CoFlame code was used to calculate the soot formation in the coflow flames with a newly developed n-heptane/n-butanol/polycyclic aromatic hydrocarbon (PAH) skeletal model and a fixed sectional soot method. The qualitative characteristics of SVF and number densities obtained from TEM images were well captured by the calculation, although the visible heights were underpredicted. Calculated peaks of soot mass fraction (SMF) and number densities show a decrease as more n-butanol is added, and all the peaks are reached around the height of 3.2 cm. Inception by the dimerization of pyrene (A4) plays the most important role in the evolution of soot number densities, while hydrogen-abstraction-carbon-addition (HACA) process dominates the soot mass addition. Lower mole fractions of A4 and C_2H_2 are responsible for the suppression of SMF and number densities with the addition of n-butanol. Condensation of A4 and fragmentation play small roles in the increase of SMF and number densities, respectively. Finally, all the particles are completely oxidized, first mainly by OH, and then by O_2.

1. Introduction

Medical researches show that ultrafine particles with size less than 100 nm are seriously harmful for human health. In addition, recent studies have found that particles in the atmosphere are likely the second most important contributor to global warming after CO_2 [1]. Among them, the particles exhausted from engines are the main sources. Therefore, more and more stringent restrictions on engine particle emissions were specified by the emission regulations over the past two decades [2]. Particle mass (PM) emission of heavy-duty diesel engine is limited to 0.1 g/kWh by Euro VI starting from 2013, besides, particle number (PN) is first limited by the regulation to 8.0×10^{11}/kWh at the same time [2]. In general, particles from engines are a complex mixture consisting of volatile (organic, sulfate, nitrate fraction) and non-volatile (soot, ash) materials, and the vast majority of them are soot particles [3]. Combustion-generated soot particles bear the fractal-like aggregate structure, and each soot aggregate is comprised of a certain number of nearly spherical primary soot particles of almost equal size [4,5]. The impact of soot particles on health and climate depends not only on the total mass, but also on their number densities.

The earlier emission targets have been achieved for the most part by in-cylinder strategies, such as higher fuel injection pressure, higher intake boost, lower intake temperature, adjustments to fuel injection timing, low to moderate levels of exhaust-gas recirculation (EGR) [6], and premixed low-temperature combustion [7]. The later targets, however, have been achieved or are expected to be achieved through a combination of both in-cylinder

---

* Corresponding author.
E-mail address: xbcheng@hust.edu.cn (X. Cheng).

https://doi.org/10.1016/j.combustflame.2018.01.024

strategies and aftertreatment devices. After years of research and development, viable aftertreatment technologies are now available, but their widespread adoption is impeded by higher economic costs, durability issues, fuel economy penalties, and greater space requirements compared to engines without aftertreatment systems [8]. Apart from environmental degradation, the world is also suffering from the crises of fossil fuel depletion. Indiscriminate extraction and lavish consumption of fossil fuels have led to reduction in underground-based carbon resources. Thus, the search for alternative fuels, which promise a harmonious correlation with sustainable development, energy conservation, efficiency and environmental preservation, has become highly pronounced in the present context [9].

Fortunately, biofuels can meet these demands. A biofuel is defined as any fuel whose energy is obtained through a process of biological carbon fixation [10]. The chemical structure of biofuels can differ in the same way that the chemical structure of fossil fuels can differ. Generally, liquid biofuels contain bioalcohols and biodiesels [11]. However, engines must be modified or redesigned to run on bioethanol or biodiesels. n-Butanol is a 4-carbon straight chain alcohol that can be produced from biomass (bio-butanol) as well as fossil fuels (petro-butanol), both of which have the same chemical properties and produce similar effects when used in engines [12]. Bio-butanol can run in any car that uses gasoline or diesel without modification of engine components [10,13]. Besides, bio-butanol has high energy density (29.2 MJ/L, only 9% below that of conventional gasoline or diesel) and less corrosion effects [14]. Thirdly, bio-butanol as a consequence of its lower polarity exhibits better miscibility characteristics with both gasoline and diesel [15]. Diesel/butanol blends require no emulsifying agents as they do not phase separate even after several days [16].

Soot emissions consistently decreased with increasing butanol content in the diesel/butanol blends due to a higher oxygen content and lower carbon content in butanol [17]. Larger numbers of experiments concerning the blending of gasoline or diesel with butanol have been conducted on engines [13,14,16–21], and results show a substantial reduction in soot emissions with butanol addition. Armas et al. [22] investigated the emission of diesel/alcohol blends on a turbocharged direct injection diesel engine during relatively cold and warm start, and reported a positive effect on the reduction in the smoke opacity and particle concentration during warm start as alcohol was added. However, the combustion instabilities and inefficient operation of the diesel oxidation catalyst lead to an increase of all regulated pollutant emissions. Gu et al. [23] studied the performance and emissions of iso-butanol and n-butanol enriched diesel fuel on a diesel engine, and found that the soot emissions from diesel/n-butanol blends were lower than those from diesel/iso-butanol blends. Liu et al. [24] pointed out that the oxygen in 80%diesel/20%n-butanol was the main factor for soot reduction in comparison with the diesel fuel, while the secondary factor for soot reduction was the improvement in fuel mixing process due to a longer ignition delay caused by lower cetane number of n-butanol, while other changes in physical and chemical properties of 80%diesel/20%n-butanol had very limited effect on soot reduction. Liu et al. [25] investigated the emission characteristics of n-butanol and soybean biodiesel in a constant volume chamber under various ambient temperatures. They observed that the normalized time integrated soot mass for B50D50 (50% n-butanol and 50% low-sulfur diesel fuel in volume) was 20–30% lower than that of neat biodiesel fuel at different ambient temperatures even though the oxygen content in both fuels was nearly the same, which meant that n-butanol was more effective in suppressing the soot formation than biodiesel.

In-cylinder combustion of diesel engine is an extremely complicated process with turbulent flow and spray in the high-temperature and high-pressure combustion chamber. Thus, the soot formation process in diesel engine is very difficult to be predicted due to a lot of influencing factors. There are many studies performed on shock tubes [26–28], flow reactors [29,30], and premixed flames [31,32]; however, the study of soot evolution in laminar non-premixed coflow flames are of special interest [33]. They have many advantages in soot investigation: (1) they enable to study very fuel rich pyrolysis regions, (2) have distinct soot growth and oxidation areas, (3) include a coupling between chemical and transport processes [34], (4) by minimizing physical effects, such as pressure, turbulence, and fuel sprays, they allow a detailed study of soot formation at a fundamental level in a multi-dimensional structure [35], and (5) they are relatively simple to model numerically [34].

Table 1
The mass flow rate, mean velocity, temperature, and mole fractions of the compositions at the inlet of the fuel stream.

Flames	B00	B25	B50	B75	B100
m(g/min)	0.165	0.176	0.188	0.201	0.214
V(cm/s)	11.47	11.66	11.86	12.07	12.29
T(K)	453	453	453	453	453
nC_7H_{16}	0.095	0.072	0.048	0.024	0
nC_4H_9OH	0	0.038	0.077	0.116	0.156
N_2	0.905	0.890	0.875	0.860	0.844
C/O	INF	25	11	6.33	4

Ethylene is the most commonly used fuel of soot investigation in laminar coflow flames, but recently some researchers investigated the soot behavior of liquid fuels, such as n-decane [35], biodiesel [36], real fuels and their surrogates [35,37–41]. However, SVF or SMF is the only focus in the vast majority of researches, and little attention was payed to soot number density [42]. In addition, to the best of the authors' knowledge, few investigations of diesel/n-butanol or diesel surrogate/n-butanol blends were conducted in laminar coflow flames, though similar flames fueled with gasoline/ethanol blends have been studied earlier by Khosousi et al. [39].

Gasoline and diesel contain a large number of hydrocarbons, and the most important components are n-heptane and toluene [43]. Therefore, n-heptane is frequently used as the chemical surrogate fuel in the calculation of engine combustion [44]. In this work, experiments and numerical calculations were conducted on n-heptane/n-butanol laminar coflow flames to investigate the effects of n-butanol addition on SVF and soot number density. A thermophoretic probe was used to sample the combustion-generated soot particles directly from the flames. The TEM images were obtained and compared among B00, B25, B50, B75 and B100 flames in terms of soot number density, soot size and therefore SVF, qualitatively. The calculations were carried out to further investigate the cumulative contributions of each stage in the evolution of soot particles to soot mass and number densities. Spacial distributions of the concentration of key species, A4 and C_2H_2, which dominate the inception and HACA process respectively, were compared and analyzed.

2. Experimental setup

2.1. Fuels

In this work, five liquid fuels were studied, i.e., pure n-heptane (> 99%, B00), pure n-butanol (>99.5%, B100), and three n-heptane/n-butanol blends, B25, B50 and B75, which consist of 25%, 50% and 75% n-butanol by volume fraction of the liquid fuel at room temperature (298 K), respectively. The mass flow of B00 fuel was 0.165 g/min, and that of other fuels were adjusted to keep the mole flow rate of carbon constant (0.01155 mol/min), as shown in Table 1 below.