Full Length Article

Experimental and kinetic investigation on soot formation of n-butanol-gasoline blends in laminar coflow diffusion flames

Fushui Liu[a,b], Yang Hua[a], Han Wu[a,*], Chia-fon Lee[a,c], Ziman Wang[a]

[a] School of Mechanical Engineering, Beijing Institute of Technology, Beijing 100081, China
[b] Beijing Electric Vehicle Collaborative Innovation Center, Beijing 100081, China
[c] Department of Mechanical Science and Engineering, University of Illinois at Urbana-Champaign, IL 61801, USA

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ABSTRACT

Bio-butanol has been considered to be an important alternative fuel to reduce soot emissions from gasoline engines, and it has the potential to overcome the disadvantages brought by low-carbon alcohols. In this study, the effect of n-butanol addition on soot formation in gasoline laminar diffusion flame is investigated respectively under the same fuel flow and the same carbon flow. The soot volume fraction distributions in the flames are measured using the two-color laser induced incandescence (TC-LII) technique. Furthermore, the But-TRF-PAH mechanism is constructed and used to simulate the formation and oxidation of soot precursors in the CHEMKIN zero-dimensional constant pressure reactor. The experimental results showed that n-Butanol addition can significantly reduce the soot formation, and the measured peak and average soot volume fraction both decrease linearly with the increasing n-butanol ratio. For a given n-butanol ratio, the reduction of soot under the same carbon flow (SCF) condition is only slightly smaller than that under the same fuel flow (SFF) condition, which indicates that the decreasing carbon mass flow due to n-butanol addition has little contribution to soot reduction. The simulated results showed that benzene is mainly produced via $C_6H_5CH_3 + H = A1 + CH_3$, and is mainly consumed via $A1 + OH = A1- + H_2O$ and $A1 + CH_3 = A1- + CH_4$. The production pathways of PAH is consistent for the toluene reference fuels (TRF) and n-butanol/TRF blends. The ratio of benzene accounts for toluene consumption is essentially constant, about 1/4. Thus, the n-butanol addition reducing the soot is mainly due to the dilution of the amount of aromatics (toluene in TRF) in gasoline. Furthermore, the amount of OH and HO_2 radicals increases with increasing n-butanol ratio, which have an important indirect effect on the soot precursors formation.

1. Introduction

Soot particulate (also known as particulate matter, PM) is one of the main emissions from IC engines, and is considered to be a serious threat to human health especially in urban area. Soot emission of traditional port fuel injection (PFI) gasoline engine is not an issue due to its stoichiometric premixed combustion mode [1]. Subsequently, passenger cars are widely equipped with gasoline direct injection (GDI) engine. Although GDI engine has advantages of high power, operational controllability, and low fuel consumption [2], it suffers severe soot emissions due to non-homogeneous air-fuel mixing [3].

Oxygenated biofuels partially or fully to replace with gasoline is considered to be an effect way to reduce soot emissions. Among various oxygenated biofuels, bio-alcohol fuel has been widely used as an alternative fuel for gasoline in transport applications. A very competitive and promising bio-alcohol is butanol. Butanol has the potential to

overcome the disadvantages brought by low-carbon alcohols, and has the advantages of high energy density (83% that of gasoline compared with 48 and 65% for methanol and ethanol), lower hydrophilicity (only 7.7 g/100 mL water), miscibility with gasoline in any proportion, and very little corrosive [4–6]. Furthermore, butanol can be produced from lignocellulose feedstock, such as straw, wheat straw, corn fiber and bagasse, and with the development of butanol fermentative process, the productivity of butanol is getting higher and higher [4,7–9]. Thus, many studies of butanol as an alternative biofuel have been conducted, in which butanol has been blended with gasoline for SI engine test, or has been studied on some fundamental flames. These studies have demonstrated that compared to methanol and ethanol, butanol is a better alternative fuel for the gasoline fuel, from the viewpoints of engine performance, combustion characteristics, and exhaust emissions.

Tornatore et al. [10] reported that addition of 40% n-butanol to gasoline is able to reduce the ultrafine carbonaceous particles in the

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exhaust without loss on engine performance. The similar results were also observed by Huang et al. [11] in a PFI spark ignition (SI) engine. Wang et al. [12] found that only 20% addition of n-butanol can significantly reduce particulate number (PN) emission on a direct injection spark ignition (DISI) engine, especially under the acceleration mode. The comparison of pure butanol and pure gasoline was studied by Irimescu et al. [13] in an optical DISI engine. They observed a significant reduction in smoke during the whole combustion process for pure butanol compared to pure gasoline. These studies have demonstrated that butanol partially or fully to replace with gasoline is an effect way to reduce soot emissions from gasoline engine.

Although the above mentioned works have primary proved the soot reducing effect of butanol on engine test bench, it is more important to conduct investigations on fundamental flames to separate the effects of combustion chemistry from engine operating conditions and realize the soot reducing mechanism. Wang et al. [14] studied the particle size distribution function of nascent soot of butanol and butane in premixed atmospheric flames by using the burner-stabilized stagnation flame technique. The results showed that the branched functional groups have the most significant effect on soot formation due to the faster formation of benzene. Moreover, under the same C/O ratio, the butanol flame has a higher soot volume fraction than the butane flame due to the earlier nucleation of soot. However, Singh et al. [15,16] found that butane isomers produce more soot than butanol isomers in an atmospheric non-premixed counterflow flames due to the effect of OH group. They also pointed out that in addition to C_3H_3, C_2H_2 also plays an important role in the PAHs formation. In addition, the effect of butanol addition on soot formation in different alkane flames has been studied. Ghiassi et al. [17] investigated the effect of n-butanol addition on the formation and oxidation of soot in n-dodecane premixed flames by using a two-stage burner. They found that n-butanol addition is able to reduce the soot particle number, but increase the stability of soot against oxidation. Jin et al. [18] investigated the effect of n-butanol addition on the PAHs formation in methane laminar coflow diffusion flames by using synchrotron VUV photoionization mass spectrometry technique. They also observed that butanol addition increases the formation of PAHs and benzene, and regarded that the increase was due to the enhanced formation of C2 species, which prompts the formation of propargyl radical and propyne. However, the larger alkanes as the main component of gasoline can decompose a larger number of C2 species. Thus, the synergistic effect of butanol addition in methane laminar diffusion flames will not occur in the gasoline laminar diffusion flames.

Considering that diffusion combustion is the main combustion mode of soot formation, and turbulent combustion is strongly influenced by the turbulent flow field, so the laminar diffusion flame is more suitable for the basic research to analyze and understand the formation of soot in diffusion combustion mode. The laminar diffusion flame is also the basis for further study on turbulence diffusion combustion. However, there is a lack of investigation on soot formation in n-butanol/gasoline laminar diffusion flames currently. Since gasoline contains n-alkanes, *iso*-alkanes and aromatics, the investigations in alkane flames [17,18] cannot reflect the case of real gasoline. Furthermore, there is a lack of the detailed 2D soot volume fraction distributions in the n-butanol/gasoline laminar diffusion flames. Thus, in this study, the effect of n-butanol blending on soot formation in gasoline laminar diffusion flames was investigated in the aspects of experiment and chemical kinetics.

In this study, a stable laminar coflow diffusion flame was obtained by a Gülder liquid burner system. The 2D quantitative distributions of soot in n-butanol-gasoline blends flames were measured by using two-color laser induced incandescence (TC-LII) technique. In order to study the effect of n-butanol blending ratio, the n-butanol content varying from 0 vol% to 80 vol%. Considering the possible effect of carbon content decreasing due to n-butanol addition, the fuel supply strategy includes the same fuel mass flow rate (SFF) and the same carbon mass flow rate (SCF). Furthermore, polycyclic aromatic hydrocarbons (PAHs) are generally known as the soot precursors, so a semi-detailed chemical
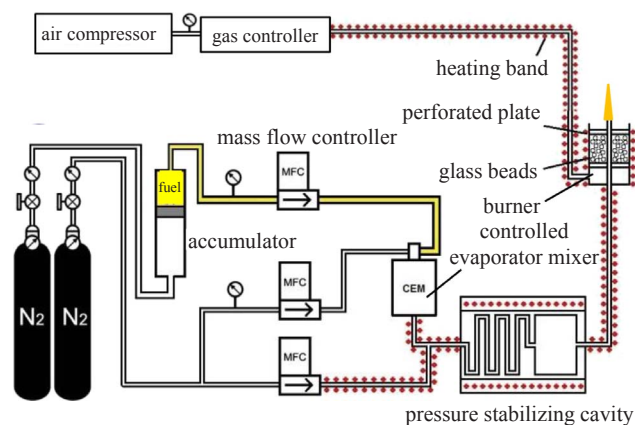

Fig. 1. The liquid burner system.

mechanism of But-TRF-PAH was developed from detailed ones. Then, the reaction pathways of soot formation was analyzed by using a zero-dimensional constant pressure reactor model in CHEMKIN package. This study could not only help to understand the effect of n-butanol blending to gasoline on soot formation in laminar diffusion flames under the SFF and SCF conditions, but also further understand how n-butanol addition to gasoline affects the soot formation.

2. Experimental setup and method

2.1. The liquid burner system

The liquid burner system shown in Fig. 1 [19] mainly comprises the Gülder burner [20], accumulator, controlled evaporator mixer (CEM), mass flow controllers (MFC), pressure stabilizing cavity, heating bands, nitrogen cylinders, air flow controller, and air compressor.

The Gülder burner is composed with two concentric tubes, the inner one with a diameter of 10.9 mm is used to supply fuel vapor, and the outer one with a diameter of 89 mm is used to provide coflow air. Glass beads and perforated plates are filled between the two tubes in order to have uniform air flow and stabilize the flame. The liquid fuel is supplied using a Tobul accumulator (3AT30-2) with a type of gas-liquid piston, whose volume is 1 L and maximum pressure is 20.68 MPa. To prevent the flame from smoking, and to ensure that the liquid fuel evaporates at about 473 K, the liquid fuel is first diluted with nitrogen, and then evaporates in a Bronkhorst controlled evaporator mixer (CEM) device. The fuel mass flow rate is controlled by a Bronkhorst Coriolis high precision mass flowmeter (CORI-FLOW), and nitrogen flow rate is controlled by a Bronkhorst thermal flowmeter (EL-FLOW). The pressure stabilizing cavity is used to buffer the disturbance of the fuel vapor and stabilize the flame. In addition, to prevent the fuel vapor from condensing [21], the tube between the CEM and the burner is heated using heating band, and the burner wall is heated using heating ring.

In the experiment, the flow rate of nitrogen was set at 0.3 L/min, (at 273 K, 1 atm) and the co-flow rate of air was set to 200 L/min (at 273 K, 1 atm), the temperatures of the heating band and heating ring were both set at 573 K. Under these conditions, stable flames with a height fluctuation of about 1–2 mm can be acquired.

2.2. Fuels

In this experiment, the fuels include pure gasoline (RON: 92, from China petrochemical gas station in Beijing), denoted as B0, and four n-butanol/gasoline blends, with the n-butanol ratio by volume of 20%, 40%, 60% and 80%, denoted as B20, B40, B60 and B80, respectively. The physical and chemical properties of gasoline and n-butanol are shown in Table 1. This experiment comprises two conditions: (1) fix the same fuel mass flow rate (at 9 g/h), denoted as SFF condition; and (2)