



Full Length Article

Effects of butanol isomers additions on soot nanostructure and reactivity in normal and inverse ethylene diffusion flames

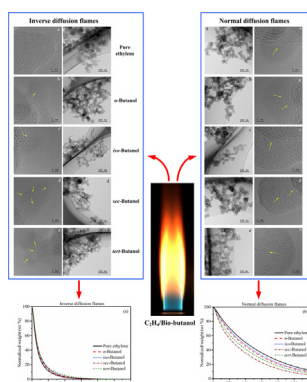
Yaoyao Ying, Dong Liu^{*}

Key Laboratory of Thermal Control of Electronic Equipment, Ministry of Industry and Information Technology, Nanjing University of Science and Technology, Nanjing 210094, People's Republic of China
Advanced Combustion Laboratory, School of Energy and Power Engineering, Nanjing University of Science and Technology, Nanjing 210094, People's Republic of China

HIGHLIGHTS

- The particles from butanol-doped IDF are highly heterogeneous in nanostructure.
- The samples from butanol-doped NDF show a typical core-shell nanostructure.
- For IDF, the reactivity ranks as *n*-butanol > *iso*-butanol > *sec*-butanol > *tert*-butanol.
- For NDF, the reactivity ranks as *sec*-butanol > *tert*-butanol > *n*-butanol > *iso*-butanol.
- A high correlation is found between soot nanostructure and reactivity.

GRAPHICAL ABSTRACT



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ABSTRACT

This paper explored the effects of butanol isomers, *n*-butanol, *iso*-butanol, *sec*-butanol, and *tert*-butanol, as fuel-side additives on soot nanostructure and reactivity in ethylene inverse diffusion flames (IDF) and normal diffusion flames (NDF). The variations of structures and oxidation rates among the soot samples derived from ten different flames were studied and compared using high resolution transmission electron spectroscopy (HRTEM), Raman spectroscopy, thermogravimetric analyzer (TGA), surface area and porosimetry analyzer, and elemental analyzer. Results demonstrated that soot from IDF with the additions of butanol isomers had irregular shapes with film-like materials and highly heterogeneous nanostructures, which showed both amorphous carbon and fullerenic lamellae. The shells presented more prevalently in *tert*-butanol-doped IDF soot, which had the longest fringe length and smallest fringe tortuosity. Whereas in the NDF as butanol isomers were individually added, the soot samples were aggregated by several tens or hundreds nearly rounded particles. All the NDF soot exhibited a typical core-shell structure with extended and planar lamellae. Soot from *sec*-butanol added NDF presented the lowest degree of graphitization with the shortest fringe length and largest fringe tortuosity. The TGA results revealed that the oxidation rates of the IDF soot particles were much higher than that of the NDF soot. Moreover, when butanol isomers were individually added in IDF, soot produced with *n*-butanol ($1.34\text{E-}03\text{ s}^{-1}$) showed the highest reactivity with the least degree of crystallization in structure, followed by *iso*-butanol ($1.31\text{E-}03\text{ s}^{-1}$), *sec*-butanol ($1.29\text{E-}03\text{ s}^{-1}$) and *tert*-butanol ($1.14\text{E-}03\text{ s}^{-1}$) in the sequence of reactivity. However, the oxidation rates of the soot samples generated from butanol isomers-doped NDF were in the order of *sec*-butanol ($4.26\text{E-}04\text{ s}^{-1}$), *tert*-butanol ($3.70\text{E-}04\text{ s}^{-1}$), *n*-butanol

^{*} Corresponding author at: Key Laboratory of Thermal Control of Electronic Equipment, Ministry of Industry and Information Technology, Nanjing University of Science and Technology, Nanjing 210094, People's Republic of China.

E-mail address: dongliu@njust.edu.cn (D. Liu).

(3.52E-04 s⁻¹) and *iso*-butanol (3.23E-04 s⁻¹) from the highest to lowest. The results confirmed a relationship on structure-property between soot nanostructure and reactivity.

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1. Introduction

It is well known that soot particles emitted by combustion systems such as furnaces and internal engines are inescapable, which are important to increase heat transfer and combustion efficiency [1]. However, soot particles have been implicated in adverse environment and human health consequences [2]. As a result, the governmental agencies are enacting increasingly rigorous regulations on the soot emissions from various combustion sources. To reduce the emissions of soot particles to the environment, combustion researchers and combustion device designers are on the way to get effective approaches by experimental and theoretical studies, including reducing dependence on traditional fossil fuels, increasing efficiency of combustion systems, and using alternative biofuels [3,4]. Biofuels, such as bio-ethanol, bio-butanol, and biodiesel, as alternatives to petroleum-based transportation fuels are of increasing interests in recent years due to the long-term promise of fuel-source regenerability and climate impacts reduction [5]. Alcohols as oxygenated compounds stand out as good alternative fuels or fuel additives on the reduction of soot emissions and have been widely investigated [3,6–8]. Butanol is a very promising fuel alternative or additive which has higher energy density (36 MJ/kg) [9], lower water absorption and better miscibility with present fuels compared to ethanol [5]. The cetane number of butanol is higher and it is less corrosive than methanol and ethanol [10]. Furthermore, there are ongoing researches for developing production processes of higher alcohols such as butanol on a commercial scale [11–13].

Butanol has a formula C₄H₉OH, and there exists four structural isomers of butanol. These four isomers (*n*-butanol CH₃CH₂CH₂CH₂OH, *sec*-butanol CH₃CH₂CHOHCH₃, *iso*-butanol (CH₃)₂CHCH₂OH, *tert*-butanol (CH₃)₃COH) have the same formula and same amount of heat energy, but due to the different molecular structures they show different properties like ignition and reaction behaviors [5,14]. Investigations of the effects of butanol on combustion fundamental characteristics and soot emissions have been extensively performed experimentally and numerically.

Chen et al. [15] conducted an experimental study on a direct injection diesel engine with exhaust gas recirculation (EGR) and port fuel injection (PFI) of *n*-butanol, and the results showed that with the PFI of *n*-butanol, the soot emissions decreased due to the large premixed combustion part and the fuel-bound oxygen content. Siwale et al. [16] evaluated the combustion characteristics and emissions of neat diesel fuel and *n*-butanol/diesel blended fuel on a high load turbo-charged diesel engine. Compared to the neat diesel combustion, soot emission showed a substantial reduction when the engine was filled with *n*-butanol/diesel blended fuel. Zhang and Balasubramanian [17] used blends of ultralow sulfur diesel with biodiesel or *n*-butanol to investigate the characteristics of particulate emissions. The results illustrated that both the blended fuels could reduce the particulate mass and elemental carbon emissions, and *n*-butanol blending was more effective. Moreover, some other investigations on *n*-butanol/diesel blends have demonstrated similar results as the aforementioned refs., showing that *n*-butanol is a potentially promising renewable alternative fuel to reduce soot emissions with the present engines [18–22]. Some previous studies have investigated *iso*-butanol effect on engine emissions [23,24] and comparisons of the impacts of butanol isomers on diesel soot emissions have been conducted as well

[25,26]. The results all concluded that the additions of butanol isomers in diesel could make a significant decrease in soot emission and a higher blending contributed to larger soot reduction, while there was very small difference between butanol isomers affecting on soot emission reduction.

In addition, sooting behavior of butanol isomers in lab-scale flame configurations has also been studied. McEnally and Pfefferle [27] investigated the sooting property of butanol isomers by adding 3500 ppm of each kind of butanol and butane isomers in coflowing methane/air flames, and found that all dopants enhanced the benzene production and butanols generated more aldehydes and ketones than butanes. Camacho et al. [28] examined the sooting characteristics in burner stabilized stagnation flames to understand the impacts of fuel structure and fuel bound hydroxyl group. The results illustrated that the branched fuels nucleated earlier than the straight-chain fuels, and under the same C/O ratio, butanol flames nucleated soot faster and had higher soot volume fractions than butane flames. Singh et al. [29] used laser diagnostics techniques to detect the influences of hydroxyl group and fuel structure on the soot formation of butanol and butane isomers in non-premixed counterflow flames, and found that butane isomers generated more soot than butanol isomers which illustrated the hydroxyl group effect on soot emission reduction.

However, a limited number of studies have been performed to investigate the soot nanostructure and reactivity with butanol isomers as fuel additives to conventional hydrocarbon fuels. Ghiassi et al. [30] studied the effects of *n*-butanol on the formation, nanostructure and oxidation behaviors of soot, which was added into *n*-dodecane in a two-stage burner. They obtained quantified fringe orientation maps and illustrated soot oxidation reactivity had a correlation with the degree of crystalline order of carbon layers.

Furthermore, it has been identified that soot structural differences at a nanoscale level can influence its oxidation reactivity depending on fuel and combustion conditions [31]. Various previous studies considering diesel or biodiesel soot [32–36], and only a few works regarding carbon blacks or soot produced in premixed or diffusion burner flames [30,37–39] have confirmed that the degree of soot orderliness of carbon lamella is inversely related with soot oxidation reactivity. To our limited knowledge, none of the literature studies have been conducted to investigate the soot nanostructure and reactivity of these four butanol isomers as fuel additives thoroughly in both inverse diffusion flames (IDF) and normal diffusion flames (NDF).

To gain the structural characteristics of carbonaceous materials, Raman spectroscopy and high-resolution transmission electron microscopy (HRTEM) are widely used. Raman spectroscopy is one of the suitable techniques for carbon material characterization and has been successfully implemented for various kinds of carbons [40–44]. Raman spectroscopy is quite sensitive to the structures and the changes of the analyzed sample. Since the Raman signal is sensitive to short range order, it can also reveal ordering degree of amorphous carbons. Raman analysis can provide a measure of disorder relative to graphitic content and is based upon semi-empirical relations for quantitative information. However, the interpretation of Raman spectra and the data processing of carbon materials generate considerable debate in the literature. To compensate for these deficiencies, HRTEM has been widely used in conjunction with Raman spectroscopy in previous studies [45–48]. HRTEM offers direct information on the graphene layers at

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