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Morphological analysis of soot agglomerates from biodiesel surrogates in a coflow burner



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ABSTRACT

The effects of unsaturation (including the C–C double bond's position in the alkyl chain) and the ester moiety in the fuel molecule on the morphology of soot particles in a laminar coflow diffusion flame are studied. The effects of the ester moiety are evaluated by comparing an n-decane flame with a biodiesel surrogate flame (composed of 50%/50% molar blend of n-decane and methyl octanoate). The effects of the unsaturation and the position of C–C double bond in the alkyl chain are analysed by comparing 1-decene, 5-decene and n-decane flames. Particles were collected thermophoretically on Transmission Electron Microscopy (TEM) carbon coated copper grids. TEM images are analysed to obtain parameters related to the size of the agglomerates such as radius of gyration, and morphological parameters such as fractal dimension and prefactor of the power-law relationship. The results show that the average primary particle diameter, the size of the agglomerates and the number of primary particles composing the agglomerates increase along the flame length to around two thirds of the flame length and then decrease as a consequence of oxidation becoming dominant over soot nucleation and growth. The fractal dimension of the agglomerates does not change significantly along the different pathlines of the flames. However, effects of the fuel chemical structure are clear. The lowest fractal dimensions are observed for the oxygenated fuel and the highest ones for the unsaturated fuels, with higher fractal dimension when the double bond is located at the edge of the molecule. The same trends are observed for agglomerate sizes: smallest agglomerates are observed for the oxygenated fuel and largest ones for the unsaturated fuels. This suggests that low soot-emitting fuels reduce their size as a consequence of oxidation while keeping their agglomeration skeletal structure.

1. Introduction

Soot is a flame-made carbonaceous nanoparticle that is considered a pollutant emission when it is released from combustion of hydrocarbon fuels (Stanmore, Brillhac, & Gilot, 2001). The bright yellow luminosity of hydrocarbon flames indicates the presence of microscopic soot nanoparticles which are formed, transported, and ultimately oxidized in the combustion flames (Megaridis & Dobbins, 1989). Soot particles emitted into the atmosphere cause increased mortality due to respiratory and cardiovascular disease (Pope, 2000), are a major factor in global warming due to the greenhouse effect (Quaas, 2011), and decrease atmospheric visibility (Horvath, 1993).

With a continuously tightening global allowance of particulate matter from hydrocarbon exhaust (Hageman, Sakai, & Rothamer,

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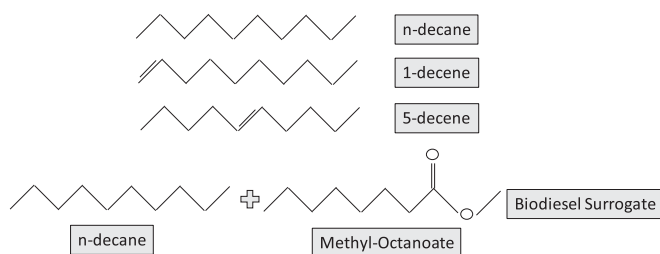


Fig. 1. Fuels used in this study.

2015), alternative fuels such as Fischer-Tropsch (F-T) diesel, vegetable oils, and biodiesel have been studied for use in transportation vehicles. These “greener” fuels have shown significant promise in reducing particulate matter emissions. Therefore, a great deal of effort has been put into determining the effects of biodiesel fuel chemistry (Herbinet, Pitz, & Westbrook, 2008) on engine performance and particulate emissions when used as a pure fuel (B100) or as a blend with conventional diesel fuel (Lapuerta, Armas, & Rodríguez-Fernández, 2008).

Biodiesel fuel consists of long chain fatty acid methyl esters (FAME) derived from renewable feedstocks such as vegetable oils, recycled cooking grease, or animal fats for use in diesel engines (Baig & Ng, 2010). Biodiesel is compatible with conventional diesel engines and can be used as a complete alternative or blended in any proportion (Lapuerta et al., 2008). Biodiesel is typically characterized by two recognizable features when compared with other fuels. These are the presence of an ester moiety and unsaturation which is indicated by the presence of one or more carbon-carbon double bonds. These features have different effects on the soot formation. The ester moiety typically decreases soot production, while unsaturated species increase the amount of soot produced. Unlike conventional diesel, biodiesel contains no aromatic species which are known to increase the amount of soot produced (Cignoli, De Iulii, & Zizak, 2001; Scrimgeour, 2005).

This study focuses on the effects of unsaturation (including the position of C-C double bond in the alkyl chain) and the ester moiety, on the soot morphology in laminar coflow diffusion flames. To investigate the effects of the ester moiety, n-decane is compared with a biodiesel surrogate which has been defined as a 50%/50% molar blend of n-decane and methyl-octanoate. To study the effects of unsaturation and that of the double bond location 1-decene and 5-decene were chosen. Both 1-decene and 5-decene contain the same number of carbon atoms as n-decane, with the carbon-carbon double bond located at the end and middle of the molecule, respectively. Fig. 1 shows the different fuels used in this study.

There is need for techniques and methods which provide in-situ information of particle morphologies, in order to improve understanding of soot formation and oxidation during combustion (Michelsen, 2017). In this study, thermophoretic sampling was followed by Low-Resolution Transmission Electron Microscopy (LRTEM) image acquisition and multiple image analysis of primary particle sizes, agglomerate sizes and fractal structure of the agglomerates. The information obtained is useful for understanding and modelling different phenomena involving soot agglomerates in both flames and post-flame flows, such as collision frequencies (with consequences on the size distributions of the emitted particles and thus on their health effects), optical properties (with consequences in radiative forcing, opacity and climate change), chemical reactivity (with consequences in soot regeneration and atmospheric activity), adsorption capability (with consequence on particulate matter composition), filtration efficiencies (with consequences in filter designs and health effects), etc.

This study is complementary to two previous studies (Kholghy & Weingarten et al., 2017; Kholghy, Afarin et al., 2017). In Kholghy, Weingarten et al. (2017), the structural effects of biodiesel on soot formation were studied by comparing the concentration, primary particle size, and primary particle number densities of the same fuels, at different flame locations. The results showed that the ester moiety in the fuel reduces soot concentration mainly by decreasing the concentration of soot growth species such as acetylene. However, unsaturation increases soot concentration by increasing both soot inception and growth species such as benzene and acetylene, respectively. A more centrally located C=C double bond further increases soot inception species such as benzene. In Kholghy, Afarin et al., (2017), multiple diagnostic techniques were used on a 1-decene flame, to compare the sensitivity of each method to soot particles with different age and maturity. It was shown that all of the optical techniques used are only sensitive to carbonized mature soot. The focus of the current study is on the structural effects of biodiesel on the agglomerate structure of soot particles in the coflow diffusion flames.

2. Experimental apparatus and methodology

2.1. Burner and vaporizer

A co-annular burner (Snelling et al., 2002) was used in this study to create an atmospheric laminar diffusion flame. The schematic of the burner is illustrated in Fig. 2. To deliver the vaporized mixture to the burner, a heated tube was used with flexible heaters at the vaporizer outlet and the burner inlet. Once the mixture entered the burner it was heated along the fuel tube using coil heaters to stop the fuel from condensing as it leaves and mixes with the air and allows for a short flame lift-off. Adding extra oxygen with air further reduces lift-off to around 1 mm, and improves the stability of the flame. The oxygen/air mixture was injected through the air annulus of the burner at 3.30 SLPM and 55 SLPM respectively (with flowmeters calibrated at 21 °C and 1 atm). Lab temperatures ranged from

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