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Formation and evolution of carbon particles in coflow diffusion air flames of vaporized biodiesel, diesel and biodiesel-diesel blends

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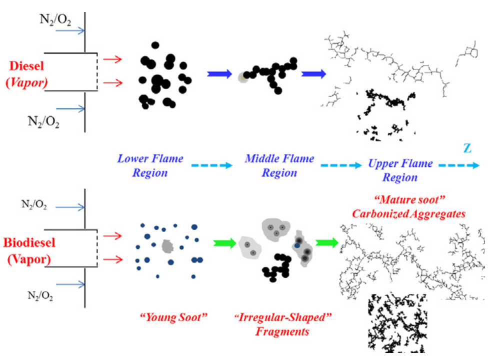
GRAPHICAL ABSTRACT



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ABSTRACT

Evolution profiles on the formation of the carbon particulate (soot) in vaporized coflow diffusion flames of biodiesel (BD), No. 2 diesel and blended fuel mixtures were obtained. The evolution profiles contain the different stages of soot formation, including soot inception, particle growth and agglomeration, and oxidation. Carbon samples were collected directly from inside the flame's yellow luminous zone along the axial direction at various heights above the burner (HAB) using a well-accepted approach. The studied BD flames were formed using canola methyl ester (CME), cotton methyl ester (COME) and soy methyl ester (SME) in their neat form (B100). The blends consisted of B80 (80% CME/20% No. 2 diesel), B50 (50% CME/50% No. 2 diesel) and B20 (20% CME/80% No. 2 diesel). The evolution profile of the No. 2 diesel flame was compared to the evolution profiles of the neat (B100) BD and blended fuel flames. Soot evolution profiles in the studied vaporized BD and diesel flames are consistent with the general trends of particle formation present during the combustion of diesel and gaseous fuels. That is, particle inception (singlet particles) present in a region at the lower part of the flame followed by particle growth and agglomeration, and the subsequent soot carbonization and oxidation in the upper regions of the flame. However, the soot evolution profiles also show that significant differences exist in the soot morphological properties of the tested BD and blended flames. The presence of "irregular-shaped" fragments such as the "liquid-like" droplets or "globules" at the different stages of soot formation is evident in these oxygenated flames. The "irregular-shaped" fragments resemble short chain-like aggregates that appear to be formed of fused particles having undefined shapes and boundaries. The "irregular-shaped" fragments resemble eutectic (solid/liquid) phases manifesting their viscous liquid nature. Some of these "irregular-shaped" structures contain embedded carbonized inclusions. By a small axial variation of the flame, the fragments are transformed into fully carbonized aggregates that are significantly larger and of complex fractal morphology (multi-branched). From the transmission electron microscopy analysis it can be suggested that the "liquid-like"

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droplets or "globule" structures serve as possible growth pathways for the formation of the fully carbonized aggregates composed of spherical particles in the upper part of the flame. The size and number density of the "irregular-shaped" fragments in the vaporized BD are much larger than those present in the flame formed from the vaporized diesel. It is also observed that as the percentage of BD is increased in the blended flames, these "irregular-shaped" structures become more pronounced. The evolution profiles also present other morphological properties of the carbon particulates (particle size and nanostructure) along the flame's axial direction.

1. Introduction

Oils extracted from vegetables, animal fats and their derivatives can be transformed into biodiesel (BD) through a transesterification process. In that process a short-chain alcohol (typically ethanol or methanol) is added to a triglyceride molecule (oil) in order to separate the fatty acid chains that are attached to a glycerol "backbone". Potassium hydroxide or sodium hydroxides are typically used to act as a catalyst to speed up the reaction. BD exhibits its potential to be an alternative form of energy to alleviate the enormous amount of diesel used in engines as the worldwide supply of petroleum is continuously diminishing. For instance, in 2016 approximately 29% of the total U.S. energy consumption was in the transportation sector [1]. In that particular sector diesel is used to power compression-ignition engines of trains, boats, trucks, public and school buses, equipment in farms and construction, tanks (military), among many others. From ~29% of energy consumption in the transportation sector; diesel fuel accounted for ~21% of the petroleum fuels consumed [1], gasoline (petroleum) ~55%, jet fuel (petroleum) ~12%, biofuels ~5%, natural gas ~4% and other ~4%. Therefore, BD is already having an impact in the transportation area. According to the U.S. EPA Moderated Transaction system data, the market for biodiesel and renewable diesel topped 2.6 billion gallons in 2017, while in 2016 the market approached 2.9 billion gallons [2]. Additionally, analogous to gasoline blended with ethanol, BD can be easily blended with diesel. BD consumption increased from 10 million gallons in 2001 to approximately 2.1 billion gallons in 2016 reflecting the growing use of BD. BD is chemically similar to diesel and both have a high number of C and H atoms. However, BD contains an oxygen molecule attached to the hydrocarbon structure which can significantly suppress or enhance the combustion by-products during its burning [3,4]. Combustor configurations (such as Diesel engines and shock tubes) have been employed to study the emissions resulting from the burning of BD [5–8]. Several research efforts have shown that BD can significantly reduce CO_2, CO, SO_x, NO_x and other volatile organic compounds in engines running with BD [9,10]. Other studies using diesel engines have been conducted for understanding the effects of BD on particulate matter (PM) and, especially, particle morphology [6,5,8,11–20]. The understanding of the underlying mechanisms of the formation of these ultrafine particulates in emerging fuels is vital for many industrial processes, as well as the atmosphere/environment and human health. Internal combustion engines or shock tubes represent closed experiments where samples of soot particles are difficult to extract during their growth process. In those BD studies most of the results in soot morphology are based on samples trapped after leaving the reaction zone, making it difficult to understand the soot properties, such as particle nucleation or inception, growth, aggregation and oxidation. However, flames have been found to be an ideal medium for understanding soot formation as samples can be easily extracted from different regions of the flame, hence allowing to obtain fundamental information of all growth steps of soot within a single medium. Studies of soot formation using various types of gaseous fuels, burner types, and flame configurations have been performed. Those flame studies have shown that the type of fuel, addition of oxygen in the oxidizer, doping of the base fuel with small concentrations of energetic fuels, and the order in which the fuel and oxidizer are introduced into a combustion chamber can all significantly contribute to variations in soot concentration and in soot morphology (appearance, size and nanostructure) [21–29]. However, only recently a few studies on the thermal and pollutant emissions have been conducted on BD formed flames [30–34].

Despite the advantages that BD offers, the usage of neat BD can have its own complications. Some well-known major drawbacks of neat BD are: i) its high viscosity compared to Diesel or other common liquid fuels; ii) its strong effect towards oxidation and thermal degradation; iii) its high cloud point that will lead to solidification of the fuel at lower temperatures in comparison to diesel. One of the simplest and most inexpensive ways to remedy some of these problems is to blend BD with Diesel. For instance, the blending of BD (characteristics of higher cloud point) with Diesel (lower cloud point) can significantly improve the practicability of BD in low temperature environments. Fortunately, biodiesel is thoroughly mixable with diesel making it possible to use their blending in order to take advantage of BD's benefits [35,36]. Therefore, many studies have been conducted with the aim of investigating the properties and emissions of various biodiesel/diesel blends in engines [7,37–41]. Similarly, the flame approach could be used to study soot formation in blended BD/diesel fuel mixture by varying the fuel concentrations. In addition to the impracticability of BDs under extreme environmental conditions there is the social issue of the "food vs. fuel" controversy. Despite the advantages of biofuels (such as the lessening of petro-fuel dependence, renewable, biodegradable, superior lubricity properties to fuel, and its environmental benefits) some concerns have been raised about the large volume of corn and other grains that are being diverted for use as biofuel feedstocks [42]. Additionally, it has been pointed out that enormous areas of farm land will be needed for making biofuels capable of competing with petro-fuel. These factors are driving the development of biofuels (i.e. BD) to be derived from feedstocks that are less controversial. These include BD made of oils from terrestrial plants such as soybeans, cotton, rapeseed, palm, among many others. In order to address some of these challenges some strategies are being developed. One proposed strategy is to obtain the oil from plants that do not need nutrient rich soil such as is used in traditional agricultural crop plants [43–45]. Another strategy is to obtain oils from plants that can grow in a few months but are full of energy-rich oils and are at the bottom of the food chain. For instance, it has been proposed that it is possible to obtain BD from a non-terrestrial plant such as algae [43,46]. Algae are composed of a broader genetic diversity that is considered photosynthetic and aquatic. Some algae groups can grow rapidly to orders of meters long producing biomass very quickly. Some of the species double in as few as six hours, many of which exhibit two doublings per day [47,48]. Most algae groups have the capacity to produce energy-rich oils. A number of microalgal species have been found to naturally accumulate high oil levels in totally dry biomass as well [49]. The algae strategy can also indirectly remedy other existing environmental issues. Being a non-terrestrial plant, algae can be grown in waste streams (i.e. municipal waste water) removing nitrates and phosphates before they are released. Algae are known for efficiently using CO_2 and hence can clean flue gases of coal and other combustible-based power plants by capturing sulfates and CO_2 from the flue gas [50–54].

In this study we report evolution profiles containing physical characteristics of soot in vaporized coflow diffusion flames along the flame centerline. The profiles are obtained using transmission electron microscopy (TEM) on the collected samples revealing the sequence of the morphological changes within a flame axial direction. The studied