



Research article

Near-azeotropic volatility behavior of hydrous and anhydrous ethanol gasoline mixtures and impact on droplet evaporation dynamics

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ABSTRACT

After fermentation, the concentration of bioethanol is only 8–12 wt%. To produce anhydrous ethanol fuel, a significant amount of energy is required for separation and dehydration. Once the azeotrope composition is reached, distillation can no longer be exploited for purification and more expensive methods must be used. Replacing anhydrous ethanol fuel with hydrous ethanol (at the azeotrope composition) can result in significant energy and cost savings during production. The goal of this study was to characterize the volatility behavior and the droplet evaporation dynamics of hydrous and anhydrous ethanol gasoline blends. Three hydrous ethanol-gasoline blends (10, 15, and 30 vol%) in which the hydrous ethanol was composed of the azeotropic proportions of ethanol and water, and three anhydrous ethanol gasoline blends (10, 15, and 30 vol%) were prepared and analyzed with the advanced distillation curve method. Distillation curves were obtained for all test fuels and distillate samples were taken during the distillation process. A droplet evaporation model validated with the distillation data was exploited to understand how the non-ideal volatility behavior of these blends, the high heat of vaporization of water, and altered fluid properties can affect the transient droplet evaporation phenomena and thus the fuel's potential to effectively mix with air in direct injection internal combustion engines. Minor differences in the distillation curves and vapor-liquid equilibrium between the hydrous and anhydrous fuels were measured. Droplet modeling results showed that the higher heat of vaporization and viscosity of water relative to ethanol can lead to significant differences in the net droplet evaporation time between the two types of blends, especially at the higher blending ratios evaluated. These results suggest that the presence of water in ethanol-gasoline blends may extend droplet lifetimes and increase the susceptibility of the fuel to form particulate matter emissions. This is the first study to use distillation methods to gain a better understanding of evaporation behavior and the role of water's non-linear vapor-liquid equilibrium on droplet evaporation dynamics.

1. Introduction

In recent years, biofuels have been used to offset the consumption of gasoline and diesel because they can be derived from renewable resources while being less harmful to the environment and humans, especially regarding greenhouse gas production [1]. Among biofuels, bioethanol has been produced in the largest quantities and is blended with gasoline at 10 vol% in nearly all the United States. The primary motivation to use ethanol is that it can be produced with low cost from renewable feedstocks. Despite having lower specific energy content than the gasoline it is replacing, the use of ethanol can lead to some improvements in combustion and emission characteristics stemming from its oxygen content. Ethanol has a high octane number, allowing engines to operate at higher compression ratios, thereby promoting fuel

efficiency. In addition, its high heat of vaporization (HoV) can lead to a charge cooling effect that can enhance volumetric efficiency through increased brake mean effective pressure [2–4]. In 2012, the U.S. Environmental Protection Agency approved ethanol-blended gasoline at volumetric concentrations up to 15 vol% for use in Model Year 2001 and newer cars, light-duty trucks, medium-duty passenger vehicles, and all flex-fuel vehicles. In the United States, the ethanol blended in gasoline is anhydrous, with a maximum water content of 1 wt%, as specified by ASTM D579 8-99 [5] for fuel ethanol for automotive spark-ignition engines. However, ethanol produced by fermentation results in a mixture with a concentration of only 8–12 wt% ethanol; the rest is composed primarily of water along with organic acids, carbon dioxide, and other trace species [6]. Thus, to produce the anhydrous ethanol fuel, a significant amount of energy is required for separation and

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dehydration. Shapouri et al. [7] conducted a study to identify the net energy value of corn ethanol. In their study, the total output energy based on the higher heating value of ethanol and the energy credits of the co-products was estimated to be around 25 MJ/L. These authors determined that the energy required for water removal to obtain anhydrous ethanol accounts for 37% (~9.5 MJ/L) of the total output energy, including distillation (23%) and dehydration (14%) processes.

Separating water and ethanol by way of distillation is an energy-intensive process, especially when distillation-based separation techniques are applied to mixtures containing > 90% ethanol. This is a result of azeotrope interactions; at the ethanol-water azeotrope point (95.6/4.4 wt% ethanol/water at 1 atm) no further separation can be achieved at constant pressure. Instead, additional energy must be provided to overcome this limitation [8,9]. Once an azeotropic mixture forms, distillation can no longer be exploited for further purification [10]. Instead, methods such as membrane-distillation hybrids, pressure-swing distillation, entrainer-addition distillation methods, and molecular sieve separation techniques are required [11,12]. These alternate methods add expense, complexity, and energy requirements. The expense of anhydrous ethanol production suggests opportunities for improvements from economic, energy, and greenhouse gas points of view. One option to address these shortcomings is to use hydrous ethanol (at the water/ethanol azeotropic composition) blended with gasoline. This could save up to 14% of the fuel energy during its production (~3.5 MJ/L) [6]. However, questions related to the water addition and its impact on engine operation, fuel economy, and fuel supply systems must be answered.

Although hydrous ethanol may cause negative long-term impacts such as lubricant deterioration and fuel system corrosion [10], its overall impact on combustion and emission characteristics has been shown to be positive. Several studies have been conducted to investigate the impact of hydrous ethanol on engine performance and emissions [10,13–19]. Generally, in comparison to anhydrous ethanol blends, hydrous ethanol blends show higher brake thermal efficiency and brake-specific fuel consumption (BSFC) with lower unburned hydrocarbon (UHC), CO, and NO_x emissions. BSFC is increased due to the lower heating value of hydrous ethanol [10]. Although the lower heating value and lower flame speed of hydrous ethanol compared to anhydrous ethanol result in a lower peak heat release rate and pressure, efficiency improvements are observed because of charge cooling effects stemming from the HoV of hydrous ethanol which decreases heat losses to the cylinder walls and allows for increased mass loading into the cylinder [13]. The HoV-influenced cooling and subsequent lower flame temperatures also leads to reduction in NO_x emissions. At low loads, the presence of water decreases the exhaust gas temperature and limits the oxidation of CO and UHC. However, at high loads, breakdown of water into hydroxyl and hydrogen radicals promotes the oxidation of CO and UHC at high temperature conditions [18]. It should be noted that these previous studies were carried out with current engine platforms designed for anhydrous fuels and did not examine the effect of tuning/optimizing engine design to leverage gains in octane numbers and charge cooling corresponding to water addition on fuel economy. Such modifications could potentially offset the reduction in overall fuel heating value when water is present.

In direct injection spark ignition (DISI) engines, the spray atomization and fuel evaporation processes play an essential role in the combustion efficiency and emission formation [20]. In several studies conducted on DISI engines, it has been observed that use of a gasoline containing moderate ethanol concentrations (10–20 vol%) increases particulate matter (PM) emissions relative to base-gasoline stemming from slowed spray/droplet evaporation dynamics resulting from the high HoV of ethanol [21–25]. The goal of this study was to characterize the volatility behavior and mixing/sooting potential of hydrous and anhydrous ethanol blends. An advanced distillation apparatus was used to obtain distillation curves for gasoline, gasoline-hydrous ethanol and gasoline-anhydrous ethanol mixtures. Distillate samples were

withdrawn at various points during distillation and their corresponding compositions were quantified, including the transient distillate water concentration. A distillation-based droplet evaporation model containing > 50 species was validated with the experimental data and used to provide insight into the spray and evaporation processes, which have been shown to play an important role in PM formation, of the hydrous and anhydrous fuel blends by tracking the changes in droplet composition and physical properties during evaporation.

2. Material and methods

2.1. Test fuels

Unleaded test gasoline (UTG-96) from Phillips 66 was used as the base fuel (E0). The base gasoline was blended with 10, 15 and 30 vol% of anhydrous and hydrous ethanol. The blends were designated as E10, E15, E30, H10, H15, and H30 corresponding to either anhydrous (E) or hydrous (H) ethanol and the blended volume percentage. Hydrous ethanol was obtained by mixing 96 vol% of anhydrous ethanol with 4 vol% of deionized water to match the reported composition for the azeotrope mixture. Ethanol (200 proof, ≥99.5%) was purchased from Pharmco-AAPER. The proof of ethanol was verified via Karl Fischer titration (described below).

2.2. Methods

A vapor pressure analyzer (Grabner Instruments Minivap VPXpert) was used to measure Reid vapor pressure (RVP) according to ASTM 5191 [26]. Each test was repeated three times.

A custom-built Advanced Distillation Curve (ADC) apparatus was used to obtain the distillation curves [27–29]. The ADC provides a number of advantages over ASTM D86 – Standard Test Method for Distillation of Liquid Fuels and was chosen for two primary reasons. First, the ADC offers the ability to sample and analyze the condensate so that the composition along with the temperature is monitored as the distillation progresses. This provides additional data to validate thermodynamic models. Secondly, the ADC measures temperature in two locations; one in the boiling fluid itself and the other in the vapor head space above the boiling fluid. This differs from ASTM D86 in which the temperature is only measured in the head space. The temperature in the boiling fluid is a more appropriate indicator of the thermodynamic state and thus provides a better comparison to thermodynamic models utilizing empirically derived vapor-liquid equilibrium data. The measurement in the head space above the boiling fluid can be used to compare to data taken according to the ASTM D86 method.

Additional details of the ADC method have been reported elsewhere [30]. In brief, a temperature-controlled heating mantle is placed around the boiling kettle containing 200 mL of the fuel blend, which is stirred to ensure a uniform composition and temperature within the boiling mixture. Temperatures of the liquid in the kettle and vapor in the distillation head are monitored with two K-type thermocouples. These thermocouples continuously record temperatures using a data acquisition system. The temperature of the heating mantle is continuously adjusted to lead the boiling fluid temperature by ~20 °C ensuring even heating throughout the distillation process. The condenser tube is chilled with water maintained at 5 °C. The apparatus is equipped with a custom sampling adapter located between the condenser tube and the volumetric receiver, which provides the ability to withdraw samples of the distillate during the distillation process. The volumetric receiver collects the condensed liquid and is calibrated to measure the distilled liquid volume. The receiver is cooled by chilled air from a vortex tube at 2 °C to prevent any vapor loss. At every 5% volume distilled, the liquid temperature in the kettle is recorded and used to create an accurate distillation curve. Distillation curves for each mixture were measured twice. Generally, the initial boiling temperature is difficult to observe and measure. In this study, the initial boiling temperature

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