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Solubility of a diesel–biodiesel–ethanol blend, its fuel properties, and its emission characteristics from diesel engine

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Abstract

In this work, we studied the phase diagram of diesel–biodiesel–ethanol blends at different purities of ethanol and different temperatures. Fuel properties (such as density, heat of combustion, cetane number, flash point and pour point) of the selected blends and their emissions performance in a diesel engine were examined and compared to those of base diesel. It was found that the fuel properties were close to the standard limit for diesel fuel; however, the flash point of blends containing ethanol was quite different from that of conventional diesel. The high cetane value of biodiesel could compensate for the decrease of the cetane number of the blends caused by the presence of ethanol. The heating value of the blends containing lower than 10% ethanol was not significantly different from that of diesel. As for the emissions of the blends, it was found that CO and HC reduced significantly at high engine load, whereas NO_x increased, when compared to those of diesel. Taking these facts into account, a blend of 80% diesel, 15% biodiesel and 5% ethanol was the most suitable ratio for diesohol production because of the acceptable fuel properties (except flash point) and the reduction of emissions.

Keywords: Diesel; Ethanol; Biodiesel

1. Introduction

For the past few decades, a lot of effort has been made to reduce the dependency on petroleum fuels for power generation and transportation all over the world. Among the proposed alternative fuels, biodiesel and diesohol have received much attention in recent years for diesel engines and could be one remedy in many countries to reduce their oil imports (since this study was done in Thailand, the focus has been placed on assisting the Thai government in reducing oil imports). Biodiesel and diesohol have many advantages over regular diesel as renewable and domestically produced energy resources. Moreover, they are recognized as environmentally friendly alternative fuels because previous studies have shown that there is a substantial reduction of CO, unburned hydrocarbons and particulate matter emission, when they are used in conventional diesel engines [1–3]. Biodiesel is an alkyl (e.g. methyl, ethyl) ester of fatty acids made from a wide range of vegetable oils, animal fat and used cooking oil via the tranesterification process. Moreover, biodiesel has been used not only as an alternative for fossil diesel, but also as an additive for diesohol – a blending of ethanol with regular diesel [4,5].

Diesohol is considered to be a candidate alternative to diesel in order to decrease the use of conventional diesel. And ethanol has been included in Thailand's national strategic plans and policies for energy [6]. Ethanol is used as an alternative fuel, a fuel extender, an oxygenate and an octane enhancer. It is expected that the use of ethanol as a gasoline-blending component can reduce the amount of petroleum-fuel imports by up to 10–15%. Moreover, the use of ethanol can also increase farmer's incomes, because it can be produced domestically from many kinds of agricultural products such as sugar cane, molasses or cassava root. This success of ethanol–gasoline blending has led to

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interest in the use of oxygenated compounds as emissions-reducing additives in diesel fuel. Moreover, the use of ethanol, particularly biomass-derived ethanol, can result in significant savings in carbon dioxide emissions. This approach offers a "no regrets" policy that reduces potential future risks associated with climate change, and it has the added benefit of economic development.

Generally, ethanol can be blended with diesel with no engine modifications required [7]. However, a major drawback in ethanol–diesel fuel blends is that ethanol is immiscible in diesel over a wide range of temperatures and water content because of their difference in chemical structure and characteristics. These can result in fuel instability due to phase separation. Prevention of this separation can be accomplished in two ways: by adding an emulsifier, which acts to suspend small droplets of ethanol within the diesel fuel, or by adding a co-solvent, which acts as a bridging agent through molecular compatibility and bonding to produce a homogenous blend [8]. Emulsification usually requires heating and blending steps to generate the final blend, whereas co-solvents allow fuels to be "splash-blended", thus simplifying the blending process [9]. Additionally, the cetane number of the blend is low, making it difficult to burn by the compression ignition technology employed in diesel engines. As a result, a number of studies have been carried out to improve the solubility of ethanol in diesel, as well as to improve the cetane number of the blends.

There are several studies concerning diesohol production and utilization. Recent studies in the US have revealed the use of additives from different manufacturers. Pure energy corporation (PEC) of New York was the first manufacturer to develop an additive package using a 2–5% dosage with 15% anhydrous ethanol, and proportionately less for 10% blends [10]. A small amount of commercially available cetane improver (<0.33% by volume) also was added to restore the cetane value of the blend. The second additive manufacturer was AAE Technologies of the United Kingdom, which has been testing 7.7% and 10% ethanol–diesel blends containing 1% and 1.25% AAE proprietary additive in different stages in the US [10]. The third manufacturer was GE Betz. They developed a proprietary additive derived purely from petroleum products; compared to the previous two, which are produced from renewable resources [10,11]. In Australia, Apace Research Ltd. [12,13] has announced the successful development of an emulsification technique using its innovative emulsifier. Their diesohol contained regular diesel fuel at 84.5 vol%, hydrate ethanol (5% water) at 15 vol%, and their emulsifier at 0.5 vol%. Engine tests for the diesohol were conducted by using a truck and a bus to compare it with regular diesel fuel. It was found that higher levels of alcohol in the diesohol maximizes the reductions in regulated exhaust emissions (HC, CO, NO_x, PM), and a reduction in net greenhouse gas emission is achieved by the use of diesohol.

Current use of diesohol in Thailand, however, only occurs on a small scale and on a *R* and *D* level. It is very difficult to determine the potential size of the diesohol market. In Thailand, there was a cooperation project between the Petroleum Authority of Thailand (PTT) Public Co. Ltd., Ford Motor Company, and the National Metal and Materials Technology Center (MTEC) to study the potential for using diesohol as an alternative to regular diesel fuel [14]. Ethanol at 10 vol%, regular diesel at 89 vol%, and an imported emulsifier (Beraid ED10 from Akzo Nobel) at 1 vol% were used for the diesohol preparation. Vehicle testing was done by using a minibus. It was observed that the fuel properties of the prepared diesohol were consistent with regular diesel, except that the flash point of the diesohol was lower.

It is evident that further studies are necessary in both diesohol production and utilization, especially in the area of domestic additive for diesohol. In particular, all automotive fuels are required to be a clear and single-phase liquid. Biodiesel was introduced in Thailand in 1971 and has been greatly discussed since 2001. The Ministry of Energy has set a target for biodiesel to replace diesel by 8.5 million liters a day by 2012, or 10% of the total diesel consumption in the country. It has given a major boost for producing biodiesel from raw materials available in Thailand, especially palm oil. Taking into account the environmental considerations, the more attractive system is the production and usage of fuel containing biodiesel and ethanol, which can both be produced in Thailand.

The objective of this research, therefore, was focused on studying the use of biodiesel (palm oil methyl esters) as an additive in stabilizing ethanol in diesel blends. The phase stability of ethanol–biodiesel–diesel three-component systems at different temperatures and different component concentrations was investigated, along with some basic fuel properties such as cetane index, density, heat of combustion, flash point, pour point, and emissions; these properties were compared with those of diesel fuel. Additionally, the long term stability of the blends was investigated.

2. Materials and methods

The phase behavior and fuel properties of the diesel–biodiesel–ethanol three-component fuel system were studied by using diesel donated by Rayong Purifier Public Co., Ltd.; ethanol 95% and 99.5% obtained from the Royal Chitralada Projects; ethanol 99.9% (J.T. Baker) and biodiesel (palm oil methyl ester) received from the Department of Naval Dockyards, Thailand.

Diesel, biodiesel and ethanol were mixed into a homogeneous mixture by a magnetic stirrer. Then, the final blend was kept in a glass vial with a screw cap for observing the physical appearance. The same procedure was carried out with other ratios of diesel, biodiesel and ethanol. Each component was varied from 0% to 100% by volume in 10% increments. In this study, the phase behavior of the three-component systems was investigated by using phase diagram [8]. All of the blends from the previous step were kept

Table 1
Fuel properties of the diesel–biodiesel–ethanol (D–B–E) system at different ratios of diesel, biodiesel and ethanol

Sample no.	%D	%B	%E	Cetane index	Flash point (°C)	Pour point (°C)	Density (g/ml)	Heat of combustion (MJ/kg)
1	90	10	0	47.99	71	6	0.8388	44.7
2	90	5	5	47.31	17.5	3	0.8313	44.5
3	90	0	10	46.05	14.5	3	0.8268	43.4
4	85	15	0	48.52	73.5	6	0.8417	44.2
5	85	10	5	47.7	14.0	3	0.8334	43.7
6	85	5	10	46.67	13.5	3	0.8313	43.6
7	85	0	15	45.81	13.0	3	0.8247	42.5
8	80	15	5	48.66	16.0	3	0.8375	43.3
9	80	10	10	46.85	15.0	3	0.8331	43.5
10	80	5	15	46.25	13.0	3	0.829	42.8
11	100	0	0	47.64	69.0	6	0.8354	45.0
12	0	100	0	55.4	122	9	0.8786	39.6
13	0	0	100	5–8	13	−117.3	0.7940	27.0

motionless for seven days at 10, 20, 30 and 40 °C to observe the physical stabilities. After seven days, the observation data were used to develop the phase diagrams at different temperatures. The phase diagram depicts the physical appearances at any ratio of the diesel–biodiesel–ethanol components by using symbols to describe the characteristics of the blends. Moreover, all samples were kept motionless further at room temperature for three months to observe the long term stability.

To study the effect of the three-component ratios on the fuel properties, ten blends of diesel with ethanol and biodiesel were used. They were obtained by mixing diesel, ethanol and biodiesel by volume in the proportions shown in Table 1. Laboratory tests were then carried out using ASTM tests standards to determine the following properties: cetane index, density, heat of combustion, flash point and pour point. All of these properties were tested by following ASTM D976, D4052, D240, D56 and D97, respectively.

Finally, the fuel blends with component ratios, as shown in Table 1, were used to investigate the emissions. The engine used in the study was a diesel generator, model DG3LE. It is a commercial single-cylinder, vertical, 4-stroke, air-cooled, direct injection diesel engine. The engine was coupled to an electrical generator through which load was applied by increasing the current to supply an electrical apparatus that was used to adjust the load. The load condition in this study was divided into four steps, which are 0%, 30%, 60% and 100% load (0 A, 3 A, 6 A and 10 A, respectively). A Motorscan Eurogas 8020 emissions analyzer was used to measure the concentration of carbon monoxide (CO), unburned hydrocarbon (HC), and nitrogen oxide (NO_x) of the exhausted gas at different loads.

3. Results and discussion

3.1. Effect of ethanol concentration on phase stability

The effect of ethanol concentration on phase stability was studied by using three purities of ethanol (95%, 99.5%, and 99.9%) at room temperature. And the results are represented in the three phase diagrams of diesel, biodiesel, and ethanol components.

The phase behavior of the diesel–biodiesel and 95% ethanol system at room temperature is shown in Fig. 1A. In case of the 95% ethanol, the diesel and the ethanol with 95% purity were insoluble because 95% ethanol, also called hydrous ethanol, has 5% water in its mixture. Due to the high polarity of water, this amount of water enhances the polar part in an ethanol molecule. Consequently, diesel, which is a non-polar molecule cannot be compatible with ethanol 95% purity. Solubility of the diesel–biodiesel pair is not limited, as well as the biodiesel–95% ethanol pair. In this case, it seemed that adding biodiesel cannot improve the intersolubility of diesel and 95% ethanol because after seven days the mixtures of the three-components had split into two phases; polar and non-polar. The reason might be that the water content in 95% ethanol has a stronger effect than biodiesel, resulting in poor emulsion stability. Therefore, 95% ethanol may not be suitable for diesohol production.

In the case of 99.5% ethanol, the intersolubility of the three-components was not limited. They could be mixed into a homogeneous solution at any ratio, as shown in Fig. 1B. Because 99.5% ethanol has lower water content than that of 95% ethanol, it is more soluble in diesel fuel than 95% ethanol. However, after three months, some ratios of the mixtures between diesel and 99.5% ethanol (60% Diesel, 40% Ethanol; 50% Diesel, 50% Ethanol; and, 40% Diesel, 60% Ethanol) became two phases, but the mixtures having biodiesel as an additive was still liquid one phase. This homogeneity was due to the fact that biodiesel can act as an amphiphile (a surface-active agent) and form micelles that have non-polar tails and polar heads. These molecules are attracted to liquid/liquid interfacial films and to each other. These micelles acted as polar or non-polar solutes, depending on the orientation of the biodiesel molecules. When the diesel fuel was in the continuous phase, the polar head in a biodiesel molecule oriented itself to the ethanol, and the non-polar tail was oriented to the diesel. This phenomenon held the micelles in a thermodynamically stable state, depending on the

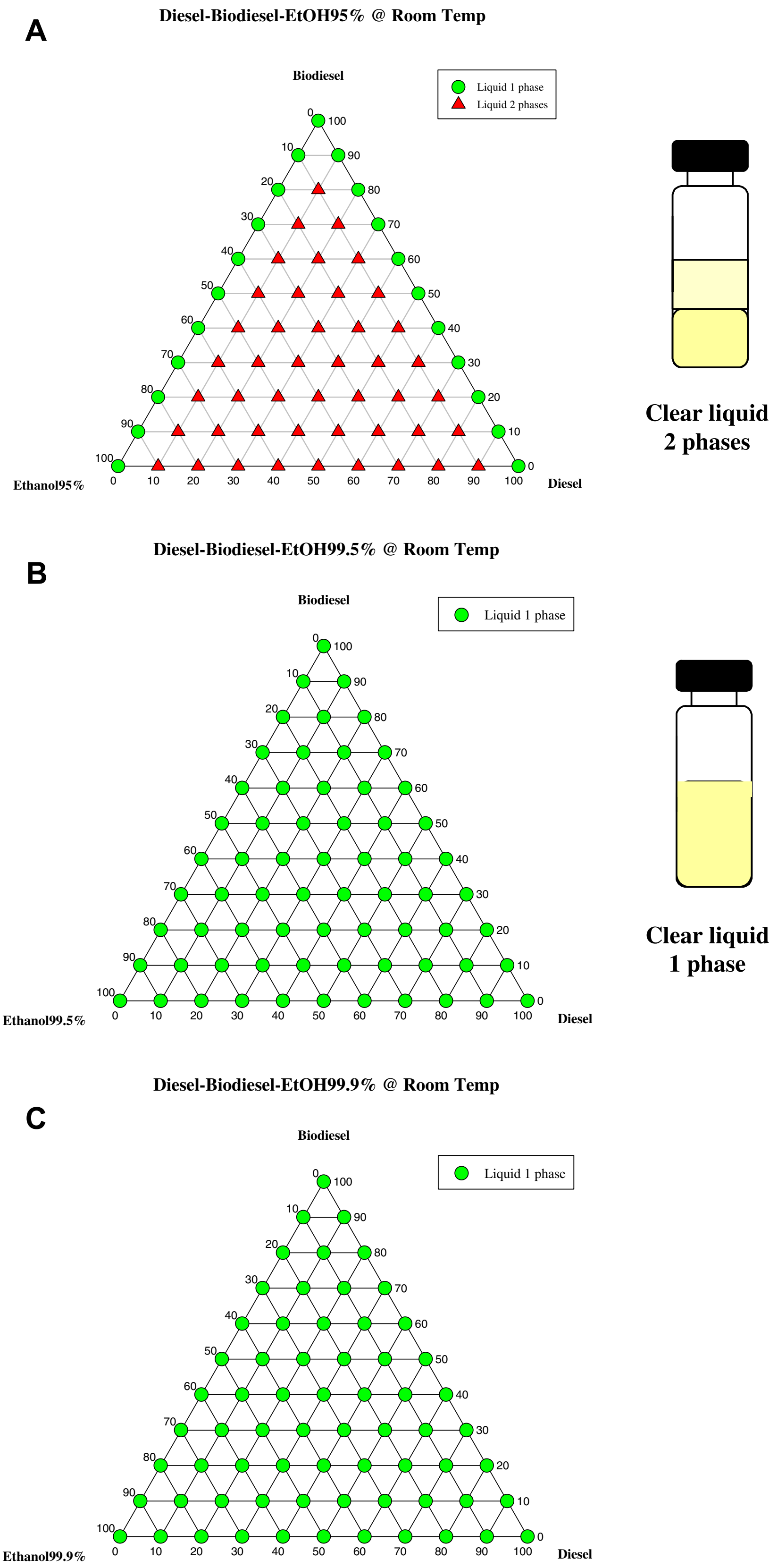

Fig. 1. Phase behavior of diesel–biodiesel–ethanol system at room temperature using ethanol at different concentrations: (A) 95%, (B) 99.5%, (C) 99.9%.

component concentrations and other physical parameters [4].

The same result was shown for the 99.9% ethanol as for the 99.5% ethanol. As shown in Fig. 1C, 99.9% ethanol and diesel could be mixed to a homogeneous solution at any ratio. However, since 99.5% ethanol is much cheaper than 99.9% ethanol and is produced in our country, it was therefore chosen to blend with diesel and biodiesel in order to study further fuel properties.

3.2. Effect of temperature on phase stability

In Fig. 2A, at the temperature of 10 °C, the tendency can be observed, where the mixtures of diesel and ethanol in the range of 20–80% by volume were in clear liquid and crystalline phases. The mixtures of biodiesel and ethanol form a true solution, which can be easily blended. Mixtures containing biodiesel from 70% to 100% without ethanol become a gel. This might be the effect of fatty acid in the biodiesel component. And the other ratios were liquid crystalline 1 phase in which crystallization occurs in the diesel and ethanol phases.

As shown in Fig. 2B, at 20 °C, almost all the blends were liquid 1 phase except the ratios of ethanol from 30% to 70% with diesel. These ratios were in liquid 2 phases in which they are totally immiscible and remain as two distinct

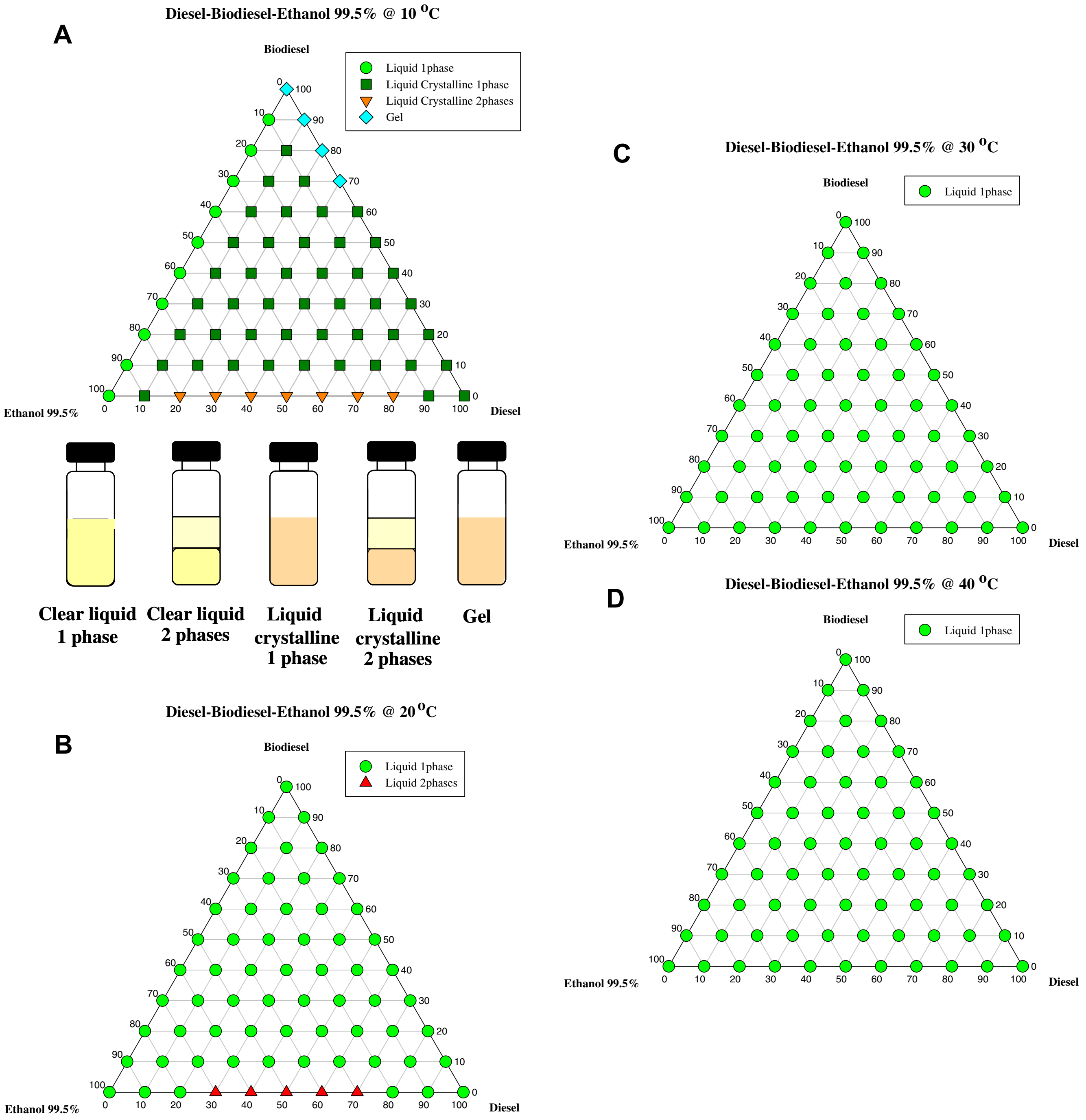

Fig. 2. Phase behavior of diesel–biodiesel–ethanol 99.5% (D–B–E 99.5%) system at different temperatures: (A) 10 °C, (B) 20 °C, (C) 30 °C, (D) 40 °C.

separate phases. Therefore, at 20 °C, fuel ethanol was completely miscible in diesel, when the diesel concentration was lower than 30% or higher than 70%. At 30 °C, all the blends were liquid 1 phase, as shown in Fig. 2C. Fuel ethanol could be mixed with diesel at any ratio. Therefore, at this temperature, there is no problem of phase separation.

Fig. 2D shows the phase behavior of the diesel–biodiesel–99.5% ethanol system at 40 °C. The result was similar to that of 30 °C, proving that diesohol blends will be stable as a liquid single phase fuel at rather high ambient temperatures (30–40 °C). Consequently, due to the normal temperature in Thailand, diesohol emulsions can be used as a liquid fuel without the problem of phase separation.

3.3. Fuel properties testing

Table 1 shows the fuel properties of the blends at different ratios of diesel, biodiesel and ethanol. It can be observed that the density of the blends decreased with an increasing of the percentage of ethanol in the blends. This is attributed to the fact that ethanol has lower density and as such will lower the density of the mixture. But, when the percentage of biodiesel was increased, the density increased, which is due to the fact that the palm oil biodiesel has a higher density than the other two components. Normally, it is recognized that higher density leads to higher flow resistance of fuel oil, resulting in higher viscosity. This finding suggests that the higher viscosity can lead to inferior fuel injection. However, all the blends had density values that were acceptable for the standard limit for high-speed diesel. These results show the same tendency as those of earlier works [5,7].

The cetane index is also in proportion to density value. It was observed that the cetane index of the diesohol mixture decreased, when increasing amounts of ethanol because ethanol itself has very low cetane, approximately 5–8. The lower the cetane index is, the poorer the ignition property will be. Cetane index also has an effect on the engine start up, combustion control, and engine performance. However, biodiesel, due to its high cetane value, could improve this property. Some of the fuel blends had a cetane index higher than base diesel. In this result, the sample consisting of 80% diesel, 15% biodiesel and 5% ethanol had the highest cetane index.

Heat of combustion is one of the most important fuel properties. The result showed that the heat of combustion of diesohol decreased, when greater amounts of ethanol and biodiesel were added, which is due to the lower heating value of biodiesel and ethanol. These results have the same trend as those reported earlier by Fernando and Hanna [4], Cheenkachorn et al. [5], and Ajav and Akingbehin [7]. Lower heating value of a fuel has a direct influence on the power output of an engine. However, it seems that the heating value of the blends containing ethanol lower than 10% were not much different from conventional diesel.

All of the blends, except samples containing 90% diesel and 10% biodiesel, and 85% diesel and 15% biodiesel, were found to have the same pour point at 3 °C, while the samples, which contained only diesel and biodiesel, without ethanol, had the same pour point as that of base diesel. The reason is that ethanol has a very low pour point and biodiesel normally has a pour point higher than conventional diesel. But all of the blends have diesel as a major component, and, therefore, the pour points of the fuel blends were found to be not much different from conventional diesel.

The flash point is the lowest temperature at which a fuel will ignite, when exposed to an ignition source. In this study, the flashpoint of diesohol was substantially different from diesel and was found to be extremely low, in the range of 12–17 °C. All of the blends containing ethanol were highly flammable with a flash point temperature that was below the ambient temperature. The flash point of the fuel affects the shipping and storage classification of fuels and the precautions that should be used in handling and transporting the fuel. In general, flash point measurements are typically dominated by the fuel component in the blend with the lowest flash point. The flash point of a diesohol mixture is mainly dominated by ethanol. As a result, the storage, handling and transportation of diesohol must be managed of in a special and proper way, in order to avoid an explosion. These facts were also discussed in earlier research [4,5,7,15].

3.4. Emissions testing

The variations of CO emission with respect to fuels at various loads are presented in Fig. 3A–C. The proportion of diesel was fixed at 90%, 85% and 80% by volume, respectively. As shown in these Figures, at low and medium loads (0%, 30% and 60% load), CO emissions of the blends were not much different from that of conventional diesel. However, at full load (100% load), CO emissions of the blends decreased significantly, when compared with those of conventional diesel. This can be explained by the enrichment of oxygen owing to the ethanol and biodiesel addition, in which an increase in the proportion of oxygen will promote the further oxidation of CO during the engine exhaust process. The result showed that the blend of 80% diesel, 15% biodiesel and 5% ethanol produced the smallest amount of CO (~0.6–0.8 vol%) at full engine load. The addition of oxygenates into the diesel fuel resulted in only a slight effect on CO emissions at low and medium load, but significantly reduced CO emissions at high or full load. It should be noted that the impact of diesel–biodiesel–ethanol on CO emissions varies with engine operating conditions and was not conclusive. Some studies reported the reduction of CO emissions by using ethanol–diesel blends and our result is comparable to the previous work reported by De-gang et al. [15]; however, opposite results were also reported [16].

Fig. 4A–C illustrate that samples containing diesel at 90%, 85% and 80% by volume, respectively, show the same

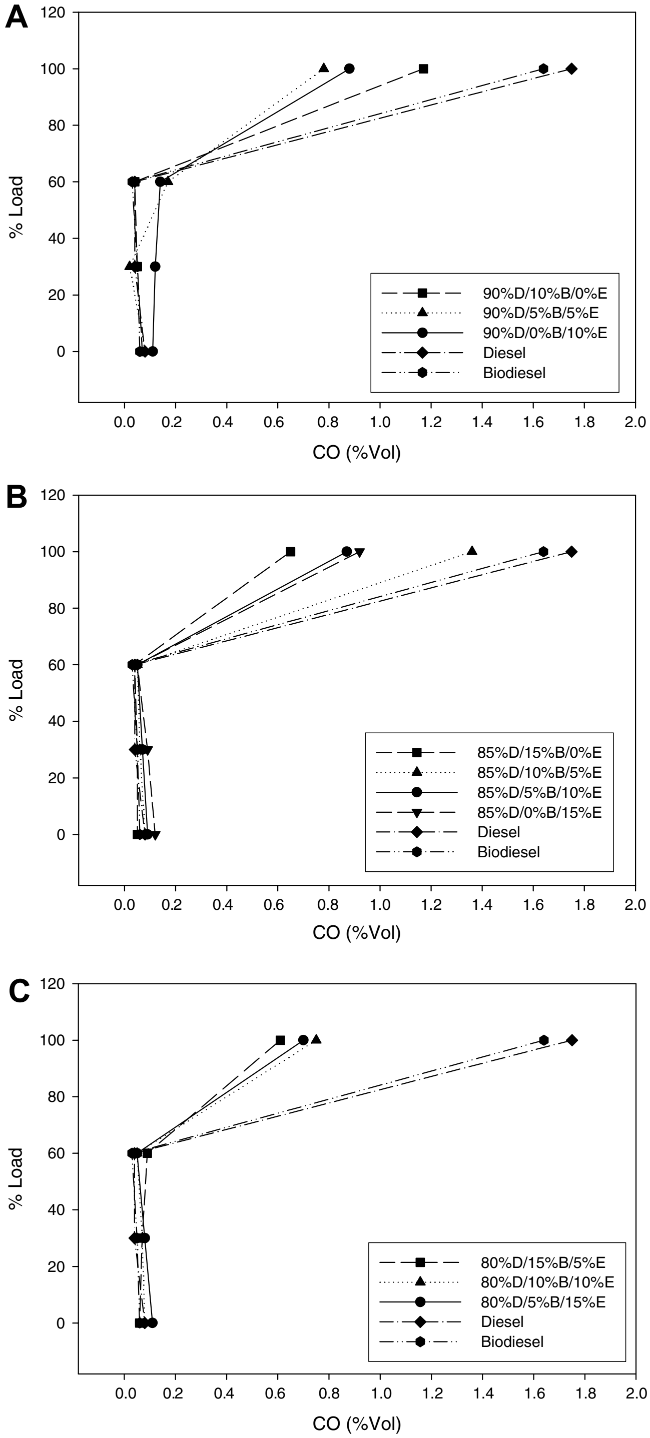

Fig. 3. CO emissions for diesel–biodiesel–ethanol (D–B–E) system under various operating conditions.

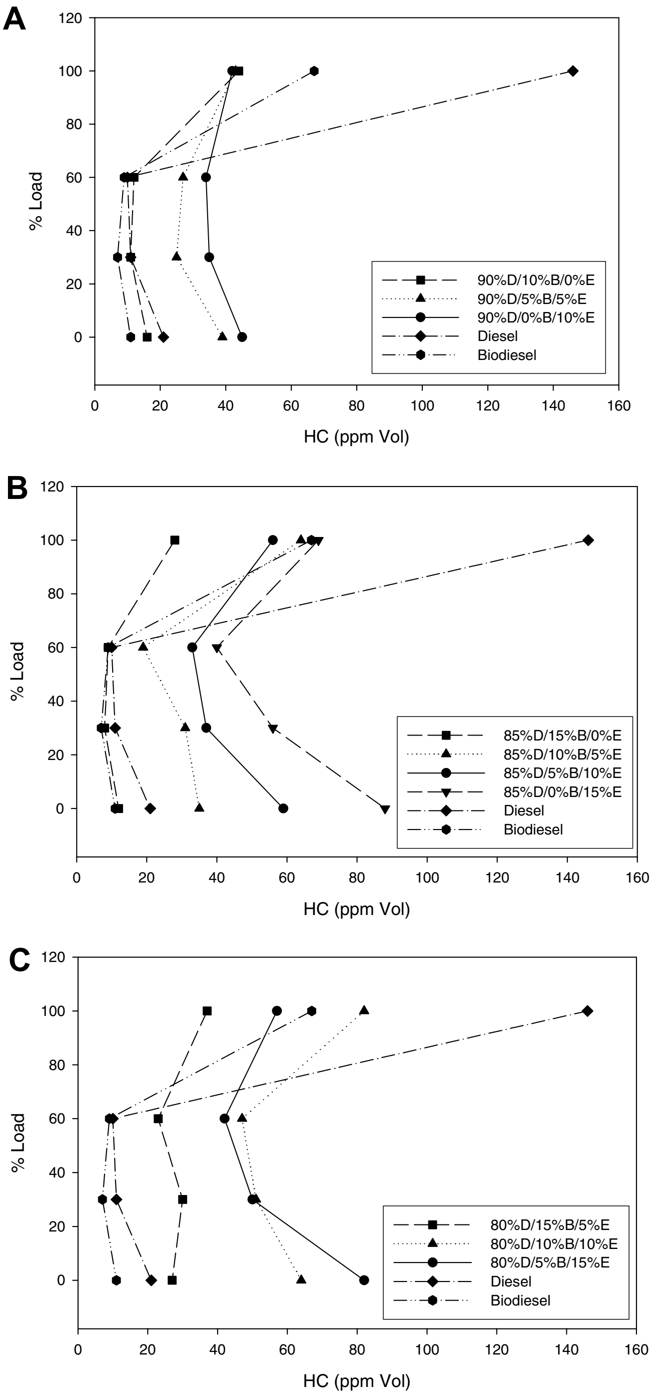

Fig. 4. HC emissions for diesel–biodiesel–ethanol (D–B–E) system under various operating conditions.

tendency as at low and medium load; the blends that contain a higher percentage of ethanol will have higher HC emission. On the other hand, it was also observed that the blends containing a higher percentage of biodiesel will have lower HC emission. This indicates that the presence of ethanol might be the essential factor for the increase of HC emissions. High HC emission means that there is some unburned ethanol emitted in the exhaust due to the larger ethanol dispersion region in the combustion chamber. Biodiesel has a higher cetane number than conventional diesel, resulting in more complete combustion in the cylinder. At medium load, diesel, biodiesel and fuel blends had lower HC emissions than those emitted from lower or higher loads. The reason is that, normally, better combustion can be achieved at a medium speed and with a medium-sized load. However, at full load, diesel had the highest HC emission. Thus, at this condition the fuel blends had less HC emissions than conventional diesel fuel. In this study, the blend of 85% diesel and 15% biodiesel had the lowest HC emission at the full load condition. Lower HC emission means that the fuel has higher combustion efficiency, resulting in higher energy output and a cleaner combustion chamber.

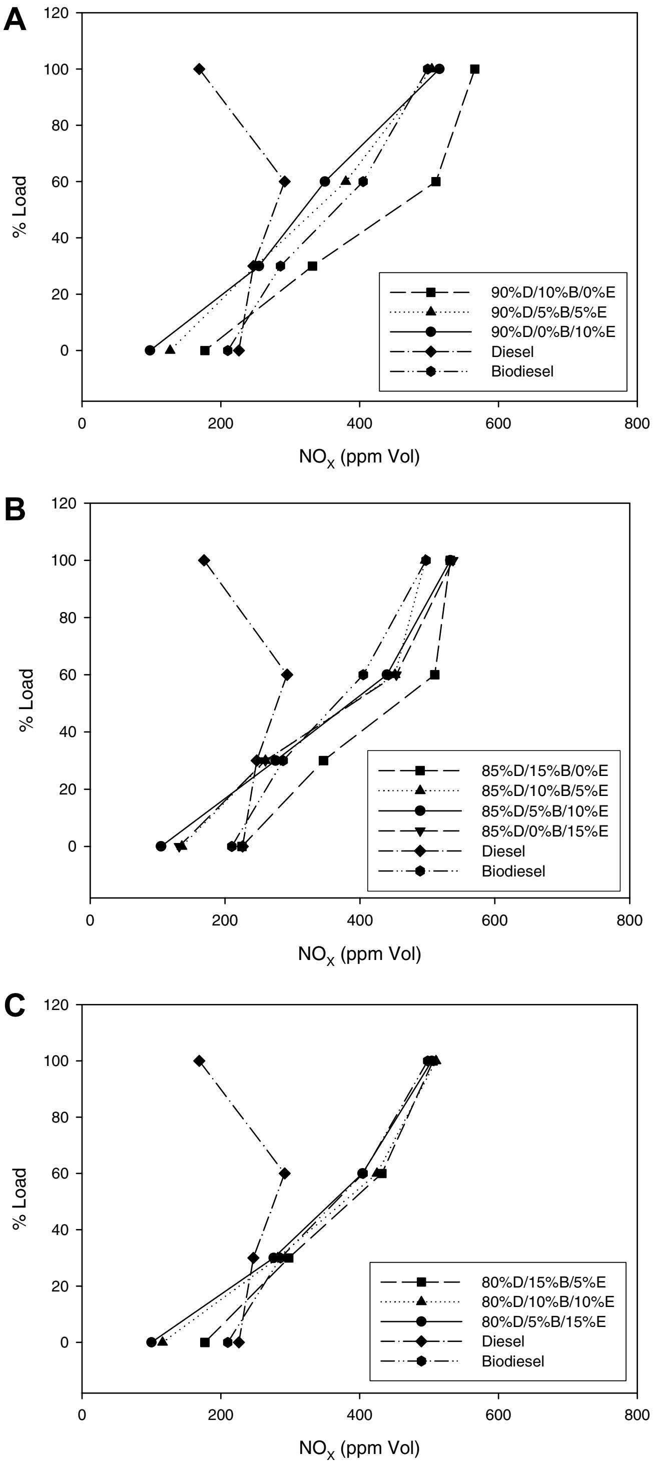

Fig. 5. NO_x emissions for diesel–biodiesel–ethanol (D–B–E) system under various operating conditions.

The NO_x emissions of the engine using different blended fuels under various engine loads are shown in Fig. 5A–C. At every diesel proportion (90%, 85% and 80% by volume), it can be observed that all fuel blends and biodiesel reduced the NO_x emissions at the no-load condition. However, at low, medium and high loads, NO_x emissions are higher for the blended fuels than that of neat diesel. Especially at full load, all fuel blends increased NO_x significantly relative to diesel fuel. The cause of the increased NO_x emissions for biodiesel is unknown; however, a number of fuel properties have been shown to effect the emission of NO_x. Normally, if we can create a more complete combustion, we can get a higher combustion temperature, which will cause high NO_x formation. Therefore, adding biodiesel and ethanol to diesel as oxygenates can enhance combustion efficiency of the fuel. Another reason for the increase of NO_x emissions is the decrease of the cetane number with the addition of ethanol. It is well known that the cetane number and fuel aromatic content are known to influence NO_x and PM emissions from diesel engines. A lower cetane number means an increase in the ignition delay and more accumulated fuel/air mixture, which causes a rapid heat release in the beginning of the combustion, resulting in high temperature and high NO_x formation [2]. The cetane number of the fuel was reduced with the increase of ethanol content in the fuel because of the low cetane number of the ethanol. It should be noted that biodiesel increased NO_x, but had a substantially higher cetane number than diesel fuel. The NO_x behavior of biodiesel blended fuels is complex and is not conclusive. Many studies indicate that oxygenate fuel blends can cause an increase in NO_x emissions. However, some studies also found no NO_x increase or even a decrease in NO_x, as reported by Lee et al. [17]. In the case of ethanol–diesel blended fuels, Ajav et al. [7] and Caro et al. [18] reported a significant benefit in terms of NO_x emissions; however, Ozer et al. (2004) [19] expressed opposite results.

4. Conclusions

The stability and fuel properties of diesohol blend were investigated in order to evaluate the potential for using biodiesel as an effective agent for diesohol and making diesohol an alternative fuel for diesel engines. The conclusions of this study are as follows:

1. Palm oil derived biodiesel could be used as an effective additive for diesohol emulsions. For diesohol production, anhydrous ethanol should be used.
2. Intersolubility of the components of diesel–biodiesel–ethanol system decreased with decreasing temperature. However, at temperatures above 20 °C there was no problem with phase separation.
3. Density and pour point of all the blends were under the standard limits for diesel fuel.
4. The high cetane value of biodiesel could compensate for the decreased cetane caused by the presence of ethanol in diesel fuel.

5. Heat of combustion of all the blends was found to be lower than that of diesel fuel alone. However, the heating value of the blends containing ethanol lower than 10% were not much different from that of conventional diesel.

In general, the diesohol blends containing 5% ethanol had very close fuel properties compared to diesel fuel. In this study, the blend of 90% diesel, 5% biodiesel and 5% ethanol had a heating value very close to that of diesel fuel. And the blend of 80% diesel, 15% biodiesel and 5% ethanol had the highest cetane index. The physical characteristics of diesohol make it technically impossible to meet the diesel fuel standard and the difference from the diesel standard does not compromise the operational integrity of an engine tuned to run on conventional diesel. Additional specifications are required for fuel properties that are unique to diesohol/fuel grade ethanol, to ensure the fuel is fit-for-purpose.

As for the emissions of the blends, it was found that CO and HC were reduced significantly at high engine load, whereas NO_x increased, when compared to that of diesel. Taking these facts into account, a blend ratio of 80% diesel, 15% biodiesel and 5% ethanol was the most suitable for diesohol production because of the acceptable fuel properties and the reduction of emissions.

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