

Impact of nanoparticles and butanol on properties and spray characteristics of waste cooking oil biodiesel and pure rapeseed oil

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Abstract

Renewable biofuels can offset greenhouse gases by replacing fossil fuels destined for internal combustion engines. However, biofuels have their own setbacks and may lead to poor combustion inside the engine cylinder. In this study, nanoparticles and butanol were blended either separately or together with waste cooking oil biodiesel and neat rape seed oil to investigate the impact of these additives on the properties and spray characteristics. The investigation comprised of three stages, with each having an effect on how the next stage of the investigation was conducted. Initially, the physicochemical characteristics of 25ppm, 50ppm, 75ppm and 100ppm concentrations of aluminium oxide and copper oxide nanoparticle blends with fossil diesel, waste cooking oil biodiesel and rapeseed oil were investigated. The results from first stage investigation showed that, in general, blends containing aluminium oxide nanoparticles gave better results for almost all the concentrations when compared with copper oxide nanoparticle blends with the same nanoparticle concentrations. Overall, waste cooking oil biodiesel blended with 100ppm aluminium oxide nanoparticle showed most promising results like the flash point of 159.3°C, kinematic viscosity @40°C of 4.66 cSt, and gross calorific value of 44.43 MJ/kg. These values were 61.6% higher, 51.3% higher and 3.2% lower than that of corresponding fossil diesel values. Subsequently, in the second stage of the study, the addition of butanol was investigated to assess its ability to enhance the emulsion of biofuel-nanoparticles blends. Four blends containing 90% biodiesel & 10% butanol, and 90% rapeseed oil & 10% butanol, with and without 100ppm Al₂O₃ were prepared. Results showed that the kinematic viscosity of the fuel blends containing 100ppm aluminium oxide nanoparticles were decreased by 0.4% and 3.3%, for 90% biodiesel & 10% butanol and 90% rapeseed oil & 10% butanol blends respectively, when compared to without the nanoparticles. The results obtained from the second stage of investigation proved that butanol acted as a surfactant and thus addition of butanol helped to improve the properties of the biofuel-nanoparticle blends. In the third stage of the study, the spray characteristics of fossil diesel, biodiesel, biodiesel + 100ppm aluminium oxide nanoparticles, rapeseed oil, rapeseed oil + 100ppm aluminium oxide nanoparticles, 90% biodiesel & 10% butanol, 90% biodiesel & 10% butanol + 100ppm aluminium oxide nanoparticles, 90% rapeseed oil & 10% butanol and 90% rapeseed oil & 10% butanol + 100ppm aluminium oxide nanoparticles were investigated. It was found that amongst all fuels, blend containing 90% biodiesel + 10% butanol + 100ppm aluminium oxide nanoparticles gave better spray characteristics; for example, the liquid sheet angle was 7.14% lower and the spray cone angle was 7.87% higher than the corresponding fossil diesel values. The study concluded that the spray characteristics and properties of biofuels could be improved by blending with both aluminium oxide nanoparticles and butanol.

Keywords: *Nanoparticle, Butanol, Biofuel, Properties, Spray characteristics, CI Engine*

INTRODUCTION

The increase in the energy demand, global warming, limited reserve of fossil fuels and refinery capacity have alarmed the world to harvest into green methods of producing energy. Scientists and researchers have been trying to find ways to reducing harmful emissions of internal combustion (IC) engines by modifying them, using alternative fuels, blending different fuels together and using additives [1]. Biofuels can be used in compression ignition (CI) engines and they may be produced domestically. However, they have their own setbacks as being highly viscous, less stable and incompatible with the existing fuel supply systems. One approach to improve the biofuels properties is by addition of nanoparticles; but currently there are concerns if the nanoparticles does not burn inside the engine cylinder and their effect on the exhaust emission gases, fuel pump, fuel filter and fuel injection characteristics [2]. Nanoparticles have a higher surface area to volume ratio, which acts as a highly reactive catalysts [3]. Aluminium oxide nanoparticles have the ability to donate their oxygen atoms from its lattice structure to the fuel which enhances combustion characteristics [4]. Lower NO_x and soot emission can be achieved by addition of nanoparticles [2, 5]; Samuel and Shefeek [6] investigated the effects of cerium oxide nanoparticles addition in fossil diesel (FD) on performance and emission characteristics of a four cylinder CI engine. An ultrasonic shaker was used to mix the nanoparticles with FD. Four blends of FD were made with 10ppm, 20ppm, 30ppm and 40ppm of nanoparticles. It was observed that the specific fuel consumption was decreased as cerium oxide nanoparticles acted as an oxygen donating catalyst which enhanced combustion characteristics. CO and nitrogen gases reduced from 2% to 1.4% and from 83% to 81.6% respectively, with 40ppm cerium oxide nanoparticles blend when compared to neat FD [6]. Gumus et al. [7] assessed the impacts of aluminium oxide nanoparticles (Al₂O₃) and copper oxide nanoparticles (CuO) on FD; they investigated the physicochemical properties, stability of nanoparticles in the FD with the use of various surfactants, and CI engine performance & emissions. Nanoparticles blends of 50ppm with FD for both nanoparticles were created using an ultrasonic mixer. The emulsion of nanoparticles in diesel was tested using two different surfactants (Sodium Silicate and Darvan-C). They reported that 2% Darvan-C showed the best emulsion layer [7]. Santhanamuthu et al. [8] investigated the performance and exhaust emissions of a CI engine with blends of polanga seed oil, FD and iron oxide nanoparticles. They observed best optimum engine performance with 100ppm iron oxide nanoparticles. D'silva et al. [9] added titanium oxide nanoparticles (TiO₂) in FD to investigate stable dispersion, engine performance and emission characteristics. They reported that the flash point, fire point, kinematic viscosity, density and calorific value were increased with the addition of TiO₂ by 41.17%, 40.74%, 6.17%, 0.43% and 0.59%

respectively [9]. About 21.28% reduction in brake specific fuel consumption (bsfc) was observed at peak load when TiO_2 were added. Other studies also reported improvement in fuels properties when blended with nanoparticles [10-12]. However, in some cases decrease in the calorific value was also reported [6]. Kinematic viscosity increased with the addition of nanoparticles, but the use of surfactants allowed the nanoparticles to emulsify in the base fuel properly, hence decrease in the viscosity was also reported [7]. Spray characteristics of neat biofuels were investigated [13-14]. Wang et al. [13] investigated the spray characteristics of biodiesels produced from waste cooking oil, palm oil and FD at various injection pressures. Biodiesels gave longer spray tip penetration with injection delays, smaller projected area, less volume and smaller spray angle than FD. With increased injection pressure, the difference between the sauter mean diameter (SMD) of biodiesel and FD was decreased. Butanol was found to be a suitable additive for biofuels for improved engine performance and reduced emission gases [15-16]. In this study, the effect of butanol addition on physicochemical and spray characteristics of biofuel-nanoparticles blends will be investigated. The study will be conducted in three stages: (i) two nanoparticles (CuO and Al_2O_3) will be used to blend separately with FD, waste cooking oil biodiesel (B100) and neat rapeseed oil (RSO100); physicochemical properties will be measured and compared to find the optimum nanoparticles blends (ii) butanol will be added into the optimum nanoparticles blends, and the effectiveness of butanol as surfactant will be assessed by comparing the properties of the butanol-nanoparticles-biofuel blends (iii) spray characteristics of the fuel blends will be measured and analysed. Finally, properties and spray characteristics will be compared to find the optimum blends for CI engine testing.

MATERIALS AND METHODS

Waste cooking oil biodiesel (B100) was acquired from a local company, and rape seed oil (RSO100) was acquired from supermarket. The nanoparticles were purchased from Sigma-Aldrich. Butanol (1 Butane) was purchased from Fisher Scientific Ltd. The study was carried out in three stages: (i) Firstly, to investigate the blends of FD, B100 (from WCO) and RSO100 with Al_2O_3 and CuO nanoparticles to find out which of the two nanoparticles was more feasible to be used and also in what concentration, (ii) Secondly, to investigate if the Butanol could be used as an emulsifier when nanoparticles were mixed with the fuel, and (iii) Finally, to investigate the effects of nanoparticles and butanol on macroscopic spray characteristics of the fuel blends. Parr 6100 Bomb Calorimeter (ASTM-D240), Setaflash series 3 plus closed cup flash point tester (ASTM-D3278), and Cannon Fenski u-tube viscosity meter (ASTM-D130) were used to measure the heating

value, flash point temperature and kinematic viscosities of the fuel blends. Kinematic viscosity was measured at @40°C using a constant temperature water bath. The density was measured using hydrometer and volume-weight method. Fuel blends samples for all the three base fuels (FD, B100 and RSO100) with 25ppm, 50ppm, 75ppm and 100ppm concentrations of both nanoparticles (Al_2O_3 and CuO) by mass were prepared. Physicochemical characteristics were measured. The results were analysed and compared, improved nanoparticles blends were taken for phase 2 study, where butanol was introduced – biofuel and butanol was mixed together and after that 100ppm the nanoparticle was added in the blend. The blends were shaken for 15 minutes so the nanoparticles could be dispersed into the blends properly. Macroscopic spray characteristics of fuel injection (liquid sheet angle and spray cone angle) were measured using optical and mechanical methods. Sealey VS2058 diesel injector nozzle POP tester and Lister Petter fuel injector (P751-62090) manufacture by Delphi were used (Figure 1). The maximum pressure of the POP tester was 600 bar and the opening pressure was 125bar. The nozzle had a single hole through which the fuel was sprayed as a full cone (Figure 1). Liquid sheet angle is defined as the angle of the fuel as it comes out of the nozzle (Figure 2), before it turns into spray particles. An optical technique was used to measure the liquid sheet angle. Nikon D3320 camera was used to take the photographs of the fuel. All the photographs were taken from a specific point marked on the ground for accuracy. The spray cone angle is the angle at which the fuels starts dispersing from liquid into tiny molecular particles when injected from an injection nozzle. A mechanical technique of using a 445×570 blotting (absorbent) paper at a distance of 302mm below the nozzle was used to find the indentation diameter on the blotting paper. The distance was selected in such a way so the fuel was allowed to form into tiny molecular



(a) Delphi injector



(b) POP tester

Figure 1 - The injector nozzle and POP tester setup

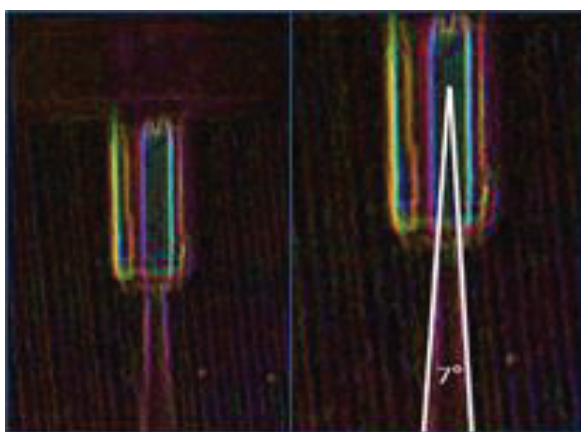


Figure 2 - Left: The edges sharpened of the magnified image, and Right: manually addition of lines on the edges to measure the liquid sheet angle, these images are for FD

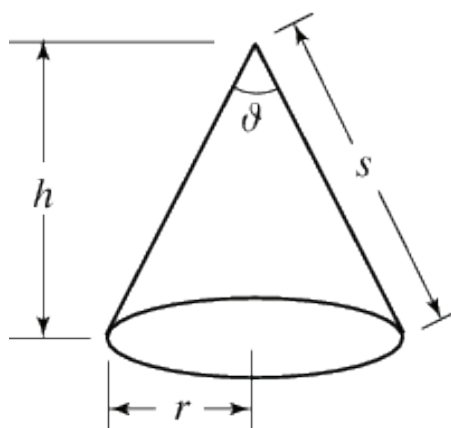


Figure 3 - Spray cone angle parameters [17]

particles. Platform was manually constructed so the blotting paper was as horizontal as possible, ensuring that indentation of the spray was accurate. The test was performed 5 times and the average value was taken. For each of the blends, the

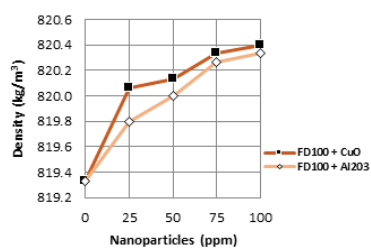
formula below was used to find the spray cone angle. Figure 3 shows the parameters of a cone used to estimate the spray cone angle using following equation:

$$\text{Spray cone angle } (^{\circ}) = 2 \times \tan^{-1} \left(\frac{r}{h} \right)$$

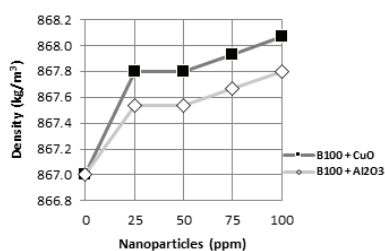
Where r is the radius (of the indentation) and h is the height (distance from the nozzle tip to the blotting paper).

RESULTS AND DISCUSSION

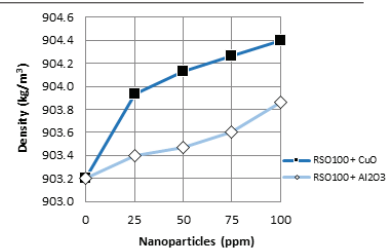
Figure 4 illustrate the physicochemical properties of the neat fuels and blends when CuO and Al₂O₃ was added. Figures 4(a-c) shows comparison of the density [@15°C] of nanoparticles blends with respect to the neat FD, B100 and RSO100 fuels. The density of CuO and Al₂O₃ are $6.4 \left(\frac{\text{g}}{\text{m}^3} \right)$ [18] and $3.7 \left(\frac{\text{g}}{\text{m}^3} \right)$ [19]; hence, density increase due to CuO addition was higher than Al₂O₃. In general, density of all blends were increased in the range of 0.07% to 0.13% as the nanoparticles concentration was increased from 25ppm to 100ppm. The flash point of neat RSO100 fuel was maximum; flash point for blends of Al₂O₃ and CuO with FD increased as the concentration of the nanoparticles increased up to 75ppm. The flash point temperature stayed constant from 75ppm to 100ppm (Figure 4(d)). Almost similar trend was also observed for B100 fuel (Figure 4(e)). In the case of RSO100 and blends, for Al₂O₃ nanoparticle, the flash point stayed constant up to 75ppm and then increased by 0.19% when the concentration was increased to 100ppm. However, for CuO, the trend was not clear possibly due to the non-homogeneous mixing of nanoparticles (Figure 4(f)).



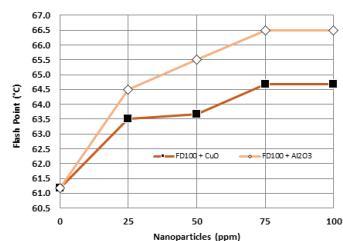
(a) Density of Fossil Diesel & blends



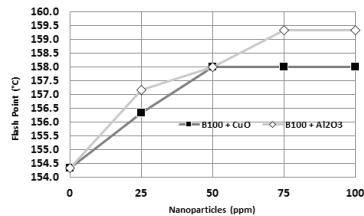
(b) Density of B100 & blends



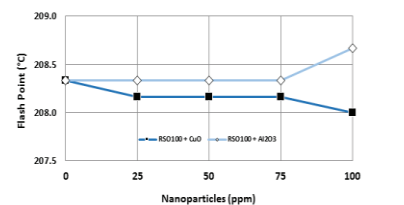
(c) Density of RSO100 & blends



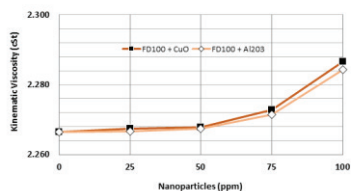
(d) Flash point of Fossil Diesel & blends



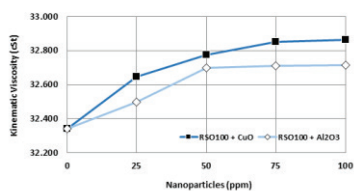
(e) Flash point of B100 & blends



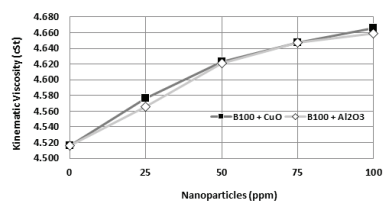
(f) Flash point of RSO100 and blends



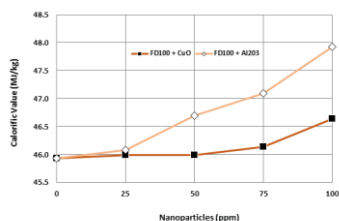
(g) Fossil Diesel & blends @40°C



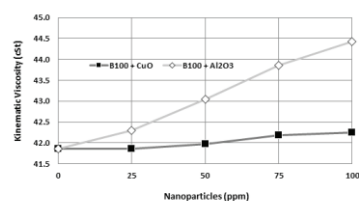
(h) B100 & blends @40°C



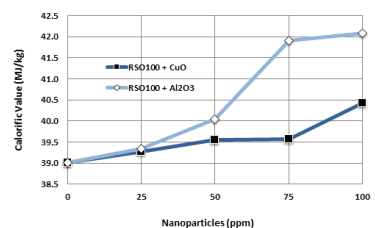
(i) RSO100 & blends @40°C



(j) HHV of Fossil Diesel & blends



(k) HHV of B100 & blends



(l) HHV of RSO100 & blends

Figure 4 – Physicochemical properties of Al₂O₃ and CuO nanoparticle blends

For all nanoparticle blends of FD, B100 and RSO100, the kinematic viscosity increased with increased concentration of nanoparticles (Figures 4(g-i)). CuO caused higher kinematic viscosity in comparison to Al_2O_3 in all the cases. It was believed that this was due to the higher density of the CuO than Al_2O_3 blends. In addition, nanoparticles increases the surface tension in the fuel blends [20], with increased concentration of nanoparticles, the surface tension would also increase and hence the viscosity. The calorific value increased with increased concentration of nanoparticles (Figure 4(j-l)). For all blends and for same concentration of nanoparticles, Al_2O_3 gave higher HHV than CuO. The increase in HHV of RSO100 when 100ppm Al_2O_3 was added was 7.3%; whereas, these values were 5.8% and 4.1% for B100 and FD respectively (Figure 4(j-l)). Furthermore, it was found that calorific value of B100 with 100ppm Al_2O_3 is almost similar to that of neat FD. Higher oxygen content in Al_2O_3 caused higher HHV than CuO, as oxygen helps in combustion. On the basis of the above results, eight more blends were prepared using 100ppm Al_2O_3 nanoparticle and 10% Butanol : 90% B100 & 10% Bu (B90Bu10), 90% B100 & 10% Bu + 100ppm Al_2O_3 (B90Bu10 + 100ppm Al_2O_3), 90% RSO100 & 10% Bu (RSO00Bu10), 90% RSO100 & 10% Bu + 100ppm Al_2O_3 (RSO90Bu10 + 100ppm Al_2O_3), 80% B100 & 20% Bu (B80Bu20), 80% B100 & 20% Bu + 100ppm Al_2O_3 (B80Bu20 + 100ppm Al_2O_3), 80% RSO100 & 20% Bu (RSO80Bu20) and 80% RSO100 & 20% Bu + 100ppm Al_2O_3 (RSO80Bu20 + 100ppm Al_2O_3).

The emulsions of the nanoparticle blends were checked after 5 days of blend preparation and it could be seen that the Al_2O_3 particles had dispersed better with the use of butanol (Figure 5).

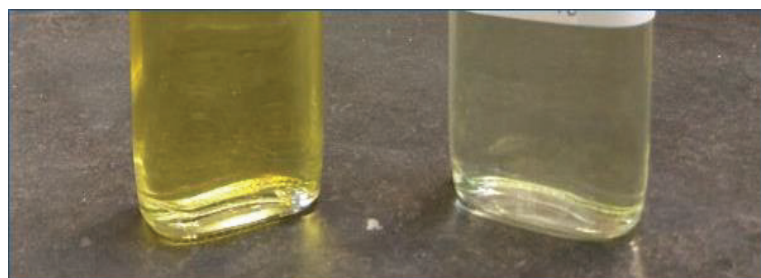


Figure 5 - Examples of a close up look of the blends after 5 days of Al_2O_3 nanoparticles addition

Figure 6 (a) shows the effect of butanol addition, for example, when 10% butanol was added to B100 (from B100 to B90Bu10) flash point was decreased by 58.1%.

The flash point of Butanol is 35°C [21] which caused this behaviour. However, this is still 5.4% higher than FD. Similar behaviour was observed with RS100, the flash point was decreased by 66.7% when 10% Bu was added (ie. from RSO100 to RSO90Bu10).

Flash point temperatures were increased when 100ppm Al₂O₃ was added. For example, flash point was increased by 3.4% for B100 fuel when 100ppm Al₂O₃ was added, and in the case of B90Bu10 fuel this was increased by 7.6%. The increase in flash point for RSO100 when 100ppm Al₂O₃ were added was 0.2% and the increase rate was 5.14% for RSO90Bu10 fuel when 100ppm Al₂O₃ was added. Furthermore, it was observed that percentage increase for B100 and RSO100 fuels when 100ppm Al₂O₃ were added was much lower than B90Bu10 and RSO90Bu10. This proved that addition of butanol helped the nanoparticles to emulsify effectively. Kinematic viscosity of butanol @40°C is 1.0039cSt [22].

When 10% butanol was added to B100, viscosity decreased by 15.2% (Figure 6(b-c)). Similar characteristics were observed with RSO100 when 10% Bu was added - viscosity @40°C was decreased by 39.7%. The increase for B100 when 100ppm Al₂O₃ were added was 3.1%. On the other hand, viscosity was decreased by 0.4% for B90Bu10 fuel when 100ppm Al₂O₃ was added. The percentage increase for RSO100 when 100ppm Al₂O₃ were added was 1.1%, whereas for RSO90Bu10 viscosity was decreased by 3.3% when 100ppm Al₂O₃ were added. So, it was established that when Al₂O₃ nanoparticles were added to B100 and RSO100 the viscosity of the blends increased due to increased surface tension.

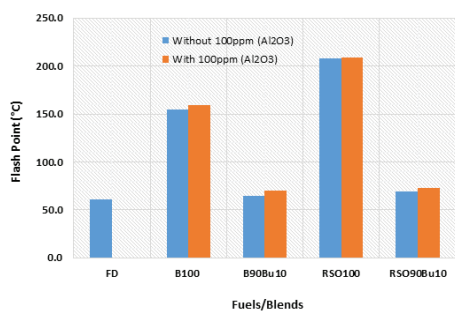
On the other hand, when 100ppm Al₂O₃ was added to B90Bu10 and RSO90Bu10 fuels, the viscosity decreased due to the fact that butanol acted as a surfactant and dispersed the Al₂O₃ into the blends properly. It was believed that the decrease in the viscosity of the biofuel + 10%Bu + 100ppm Al₂O₃ blends would give better spray characteristics than only butanol-biofuel blends.

The calorific value was decreased by 6.1% when 10% butanol was added to B100 – Figures 6(c) and 6(d). This was due to the low calorific value of butanol being $37.334 \frac{\text{MJ}}{\text{kg}}$ [21]. The same happened with RS100 when 10% Bu was added (from RSO100 to RSO90Bu10), the calorific value was decreased by 2.8%.

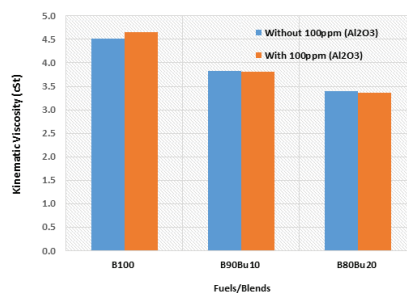
In general, for all blends the calorific value increased when 100ppm Al_2O_3 were added. The increase in HHV for B100 and B90Bu10 fuels were 5.8% and 9.6% respectively when 100ppm Al_2O_3 were added. On the other hand, these values were 7.2% and 8.2% for RSO100 and RSO90Bu10 when 100ppm Al_2O_3 were added – Figure 6(d). These results proved that rate of increase in HHV were higher when nanoparticles were added in the biofuel –butanol blends instead of only neat biofuels. Hence, it was evident that the butanol was able to help catalyse the combustion process much better in the nanoparticles blends. Macroscopic spray characteristics with and without Al_2O_3 nanoparticles are shown in Figure 7.

When 10% Butanol was added to B100 (from B100 to B90Bu10), liquid sheet angle stayed the same. On the other hand, when 10% Butanol was added to RSO100 (from RSO100 to RSO90Bu10), the liquid sheet angle increased by 25% (Figure 7(a)). Furthermore, when Bu was added to the biofuels with Al_2O_3 blends, the liquid sheet angle increased by 23.1% for B90Bu10 + 100ppm Al_2O_3 when compared to B90Bu10; and 11.1% for RSO90Bu10 + 100ppm Al_2O_3 when compared to RSO90Bu10. The decrease for B100 when 100ppm Al_2O_3 were added was 40%, and this value was 16.7% for RSO100 when 100ppm Al_2O_3 were added (Figure 7(a)).

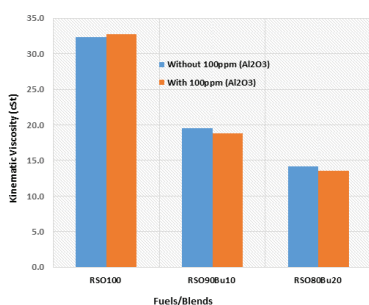
The results illustrated that overall, Butanol - biofuel - nanoparticle blends gave increased liquid sheet angle; this will eventually lead to improved combustion inside the engine cylinder and lower fuel consumption. The spray cone angle increased by 0.19% when 10% Butanol was added to B100 (from B100 to B90Bu10) – Figure 7 (b). Whereas, this was decreased by 21.4% in the case of RSO100. When Bu was added to the biofuels with Al_2O_3 blends, the spray cone angle increased by 19.5% for B90Bu10 + 100ppm Al_2O_3 when compared to B90Bu10, and by 22.3% for RSO90Bu10 + 100ppm Al_2O_3 when compared to RSO90Bu10. The spray characterisation results proved that butanol - nanoparticle blends gave better spray characteristics than only nanoparticles-biofuels blends. The increase in the liquid sheet angle and the spray cone angle would mean that the atomisation of the fuels would be better as more fuel particles will get in contact with air.



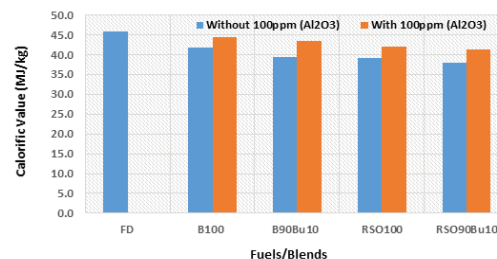
(a) Flash point with & without 100ppm Al₂O₃



(b) Viscosity @40°C with & without 100ppm Al₂O₃

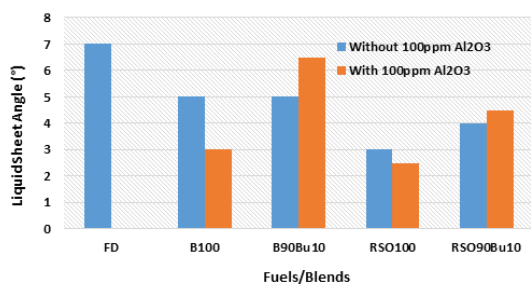


(c) Viscosity @40°C of RSO & butanol blends - with & without the addition of 100ppm Al₂O₃

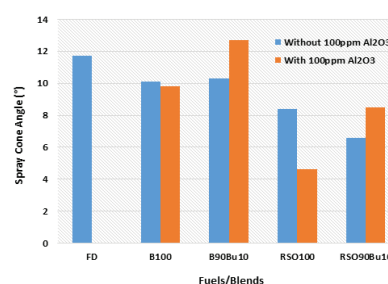


(d) Calorific value of blends with & without 100ppm Al₂O₃

Figure 6 – Comparison of biofuels (and blends) properties with and without Al₂O₃ nanoparticles



(a) Liquid sheet angle of blends with & without 100ppm Al₂O₃



(b) Spray cone angle of blends with & without 100ppm Al₂O₃

Figure 7 – Spray properties of fuels with and without Al₂O₃ nanoparticles

CONCLUSIONS

Two nanoparticles (Al_2O_3 and CuO) were used in this study to investigate the physicochemical properties and spray characteristics of the biofuel blends. It was found that the addition of nanoparticles increased the density, flash point temperature, kinematic viscosity and heating value of the fuels when mixed separately with neat FD, neat B100 and neat RSO100.

The density of the Al_2O_3 blends were lower than CuO blends. The flash point were much higher for Al_2O_3 blends in comparison to CuO blends. The viscosity was increased with both nanoparticles; however, Al_2O_3 gave smaller increase than CuO nanoparticles. It was proved that addition of Al_2O_3 gave better properties than CuO nanoparticles.

For internal combustion engines application, increase in the density and the viscosity would produce negative effects on engine fuel supply systems and combustion; whereas, on the other hand, increased flash point and the heating value would produce positive effects on fuel systems and combustion.

Hence, in the second stage of the study, the effect of butanol addition in Al_2O_3 nanoparticles-biofuels were assessed to see how butanol addition affect physicochemical properties and spray characteristics of the blends. It was observed that Butanol-biofuel- Al_2O_3 blends improved physicochemical properties and spray characteristics when compared to only biofuel- Al_2O_3 blends. The spray characteristic results showed that both spray cone angle and the liquid sheet angle parameters were improved significantly for Al_2O_3 - butanol - biofuel blends. The study concluded that addition of butanol helped to emulsify Al_2O_3 nanoparticles effectively in the blends; and butanol can be used as a surfactant for emulsification of Al_2O_3 nanoparticles in biofuels. Measurement of surface tension, engine performance and exhaust emission analysis are recommended as further work. Use of other nanoparticles and ethanol is another area of further investigation.

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