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Effect of Nano Additives Application and Strategy of Injection on Particulate Characteristics in Engine Operated with Biodiesel Blends

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A B S T R A C T

Recent developments in nano additives and injection strategies of fuel are effective techniques used in diesel engines to decrease exhaust pollutants and boost engine performance. The injection effect strategy of fuel with titanium dioxide (TiO_2) application on exhaust emissions and particulate matter (PM) characteristics in common-rail direct ignition (CRDI) diesel engines for biodiesel blends was experimentally examined. Experimental test results indicated that usage of TiO_2 into the B100, B20 and B30 enhanced the decline in CO, THC and NO_X than to the diesel without additives. PM number and concentration decreased by 13.54%, 22.73% and 32.68% from the combustion of B100+TiO_2, B20+TiO_2 and B30+TiO_2, respectively, compared to the nano additives absence into the fuel. Furthermore, the rate of soot oxidation, mass and weight significantly increased higher from the biodiesel blends than the diesel. It indicated that the internal structure form of soot particles produced from B100+TiO_2, B20+TiO_2 and B30+TiO_2 are oxidised earlier at lower temperatures in comparison with diesel. Regarding the TEM images, it is indicated that soot particles emitted from oxygenated fuels are easier to oxidise at low temperatures and quick time compared with diesel. The fuel injection strategy and both oxygen-bond from nano additives and fuel properties are beneficial for improving the soot oxidation and at the same time decreasing emitted PM.

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1. Introduction

In the last years, nanoparticles mixed with diesel engines have received massive interest due to nano-fuel blending as promising qualities (catalyst activity and thermal conductivity) which promote the reduction in engine emissions and enhance engine performance [1]. In recent years, many studies focused on using nano additives into the oxygenated fuels to reduce the pollutants emitted from internal combustion engines [2, 3]. It is reported that using a lower blend percentage (10% and 30%) of biodiesel with diesel fuel enhances the reduction of pollutant emissions and saves the environment [4, 5]. The nanoparticles of zinc oxide and titanium dioxide (TiO_2) nanoparticles were added to the Calophyllum Inophyllum biodiesel as stated in Nanthagopal *et al*. [6]. They found that emissions reduction and enhanced diesel engine execution can be accomplished by the addition of nanoparticles into biodiesel. Additional work by Prabhu *et al*. [7] investigated that the emissions of HC, nitrogen oxides (NO_X) and soot nanoparticles decreased with doping of Cerium oxide (CeO_2) nanoparticles into the biodiesel (type of Jatropha). Venu *et al*. [8] observed that the HC, NOx, CO and the emissions of smoke decreased by 5.69%, 9.39%, 11.24% and 6.48% when adding 20 ppm alumina nano additive (Al_2O_3 n) to the blend of diesel-biodiesel-ethanol compared to the neat blend. The inclusion of nanoparticles (TiO_2) into the blends of biodiesel leads to a decrease in hydrocarbons (HCs) compared to biodiesel without additives [9].

Particulate matter (PM) and emissions of NO_X are considered a major concern in diesel engines for the recent investigations on engine emissions. Different technologies of the common-rail fuel system, after-treatment system, nano additives and gas recirculation of exhaust (EGR) are applied in diesel engines to fulfil the stringent regulations of emission by reducing exhaust emissions [10, 11]. It is mentioned that the injection strategy of fuel improved the particulate emissions and temperature of the combustion and exhaust [12]. It is stated in the work by Yoon *et al*. [13] that late fuel injection is suitable for exhaust gas catalysts. The critical emissions of smoke opacity were decreased as mentioned in a study by Hotta *et al*. [14] for different injection timing of fuel. They found that the effective reduction in smoke opacity can occur with early injection timing of fuel in the combustion cycle. Hardy *et al*. [15] indicated that the combustion efficiency and PM reduction enhanced with the advanced injection strategy of fuel. Several researchers have recently stated that the emissions of soot can be declined by 40% through presented fuel injection without increasing NO_X emissions [16]. Park and Bae [17] examined that CO and total hydrocarbons (THCs) reduced when applying the late injection strategy of fuel. In contrast, other studies from Fayad *et al*. [18] and Yamamoto *et al*. [19] documented that the delayed injection increases the CO and THC from the burning of different fuels in diesel engines. Recent research has explored the influence of fuel injection on the speciation of hydrocarbons (HC) produced during diesel combustion [20]. Recent work [21] examined the injection effect of fuel on gaseous emission composition and EGR in engines from n-butanol blend combustion. The study outcome displayed that the THC and CO are significantly reduced with early fuel injection [18]. Recently, biodiesel has gained significant attention as an alternative energy source, mostly because its features closely resemble those of mineral diesel compared to other renewable fuels [22]. Higher O_2 (oxygen content) in the characteristics of biodiesel offers a sufficient reduction in CO and HC as well as PM [23, 24]. Furthermore, more attention received on applying alcohol blends in internal combustion engines as a beneficial alternative to diesel fuel [25]. In addition, alcohol molecules combustion produces lower levels of soot particles, THC and CO.

Serious problems linked to environmental and health issues were reported concerning fuel combustion. Also, the latest studies documented that the first generation of biodiesel emitted lesser HC, CO and PM than pure diesel [24, 26]. It was also obtained that the polycyclic aromatic hydrocarbon (PAH) and soot pressures diminished with combustion biodiesel. Furthermore, the alcohol-diesel blend exhibited the same trend of soot and PAH reduction [27, 28]. Soot particles number produced from the combustion cycle are significantly reduced when the engine fuelling with butanol-diesel fuel [12, 29-32]. When using pure biodiesel in the engine, the critical emissions of NO_X decreased by 10% compared to the diesel [33]. High temperature produced during biodiesel combustion leads to a notable increase in NO_X emissions [24, 34]. The literature lacks a clear consensus regarding the threshold for critical emissions of NO_X inside the combustion cycle, in some cases, the researchers stated that alcohol blend combustion increases NO_X emissions and others mentioned that decreases depending on engine operating conditions and injection strategies [35]. It is documented that the injection strategy of fuel is one of the methods to control emissions of PM and NO_X. It is confirmed that UHC, NO_X and smoke decreased by 60 %, 13 % and 70 with adding graphene oxide nano-additives (GO) to the biodiesel blends [1]. The previous study [36] stated that the injection strategy of fuel and using nano additives can be beneficial for particulate reduction. Also, the injection strategy of fuel assists decreases the formation of particulate in the combustion cycle and helps in decreasing the particulate over the tailpipe. The effect of diesel/alcohol blends and GO nanoparticle additives in CI engines was examined by Ahmed *et al*. [37]. They obtained that significant reduction in CO by 40%, UHC by 50%, and smoke by 20% from the combustion of diesel/alcohol blends with GO nanoparticles. The Work [38] documented that the improved soot oxidation rate characteristics can be achieved with late injection of fuel. The soot emissions were reduced by approximately 30% with fuel injection without an increase in emissions of NO_X. The inclusion of various concentrations (10 mg/L, 20 mg/L, 30 mg/L, 40 mg/L and 50 mg/L) of Al_2O_3 nanoparticles into the biodiesel blend result in promising reduction in HC, CO and NO_X emissions by 80%, 60% and 70%, respectively. It is stated in different studies on effect injection strategy of fuel, but still unclear how the exhaust emissions and soot particles can be affected by injection strategy of fuel. In addition, nano additives effect and injection strategies of fuel on the PM and emissions characteristics were not well covered in the literatures. Hence, this work focuses on inspect the impacts injection strategy of fuel and nano additives with oxygenated fuel blends on emissions and PM features. Furthermore, provide experimental data for the injection effect of fuel on characteristics (number and concentration) of PM and soot oxidation activity. The number of soot emissions and soot nanostructure of soot particles were also highlighted with burning diesel, B100, B20 and B30.

2. Experimental Procedure

2.1 Fuels and equipment

In this study, Al-Doura Refinery provides diesel fuel to use as a base fuel. Alcohol and biodiesel were fed to the engine as substitute fuels and mixed with diesel fuel. Sunflower oil is the source of biodiesel fuel and is supplied by Vegetable Oils Company for experimental testing. Further, the different features of diesel, B100, B20 and B30 are presented in **Table 1**. For tests, the different blends of oxygenated fuels were selected as B100, B20 (80% diesel fuel and 20% biodiesel) and B30 (70% diesel, 15% biodiesel and 15 % butanol). The blends of alternative fuels were mixed at the same time in the experiment to enrich the properties of fuels and avoid any incidence of agglomeration during the tests.

Table 1: Specifications of tested fuels [24, 39]

Properties	Diesel	Biodiesel	B20	B30
Chemical formula	$C_{16}H_{34}$	$C_{19}H_{36}O_2$		
Derived cetane number	51.8	62	53.4	56.3
Latent heat of vaporization (kJ/kg)	242	216	-	-
bulk modulus (MPa)	1410	1554	-	-
density at 15 °C (kg/m^3)	844.3	896.1	861.6	873.4
Calorific value (MJ/kg)	45.80	38.90	52.43	54.67
Flash & Fire point (°C)	65-70	157-162	82-85	96-98
Water content by coulometric KF (mg/kg)	40	170	387.3	392.4
kinematic viscosity at 40 °C (cSt)	2.77	5.0	3.322	3.83

An analysis of exhaust gas was carried out to record the exhaust emissions emitted from the research engine. Further, the emission variations were registered and documented during the test (inside the cylinder) and from the exhaust pipe. The nano characteristics of PM were measured using a "scanning mobility particle sizer (SMPS)". The "transmission electron microscopy (TEM)" was employed to analysis the characteristics of soot particle morphology. The oxidation reactivity results from nanoparticles of soot emissions (soot mass and weight) were obtained using Thermogravimetric analysis (TGA). The results in the TGA were under exothermic reaction. The soot particulate properties were calculated using soot images through digital image analysis software [40].

2.2 Nano additives preparation

The nanoparticles of titanium dioxide were prepared according to the method of sol-gel. The oxygenated fuel blends (B100, B20 B16) were prepared as mentioned in the above section. The nanoparticles were added to the B100+ TiO_2, B20+ TiO_2 and B30+ TiO_2 to prepare in the concentration of 100 ppm. The thermal stability of nanoparticles in the current study was calculated to enhance the properties of nano additives. The nano fuel blends were stirred for 25 minutes continuously in ultrasonication and a stirrer to avoid the nanoparticle's agglomerate. TiO_2 properties are found in **Table 2** including the thermal conductivity and stability of these particles [41]. In addition, the nano fuel was also blended with surfactant to improve the homogeneity and stability of nano fuel blends.

Table 2: Specifications of TiO_2 nanoparticles [42]

Property	Specifications
Chemical formula	TiO_2
Appearance	White powder
Purity (%)	99.9
Average particle size (nm)	30–50
Surface area (m^2/g)	>40
Bulk density (%)	0.40
Loss on drying (≤ %)	0.90
Si ≤ ppm	0.003
Fe ≤ ppm	0.003

2.3 Setup of research engine and test conditions

A diesel engine with a 4-cylinder, CR injection system of fuel, water-cooled was carried out as found in **Fig. 1**. The strategy of fuel injection was monitored within the test by using the CRFI system. Different specifications of the research engine were explained in this section. The further details of the engine are a direct injection (DI), 4-Stroke, with 100mm and 110 mm bore and stroke, respectively. The speed from 1000 to 2000 rpm is considered for the engine with 3.6 L displacement. Further, the injection range of fuel pressure is between 500 to 1500 bar, while the affected pressure of the engine is less than 7 (bar). An E-type thermocouple was employed to obtain the temperature of the exhaust as well as to confirm that the engine was fully warm. Before collecting any data from the engine, it was operated using diesel fuel for 30 minutes to ensure it reached the optimal operating temperature. PC was used to record the data of exhaust temperatures, IMEP (indicated mean effective pressure) and engine torque for fuel types.

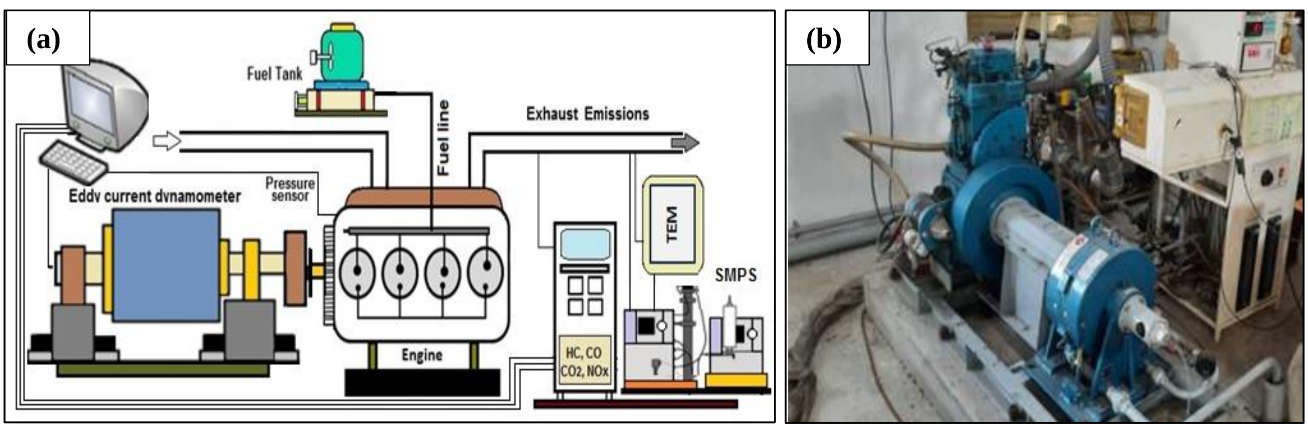

Figure 1: Research engine setup and tools for (a) schematic diagram and (b) real image of the research engine.

For experiments, at 1500 rpm, the engine speed was fixed, which is considered the middle speed of vehicle engines on city roads. IMEP and injection pressure of fuel were fixed at 4 bar and 550 bar, respectively. Pure fuel diesel was tested as the first fuel in the engine to ensure all equipment working properly and for the heating process. Three test measurements were carried out to average the results to avoid experimental error.

3. Results and Discussion

3.1 Analysis of gaseous emission

During combustion, the measure of inefficiency is represented by hydrocarbon emissions. Incompletely burnt fuel particles can be emitted into two forms either in solid state (PM) or in gaseous state (organic compounds) [43]. Unburned HC formation can be formed in CI engines such as through various possible sources such as crevice areas and cylinder piston interface as well as from fuel trapped in the nozzle. Incomplete evaporation of fuel mixture is another way to produce THC emissions. **Fig. 2** shows the effect of incorporating loaded nano additives TiO_2 into the fuel blends and late injection strategy on emissions of carbon monoxide (CO) as shown in **Fig. 2a**, total HC as shown in **Fig. 2b** and NO_X as shown in **Fig. 2c**. According to the figure, it can be inferred that adding TiO_2 to the pure biodiesel (B100) and biodiesel blends (B20 and B30) leads to a notable decrease in emissions of CO, THC and NO_X than to diesel fuel. This is due to the addition of nanoparticles of TiO_2 and higher oxygen content from biodiesel [44]. The addition of nano additives improves combustion within the cylinder, resulting in enhanced oxidation of exhaust emissions. Moreover, biodiesel blends (B20 and B16) produce lower levels of emissions compared with B100, even with the addition of nanoparticles to the biodiesel. The addition of TiO_2 with oxygen content and injection strategy helped decrease the emissions produced from B20 and B30 compared with B100. For biodiesel blends, B30 emitted less total HC, CO and NO_X compared to B20 and B100. The content of oxygen-bond in the nano additives and the mixture of butanol and B100 enhance the reduction of emissions from B30 in comparison with other nano fuels. High temperatures prevailing inside the cylinder are the main reason for the release of NO_X emissions [45]. The adding TiO_2 into the blends combined with a late injection strategy, significantly reduces NO_X emissions compared to conventional diesel fuel (**Fig. 2c**). This reduction is attributed to the lower cylinder chamber temperature, resulting from a decrease in the total heat capacity of the working gas. Furthermore, among the tested blends, B30 demonstrates the most effective reduction in NO_X emissions, as shown in **Fig. 2c**. This improvement is primarily due to the lower flame temperature and reduced oxygen concentration in the fuel when using the delayed injection strategy.

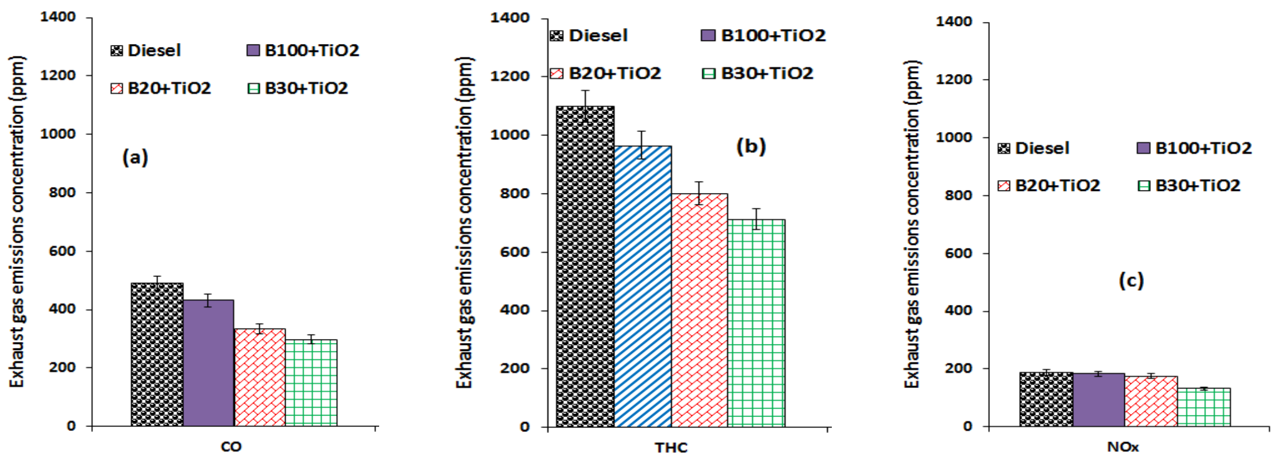

Figure 2: Exhaust gas emissions analysis of (a) CO, (b) THC and (c) NO_X for B100+TiO_2, B20+TiO_2, B30+TiO_2 and diesel

3.2. PM size distribution

Metal components and nanoparticles of soot are the main components of PM. The mechanism of formation and size of PM are greatly affected by the mechanism process of combustion. Some authors have stated that the amount of formation and soot size are related to different parameters such as pressure, temperature, fuel properties, time, and oxygen content [46]. PM oxidation activity is directly affected by PM structure and components. Adding nanoparticles into the B100, B20 and B30 enhanced the reduction of concentration and number of PMs by 13.54%, 22.73% and 32.68%, respectively, as depicted in **Fig. 3**. The content of soot particles inside PM reduced with increasing the mixing ratio of biodiesel blends. Oxygen concentration in the blends of fuel could be the major reason for the soot decline, which caused limited soot nucleation in the formation process and reduced locally fuel-rich regions. The average diameter of PM is in the range of 17 and 30 nm based on the engine operation conditions and fuel characteristics. Biodiesel blends with TiO_2 lead to a significant reduction in the number and concentration of particulates compared to the B100 and diesel. The positive impact of TiO_2 and oxygen inside fuel helps in better oxidation of PM through the combustion cycle and tailpipe. The number of PMs increased in the case of diesel because of the sulfur content and heavy hydrocarbons in the fuel.

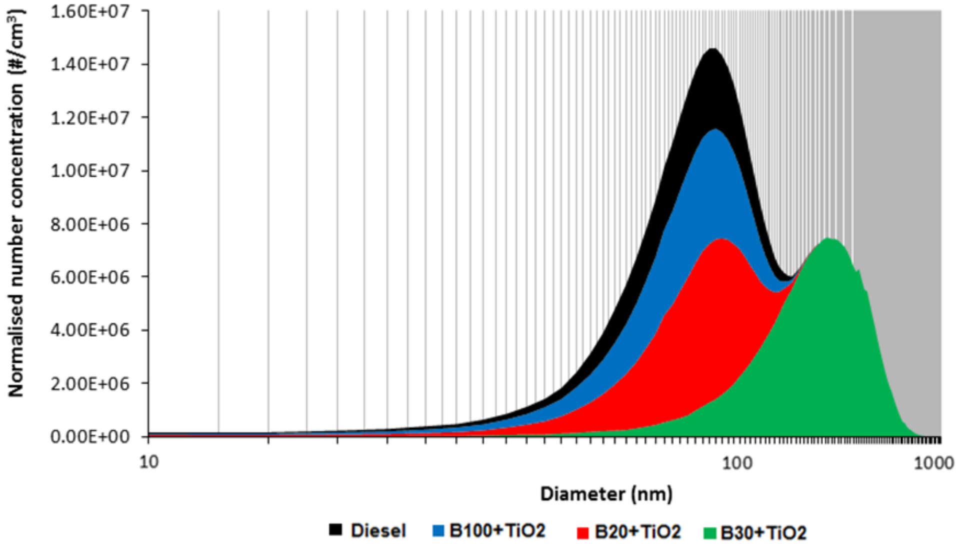

Figure 3: PM concentration and number for B100+TiO_2, B20+TiO_2, B30+TiO_2 and diesel

3.3 Oxidation reactivity

PM formation during combustion occurs through two primary pathways: the pyrolysis products and the oxidation of fuel molecules. **Fig. 4a** shows that the BI00, B20 and B30 improved the oxidation level in PM than to the absence of nano additives in diesel. Improved combustion efficiency, nano additives and higher oxygen (O_2) content cause the significant reduction in oxidation reactivity of PM. The soot mass was reduced from the burning of oxygenated fuels with TiO_2 nanoparticles under various temperatures of oxidation compared with diesel. This may be because of active oxygen content and less aromatic HC in the fuel blends which leads to increasing the potential particle oxidation of soot producing less soot mass, and then reducing the total PM. It is mentioned that most formation of carbonaceous products is oxidized at an early stage during combustion. In contrast, microscopic solid carbonaceous particles cannot undergo oxidation due to the presence of non-volatile species formed during the nucleation mode in diesel combustion. These particles are likely composed of ash derived from metallic additives.

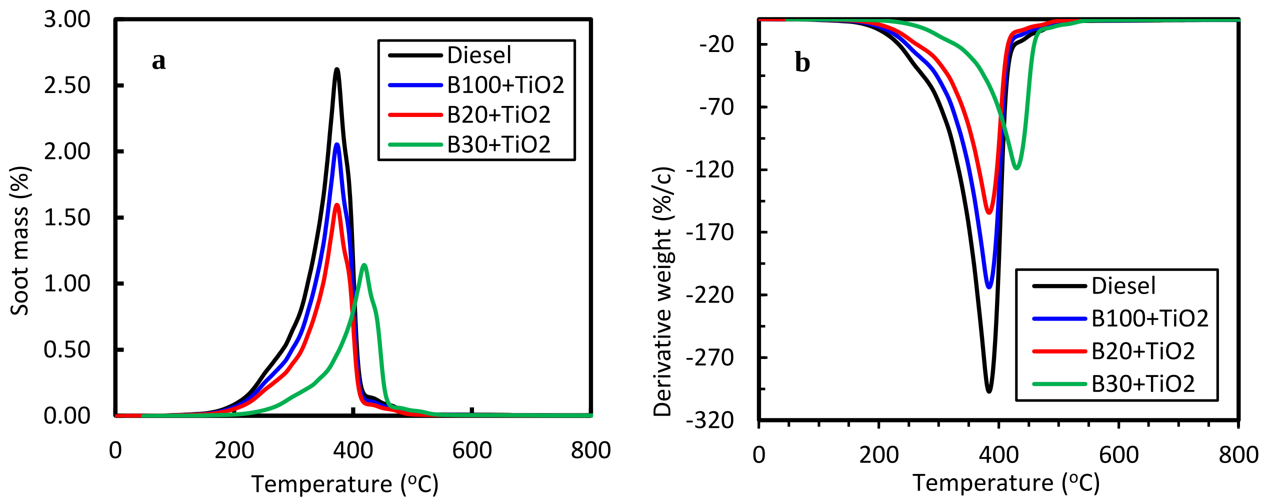

Figure 4: a) TGA analysis of soot mass for B100+TiO_2, B20+TiO_2, B30+TiO_2 and diesel. b) TGA analysis of soot weight for B100+TiO_2, B20+TiO_2, B30+TiO_2 and diesel.

The findings of TGA analysis for soot weight can be shown in **Fig. 4b** for B100, B20, B30 and diesel fuel. In general, the weight losses of soot particles increased with increasing the temperature for all tested fuels. In addition, the soot particle weight losses increased with adding TiO_2 to the B100, B20 and B30 compared to diesel without nano additives. These results agree with results found in Fig. 4, the trend of the results caused due to active oxygen in TiO_2 and biodiesel blends [47], which is beneficial for losing the weight of soot particles and decreasing the total PM as depicted in **Fig. 5**.

3.4 Soot Nanoparticles Structure

The results of biodiesel blends, B100 and diesel show that the size distribution of soot particle number declined with B20+TiO_2, and B30+TiO_2 than to the diesel and B100+ TiO_2 (**Fig. 5**). Lower production in the number of particles of soot from the oxygenated fuels with nano additives (**Fig. 5b, c,** and **d**) than to the diesel without nano additives (**Fig. 5a**). The diameter of soot particles is also reduced by adding TiO_2 to the pure diesel, B20 and B30 compared to diesel without nanoparticles of TiO_2. The effect of fuel on soot structure and particulate, numerous studies [48] have been studied for diesel fuel in diesel engines. **Fig. 6** shows the different forms of the internal structure of soot particles obtained by TEM images from the combustion of B100+ TiO_2, B20+TiO_2, B30+TiO_2 (**Fig. 6b, c,** and **d**) and diesel (**Fig. 6a**) without nano additives. The internal structure of oxygenated fuels with nano additives has the non-uniform distribution of internal carbon layers and some hollow inside soot particles which give a good chance of oxidation of the soot particles in quick time and low temperatures [49]. The attractive properties of biodiesel blends (e.g. oxygen-bond) enhance the ability of oxidation inside the graphene layers structure of particles, which increases the oxidation of PM and declines the PM formation.

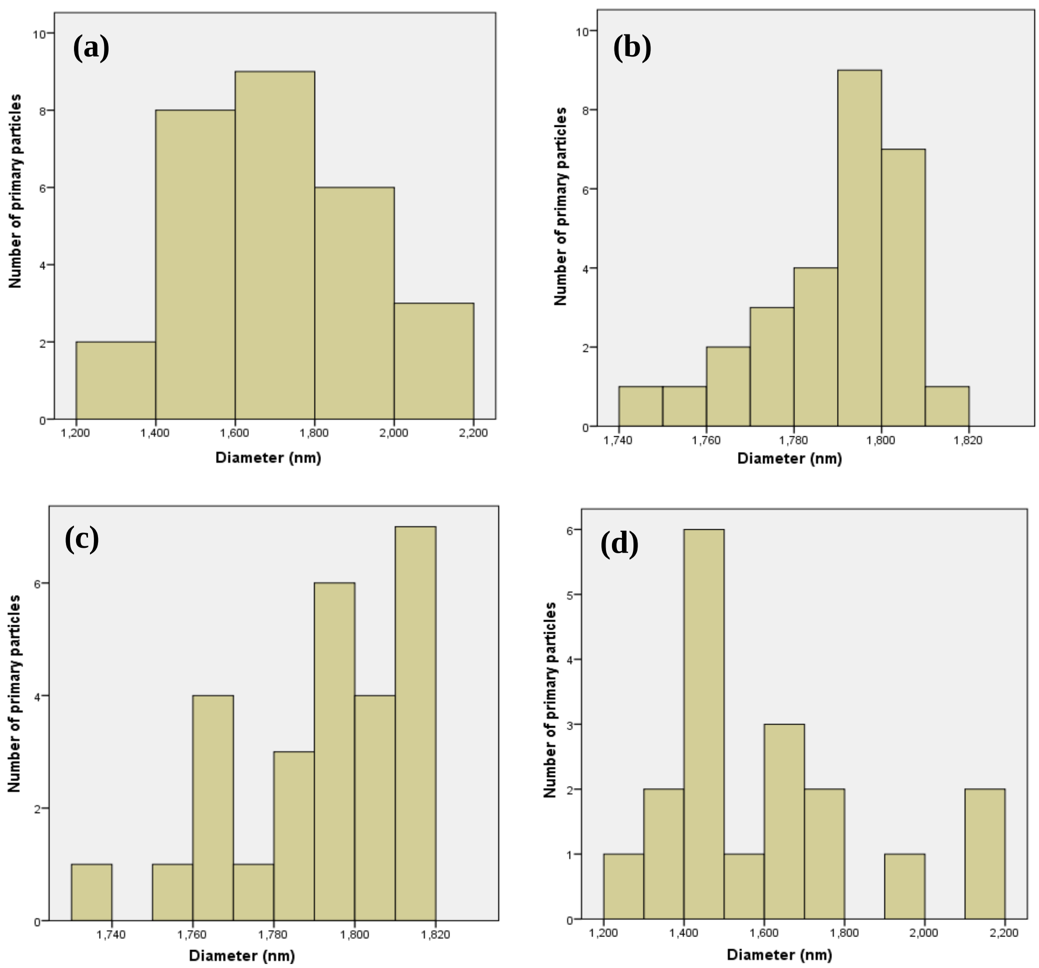

Figure 5: Number of soot particles for (a) diesel, (b) B100+TiO_2, (c) B20+TiO_2 and (d) B30+TiO_2

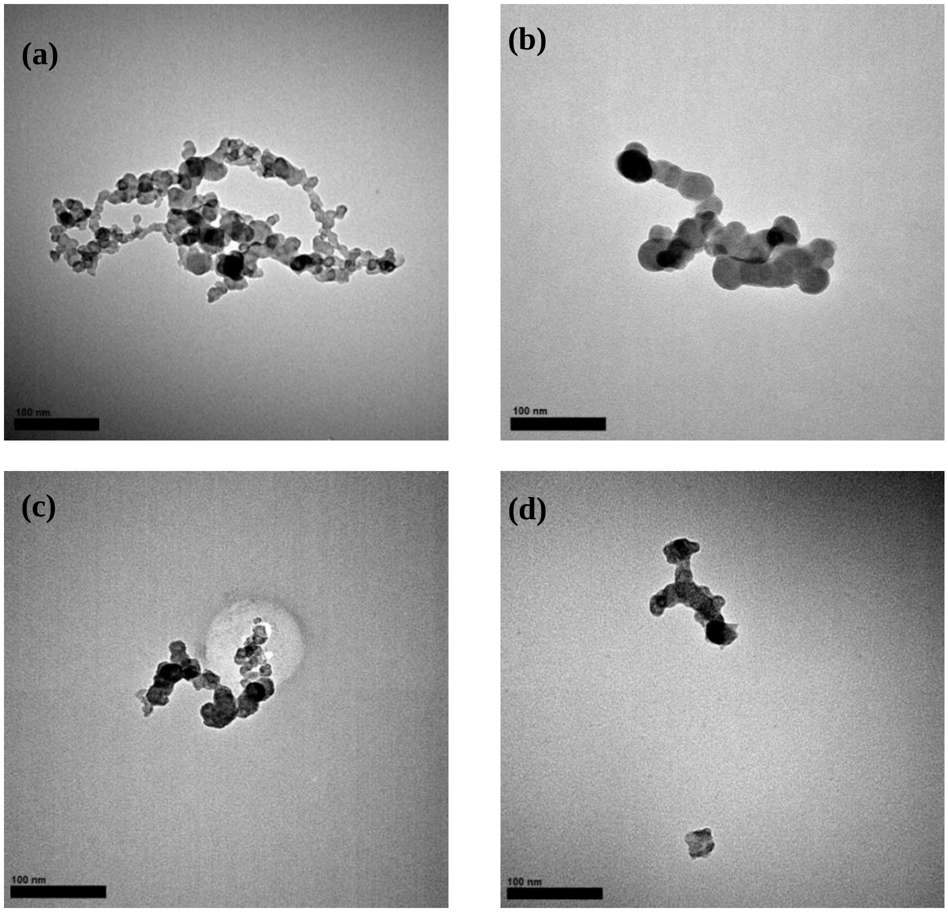

Figure 6: TEM images for (a) diesel, (b) B100+TiO_2, (c) B20+TiO_2 and (d) B30+TiO_2

4. Conclusions

The twofold influence of the injection strategy of fuel and nano additives (TiO_2) on emissions and PM characteristics was explored in the current study for different biodiesel blends. Also, the characteristics of oxidation activity and nanoparticle structure of soot particles from B100, B20, B30 and diesel combustion were investigated. From the experiments study, the rich potential of nano additives in the B100, B20 and B30 offers multiple benefits by decreasing the emissions of NO_X, THC and CO. Furthermore, NO_X emissions generated from blends of B20 and B30 with TiO_2 nanoparticles were found to be lower than those from B100 and diesel. In both B20 and B30, the PM concentration and number showed a greater reduction with adding TiO_2 compared to the B100+TiO_2 and diesel. For particulate emissions, the reduction of concentration and number of PM was found in B100, B20 and B30 by 13.54%, 22.73% and 32.68%, respectively, from both the injection effect of fuel and nano additives. The oxidation reactivity of soot increased with the presence of TiO_2 and the injection strategy of fuel for oxygenated fuel in comparison with diesel. It was observed that the injection strategy of fuel increases the potential particle oxidation of soot and enhances the oxidation reactivity for all fuels, but this reduction is clearly in oxygenated fuels. High rate of soot oxidation for both mass and soot weight with adding of TiO_2 to the oxygenated fuels than to the neat diesel. It was found that the internal structure of soot particles is easier to oxidise from B100+ TiO_2, B20+TiO_2, and B30+TiO_2 at lower temperatures. The effect of adding different concentrations (25 ppm, 50 ppm, 75 ppm, 100 ppm) of nano additives into the renewable fuel blends and different proportions of exhaust gas recirculation (EGR) on PM characteristics will be good points for future study.

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Conflict of Interest

The authors declare that they have no conflict of interest.

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