METAL OXIDES AS SOLUBLE NANO CATALYST ON BIODIESEL: A REVIEW

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Abstract. *Nano particles of metal oxide developed as soluble nano additive in liquid fuels to improve fuel quality. One application of nano metal oxide particles is an additive to biodiesel. Biodiesel is an alternative fuel that can reduce dependence on fossil fuels. Pure biodiesel has a relatively lower calorific value compared to fossil fuels. Low calorific value results in increased brake specific fuel consumption. Moreover, biodiesel has a higher density and viscosity compared to fossil fuel. The content of carbon monoxide (CO), unburned hydrocarbons (HC) and nitrogen oxide (NOx) in exhaust gases with biodiesel is higher than fossil fuels. Metal oxide nanoparticles are added to biodiesel between 6 to 80 nm with concentrations about 50 to 500 ppm. Addition of metal oxide nanoparticles to biodiesel can improve brake thermal efficiency, reduce brake specific fuel consumption, carbon monoxide (CO), unburned hydrocarbons (HC), nitrogen oxide (NOx) and improve carbon dioxide (CO_2) emission due to the catalytic effect of metal oxide nanoparticles. Metal oxide acts as an oxidation catalyst thereby reduce the carbon combustion activation temperature and thus enhances hydrocarbon oxidation, promoting complete combustion. Nanoparticles that are often used in various studies are nickel (II) oxide (NiO), cerium (IV) oxide (CeO_2), titanium oxide (TiO_2), zinc oxide (ZnO), aluminum oxide (Al_2O_3), and silicon dioxide (SiO_2). This review paper describes the progress and development of nano metal oxide applications as additives for biodiesel, and the discussion in this paper is divided into 3 main topics, including the effects of nanoparticles on the properties of biodiesel, engine performance, and emission characteristics.*

Keywords: *nanoparticles; catalytic effect; biodiesel; engine performance; fuel consumption; exhaust gases*

1. Introduction

The world's energy needs still depend on fossil fuels. Liquid fuel is one of the fuels that continues to increase in consumption. The world's consumption of liquid fuels is estimated to increase from 95 million barrels per day (b/d) in 2015 to 113 million b/d in 2040. Liquid fuel consumption is increasing in the industrial and transportation sectors (Energy, 2017). Bioenergy is promoted to reduce dependence on non-renewable (fossil) fuels and to reduce greenhouse gas emissions. Bioenergy is a non-fossil fuel produced from plants and animals (biological processes). Utilization of bio energy currently reaches 50 EJ/year or about 10% of global energy needs. The

potential of major global bioenergy techniques by 2050 is estimated to reach 160-270EJ/year (Haberl et al., 2010). One of the most widely developed of bioenergy currently is biodiesel.

Biodiesel is an environmentally friendly alternative fuel for ignition compression engines. Biodiesel has better properties than petrodiesel because it is made from renewable materials such as vegetable and animal oils, is non-toxic and biodegradable converted by soil and water borne organisms, and is free of sulfur and aromatics (Demirbas, 2005). Biodiesel or Fatty Acid Methyl Ester (FAME) can be made from various vegetables or animal oils (Fukuda et al., 2001; Van Gerpen, 2005). Triacylglycerides (TAG) or often called triglycerides are vegetable oils or animal oils used to produce biodiesel. In general, transesterification is a biodiesel production process. It is the reaction between triglycerides and alcohol in the presence of a catalyst to produce fatty acid alkyl esters. Glycerin is referred to as glycerol that is a by-product of transesterification, and methanol is the most commonly used alcohol (Hoekman & Robbins, 2012).

The use of biodiesel in compression ignition engines can reduce smoke opacity but increase Brake Specific Fuel Consumption (BSFC) (Özener et al., 2014). The smaller calorific value of biodiesel compared to petrodiesel causes an increase in fuel consumption. Biodiesel requires more fuel consumption to get the same power output as petrodiesel (Rahman et al., 2013). In addition, the use of biodiesel also reduces Brake Thermal Efficiency (BTE). Unburned hydrocarbons (HC), Carbon Monoxide (CO) in biodiesel were found to be lower than petrodiesel (Chauhan et al., 2011). However, Nitrogen Oxide (NOx) contained in exhaust gas was significantly increased when using biodiesel compared to petrodiesel (Ashok et al., 2017; Dhar et al., 2012; How et al., 2018; Raheman & Ghadge, 2007). Biodiesel has five main long carbon chains including, FAME: methyl palmitate (MP, $C_{17}H_{34}O_2$) and methyl stearate (MS, $C_{19}H_{38}O_2$) as saturated FAME, methyl oleate (MO, $C_{19}H_{36}O_2$), methyl stearate (MS, $C_{19}H_{38}O_2$) as saturated FAME, methyl oleate (MO, $C_{19}H_{36}O_2$), methyl linoleate (ML, $C_{19}H_{34}O_2$) and methyl linolenate (MLE, $C_{17}H_{32}O_2$) as unsaturated FAME (Herbinet et al., 2008). Biodiesel contains bound oxygen thereby suppressing the formation of soot particles. The soot particles result in reduced radiant heat transfer thereby increasing the reaction temperature and increasing NOx formation (Varatharajan & Cheralathan, 2012).

Catalytic combustion is widely used in various fields, especially in the use of efficient and low pollutant fossil fuels. Catalytic combustion can increase efficiency and reduce NOx pollutant emissions (Gong et al., 2018). Catalytic combustion is also often used in small-scale combustion. The catalyst can increase the flame stability limit. The fire is more stable even though it is in a very small space and far from the stoichiometric Air Fuel Ratio (AFR) (Wang et al., 2010; Wierzbicki et al., 2014). In internal combustion engines, it is impractical to add catalyst to the combustion chamber. To improve performance and to reduce internal combustion engine

emissions, the catalyst is mixed directly in the liquid fuel. Catalyst is added in nano-sized liquid fuel. Nano-sized metal catalysts can dissolve and mix in liquid fuels. Catalysts in liquid fuels are often referred to as nano-additives (Wickham et al., 2006).

Nano metal oxide is often used to improve performance and to enhace biodiesel exhaust gases. From the background above, this paper aims to provide an overview of several things are the use of nano metal oxide as a soluble nano catalyst, the effects among soluble nano catalyst on biodiesel properties, soluble nano catalyst on biodiesel performance, and soluble nano catalyst on biodiesel exhaust gases. This paper is a narrative review by comparing several studies to find conclusions and match them with existing theories.

2. Present state of the art: nano metal oxide

Metal oxide belongs to an important group of engineered nanoparticles, besides being used in liquid metal additives, nano metal oxide is widely used in cosmetics and sunscreens, coatings and textiles that clean themselves. Other applications include its use as an air fuel and battery processing agent and newer car catalytic converters (Puzyn et al., 2011). Furthermore, ultrasound is used to mix biodiesel with nano metal oxide. Furthermore, ultrasound is used to mix biodiesel with nano metal oxide. Figure 1 shows a soluble nano catalyst mixed with biodiesel using an ultrasonicator.
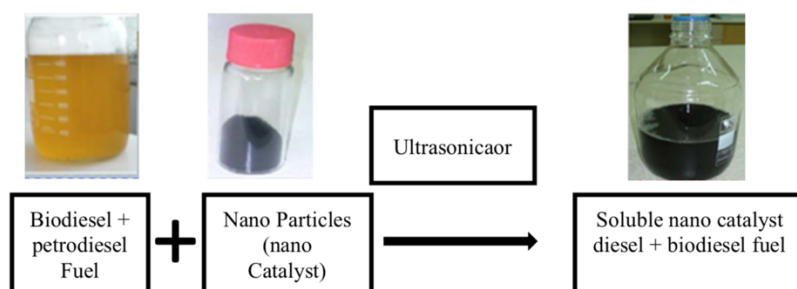

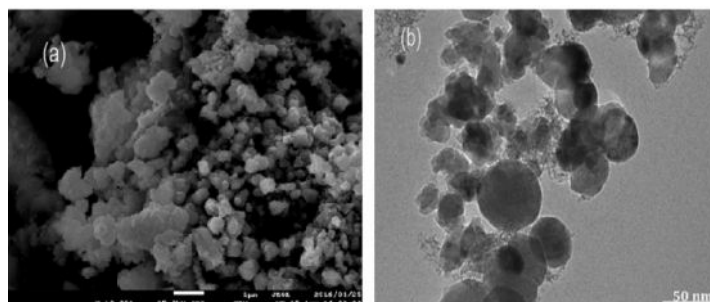Figure 1. Soluble nanocatalyst mixed with using an ultrasonicator (Najafi, 2018)

Figure 2. SEM (a) and TEM (b) image of TiO$_2$ nano particles (Yuvarajan et al., 2018)

Nanoparticles have properties that depend on their dimensions. Commonly used methods to characterize dimensions include transmission electron microscopy (TEM), scanning electron microscopy (SEM), atomic force microscopy (AFM), and dynamic light scattering (DLS) (Eaton

et al., 2017). Figure 2 shows the results of the SEM and TEM characterization of TiO2 nanopartices.

X-ray Diffraction (XRD) is used to confirm that nanoparticles are nano metal oxides. Figure 3 shows the XRD results from NiO nano particles, where Ni-O particle patterns are found in the range of 20 ° -80 ° (Srinidhi et al., 2019).
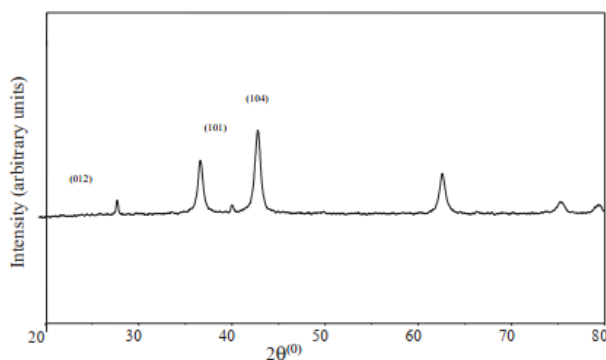

Figure 3. XRD of NiO (Srinidhi et al., 2019)

2.1 Fuel properties

The nano metal oxides used as soluble nano additives in biodiesel are 6 to 60 nm in size with a concentration of 10 to 300 ppm. The parameters measured in biodiesel that have been added or not yet added by nano particles are density, viscosity, calorivic value and centane number with ASTM D7467 standard. Various combinations of concentration and particle size that have been used as biodiesel additives in various studies are shown in Table 1.

The higher the concentration of nano particles in biodiesel the higher the density of biodiesel. But in other studies, the density is not so affected by the amount of nano particle concentration in biodiesel (Krupakaran et al., 2016).

Likewise with viscosity. The higher the concentration of nano particles, the higher the viscosity on biodiesel (Anand, 2013) (Mahalingam & Ganesan, 2018) (Yuvarajan et al., 2017) (Nanthagopal et al., 2017) (Gunasekar et al., 2017). However, other studies state that the higher the concentration of nano particles, the lower the viscosity of biodiesel (Krupakaran et al., 2016) (Nithya et al., 2017) (Karthikeyan & Prathima, 2016).

Flash points in the study (Anand, 2013) decreased with increasing concentration of nano particles. Whereas other research states that the higher the concentration of nano particles, the flash point is increasing (Krupakaran et al., 2016) (Nanthagopal et al., 2017) (Karthikeyan & Prathima, 2017a).

At calorific value, the higher the concentration of nano particles, the higher the heat value of biodiesel (Anand, 2013) (Krupakaran et al., 2016). While other studies state that the higher the concentration of nano particles, the lower the heating value of biodiesel.

Of the ten studies that have been reviewed, the Cetane number has consistency. The higher the concentration of nano particles, the higher the biodiesel cetane number (Anand, 2013) (Krupakaran et al., 2016) (Yuvarajan et al., 2017) (Nithya et al., 2017) (Nanthagopal et al., 2017) (Karthikeyan & Prathima, 2016) (Gunasekar et al., 2017). From all references reviewed, the addition of nano particles does not significantly affect the density, viscosity, and flash point values, but the cetane number has a relationship with the addition of nano particles. The higher the concentration of nano particles, the higher the value of biodiesel cetane number.

Tabel 1. Summary of fuel properties of biodiesel with soluble additive nano metal oxide

Biodiesel	Nano metal oxide	Size (nm)	Concentrations (ppm)	Density (kg/m^3)	Viscosity (cSt)	Flash point (°C)	Calorific value (mJ/kg)	Cetane number	Ref.
J. Curcas	Al$_2$O$_3$	Rata rata 51	0	895	5.25	85	38.88	53	(Anand, 2013)
			25	896	5.31	84	39.22	54	
			50	897	5.35	82	39.53	56	
J. Curcas 20% + Diesel 70% + Ethanol 10%	Al$_2$O$_3$		25	837.2 (20 °C)	2.57	22	39.137	54	(Venu & Madhavan, 2016)
Palm	γ Al$_2$O$_3$	6 - 12	0		4.8	130	38.49	51	(Krupakaran et al., 2016)
			25		4.76	160	38.52	55	
			50		4.74	168	38.56	59	
			75		4.73	172	38.582	62	
			100		4.73	180	38.59	63	
Ruber seed B20	Al$_2$O$_3$		10		4.1		42		(Mahalingam & Ganesan, 2018)
			20		4.2		41		
			30		4.2		41		
mustard	TiO$_2$		100		4.34		37.854	54	(Yuvarajan et al., 2017)
			200		4.38		37.652	57	
C. inophyllum B20	TiO$_2$		40	844.5	3.72		41.935	53.94	(Praveen et al., 2017)
Canola B20	TiO$_2$		0	915	4.8			42	(Nithya et al., 2017)
			300	840	3.4			56	
C. inophyllum	TiO$_2$		0	868.6	4.72	122	38	52	(Nanthagopal et al., 2017)
			50	869.2	4.73	123	37.12	53	
			100	870.4	4.75	124	37.54	55	
Algae	TiO$_2$ SiO$_2$		0	0.816 (gm/cc)	3.12	60.49	40.422	47	(Karthikeyan & Prathima, 2017b)
			50	817	3.03	62.45	42.6	48	
			100	817	3.01	63.61	44	48	
C. inophyllum	TiO$_2$		0	815	3.1		43	45	(Gunasekar et al., 2017)
			100	870	5.1		38	52	
	TiO$_2$ + Ethanox		100	902	4.7		38	56	
			200	904	4.6		39	58	
			300	910	4.8		39.2	59	
C. inophyllum	γ Al$_2$O$_3$		0	855.8	5.5	101	43.10		(Pambudi et al., 2021)
			100	864.1	5	100	43.95		
			200	853.5	4.4	104	43.73		
			300	855.3	3.6	97	44.25		

3. Engine performance

3.1 Brake specific fuel consumption (BSFC)

Brake specific fuel consumption (BSFC) is a complete measurement of fuel consumption in the combustion chamber that shows the vehicle's fuel consumption and power produced in a certain time. BSFC is expressed in units of mass (gr / kWh). In fact, fuel consumption will be greater in conditions that are not ideal combustion (Mirzajanzadeh et al., 2015). Nano cerium oxides (CeO2)

particles can reduce brake specific fuel consumption (BSFC) on biodiesel rice bran. Biodiesel 20% and petrodiesel 80% (B20), Biodiesel B20 + CeO_2 50 ppm, Biodiesel B20 + CeO_2 100 ppm were tested using 1 cylinder diesel engine test equipment. Biodiesel without the addition of CeO_2 nano particles has a higher brake specific fuel consumption (BSFC) value compared to biodiesel with CeO_2 nano particles (Karthikeyan et al., 2016). Another research states that the addition of cerium oxide on amide-functionalized multiwall carbon nanotubes (MWCNT) 30, 60, 90ppm to biodiesel of used cooking oil biodiesel 5% (B5) and biodiesel 20% (B20) can reduce BSFC by 0.42%, 0.84% and 3.09% on B5 biodiesel, and 0.34 %, 1.49%, and 4.51% in B20 biodiesel.

Nano CeO_2 provides oxygen molecules in a chain reaction causing the burning of hydrocarbon and carbon that are not completely burned. Burning more fuel in the cylinder produces more energy. Additionally, CeO_2 nano prevents the deposition of non-polar components in the cylinder wall and burns residual carbon. Nano catalysts in fuels produce millions of nano clusters that explode to decompose and to destroy deposits and to prevent the formation of deposits in the cylinder walls (Mirzajanzadeh et al., 2015).

4. Emission characteristic

Diesel engines have exhaust gas emission parameters including nitrogen oxide (NOx), carbon monoxide (CO), and hydrocarbons (HC).

4.1 Nitrogen oxides (NOx)

Nitrogen oxide is formed due to the presence of nitrogen in the air. Nitrogen reacts with oxygen during the combustion process which affects the amount of NOx content in exhausted gas emissions, among others, combustion temperature, combustion activation energy, and the equivalent ratio in combustion (Mofijur et al., 2013). Biodiesel exhaust gas emissions, containing more NOx compared to petrodiesel due to biodiesel containing oxygen so that the combustion of biodiesel involves more oxygen content and causes the combustion temperature to increase. So that the NOx in the exhaust gas increases (Tüccar et al., 2014).

Metal oxides in biodiesel add local oxygen content in combustion and with the addition of nanoparticles, which increases the duration of diffusion controlled combustion that increases NOx emissions (Aalam et al., 2015) using the additive Al_2O_3 biodiesel ziziphus B25 the higher the concentration of nano particles, the higher the NOx in the flue gas, which is 0 ppm = 3,148 g / kWH, 25 ppm = 3,456 g / kWh, and 3,729 g / kWh. However, other studies have revealed that the higher the concentration of nano particles, the lower the NOx concentration (Anand, 2013) Al_2O_3 jatropha biodiesel, (Pandian et al., 2017) TiO2 Mahua biodiesel, (Yuvarajan et al., 2018) TiO2 Mustard biodiesel, (Radhakrishnan et al., 2018) Al_2O_3 cashew biodiesel. They are of the opinion

that, however, from the whole NOx biodiesel research with additive nano metal oxide there is still more compared to petrodiesel.

4.2 Carbon monoxides (CO)

The formation of CO emissions is due to the lack of oxygen in combustion, uneven mixing of air and fuel, and lack of combustion processes. From several references that have been reviewed, CO emissions decrease along with the addition of nano particles to biodiesel or a mixture of biodiesel and petrodiesel. With the addition of 200 ppm TiO_2 nano particles to neat mahua biodiesel oil (BD100) can reduce CO emissions by 9.3% in peak brake power (Pandian et al., 2017). Zizipus jujube biodiesel methyl ester blended fuel (ZJME25) and nano particle AL_2O_3 by 25ppm at full load can reduce CO by 22.66% (Aalam et al., 2015). Cashew nut sold BD100 biodiesel can reduce CO emissions by 57.70% compared to petrodiesel. The addition of AL_2O_3 to BD100 biodiesel cashew nut can reduce CO emissions by 62.29% when compared to petrodiesel and can reduce by 10.85% when compared to BD100 biodiesel cashew nut without AL_2O_3 (Radhakrishnan et al., 2018).

Nano metal oxide particles as additives on biodiesel can reduce CO emissions due to the active catalytic effect on metal oxides. The catalytic effect on nano metal oxide increases the rate of mixing of fuel with air during the combustion process, so that the catalyst activity reduces the formation of CO and converts it to CO_2 (Pandian et al., 2017).

4.3 Hydrocarbon (HC)

Hydrocarbon (HC) is formed due to incomplete fuel combustion. Along with increasing power in diesel engines hydrocarbon (HC) emissions will also increase because more fuel injection is needed to maintain a constant speed at high power. Insufficient oxygen content in incomplete combustion can result in increased amounts of hydrocarbon (HC) in the flue gas (Annamalai et al., 2016).

The oxygen content in biodiesel contributes to HC oxidation thereby reducing the HC content in the flue gas. The addition of nano metal oxide can also increase the oxygen content in biodiesel. Moreover, tthe addition of Al_2O_3 nano metal oxide to zizipus jujube biodiesel methyl ester blended fuel (ZJME25) can reduce HC emissions by 25% at 25ppm and up to 36.11% at 50ppm addition compared without using nano metal oxide (Aalam et al., 2015). Cashew nut sold BD100 biodiesel can reduce HC in exhaust emissions by 20.83% compared to petrodiesel. And with the addition of nano Al2O3 to biodiesel biodiesel BD100 can reduce HC content in exhaust gas by 27.08% compared to petrodiesel.

5. Summary

This review provides an overview of the development of the use of nano metal oxide as a soluble nano catalyst in biodiesel. The conclusions of the above review are the addition of nano particles does not significantly affect the density, viscosity, and flash point values, but the cetane number has a relationship with the addition of nano particles. It means that the higher the concentration of nano particles, the higher the value of biodiesel cetane number. In other hand, the addition of nano metal oxide can reduce the brake specific fuel consumption (BSFC) of biodiesel because nano metal oxide contains oxygen molecules which can oxidize hydrocarbons and carbon monoxide. More fuel burned causes the energy produced is also greater and decreases fuel consumption. Moreover, the addition of nano metal oxide to biodiesel and its mixture did not significantly influence NOx, but CO and HC dropped significantly due to the catalytic effect of nano metal oxide on combustion in the combustion chamber.

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