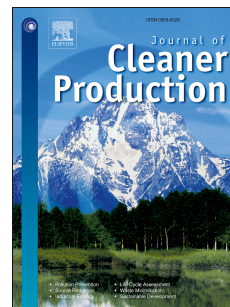


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Review of synthesis, characteristics and technical challenges of biodiesel based drilling fluids

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Review of Synthesis, Characteristics and Technical Challenges of Biodiesel Based Drilling Fluids

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Abstract

Over the last decades, biodiesel-based drilling fluids have drawn attention of researchers due to their biodegradability, low cost, high flash point compared to other synthetic-based drilling fluids. This review study discusses the development of biodiesel-based drilling fluids and analyzes the characteristics of biodiesels (based on the required range of values for their properties) that lead to better understanding in choosing the proper biodiesel for the drilling fluids. Although, using biodiesels as the base oil in drilling fluids, showed a favourable performance, it still poses some technical challenges such as excessive viscosity, poor pour point, low hydrolytic stability and water-in-biodiesel emulsion stability. In this study, recommended solutions including chemical modification of biodiesel's molecular structure, additives and mixing with other oils are presented. It was highlighted that the modification of biodiesel structure or the use of additives can be expensive and promote incompatibility with the other drilling fluids components. On the other hand, blending biodiesels with other base oils, showed promising solutions for the challenges of high viscosity, poor pour point and hydrolytic stability. It was found that the environmentally-friendly oils to mix with biodiesels, are synthetic esters, paraffins and linear alpha olefins. Furthermore, recent studies have suggested the use of nanomaterials (with hydrophobic characteristics) to stabilize water-in-biodiesel emulsion drilling fluids.

Keywords: Biodegradability, Biodiesel, Drilling Fluids, Ester, Nanomaterials, Pickering

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Abbreviations:

SBDF	Synthetic based drilling fluid	RBODB	Rice bran oil derived biodiesel	ARD	Absolute relative deviation	CB	Canola biodiesel	BWCO	Biodiesel from waste cooking oil
ECD	Equivalent circulation densities	CODB	Corn oil derived biodiesel	DODF	Diesel oil-based drilling fluid	ECETOC	European Centre for Ecotoxicology and Toxicology of Chemicals	DHFA	Dihydroxy fatty acid
OBDF	Oil-based drilling fluids	PFAME	Palm oil fatty acid methyl ester	LTODF	Low toxic oil-based drilling fluid	LAO	Linear alpha olefin	PP	Palm-based polyol
BBDF	Biodiesel based drilling fluid	SASO	Biodiesel based on almond seed oil	OLBDF	Olefin base drilling fluid	BOD	The biochemical oxygen demand	HLB	Hydrophilic-lipophilic balance
EBDF	Ester based drilling fluid	IMME	Indian mango methyl ester	LP	Linear paraffins	COD	The chemical oxygen demand	AV	Apparent viscosity
PV	Plastic viscosity	API	American petroleum institute	SMEBDF	Soybean oil methyl ester-based drilling fluid	PMA	Polymethacrylate	ES	The electrical stability
YP	Yield point	DSC	Differential scanning calorimetry	CBBDF	Canola BBDF	EO	Ethylene oxide	PAG	Polyalkylene glycols
LPLT	Low pressure Low temperature	HPHT	High pressure High temperature	WBDF	Water based drilling fluid	LCM	Lost circulation materials	PO	Propylene oxide
FCT	Filter cake thickness	FL	Filtrates loss	GS	Gel-strength	MWCNT	Multi-walled carbon nanotubes	TAN	Total acid number

1. Introduction

Synthetic-based drilling fluids (SBDFs) attract much attention among all other types of drilling fluids as SBDFs seems able to make the challenging balance between the performance and biodegradability in drilling operations (Praveen Kumar Jha, Mahto, and Saxena 2013; Swaco 2006). SBDFs were developed because the diesel and mineral oils failed to satisfy the new environmental requirements of biodegradability for the drilling cuttings discharge (Praveen K. Jha, Mahto, and Saxena 2017; Praveen Kumar Jha, Mahto, and Saxena 2014).

Diesel and mineral oils are still used in drilling processes in some countries because of their performance (high stability and lubricity) and viscosity profile (Agwu, Okon, and Udoh 2015). However, it should be reminded that mineral oils and diesel have low biodegradability, and high toxicity and handling costs. Since the eighties, many countries have banned using diesel and highly toxic mineral oils worldwide (e.g., the Gulf of Mexico and the North Sea). That is because they can cause damages to terrestrial, coastal, and aqua habitats and polluting water supplies especially those based on diesel, are highly persistent in sediments (Agwu, Isemin, and Akpabio 2015; Nelson, Liu, and Sommers 1984; S. Li, Osisanya, and Haroun 2016; Wai Li et al. 2016). The US Environmental Protection Agency put limitation guidelines and precise specifications to control cuttings discharge and the North Sea is urging on zero levels of cuttings discharge (United States Environmental Protection Agency 2000). Subsequently, the produced cuttings have to be transported onshore before release. Cuttings transportation increases operational costs as part of discharge management and environmental protections (E. Van Oort et al. 2004; Okie-Aghughu, Aluyor, and Adewole 2013; Dosunmu and Ogunrinde 2010). Similarly, the Arabian governments laid down corporate rules for offshore drilling operations, which insist on the transportation of all drill cuttings to onshore rather than discharging them offshore (Dosunmu and Ogunrinde 2010).

Consequently, vegetable oil-based drilling fluids have been considered as an alternative for diesel and highly toxic mineral oils (Aboulrous et al. 2015; SA, n.d.). They are biodegradable and have remarkably low toxicity levels (Fechhelm, Gallaway, and Farmer 1999). They are also less volatile due to their high molecular weights and high flash points (Atabani et al. 2013; Onuh et al. 2017; Growcock and Patel 2011). Still, their performance falls short in comparison with diesel and mineral oils due to their high viscosities. The seabed low temperature raise the viscosity of vegetable oil-based drilling fluids by three to four times. During circulation and drilling, the

viscous drilling fluid creates a high Equivalent Circulation Densities (ECD) which may fracture the formation and cause fluid loss (R Schlemmer et al. 2003; Robert Schlemmer, Khor, and Chandran 2007; McLean and Addis 1990). Therefore, in research studies, the main goal in designing environmentally friendly substitutes for oil-based fluids is to formulate a synthetic oil that can offer the same performance as diesel and mineral oils while it has improved bioaccumulation and degradation rate (Candler et al. 1995; American Petroleum Institute 2010; Burrows et al. 2001).

As a result, synthetic- or pseudo-oil-based drilling fluids (SBDFs) are introduced for drilling operations. It was reported that SBDFs are less toxic than oil-based drilling fluids (OBDFs), and they degrade faster than other oils. The cost of preparing a synthetic drilling fluid (SBDF) is high in comparison to oil-based drilling fluids (OBDFs) (Carpenter JF 2001; Dosunmu and Ogunrinde 2010), but SBDFs can minimize drilling time by 30–50% and consequently reduce the overall cost of the drilling a well (K 1996).

There are mainly five types of synthetic oils used in drilling fluids, esters, ethers, acetals, hydrocarbons (including linear paraffins, linear alpha-olefins, poly alpha-olefins, and internal olefins), and paraffin (G. Chen et al. 2003). Nowadays, all synthetic oils have been eliminated from use in drilling operations except esters, olefins, and paraffins; this is due to the negative environmental impacts they can have (Sinha et al. 2017; Ewy and Cook 1990; Coelho et al. 2005; DARLEY HCH 1969; Eric van Oort 2003; Lyons, Plisga, and Lorenz 2015). Also, SBDFs have an advantage over the vegetable oil-based drilling fluids as their rheology, and physical properties can be controlled by adjusting their chemical structure (Fornasier et al. 2017). Over the recent years, many studies have focused on biodiesels or vegetable oil-based esters as base oil for drilling fluids. Biodiesel is a long chain fatty acid monoester that is produced by the chemical reaction between oil or lipids with alcohol in presence of alkali as sodium hydroxide or potassium hydroxide. Biodiesel-based drilling fluids (BBDFs) have shown advantages over other ester-based drilling fluids because of their low cost, biodegradability, high thermal stability. However, BBDFs have some technical challenges such as the base oil hydrolysis, poor pour point and high viscosity.

In the rest of this review study, discussions on the development of biodiesel-based drilling fluids (BBDFs), requirements, biodegradability are presented. Then the potential solutions for the

BBDFs technical issues, were summarized that include the application of nanomaterials in emulsion stability issue.

2. Synthesis and Development of Biodiesel Based Drilling Fluids

Several studies were conducted on development biodiesel or esters based on vegetable oils to act as synthetic oils for ester-based drilling fluid (EBDF). The biodiesel can be prepared by more than one method such as pyrolysis, dilution with hydrocarbons blending, microemulsion, and transesterification. The most common used method is the transesterification. Generally, the transesterification is the reaction between vegetable oil with alcohol to produce biodiesel in the presence or absence of catalyst as shown in Table 1 (Growcock and Patel 2011; Photaworn, Tongurai, and Kungsanunt 2017).

Table 1: Biodiesel production by transesterification method (Singh and Singh 2010; Sharma, Singh, and Upadhyay 2008)

<i>Sample</i>	<i>Alcohol</i>	<i>Catalyst</i>	<i>Ration of alcohol to oil</i>	<i>Temperature (°C)</i>	<i>Yields (%)</i>
<i>Madhuca indica</i>	Methanol	H ₂ SO ₄ (1%, v/v)	0.3–0.35 (v/v)	60	98
<i>Peanut oil</i>	Methanol	NaOH	–	50	90
<i>Rubber seed oil</i>	Methanol	H ₂ SO ₄ (0.5%, v/v)	6:1	45	–
<i>Canola oil</i>	Methanol	NaOH (1%, w/w)	6:1	45	98
<i>Microalgae</i>	Methanol	Sulfuric acid	56:1	30	60
<i>Pongamia pinnata</i>	Methanol	KOH (1%, w/w)	10:1	105	92
<i>Soybean oil</i>	Supercritical methanol	Absence of catalyst	24 and CO ₂ /methanol = 0.1	280	98
<i>Sunflower oil</i>	Supercritical methanol and ethanol	No catalyst	40:1	200–400 (pressure 200 bar)	78–96
<i>Rice bran</i>	Methanol	Sulfuric acid	10:1	60	<96

Biodiesel (especially linear chain ones) are stable at standard conditions, but they are susceptible to hydrolysis under the harsh subsurface conditions i.e., high temperatures, high pressure, basic or acidic environments. These conditions will act as a catalyst for biodiesel hydrolysis (Helstrup, Chen, and Rahman 2004). Their rapid hydrolysis is due to their chemical structure as they contain an active carbon atom attached to two oxygen atoms and two terminal alkyl groups which can hydrolyze to produce the corresponding acid and alcohol, as shown in Figure 1.

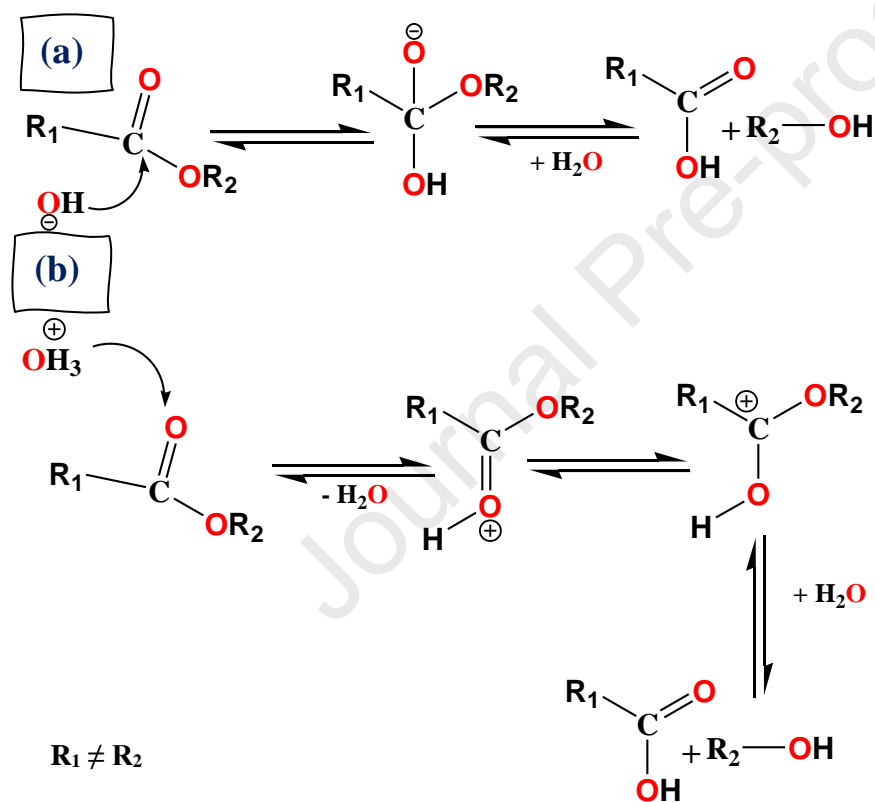


Figure 1: hydrolysis process of biodiesel under, (a) basic conditions and (b) acidic conditions

Generally, the hydrolysis process seems to be a significant disadvantage of using any esters in drilling fluids. On the other hand, it can also be an advantage as it offers fast biodegradation to

EBDFs according to different laboratory tests and seabed conditions (MI Swaco 2006). The other main merit of using EBDFs is their tolerance to acidic gases and cement contamination. Moreover, they do not damage polymeric components in drilling tools, as esters have a low aniline point (Swaco 2006).

Initially, synthetic esters were extensively used in the Gulf of Mexico and the UK sector of the North Sea. Drilling companies tend to blend the ester with synthetic hydrocarbons (olefins) to attain required drilling fluid performance (Swaco 2006; Growcock and Patel 2011). Researchers have synthesized esters from carboxylic acids and alcohol and used them as base oil for drilling fluids. Table 2 summarizes the synthetic esters used in drilling fluids.

Table 2: synthetic esters used as base oil for drilling fluids [BAT: Before Applying Temperature, AAT: After Applying Temperature]

Carboxylic acid	Alcohol	PV, cP		YP lb/100ft ²		Gel strength 10 sec. lb/100 ft ²		Temp. conditions	Lime content (g)	Reference
		BAT	AAT	BAT	AAT	BAT	AAT			
Lauric acid	Propanol	10	8	5	2	3	2	200 °F	7	Orji et al. (I. Orji, Ibezim-Ezeani, and Akaranta 2018)
Lauric acid	Isopropanol	18	15	12	7	7	5	200 °F	7	Orji et al. (I. Orji, Ibezim-Ezeani, and Akaranta 2018)
Octanoic acid	Ethanol	10	9	3	3	3	1	200 °F	7	Orji et al. (K. Orji, Ibezim-Ezeani, and Akaranta 2016)
Acetic acid	Octanol	17	7	3	1	2	1	200 °F	7	Orji et al. (K. Orji, Ibezim-Ezeani, and Akaranta 2016)
Oleic	Iso-butanol	46	41	35	32	21	29	Hot rolling for 16 h at 125 °C	1	Muller at al. (Muller H, Herold C, Von Tpavicza S, Nuess M, Zoellner W 1993)

<i>Oleic</i>	Methanol	21	23	6	6	7	9	Hot rolling for 48 h at 125 °C	2	Muller and Hrold (Muller H, Herold C 1992)
<i>Lauric</i>	n-hexanol	28	28	11	14	7	8	Hot rolling for 16 h at 125 °C	1	Muller at al. (Muller H, Herold C, Von Tpavicza S, Grimes DJ, Braun J-M 1992)
<i>Caprylic</i>	2-Ethylhexanol	21	22	15	11	8	5	Hot rolling for 16 h at 125 °C	1	Muller at al. (Muller H, Herold C, Von Tpavicza S, Nuess M, Zoellner W 1993)
<i>Capric</i>	2-Ethylhexanol	26	31	14	14	8	9	Hot rolling for 16 h at 125 °C	1	
<i>Capric</i>	n-octanol	26	28	17	14	8	7	Hot rolling for 16 h at 125 °C	1	
<i>Iso-nonanic</i>	n-octanol	27	29	12	13	7	7	Hot rolling for 16 h at 125 °C	1	
<i>Caprylic/capric</i>	Isotridecyl	44	55	13	24	8	10	Hot rolling for 16 h at 125 °C	2	
<i>Acetic</i>	Isotridecyl	30	29	13	17	8	8	Hot rolling for 16 h at 125 °C	2	Muller at al. (Muller H, Herold C, Von Tpavicza S, Nuess M 1994)

Mueller et al. studied the performance EBDFs with different esters that synthesized from synthetic acids and alcohol. The esters synthesized from different fatty acids with normal and branched

hexyl alcohols. These esters have low viscosity and exhibited a stable EBDF rheological property even after dynamic ageing at 125 °C for 16 hours (Carlson and Hemphill 1994; Photaworn, Tongurai, and Kungsanunt 2017; Muller H, Herold C, Von Tpavicza S 1993; Muller H, Herold C, Von Tpavicza S, Grimes DJ, Braun J-M 1992; Muller H, Herold C, Von Tpavicza S, Nuess M, Zoellner W 1993). One of the typical esters used in drilling fluids is 2-Ethylhexyl dodecanoate (Muller H, Herold C, Von Tpavicza S 1993; Muller H, Herold C, Von Tpavicza S, Grimes DJ, Braun J-M 1992; Muller H, Herold C, Von Tpavicza S, Nuess M, Zoellner W 1993). However, the ester resulted from branched longer alcohols (C-atoms more than 8) and medium-chain acids failed to fulfil the technical requirements (Muller H, Herold C, Von Tpavicza S, Nuess M, Zoellner W 1993). For example, EBDF based on isotridecyl caprylic ester exhibited excessive viscosity.

On the other hand, the esters resulted from a short saturated carboxylic acid and branched longer alcohol, exhibited high hydrolytic stability and low viscosity (Muller H, Herold C, Von Tpavicza S, Nuess M 1994). For example, EBDFs based on isotridecyl acetate ester showed stability even after dynamic ageing for 72 hours at 125 °C (Muller H, Herold C, Von Tpavicza S, Nuess M 1994). However, all investigated EBDFs were formulated with a minimum lime content (1 or 2 g) to prevent the hydrolysis process during ageing.

Generally, the drilling fluid properties depend on the structure of ester and the chain length of the alcohol and the carboxylic acid moieties. For example, the drilling fluid that contain branched-chain ester as isopropyl laurate (filtrate after 30 minutes = 3.5 ml) exhibited a less drilling fluid loss than the drilling fluid that contain linear chain ester as methyl laurate (filtrate after 30 minutes = 6.2 ml) (R. A. Ismail, Kamis, and Saik Foo 2000; A. R. Ismail, Kamis, and Foo 2001). Habib et al. (Abdul Habib et al. 2014) studied 2-Ethylhexyl oleate based drilling fluid which showed excessive viscosity. On the other hand, 2-Ethylhexyl laurate-based drilling fluid exhibited better drilling fluids performance after 16 hours of dynamic ageing at 250 °C (Jassim et al. 2016).

Due to the high cost of the synthetic esters, esters derived from vegetable oils or biodiesels were suggested to be used as base oil for drilling fluids. Moreover, biodiesels have surpassed expectations as they have all the oils' favourable properties, and there are no excessive viscosities (Friedhelm and Conn 1996; Setyawan 2018). Table 3 showed ester derived from vegetable oils used as base oils for drilling fluids. Esters also have no toxicity effect on marine habitats. Vegetable

oil-based esters biodegrade in the aqua environment, and they have no risk to human health. EBDFs fulfil the US Environmental Protection Agency standards and EPA's requirements for the biodegradation test (United States Environmental Protection Agency 2000). They also have a high flash point and low vapour pressure, so they will not cause fires (Sulaimon, Adeyemi, and Rahimi 2017; von Tapavicza 2005; Yassin and Kamis 1990; Amin et al. 2010; Abdul Razak Ismail and Engineering 2001; Degouy et al. 1993; Peresich, Burrell, and Prentice 1991). Mueller et al. (Muller H, Herold C, Von Tpavicza S 1993) used iso-butyl rapeseed oil ester as synthetic oils for the EBDF. These esters have a low viscosity but low thermal stability that caused high plastic viscosity (PV) after dynamic ageing for 16 hours at 125 °C. However, both methyl rapeseed oil ester and methyl soybean oil ester exhibit more stable drilling fluid performance even after ageing (Muller H, Herold C 1992).

Table 3: Biodiesels used in drilling fluids (Yield point (YP) and gel strength (after 10 sec. (GEL)) were in lb/100 ft² and plastic viscosity (PV) was in cP) [BAT: Before Applying Temperature, AAT: After Applying Temperature]

<i>Oil type</i>	Alcohol	Emulsion Type	PV		YP		GEL		Temperature	Lime Content	Acid Number of biodiesel (mg KOH/g)	Reference
			BAT	AAT	BAT	AAT	BAT	AAT				
<i>Mango seed oil</i>	Methanol	Oil –in– Water (O/W) emulsion	23	23	16	15	7	6	Aging for 20 h at 110 °C	No lime added	0.7	Kumar et al. (Kumar et al. 2020)
<i>Palm oil</i>	Methanol	Water–in– Oil (W/O) emulsion	—	—	17	19	22	17	Aging for 16 h at 140 °C	No lime added	Not Available	Said and El-sayed (Said and El-Sayed 2018)
<i>Palm Kernel Oil</i>	2-ethyl hexanol	Water–in– Oil (W/O) emulsion	37	30	16	14	7	5	Aging for 16 h at 125 °C	1.5 g	< 0.3	Muller at al. (Muller H, Herold C, Von Tpavicza S, Grimes DJ, Braun J-M 1992)
<i>Soybean oil</i>	Methanol	Water–in– Oil (W/O) emulsion	19	23	11	6	7	6	Aging for 48 h at 125 °C	2 g	≤ 1.0	Muller and Herold (Muller H, Herold C 1992)
<i>Rapeseed oil</i>	Methanol	Water–in– Oil (W/O) emulsion	21	25	6	6	6	7	Aging for 48 h at 125 °C	2 g	≤ 1.0	Muller and Herold (Muller H, Herold C 1992)

<i>Rapeseed oil</i>	Iso-butanol	Water-in- Oil (W/O) emulsion	35	62	21	24	12	12	Aging for 16 h at 125 °C	2 g	< 1.0	Muller et al. (Muller H, Herold C, Von Tpviczka S 1993)
<i>Canola oil</i>	Methanol	Water-in- Oil (W/O) emulsion	24	19	42	36	9	5	ABT was 60 °C and AAT was at 100 °C	NA	Not Available	Soomro et al. and Paul and Adewale (Soomro, Soomro, and Brohi 2020; Paul and Adewale 2018)
<i>Almond seed oil</i>	Methanol	Water-in- Oil (W/O) emulsion	24	20	18	15	7	6.8	Aging for 16 h at 140 °C	10.5 g	0.78	Oseh et al. (Oseh et al. 2019)
<i>Rice bran oil</i>	Not Available	Oil -in- Water (O/W) emulsion	—	—	—	—	—	—	The rheological parameters weren't investigated.	1 g	Not Available	Ismail et al. (Abdul Razak Ismail, Ismail, et al. 2014)
<i>Corn oil</i>	Not Available	Oil -in- Water (O/W) emulsion	—	—	—	—	—	—	The rheological parameters weren't investigated.	1 g	Not Available	Ismail et al. (Abdul Razak Ismail, Ismail, et al. 2014)

Also, one of the most promising biodiesel, used as base oil for a W/O emulsion drilling fluid, is palm oil fatty acid methyl ester (PFAME) (Said and El-Sayed 2018). PFAME showed stable rheological profile before and after hot rolling up to 150°C for 16 hours as shown in Figure 2. Also, it showed stable rheological parameters at different temperature ranging from 15 °C to 65 °C. However, there was a change in the rheological profile after ageing at 177 °C, due to the hydrolysis of PFAME and degradation of the drilling fluids (Sulaimon, Adeyemi, and Rahimi 2017). Moreover, PFAME based drilling fluids showed an increase in rheology in case of contamination by sea water.

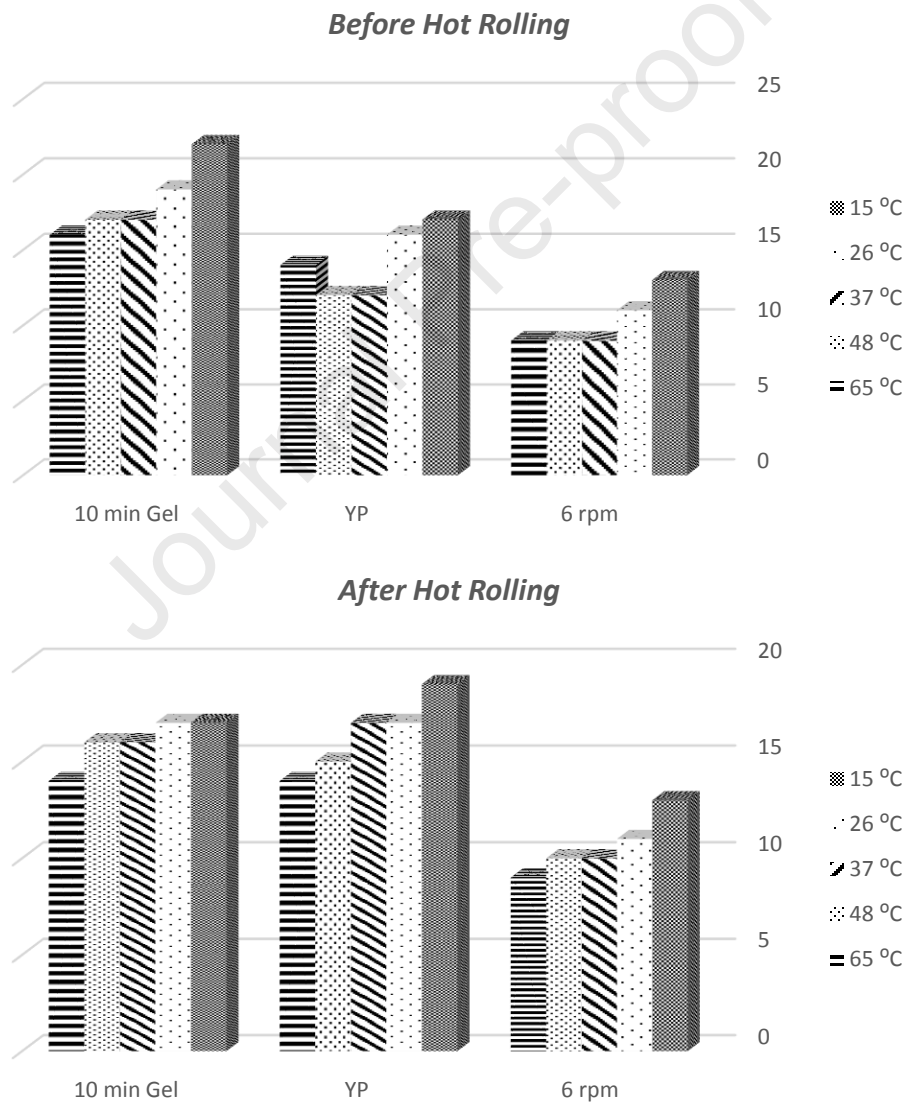
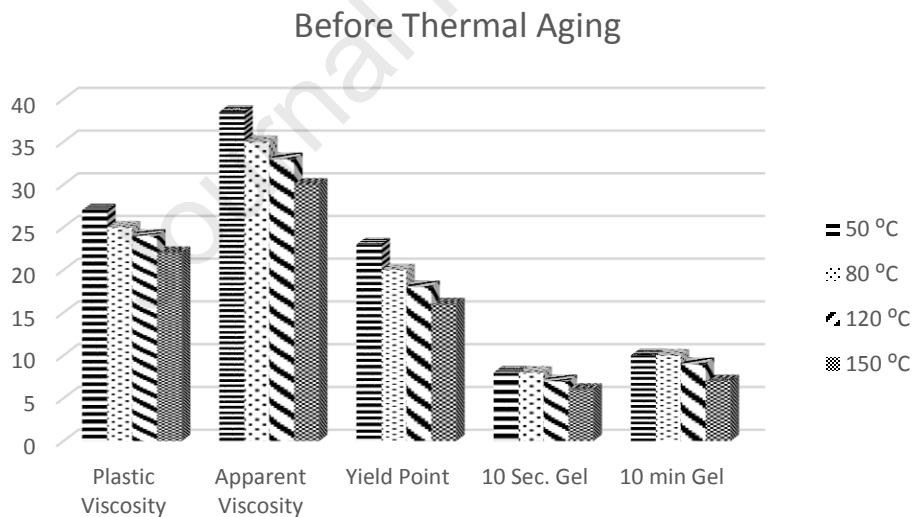


Figure 2: the rheological properties for PFAME drilling fluid before and after hot rolling [permission to reuse from Said and El-Sayed, 2018, (Said and El-Sayed 2018)] (Yield point and gel strength (after 10 min. (GEL)) were in $lb_f/100\ ft^2$)

Moreover, Oseh et al. used biodiesel based on almond seed oil (SASO) as a base oil for W/O emulsion drilling fluid (Oseh et al. 2019). The experimental rheological data (such as; PV, YP, YP/PV ratio, 10-sec and 10-min gel strengths) show that SASO based drilling fluid has stable rheological parameters at different temperatures (50, 80, 120 and 150 °C) whether it was before and after thermal ageing for 16 hours at 50 °C as shown in Figure 3. Also, SASO- based drilling fluids showed less shale swelling compared to diesel-based drilling fluids and this is because the biodiesel molecules have larger polarities than diesel molecules, due to the presence of carbonyl groups in biodiesel. This causes the biodiesel to have a strong interaction with the shale sample and reduced its swelling rate.



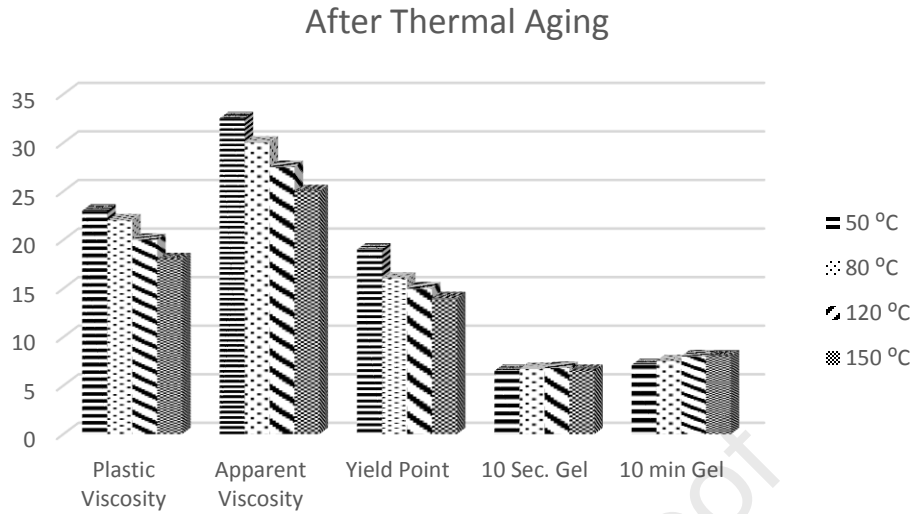


Figure 3: temperature effect on the rheological properties of SASO drilling fluid before and after ageing [permission to reuse from Oseh et al., 2019, (Oseh et al. 2019)] (Yield point and gel strength (after 10 sec. and 10 min.) were in $lb/100\text{ ft}^2$, plastic and apparent viscosity were in cP)

Kumar et al. (Kumar et al. 2020) used the Indian mango methyl ester (IMME) as a base oil for O/W emulsion drilling fluids. The IMME drilling fluid system showed promising chemical, physical, rheological, and filtration properties compared to diesel and mango seed oil. The new IMME shows no toxicity and the IMME based drilling fluid showed superior performance compared to the diesel-based drilling fluid as shown in Figure 4. Also, it was observed that the fluid loss was reduced by more than 50% as IMME drilling fluid (3 mL) compared to the commercial (diesel)-based drilling fluid (8 mL), that is required property while drilling through sensitive formations.

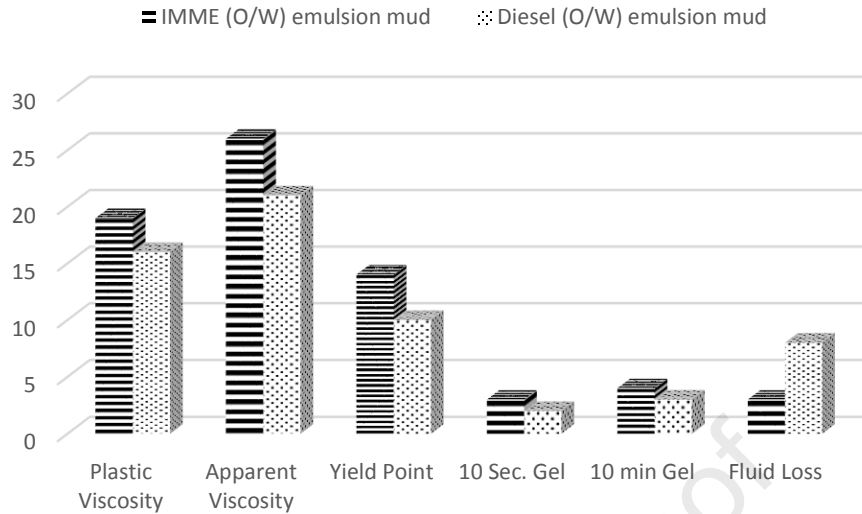


Figure 4: the rheological properties of IMME- and diesel-based drilling fluid [permission to reuse from Kumar et al., 2020, (Kumar et al. 2020)] (Yield point and gel strength (after 10 sec. and 10 min.) were in $lb/100 ft^2$, plastic and apparent viscosity were in cP and fluid loss was in ml)

Most of the above mentioned studies faces one common issue that the formulation of drilling fluids usually contains lime to activate the emulsifier (Muller H, Herold C, Von Tpavicza S 1993; Muller H, Herold C, Von Tpavicza S, Grimes DJ, Braun J-M 1992; Jassim et al. 2016). If lime content increases to 3 g per 350 ml of mud, the EBDF will have unstable properties due to ester's hydrolysis (Muller H, Herold C, Von Tpavicza S 1993). However, Oseh et al avoid this issue by using almond oil biodiesel with acid value around 0.7 mg KOH/g oil. The lime content was 10.5 g and it used to neutralize the free fatty acids (Oseh et al. 2019). Also, Patel and Friedheim (Patel AD 2003) suggested using a lime-free EBDF to avoid this problem. They used different oil/water ratios and different drilling fluid weights. The lime-free EBDF exhibited a stable drilling fluid performance up to 177 °C (Patel AD 2003; A. D. Patel 1999; Arvind D. Patel 1999; Muller H, Herold C 1993).

Another issue that faces the EBDF is the poor flowability at low temperatures, BBDFs have poor pour points compared to the other EBDF. However, this can be improved by changing the chemical structure through chemical reactions such as transesterification, but this method is not favourable for oxidative stability (Srivastava and Prasad 2000). Other than that, pour points can also be enhanced by the addition of pour point depressants. Also, The ester pour point properties can also

be improved by mixing with other base oil as olefins (Razali et al. 2018). These technical issues and its possible resolutions are discussed in later section of this review.

3. Biodiesel Characteristics as Base Oil in Drilling Fluids

According to the American Petroleum Institute (API) standards ((API) 2011), there are tests to evaluate the properties of any base oil before using them in drilling fluids, such as; specific gravity, kinematic viscosity, low-temperature fluidity (pour point), and flash point. Therefore, typical base fluids should have low viscosity, low pour point, and high flash point. Furthermore, biodiesel physical properties can be adjusted by managing the length and the degree of branching of the alcohol moiety and the saturation degree of the oil moiety (Candler et al. 1995). In this section, the requirements needed for biodiesel to act as base oil for drilling fluids, were discussed.

3.1. Density (Specific Gravity)

The specific gravity for the base fluid can affect the drilling fluid density. Drilling fluid density is necessary to control of formation pressures, and an increase in drilling fluid density increases the drilling fluid's capacity to carry drilled cuttings (Candler et al. 1995; (API) 2011). The specific gravity requirement for any base oil, should be less than 0.95 g/ml (Mesher Shaun 2003). The density for most biodiesels don't change from its parent vegetable oils as presented in Table 4.

Table 4: specific gravity for some vegetable oil-based esters (biodiesel) and their corresponding vegetable oils

<i>Biodiesel</i>	Density, g/ml	Corresponding vegetable oil	Density of vegetable oil, g/ml	Reference
<i>Castor methyl ester</i>	0.924	<i>Castor oil</i>	0.78	(Keera, El Sabagh, and Taman 2018)
<i>Mango seed oil methyl ester</i>	0.88	<i>Mango seed oil</i>	0.911	(Kumar et al. 2020; Vijayaraj and Sathiyagnanam 2016)
<i>Jatropha oil ester</i>	0.877	<i>Jatropha oil</i>	0.905	(Atabani et al. 2013)

<i>Palm oil methyl</i>	0.859	<i>Palm oil</i>	0.899	(Atabani et al. 2013)
<i>Palm Kernel Oil 2-ethyl hexyl</i>	0.861	<i>Palm Kernel Oil</i>	0.891	(Muller H, Herold C, Von Tpavicza S, Grimes DJ, Braun J-M 1992; MUSA, n.d.)
<i>Coconut oil ester</i>	0.866	<i>Coconut oil</i>	0.908	(Atabani et al. 2013)
<i>Rapeseed oil isobutyl</i>	0.872	<i>Rapeseed oil</i>	0.901	(Muller H, Herold C, Von Tpavicza S 1993; Atapour and Kariminia 2011)
<i>Rapeseed oil methyl</i>				(Muller H, Herold C 1992; Atapour and Kariminia 2011)
<i>Soybean oil methyl</i>	0.869	<i>Soybean oil</i>	0.907	(Muller H, Herold C 1992; Atabani et al. 2013)
<i>Canola oil methyl</i>	0.866	<i>Canola oil</i>	0.904	(Paul and Adewale 2018; Atabani et al. 2013)
<i>Almond seed oil ester</i>	0.894	<i>Almond seed oil</i>	0.915	(Oseh et al. 2019; Atapour and Kariminia 2011)
<i>Corn oil biodiesel</i>	0.93	<i>Corn oil</i>	0.916	(Abdul Razak Ismail, Ismail, et al. 2014; Veljković et al. 2018)
<i>Rice bran oil biodiesel</i>	0.89	<i>Rice bran oil</i>	0.913	(Abdul Razak Ismail, Ismail, et al. 2014; Zaidel et al. 2019)

3.2. Kinematic Viscosity

The base oil's kinematic viscosity can indicate the drilling fluid ability to carry cuttings and other drilling solids to the surface while it is pumpable (Burrows et al. 2001). Low viscosity synthetic oils lead to better pumpability, reduction in the viscous drag and less cost for total drilling fluid (Eckhout et al. 2000; Dawson RB 1995), (Talalay et al. 2015). It is also an indication of the needed volume of the ester to formulate the SBDF.

Kinematic viscosity increases with increasing the chain length, branching degree (molecular weight), and unsaturation level within the chain of biodiesel. Biodiesels have high kinematic

viscosity, however, the viscosity can be controlled by changing the molecular structure of the used alcohol (R. 2020). For instance, low molecular weight biodiesels synthesized from linear alcohol, have low viscosity. (R. 2020; Murphy and Zisman 1950; Pitt 1990; Fechhelm, Gallaway, and Farmer 1999).

In general, low kinematic viscosity for the base fluid is desirable and the preferred viscosity value is in range of 2.3-5 cSt (Yassin and Kamis 1990)- (Mesher Shaun 2003). In another study, researchers found that the base oil should have a kinematic viscosity at the range of 1 to 6 cSt at 40 °C (Ellice M, Helmy S 1996). In another study, Patel et al. suggested that hydrocarbon-based synthetic oil should have a viscosity in the range of 1.5 – 3.4 cSt (Patel AD, McGlothlin RE, Bleier RD 1993). Generally, esters should have the same kinematic viscosity as hydrocarbon-based synthetic oil (olefins or paraffin) to exhibit good drilling fluids performance (R. A. Ismail, Kamis, and Saik Foo 2000; A. R. Ismail, Kamis, and Foo 2001; Amin et al. 2010; Abdul Habib et al. 2014; Jassim et al. 2016; Magathevan T, Nur'aini S, Yusuf R, Nik R 2014; Lyons, Plisga, and Lorenz 2015; Goncalves JT, De Oliveira MF 2007). Internal olefins have a lower viscosity than linear olefins due to the presence of a central olefin bond (Swaco 2006). The viscosity of esters varies with temperatures and pressures conditions through the drilling process due to carbon-oxygen double linkages (Candler et al. 1995).

The high viscosity of vegetable oils, especially at low temperatures, can cause high equivalent circulating densities (ECD) (Swaco 2006). Biodiesel or vegetable-based synthetic esters have surpassed expectations as they have the preferred oil's properties without the high viscosity (Yassin and Kamis 1990). Biodiesels exhibit significantly lower viscosity than their corresponding vegetable oils due to their low molecular weight. Table 5 presented kinematic viscosity for some vegetable oil-based esters and their corresponding vegetable oils.

Table 5: kinematic viscosity of vegetable oil-based esters (biodiesel) and their corresponding vegetable oils

<i>Biodiesel</i>	<i>Viscosity @ 40°C, cSt</i>	<i>Corresponding vegetable oil</i>	<i>Viscosity of vegetable oil @ 40°C, cSt</i>	<i>Reference</i>
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<i>Castor oil ester</i>	11.13	Castor oil	580	(Keera, El Sabagh, and Taman 2018)
<i>Mango seed oil methyl</i>	3.9	Mango seed oil	3.6	(Kumar et al. 2020; Vijayaraj and Sathiyagnanam 2016)
<i>Jatropha oil ester</i>	4.94	Jatropha oil	48.09	(Atabani et al. 2013)
<i>Palm oil methyl</i>	4.69	Palm oil	41.93	(Atabani et al. 2013)
<i>Palm Kernel Oil 2-ethyl hexyl</i>	7 @ 20 °C	Palm Kernel Oil	28.89	(Muller H, Herold C, Von Tpvavicza S, Grimes DJ, Braun J-M 1992; MUSA, n.d.)
<i>Coconut oil ester</i>	3.14	Coconut oil	27.64	(Atabani et al. 2013)
<i>Rapeseed oil isobutyl</i>	24 @ 5 °C	Rapeseed oil	37.6	(Muller H, Herold C, Von Tpvavicza S 1993; Atapour and Kariminia 2011)
<i>Rapeseed oil methyl</i>	4.37			(Muller H, Herold C 1992; Atapour and Kariminia 2011)
<i>Soybean oil methyl</i>	4.37	Soybean oil	31.74	(Muller H, Herold C 1992; Atabani et al. 2013)
<i>Canola oil methyl</i>	4.53	Canola oil	35.71	(Paul and Adewale 2018; Atabani et al. 2013)
<i>Almond seed oil ester</i>	4.31	Almond seed oil	34.2	(Oseh et al. 2019; Atapour and Kariminia 2011)
<i>Corn oil biodiesel</i>	6.18	Corn oil	32	(Abdul Razak Ismail, Ismail, et al. 2014; Veljković et al. 2018)
<i>Rice bran oil biodiesel</i>	4.68	Rice bran oil	39.8	(Abdul Razak Ismail, Ismail, et al. 2014; Zaidel et al. 2019)

3.3. Flash Point

Synthetic oil should have a higher flash point than the surface temperature which is greater than 70 °C (Shaun Mesher, Chester Schesnuik 2003) during the drilling process. Flash point is

imperative to ensure conformance with health and safety requirements. The US Department of Transportation stated that fluids with flash points more than 90 °C should be considered low fire-risk fluids (Burke, C. J., & Veil 1995). Generally, the ester bond exhibits polarity which results in a higher flash point than a hydrocarbon with the same molecular weight. Consequently, the use of esters or biodiesels in SBDF minimizes the risk of fire.

Chemical structures and molecular weights are the primary factors to determine the value of flash point. Flash point increases with linearity and molecular weight (Srivastava and Prasad 2000; Atabani et al. 2013). Additionally, the biodiesel synthesized from linear compounds have a higher flash point than branched ones with the same molecular weight. Flash point decreases with the increase in branching due to reduction in the intermolecular dispersion forces and consequently preventing close packing (Murphy and Zisman 1950; Pitt 1990). Moreover, branching causes reduction in the surface area which makes the molecules more compact (Yasa et al. 2017; Kemnitz et al. 2010). Table 6 shows the flash point values for ester derived from vegetable oils and their corresponding vegetable oils. Esters derived from vegetable oils such as, palm oil, coconut oil, and canola oil have a lower flash point than their corresponding vegetable oils (above 200 °C).

Table 6: flash point of esters derived from vegetable oils (biodiesel) and their corresponding vegetable oils

<i>Ester</i>	Flash point, °C	Corresponding vegetable oil	Flash point of vegetable oil, °C	Reference
<i>Castor oil ester</i>	217	<i>Castor oil</i>	292	(Keera, El Sabagh, and Taman 2018)
<i>Mango seed oil methyl</i>	252	<i>Mango seed oil</i>	263	(Kumar et al. 2020; Vijayaraj and Sathiyagnanam 2016)
<i>Jatropha oil ester</i>	186	<i>Jatropha oil</i>	258	(Atabani et al. 2013)
<i>Palm oil methyl</i>	178	<i>Palm oil</i>	252	(Atabani et al. 2013)
<i>Palm Kernel Oil 2-ethyl hexyl</i>	165	<i>Palm Kernel Oil</i>	205	(Muller H, Herold C, Von Tpavicza S, Grimes DJ, Braun J-M 1992; MUSA, n.d.)

<i>Coconut oil ester</i>	118.5	<i>Coconut oil</i>	264	(Atabani et al. 2013)
<i>Rapeseed oil isobutyl</i>	180	<i>Rapeseed oil</i>	250	(Muller H, Herold C, Von Tpavicza S 1993; Atapour and Kariminia 2011)
<i>Rapeseed oil methyl</i>	150			(Muller H, Herold C 1992; Atapour and Kariminia 2011)
<i>Soybean oil methyl</i>	202	<i>Soybean oil</i>	280	(Muller H, Herold C 1992; Atabani et al. 2013)
<i>Canola oil methyl</i>	186	<i>Canola oil</i>	290	(Paul and Adewale 2018; Atabani et al. 2013)
<i>Almond seed oil ester</i>	169	<i>Almond seed oil</i>	250	(Oseh et al. 2019; Atapour and Kariminia 2011)
<i>Corn oil biodiesel</i>	87	<i>Corn oil</i>	254	(Abdul Razak Ismail, Ismail, et al. 2014; Veljković et al. 2018)
<i>Rice bran oil biodiesel</i>	188	<i>Rice bran oil</i>	210	(Abdul Razak Ismail, Ismail, et al. 2014; Zaidel et al. 2019)

3.4. Pour Point (Cold Flow Property)

Rheological properties of drilling fluids at low temperature depend on the pour point values, which is also known as the lowest temperature at which the drilling fluid is capable of flowing. It is an essential factor for cold or arctic drilling conditions, the preferred pour point value is less than -30°C (Shaun Mesher, Chester Schesnuik 2003). Pour points are usually measured using DSC (Differential Scanning Calorimetry) under ASTM D97 (ASTM International: West Conshohocken 2017). Synthetic oils with low pour points have good low-temperature flowability (Ellice M, Helmy S 1996).

Biodiesels have poor pour point compared to diesel. However they have good pour point compared to the pure vegetable oils which lose their fluidity below -10 °C (Swaco 2006). Table 7 presents values of pour point for some vegetable oil-based esters and their corresponding vegetable oils.

Table 7: pour points of some vegetable oil-based esters and their corresponding vegetable oils

<i>Ester</i>	Pour point, °C	Corresponding vegetable oil	Pour point of vegetable oil, °C	Reference
<i>Castor oil ester</i>	-16	<i>Castor oil</i>	15	(Keera, El Sabagh, and Taman 2018)
<i>Mango seed oil methyl</i>	-33	<i>Mango seed oil</i>	-9	(Kumar et al. 2020; Vijayaraj and Sathiyagnanam 2016)
<i>Jatropha oil ester</i>	10	<i>Jatropha oil</i>	21	(Atabani et al. 2013)
<i>Palm oil methyl</i>	15	<i>Palm oil</i>	23.6	(Atabani et al. 2013)
<i>Palm Kernel Oil 2-ethyl hexyl</i>	-5	<i>Palm Kernel Oil</i>	21	(Muller H, Herold C, Von Tpavicza S, Grimes DJ, Braun J-M 1992; MUSA, n.d.)
<i>Coconut oil ester</i>	4	<i>Coconut oil</i>	24	(Atabani et al. 2013)
<i>Rapeseed oil isobutyl</i>	-15	<i>Rapeseed oil</i>	10	(Muller H, Herold C, Von Tpavicza S 1993; Atapour and Kariminia 2011)
<i>Rapeseed oil methyl</i>	-10			(Muller H, Herold C 1992; Atapour and Kariminia 2011)
<i>Soybean oil methyl</i>	1	<i>Soybean oil</i>	13	(Muller H, Herold C 1992; Atabani et al. 2013)
<i>Canola oil methyl</i>	-9	<i>Canola oil</i>	15	(Paul and Adewale 2018; Atabani et al. 2013)
<i>Almond seed oil ester</i>	-9	<i>Almond seed oil</i>	14	(Oseh et al. 2019; Atapour and Kariminia 2011)
<i>Corn oil biodiesel</i>	0	<i>Corn oil</i>	15	(Abdul Razak Ismail, Ismail, et al. 2014; Veljković et al. 2018)
<i>Rice bran oil biodiesel</i>	-3	<i>Rice bran oil</i>	11	(Abdul Razak Ismail, Ismail, et al. 2014; Zaidel et al. 2019)

Pour point of BBDF depend on the composition of the fatty acid in the biodiesel. For instance; long carbon chain fatty acids have a negative effect on pour point value of biodiesel. Long carbon

chain fatty acids have a higher crystallization's temperature compared to the shorter carbon chain fatty acids. Ramos et al. (Ramos et al. 2009) found that the peanut oil biodiesel have poor pour point and that is because of the presence of long carbon chain of lignoceric (22 carbon atoms) and behenic acids (24 carbon atoms) and the high level of saturation (no double bond in composition).

Additionally, It is well known that saturated fatty acids such as lauric, palmitic and stearic have a poor pour point than unsaturated fatty acids (as, linoleic and oleic) which have a three dimension structure (R. 2020). Unsaturated fatty acid molecules have high energy, thereby weakening the intermolecular attraction force and crystallize at a lower temperature compared to saturated fatty acid molecules (Vijayaraj and Sathiyagnanam 2016). Consequently, biodiesel derived from the unsaturated fatty acids have a higher pour point than those derived from the saturated fatty acids (Salih, Salimon, and Yousif 2012; 2011; Salimon, Salih, and Yousif 2012). For example, biodiesel derived from palm oil or coconut oil have a poor pour point than esters derived from soybean oil because palm oil and coconut oil contains high saturated fatty acids such as lauric and palmitic fatty acid (Verma, Sharma, and Dwivedi 2016; Govindapillai, Jayadas, and Bhasi 2009). In contrast, soybean oil has a low pour point as it contains a high unsaturated fatty acid content as oleic and linoleic (Ali, Hanna, and Cuppett 1995; Adhvaryu and Erhan 2002). Also, The degree of branches in the chain of fatty acids have a positive effect on the pour point of biodiesel as the branched biodiesel has a 3D configuration, that increase the thermodynamic force needed for crystallization, and consequently, reduction in the crystallization's temperature of the biodiesel (G 2016). This fact is matched with the values in Table 7, as the pour point of rapeseed oil isobutyl is $-15\text{ }^{\circ}\text{C}$ while rapeseed oil methyl has a pour point of $-10\text{ }^{\circ}\text{C}$

In addition to the oil composition, the presence of transesterification intermediates (such as triglycerides, diglycerides and monoglycerides) in biodiesel, can deteriorate cold flow properties of biodiesel. As the transesterification intermediates have high molecular stacking due to the presence of hydroxyl moiety in their molecules which consequently induce intermolecular force mainly due and therefore affect negatively on biodiesel cold temperature properties (Fernando et al. 2007; Ma and Hanna 1999). Also, the methods of biodiesel production, may affect its pour point. Transesterification of frying oil and rapeseed oil using bio-based alcohol (biobutanol), enhance its pour point values according to Bouaid et al. (Bouaid, Martinez, and Aracil 2009). Seames et al. (Seames et al. 2010) used thermal cracking process to produce canola oil-based biodiesel with better cold

flow properties. Researchers have developed many methods to overcome the poor pour point of biodiesel. These methods were discussed in Section 5.2.

Moreover, these previous physiochemical properties of biodiesel can be predicted by various models. These models depend mainly on the type of biodiesel's feedstock (oils or animal fats) as the type of fatty acids are the key for the model's calculations (Saxena, Jawale, and Joshipura 2013). These models are tabulated in Table 8.

Table 8: models for prediction of biodiesel's properties

Property	Name of Model	Model Equation	Requirements for Models	Source of Biodiesel	%ARD*	Reference
Density	Luis et. al.	$\rho = 0.8463 + \frac{4.9}{Mi} + 0.0118N$	ρ = Density of FAME M_i = mol. Wt. of i^{th} FAME, N = no. of double bonds	Beef Tallow, Soybean Oil	N.A.	(Ramírez-Verduzco, Rodríguez-Rodríguez, and Jaramillo-Jacob 2012)
	Allen et. al.	$\ln \mu_m = \sum_{i=1}^n y_i \ln \mu_i$	μ_m = Mean viscosity of mixture μ_i = Viscosity of pure component 'i' y_i = Mass fraction of component 'i'	Coconut oil, Palm oil, Rapeseed oil, Peanut oil, Soybean oil, Canola oil	8.04	(Tesfa et al. 2010)
Viscosity	Ceriani et. al.	$\ln \mu_i = \sum_k N_k \left[A_{1k} + \frac{B_{1k}}{\left(\frac{T}{K}\right)} - C_{1k} \ln\left(\frac{T}{K}\right) - D_{1k} \left(\frac{D}{K}\right) \right]$	N_k = No. of groups k in a molecule i M = Component mol. Wt. A_{1k} , B_{1k} , C_{1k} , D_{1k} = Obtained from the regression of the experimental data.	N.A.	7.62	(Nasab et al. 2018)
	Chang and Liu	$\eta = 0.235N_C - 0.699N_{DB} - 3.648$	η = Viscosity of biodiesel N_C = weighted avg. no. of carbon atoms, N_{DB} = weighted avg. no. of double bonds	Palm oil, Olive oil, Peanut oil, Rapeseed oil, Soybean oil, Sunflower oil, Grape oil, Almond oil, Corn oil	7.60	(Gopinath A, Puan S 2009)
	Yung et. al	$\eta = 0.235N_C - 0.648N_{DB}$	η = Viscosity of biodiesel	Soybean oil	5.45	(Su et al. 2011)

		NC = weighted avg. no. of carbon atoms NDB = weighted avg. no. of double bonds			
	Luis et. al.	$\ln \eta_i = 12.503 + 2.496 \ln M_i - 0.178N$	η = Viscosity of biodiesel NC = weighted avg. no. of carbon atoms NDB = weighted avg. no. of double bonds	Beef Tallow, Soybean Oil	2.57 (Ramírez-Verduzco, Rodríguez-Rodríguez, and Jaramillo-Jacob 2012)
<i>Pour point</i>	Yung et. al.	$PP = 18.88N_C - 1.000U_{FAME}$	PP = pour point of biodiesel, N_C = weighted avg. no. of carbon atoms, U_{FAME} = Composition of FAMEs in Biodiesel	Soybean oil	1.43 (Su et al. 2011)
	Sarin et. al.	$PP = 0.571P_{FAME} - 12.24$	PP = pour point of biodiesel, P_{FAME} = Content of Palmitic acid methyl ester	Palm oil, Jatropa oil, Pongamia oil	1.56 (Sarin et al. 2009)
<i>Flash Point</i>	Yung et. al.	$T_f = 23.362N_C + 4.854N_{DB}$	T_f = Flash Number of Biodiesel, N_C = weighted avg. no. of carbon atoms, N_{DB} = weighted avg. no. of double bonds	Soybean oil	1.81 (Su et al. 2011)

* Percentage ARD (Absolute Relative Deviation) is calculated by

$$\% \text{ARD} = \frac{1}{N} \sum_i^N \left| \frac{X_{exp} - X_{cal}}{X_{exp}} \right| \times 100 \text{ where } X_{exp} = \text{Experimental value, } X_{cal} = \text{Calculated value, } N = \text{Number of experimental value.}$$

4. Performance of BBDFs

Performance of drilling fluids is key for the success of the drilling operation. The performance of drilling fluids can be determined by many factors such as fluids stability under harsh conditions of temperatures and pressures, cuttings carrying capacity (or hole cleaning), drilling fluid waste management and biodegradability, and their cost. The detailed discussions on the key factors are presented in the following sections.

4.1. Stability under Harsh Conditions

BDFs have shown a competitive high temperature performance compared to commercial ODFs. As shown in Figure 5, palm oil biodiesel-based drilling fluids (PFAME) performance is comparable to the commercial base oil (Escaid-110) and within acceptable range. Also, PFAME showed a competitive performance in the HTHP filtration test compared to the drilling fluid formulation from commercial base oil (palm oil). The amount of filtrate collected in 30 minutes was 3.9 ml at 149°C and 1000 psi differential pressure, and the filter cake had a thickness of 0.5×1/32 inch compared to palm oil based drilling fluids which produced a thicker filter cake (Said and El-Sayed 2018).

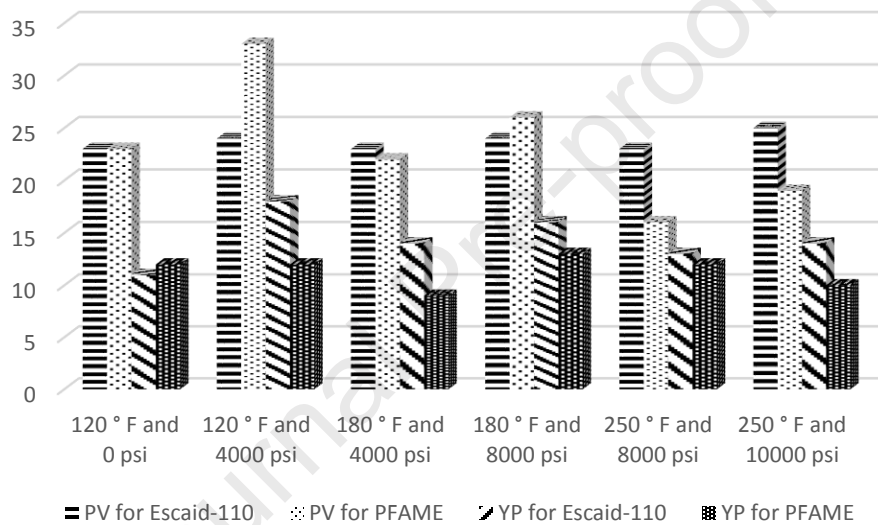


Figure 5: Rheological properties of biodiesel (PFAME) and commercial base oil (Escaid-110) under high temperatures and high pressures. (Yield point (YP) was in $lb_f/100 ft^2$, plastic viscosity (PV) was in cP)

Moreover, biodiesel based on almond seed oil (SASO) showed a competitive rheological results compared to diesel based drilling fluids (commercial used drilling fluid) as shown in Figure 6 (Oseh et al. 2019). SASO based drilling fluid showed comparable results with diesel based drilling fluids in terms of filtration properties as presented in Figure 7. The experimental data were measured at different temperatures (120 and 150 °C) before and after thermal ageing for 16 hours at 50 °C. The electrical stability value for SASO based drilling fluid was 1123 Volts while the value for diesel based drilling fluid was 927 Volts at temperature 120 °C.

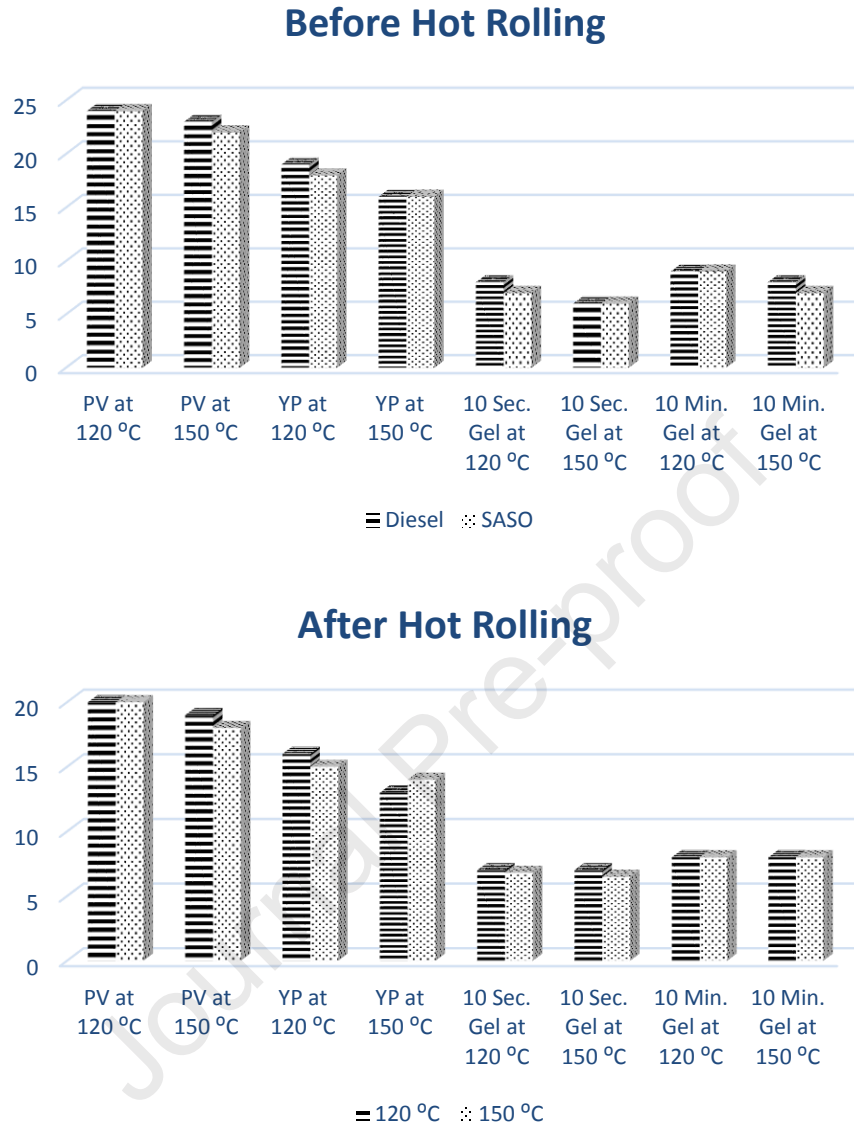


Figure 6: Rheological properties of biodiesel (SASO) and commercial base oil (diesel) before and after thermal aging. (Yield point (YP), 10 Sec. Gel and 10 Min. Gel were in lb/100 ft², plastic viscosity (PV) was in cP)

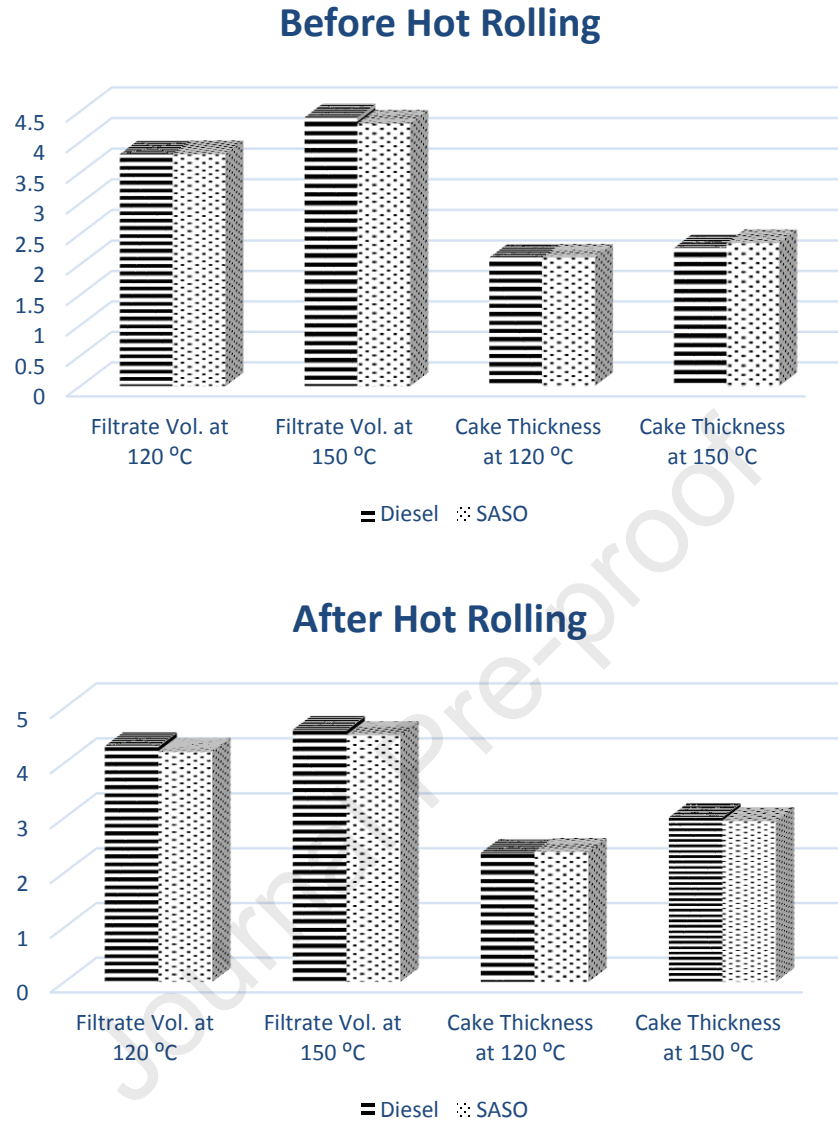


Figure 7: Filtration properties of biodiesel (SASO) and commercial base oil (diesel) before and after thermal aging. (Filtrate Vol. was in ml and Cake Thickness was in mm)

Generally, biodiesel-based drilling fluids exhibited higher thermal stability compared to the other synthetic based drilling fluids. For example, palm oil methyl EBDP showed satisfied rheological properties at 135 °C (Magathevan T, Nur'aini S, Yusuf R, Nik R 2014). In contrast, the glycerol palm oil based drilling fluid exhibited a remarkable reduction in viscosity (the viscosity reading at 600 rpm reduced from 200 to 42 cp) at 150 °C (Marbun et al. 2013).

4.2. Hole Cleaning

The performance of drilling fluids can be determined by the capability to carry the cuttings out of the borehole (Lyons, Plisga, and Lorenz 2015; Adeboye and Oyekunle 2016; Husin et al. 2015). Hole cleaning indicates the drilling fluids' capacity to transport and suspend the cuttings and the other solids. Hole cleaning relies on many parameters such as, drilling fluid properties (as rheology and density), annular velocity, well design, rotary speed, and cuttings characteristics (shape, density, size and aggregation) (Adeboye and Oyekunle 2016; Kenny and Hemphill 1996; Valluri et al. 2006). Hole-cleaning is especially important in horizontal and highly deviated drilling (Piroozian et al. 2012; Caenn and Chillingar 1996).

Generally, the ester-based drilling fluids showed superior ability to suspend and carry cuttings in comparison to the diesel oil-based drilling fluid (DODF) and low toxic oil-based drilling fluid (LTODF). Based on a study by Kenny and Hemphill, the EBDF exhibited a stable flow index while the flow index for DODF and LTODF reduced by approximately 9% (Kenny and Hemphill 1996). The flow index reduction means that the fluid tends to flow less on the narrow side and eventually will cause lost circulation, excessive drag, inefficient hole cleaning, and barite sag (Kenny and Hemphill 1996; Song et al. 2017). Ester-based drilling fluids contributes less excessive drag as it has much fewer friction factors than DODF and LTODF (Valluri et al. 2006). The lubricating properties of synthetic oil (ester) is the main reason for the lower friction factors readings.

There was a comparison between the hole cleaning potential of olefin base drilling fluid (OLBDF) and ester-based drilling fluids (EBDF). According to this study, OLBDF possesses a much less 6 rpm reading than EBDF (Eckhout et al. 2000). Furthermore, OLBDF exhibited a lower yield point compared to the EBDF with similar density. This is an indication that EBDF performs better than OLBDF in terms of transporting cuttings due to its higher yield point (Amin et al. 2010). The oil/water ratio is the main factor in changing the gel strength and yield point (Sauki, Shah, and Bakar 2015; A. R. Ismail, Kamis, and Foo 2001).

There are very limited studies in the area of hole cleaning by biodiesel-based drilling fluid (BBDF). However, according to the swab and surge simulations done by Said and El-sayed (Said and El-Sayed 2018), the palm oil methyl ester based drilling fluid maintained a flat rheology profile, and consequently the ECD improved. The palm oil methyl ester-based drilling fluid showed a good

hole cleaning in different zones of the well at different temperatures and also at the low annular velocity region in the marine riser.

Nanoparticles have shown improvement in the hole cleaning function of water-based drilling fluids. Based on a recent study conducted by Boyou et al 2019 (Boyou et al. 2019) they hypothesized that interparticle interactions between nanoparticles and cuttings (sands) are higher in nano-enhanced water-based drilling fluids. This is due to higher surface area of nanoparticles that contribute to higher electrical charges on their surfaces and hence support the suspension of the cuttings in the mud. This could increase drag and lift forces on cuttings to overcome gravitational and cohesion forces. As of yet, there has been no investigation on the use of nanoparticles in BBDFs and analyzing their hole cleaning performance.

4.3. Biodegradability and Ecotoxicity of BBDF:

Generally, esters can be readily biodegradable in anaerobic conditions because of oxygen atoms in their structure (Peresich, Burrell, and Prentice 1991; Caenn and Chillingar 1996). Ester linkages (internal oxygen atoms) and carbon-carbon double bonds (unsaturated olefins) are more biodegradable than carbon-carbon single bonds (saturated olefins and paraffin). In any ester's biodegradation, the first resultants are the corresponding alcohol and acid moieties through the hydrolysis process by extra-cellular esterase enzymes (Getliff, J., Roach, A., Toyo, J., & Carpenter 1997). The final products are combined gases (CO₂ and methane) (David Herman and Roberts 2006), as shown in Figure 8. The olefins biodegradation pathway is the same as the ester (Figure 8).

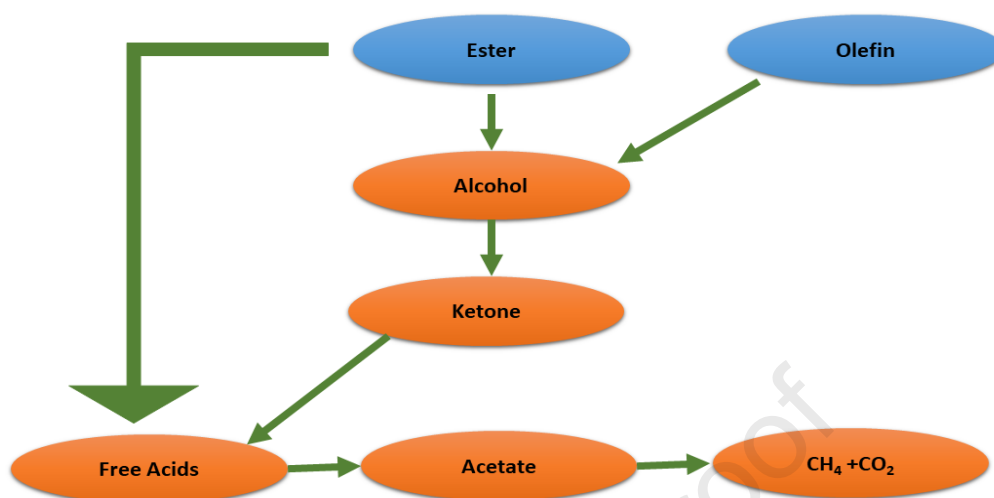


Figure 8: schematic representation of the anaerobic biodegradation pathway of esters and alkenes

Based on samples collected from the North Sea offshore, the waste of ester-based drilling fluids can be degraded up to 80% after one month (Peresich, Burrell, and Prentice 1991). According to a screening test done by ECETOC (European Centre for Ecotoxicology and Toxicology of Chemicals), it was found that ninety percent of esters can be degraded into methane gas and carbon dioxide in comparison to linear alpha olefin (LAO) (Steber, Herold, and Limia 1995; Blanchet, Audibert, and Argillier 1998). Ester traces in sediments near platforms were much lower when compared other synthetic-based drilling (Tait et al. 2004). These findings support the theory that esters can be rapidly biodegraded in sediments.

Linear Olefins and Internal Olefins have almost similar anaerobic biodegradation rates. Furthermore, the rate at which they anaerobically biodegrade is extensively faster than paraffin. In an anaerobic condition, Linear Olefins IO and LAO exhibit good biodegradability at a rate of 56% and 64%, respectively, (Candler, Rabke, and Leuterman 1999; D. Herman and Roberts 2005). In another study, the ester-based drilling fluids take 11 months to completely degrade after post-drilling operations, whereas the PAO and LAO based drilling fluids take more than 2 years (Candler et al. 1995; Neff, McKelvie, and Ayers 2000; Daan et al. 1996). However, unsaturated olefins are more biodegradable than saturated ones (Candler et al. 1995). In aerobic

biodegradation, the n-paraffin degrades at different rates from 23% to 86% (Neff, McKelvie, and Ayers 2000), while diesel degrades at a rate of 60% (Marchal et al. 2003). In anaerobic conditions, n-paraffin slowly degrades by 5%, while diesel, by 3% (Candler, Rabke, and Leuterman 1999; D. Herman and Roberts 2005).

The biodegradation rate of paraffin and olefins decreases with increasing chain length and molecular weight. Consequently, high molecular weight of synthetic oil such as PAOs are less biodegradable than lower molecular weight ones such as, IOs (Candler et al. 1995). The primary degradation mechanism of synthetic base oil is through microbes' metabolism to produce harmless byproducts (carbon fragments) that can be a source of nutrition, as shown in figure 8.

Additionally, Herman & Roberts studied the relationship between the biodegradability of commercial esters and their chemical structure (such as chain length, branching, and the unsaturation level) (David Herman and Roberts 2006). These studies conducted an anaerobic closed bottle test on marine sediment samples from Galveston Bay in Texas.

They found that the unbranched ester biodegraded faster than branched ester. The ethyl oleate (C18 unsaturated ester) biodegraded faster than the ethyl octadecanoic (C18 saturated). The fastest biodegradation was hexyl octanoate with 87%. They also found that esters with total carbon atoms from 12 to 18 gave the best biodegradability. That means the short chain synthetic esters are more biodegradable than biodiesel, which is normally more than 18 carbon atoms. However, biodiesel biodegraded faster than diesel. For instance, the almond seed methyl ester (SASO) based drilling fluid showed higher biodegradation than diesel-based drilling fluid. According to Oseh et al., the aerobic biodegradation of SASO-based drilling fluid presented 83% and 78% biodegradation with *Penicillium* sp. and *Staphylococcus* sp., respectively. In contrast, diesel only exhibited 23.7% and 25.2% biodegradation with *Staphylococcus* sp. and *Penicillium* sp. respectively. The anaerobic biodegradation showed that the SASO-based drilling fluid biodegraded by a rate of 62% and 60% with *Penicillium* sp. and *Staphylococcus* sp., respectively, while the diesel-based drilling fluid biodegraded by a rate of 31% and 29%, respectively (Oseh et al. 2019).

Also, corn oil derived biodiesel (COBD), rice bran oil derived biodiesel (RBODB) and palm oil methyl ester were categorized as non-toxic substances compared to diesel and Sarapar as their LC50 value were above 10,000 ppm (Abdul Razak Ismail, Ismail, et al. 2014) as in table 9.

Table 9: LC50 values for biodiesel and commercial base oils using juvenile of seabass and red snapper

Drilling Fluids	Test Specification	Juvenile Of Seabass	Juvenile Of Red Snapper
Diesel (Commerical base oil)	10,000 ppm	5,800 ppm	7,080ppm
Sarapar (Commerical base oil)	10,000 ppm	8,700 ppm	10,700ppm
Palm Oil Metyl Ester	10,000 ppm	28,800 ppm	22,900ppm
Corn Oil Derived Biodiesel (CODB)	10,000 ppm	35,400 ppm	33,800ppm
Rice Bran Oil Derived Biodiesel (RBODB)	10,000 ppm	40,700 ppm	39,000ppm

4.4. An Economic Evaluation of Biodiesel Based Drilling Fluids:

Usually, the synthetic-based drilling fluids' formulation contains 60% to 80% of the base oil. Thus, the cost of synthetic drilling fluids depends mainly on the price of the base oil. The most highly cost base oils are acetals and synthetic linear paraffin. Acetals are considered the highest in price because of their complicated synthesis route as they require the reaction between one mole of aldehyde and two moles of alcohol in the presence of acid and then elimination of water (Swaco 2006). On the other hand, paraffin is prepared by two methods. The first is catalytic hydrogenation of carbon monoxide (low-pressure Fischer-Tropsch synthesis) (Vik, E.A., n.d.). The second is a multistep refinery process (from a petroleum feedstock). Drilling cuttings from linear paraffins (LP), prepared by Fischer-Tropsch synthesis, are permitted by EPA for offshore discharge in the Gulf of Mexico (Fechhelm, Gallaway, and Farmer 1999). Olefins consider cheaper than other synthetic oils. Olefins starting materials are less expensive than esters (Swaco 2006). Olefins prepared by the oligomerization of ethylene using triethyl aluminium as a catalyst at high temperatures (Swaco 2006). The order of base oils regarding cost is: Acetals > Fischer-Tropsch Paraffin > Petroleum based Paraffin > Synthetic esters > Olefins > Biodiesel

Recently, biodiesels or esters derived from vegetable oils become competitive with olefins in term of cost and drilling fluid performance. For example, the water/oil ratio in soybean oil methyl ester-based drilling fluid (SMEBDF) was 40/60 vol.%. This ratio was also achieved using additives such

as pour point depressant, antioxidant, and emulsifier. SMEBDF exhibited a proper drilling fluid performance in deepwater operation at temperature 46 °C (Goncalves JT, De Oliveira MF 2007).

The amount of biodiesel in the drilling formulation is less than 70%, reducing the cost of drilling fluid. The cost of biodiesel is even more competitive to diesel. Based on laboratory study, the cost of methyl ester from almond seeds (~83\$/bbl) is less than diesel (136.8 \$/bbl) (Oseh et al. 2019; Soomro, Soomro, and Brohi 2020). Figure 9 shows the preparation cost of the methyl ester of almond seed oil (SASO) BBDF and canola BBDF (CBBDF).

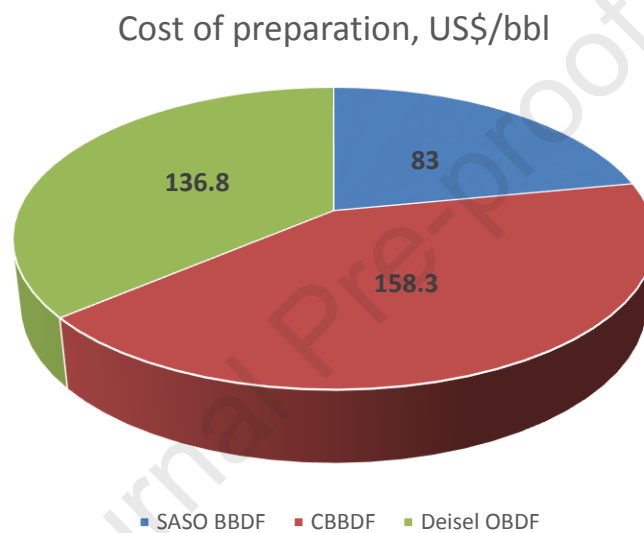


Figure 9: the preparation cost of the diesel-, SASO biodiesel- and canola biodiesel-based drilling fluids.

Another important factor in determining the cost analysis of synthetic oil is waste management. For example, the preparation cost of CBBDF is higher than diesel; however, the cost of its waste management is less than diesel (Soomro, Soomro, and Brohi 2020). Moreover, the price of waste management of Canola biodiesel (CB), SASO based drilling fluid are much less compared to diesel cost (Oseh et al. 2019; Soomro, Soomro, and Brohi 2020) as shown in Table 10.

Table 10: economic evaluation for Canola biodiesel (CB), SASO and diesel-based drilling fluids

Retail prices

Diesel **SASO** **CBBDF**
OBDF **BBDF** (Soomro,
(Oseh et al. 2019) (Oseh et al. 2019) Soomro,
and
Brohi
2020)

<i>Transportation of drilling effluents to disposal site (\$/bbl)</i>	3.0	3.0	3.0
<i>Commercial disposal of drilling effluents (\$/bbl)</i>	57.0	28.5	33
<i>Transportation + commercial disposal cost of drilling effluents (\$/bbl)</i>	60.0	31.5	38
<i>The total cost of wastes management of 1 bbl of drilling muds (\$/bbl)</i>	60.0	31.5	38

5. Technical Challenges Associated with BBDFs:

The structure and oil/fat source of biodiesel can result in some technical issues such as excessive high viscosity, poor pour point and low hydrolytic stability. In this section, these technical issues and possible solutions are discussed.

5.1. Excessive High Viscosity

The high viscosity of BBDF often occurs due to partial hydrolysis as the reaction of fatty acid anions and salt forms soap like structures that act as stabilizing agents (Muller H, Herold C 1993). Hydrolysis can be minimized by adding small concentrations of antioxidants. Furthermore, a study reported that the addition of oleophilic based amines to BBDF was able to combat its high viscosity even after 16 hours of static ageing (Muller H, Herold C 1993). These oleophilic base amines act as an alkaline reserve in the drilling fluid formulation. These additives were used with soybean methyl EBDF and reduced the viscosity to acceptable values (Goncalves JT, De Oliveira MF 2007). In another study, d-limonene reduced the thickening issue of BBDF and maintained its flowability, especially under arctic conditions. It also acts as a pour point depressant\

5.2. Poor Cold Flow Properties

BBDF has poor pour point compared to other SBDFs. As, biodiesels have poor flowability at low temperature. Researchers have many attempts to improve the cold flow of biodiesel. Based on the fact that the branched chain biodiesels have better cold flow properties than the straight chain biodiesels, researchers have tried to enhance the cold flow property of biodiesel by

transesterification of straight chain fatty acid with branched alcohols (Lee, Johnson, and Hammond 1995) or isomerization reaction (C. Yori et al. 2006). Lee et al. (Lee, Johnson, and Hammond 1995) proposed Trans-esterification of fatty acids with branched chain alcohols instead of methanol, to enhance the cold flow of biodiesel. Lee et al. (Lee, Johnson, and Hammond 1995) found that isopropyl soyate had better cold flow property than propyl soyate, however the use of highly branched alcohols caused incomplete trans-esterification reaction and lower yields of biodiesel. Also, the use of branched alcohols resulted in high increase in biodiesel production cost as the branched alcohols are more expensive than methanol. The isomerization overcome this high production cost, resulted from trans-esterification with branched chain alcohols, Yori et al (C. Yori et al. 2006) improve the cold flow quality of biodiesel confirmed by isomerizing soy oil with methanol and using solid acid crystals upto 275 °C. Also, pour point values can be enhanced by further chemical modification such as epoxidation (Shaun Mesher, Chester Schesnuik 2003).

Additionally, Researchers have used winterization techniques to enhance the cold flow properties of biodiesel (Fernando et al. 2007). Winterization method is proceed by gradually cooling the biodiesel, and the formed crystal of high melting point are then separated from the biodiesel by filtration (Fernando et al. 2007). The formed crystals are mainly crystals of the saturated fatty esters and consequently the produced biodiesel has a lower content of saturated fatty esters. Dunn et al. (Dunn, Shockley, and Bagby 1996) found that that the cold flow property of methyl soybean biodiesel was improved by winterization. The percentage content of methyl oleate and linoleate was increased due to reduction in the content of methyl palmitate by a factor of three. However, this particular method increases the production cost because of the low yield of the biodiesel production and the complicated long procedure (Knothe, Krahl, and Van Gerpen 2010).

Other than that, pour points can also be modified by the addition of pour point depressants. This method is more favoured economically and technically than other methods in enhancing the cold flow properties of biodiesel. Examples of pour-point depressants are 2-Ethylhexyl oleate, isobutyl oleate, trimethylolpropane trioleate, diisodecyl adipate, pentaerythritol tetraoleate, and glycol-based materials (Erhan et al. 2008; Erhan, Sharma, and Perez 2006). The pour point depressants work by gathering on the formed crystals' surface to prevent the crystal growth and the formation of gel network (Soni, Kiranbala, and Bharambe 2008). Wang et al. (Wang, Cao, and Han 2014) examined the effect of polymethacrylate (PMA) on cold flow properties of biodiesel from waste

cooking oil (BWCO). The results showed that the addition of 0.04% polymethacrylate decreased the PP of BWCO by 8 °C. Ming et al. (Ming et al. 2005) used Tween-80, dihydroxy fatty acid (DHFA), and palm-based polyol (PP), to improve the cold flow property of palm oil biodiesel. The result showed that the addition of 1.0 vol. % DHFA has the most reduction effect to the PP of palm oil biodiesel blends.

Another type of additives is the ozonized oil which prepared by exploring the vegetable oil to a mixture of oxygen and ozone. Ozonized oil is preferred to be prepared from the same vegetable oil as the biodiesels. However, This technique may decrease the flash point of the biodiesel (El Rafie and Attia 2008).

5.3. Low Hydrolytic Stability

The ability of ester to stand against the water or moisture is called hydrolytic stability (R. 2020). Ester is hygroscopic compounds that can attract and keep water molecules. Due to this hygroscopic nature, most ester can hydrolyze easily at low temperatures (Fregolente, Wolf Maciel, and Oliveira 2015). The hydrolytic stability of esters depend on its chemical structure and properties such as polarity, acid value, moisture content, and hydroxyl number (R. 2020; Boyde 2000; Ingold 1930; Taft 1952; Bender 1960).

Under basic conditions, biodiesel can be hydrolyzed to alcohol and fatty acids anions. Calcium soap forms due to the reaction between the fatty acid anions and lime present in the drilling fluid formulation. These calcium soaps have a high value of HLB (hydrophilic-lipophilic balance), which causes destabilization and deformation of water in oil emulsion and consequently affects the rheological properties of the drilling fluids (Muller H, Herold C, Von Tpavicza S, Nuess M, Zoellner W 1993; Muller H, Herold C, Von Tpavicza S, Grimes DJ, Braun J-M 1992; Muller H, Herold C, Von Tpavicza S, Nuess M 1994; Muller H, Herold C 1992). Also, an excessive amount of lime can cause a disturbance in the BBDF rheology, especially at high temperature [86].

Hydrolysis reaction may also occur under acidic conditions like the presence of carbon dioxide or hydrogen sulfide. (Galindo et al. 2013; Rydholm, Anseth, and Bowman 2007; Mundle et al. 2010; Z. Zhang et al. 2018). Under anaerobic conditions, the enzyme can hydrolyze the ester (Ghattas et al. 2017). It is easy to detect biodiesel-based drilling fluids' hydrolysis after dynamic ageing (Sulaimon, Adeyemi, and Rahimi 2017). Moreover, the increment in alkalinity can cause an

increment in the apparent viscosity (AV) but lower the electrical stability (ES). These low values of electrical stability indicates weak emulsion stability of drilling fluids (Growcock, Ellis, and Schmidt 1994). Consequently, the ester can be utilized in drilling fluids with minimal lime content at temperatures around 300°F (Eckhout et al. 2000; Goncalves JT, De Oliveira MF 2007; Sulaimon, Adeyemi, and Rahimi 2017; Amin et al. 2010; Peresich, Burrell, and Prentice 1991). The hydrolytic stability of ester can be measured by a beverage bottle test (ASTM 2015). The hydrolytic activity for ester-based drilling fluids can be evaluated by the presence of alcohol in the gas chromatography (Arvind D. Patel 1999; W. Li et al. 2016).

However, researchers overcame hydrolytic stability by preparing the biodiesel from branched alcohol (R. 2020). Example for the branched alcohol is 2-ethylhexanol, isoamyl alcohol etc. These branches are vital as it causes a steric effect (Boyde 2000). Wagner et al. introduce sterically hindered branches that can enhance the hydrolytic stability of ester (Wagner, Luther, and Mang 2001). A steric effect influences the reaction rate as the atoms occupy space in the molecule, as Cleaves suggested (Cleaves 2015). Short-chain and low molecular weight biodiesel are easily hydrolyzed compared to branched biodiesel. The biodiesels have a high degree of branching and high molecular weight, thus causes a steric effect which prevents hydrolysis (Boyde 2000), (Fink 2012; Muller H, Herold C, Von Tpacicza S, Nuess M 1994).

Researchers have used anti-hydrolysis additives to overcome the hydrolysis issues related to biodiesel. Some of these additives include, carbodiimides, polycarbodiimides or surface-modified macromolecules (J P 2013; Stloukal et al. 2016; Tang et al. 1997). Stloukal et al. studied the effect of Carbodiimide additive (bis 2,6-diisopropylphenyl carbodiimide) as an anti-hydrolysis additive (Stloukal et al. 2016). However, it has not been investigated using any ester-based drilling fluid formulations. According to Greaves and his colleagues, polyalkylene glycols (PAG) were used to reduce ester's hydrolysis rate (Greaves and Zaugg Hoozemans 2018). They enhanced the hydrolytic stability of two vegetable oils (canola oil and sunflower oil) and three synthetic esters (estolide, saturated trimethylolpropane ester and TMP trioleate) using a polyalkylene glycol (PAG). Their findings stated that the hydrolytic stability for canola oil enhanced when copolymers: ethylene oxide (EO), propylene oxide (PO) (ratio 30:70), was added, as shown in figure 10. The increase in TAN (total acid number) for each ester indicates the hydrolysis rate, as shown from the ASTM D2619 test method. In another study, Daoust et al. using a second oil phase

to avoid the hydrolysis process (Daoust, Jennifer A., Lecomte, Jean-paul H., Liles, Donald Taylor, Liu, Yihan, Roggow II 2018). They used W/O/W or O/W/O emulsion instead of W/O or O/W emulsion and the second oil was silicon-based oil. Three phases emulsion wasn't investigated in any synthetic-based drilling fluids so far.

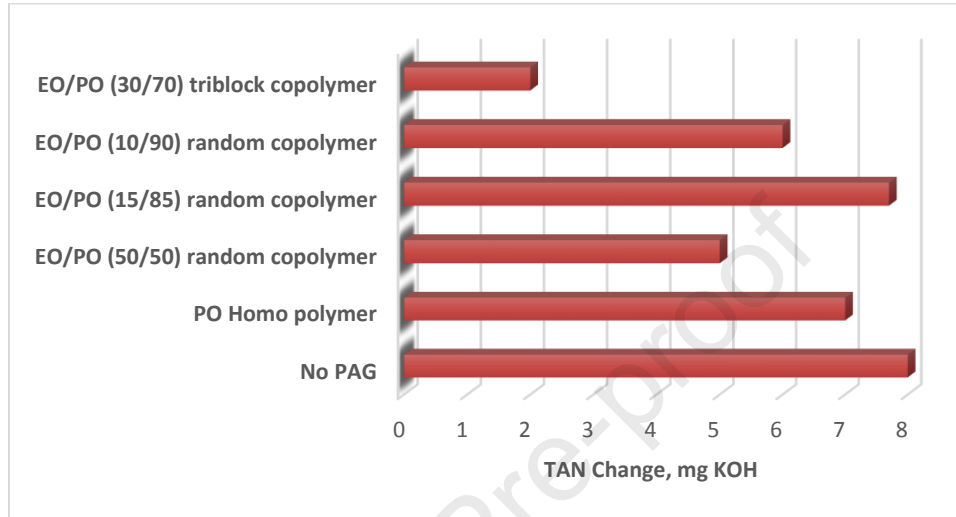


Figure 10: hydrolytic stability enhancement for canola oil using 10% PAGs. [permission to reuse from Greaves and his colleagues (Greaves and Zaugg Hoozemans 2018)]

5.4. Water-in-Biodiesel Emulsion Stability:

In general, water in oil emulsions in OBDF and SBDF possess emulsion stability challenges especially in HPHT conditions. Nanoparticles exhibit good emulsion performance as Pickering emulsion stabilizers in OBDF and SBDF due to their unique properties such as; high surface area, shape and structure, tunable wettability as shown in Table 10 (Kazemzadeh, Sharifi, and Riazi 2018; Guo and Zhang 2019), rheology modifier as well as stability in harsh conditions [212-215].

Table 10: summary of nanoparticles used in OBDF and SBDF

Authors	Type of Mud	Type of Oil	Nanoparticles	Nanoparticles Function	Results
Madkour et al. (Madkour et al. 2016)	OBDF	Diesel oil	Graphene and Carbon nanotubes	Pickering emulsion stabilizer and Filter loss agent	The study tested 0.5 wt % of graphene, and multi-walled carbon nanotube (MWCNT) reduced filtration loss under HPHT at 302°F and 500 psi.

Contreras et al. (Alsaba 2014)	OBDF (invert emulsion)	Not specified	Iron and Calcium based NPs	Pickering emulsion stabilizer and Filter loss agents	At HTHP, the iron-based NPs are more compatible than calcium-based ones with lost circulation material (LCM). At LPLT, iron-based NPs caused more reduction in filtration loss than calcium-based ones.
Anoop et al. (Anoop et al. 2014)	OBDF	Mineral oil	nano-silica (20 nm)	Pickering emulsion stabilizer and Rheology modifier	2.0 vol% of nano-silica (20 nm) exhibited high plastic viscosity at ambient conditions and stabilized the rheological properties under HPHT conditions
Wagle et al. (Wagle, Al-Yami, and Al Abdullatif 2015)	OBDF	Not Specified	Not specified	Pickering emulsion stabilizer and Rheology modifier	NPs enhanced the rheological performance of drilling fluid under HPHT (250 and 302o F) conditions
Katende et al. (Katende et al. 2019)	OBDF	Sarapar 147	silica nanoparticles	Pickering emulsion stabilizer and Rheology modifier	The concentration of 0.5 ppb nanosilica enhanced the rheological properties of OBDF
Zakaria et al. (Zakaria, Husein, and Hareland 2012)	OBDF (invert emulsion)	Not specified	Not specified	Filter loss agent and Pickering emulsion stabilizer	0.74% wt % of (20-40 nm diameter) NPs reduced filtration loss up to 70% under LPLT condition
Zhang et al. (J. Zhang et al. 2015)	OBDF	Not specified	Not specified	Pickering emulsion stabilizer and Filter loss agent	2.0wt% of NPs reduced fluid loss and filter cake thickness under HPHT condition at 194° F.
Sharma et al. (Sharma, Ravi, Lukasz Grzybek 2016)	OBDF (invert emulsion)	Mineral oil	silica particles (particle size of 1 nm to 500 nm)	Pickering emulsion stabilizer	The nanoparticles mixed with the brine solution to densify the brine. Nanoparticles (NPs) and acted as Pickering emulsion stabilizers in invert emulsion drilling fluids
Liu et al. (Liu et al. 2020)	OBDF (invert emulsion)	Diesel oil	A combination of fumed silica nanoparticles (NPs) and amine surfactant	Pickering emulsion stabilizer	The combination used as smart or pH-responsive Pickering The produced stable water-in-oil emulsions form only at pH of 2-12.
Ismail et al. (Abdul Razak Ismail, Rashid, et al. 2014)	SBDF	Ester	multi-walled carbon nanotubes (MWCNT)	Rheology and filtration modifier	They found that there are no significant changes in the rheological and filtration properties. They suggested using higher concentrations to get the desired effect on the ester-based drilling fluid's rheological and filtration properties.
Yusof et al. (Yusof and Hanafi 2015)	SBDF	Not specified	silicon dioxide (with 10-20 nm diameter)	Pickering emulsion stabilizer and Filter loss agent	40 wt% of silicon dioxide (with 10-20 nm diameter), reduced the filter cake thickness and fluid loss under HPHT (275, 351, 450 °F) conditions.
Kevin and Mahd (Kevin and	SBDF	Not specified	nano-silica	Pickering emulsion stabilizer and Shear-thinning agent	40 % of nano-silica needed to improve the rheological properties of SBDF in terms of shear-thinning behaviour

Mahmud 2019)					
Ghosn et al. (Ramy Ghosn, François Mihelic 2017)	SBDF	Olefin	Commercial Hydrophobic Silica NPs	Pickering Emulsion Stabilizers	<ul style="list-style-type: none"> The stability of the emulsion increases with increasing the hydrophobicity of the nanoparticles. After ageing for 16 h at 177 °C and 500 psi, the prepared drilling fluids showed high stability
Fan et al. (Fan et al. 2021)	SBDF	Paraffin	methyl grafted silica	Pickering Emulsion Stabilizers	Stabilizing the emulsion drilling fluids at low temperatures
Zaboli et al.(Dargahi-Zaboli, Sahraei, and Pourabbas 2017)	SBDF (invert emulsion)	Plyalphaolefins (poly 1-decene)	octadecyltrimethoxysilane-modified silica nanoparticles	Pickering Emulsion Stabilizers	The emulsion drilling fluids showed an excellent and stable rheological profile even after ageing at 120 °C.
Agarwal et al.(Agarwal et al. 2013)	SBDF (invert emulsion)	Plyalphaolefins (poly 1-decene)	A combination of hydrophobic nanoparticles and organically modified nanoclay	Pickering Emulsion Stabilizers	The invert emulsion showed high-temperature stability rheology even after ageing at 225°C for 96 h.
Ahmed et al.(Ahmed et al. 2021)	SBDF (invert emulsion)	Ester (ethyl octanoate)	A combination of aluminium oxide nanorods and commercial emulsifier	Pickering Emulsion Stabilizers	The nano-enhanced drilling fluids were stable (no separation in emulsion) after 16 hour of static aging unlike the base SBDF.

Solid particles that stabilizes the emulsion, whether solely or with the help of emulsifiers, are called Pickering emulsion stabilizers (Pickering 1907). These Pickering particles cause a steric barrier between the aqueous droplets, thus preventing coalescence (X. Chen et al. 2017). Furthermore, it was found that solid particles stabilized emulsions are thermodynamically and kinetically stable. (Ramy Ghosn, François Mihelic 2017). Several studies on particle-stabilized emulsions have shown that thermodynamic and kinetically stable Pickering emulsions were prepared using nanoparticles (NPs) from various materials such as gold (Larson-Smith and C. Pozzo 2012), biomaterials (Lam, Velikov, and Velev 2014), latex (Binks and Whitby 2005), carbonyl iron (Melle, Lask, and G. Fuller 2005), silica (Binks and Whitby 2005) and titanium dioxide (T. Chen, Colver, and Bon 2007) The characteristics of these particles that contribute uniquely relative to conventional emulsifiers include irreversible interfacial adsorption, stability against coalescence, sedimentation, flocculation and creaming, along with the ability to stabilize emulsions with large droplet size (up to several millimeters) (Xiao, Li, and Huang 2015). Depending on the hydrophobicity degree, nanoparticles can attach to the oil-water interface that improves the drilling fluids' emulsion stability (Fan et al. 2021; Dargahi-Zaboli, Sahraei, and

Pourabbas 2017). For particles-stabilized emulsions, contact angle (θ) is the equivalent hydrophilic-lipophilic balance for surfactants (Griffin 1949). Also, the empirical Finkle rule which used for pickering emulsion, is equivalent for the Bancroft rule used for surfactants stabilized emulsions (Finkle, D. Draper, and H. Hildebrand 2002; D. Bancroft 2002). Infact, the contact angle (θ) is directly linked to the type of the stabilized emulsion (O/W or W/O) (Aveyard, Binks, and Clint 2003; Albert et al. 2019). When $\theta < 90^\circ$, particles are hydrophilic and favor to stabilization of the O/W emulsions as most of particles is immersed in the water phase. In contrast, when contact angle $> 90^\circ$, particles are hydrophobic and consequently stabilize the W/O emulsion (figure 11).

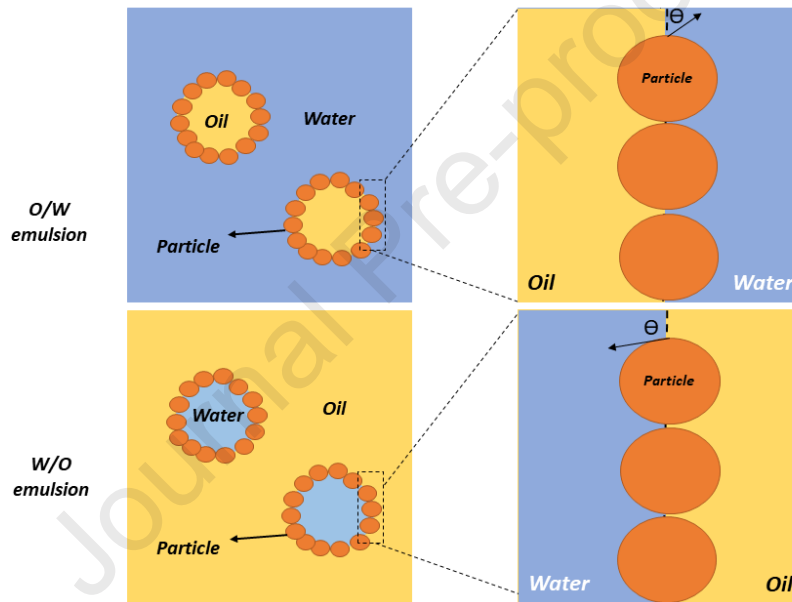


Figure 11: schematic representation of an O/W and a W/O Pickering emulsion

Moreover, these Pickering emulsion drilling fluids are expected to resolve the untapping unconventional wells (Ramy Ghosn, François Mihelic 2017). There are reports of different types of nanoparticles used to stabilize the emulsion of OBDF and SBDF. For instance, Sharma et al. used silica nanoparticles (particle size of 1 nm to 500 nm) to stabilize the OBDF emulsion; no further emulsifier or surfactant was applied in this study (Sharma, Ravi, Lukasz Grzybek 2016). The nanoparticles were mixed with the brine solution to densify the brine. Nanoparticles (NPs) have been used as Pickering emulsion stabilizers in emulsion drilling fluids (Pickering 1907; X.

Chen et al. 2017; Destribats et al. 2014). As NPs have strong ability to adsorb irreversibly at the oil/water interface that covers each dispersed droplet (Katende et al. 2019; X. Chen et al. 2017).

The research in stabilizing OBDF emulsions by combining nanoparticles and surfactants is still limited (Liu et al. 2020). Liu et al. studied the interactions between fumed silica nanoparticles (NPs) and amine surfactant as smart or pH-responsive Pickering emulsions (Liu et al. 2020). The stable OBDF formed measured a pH of only 2~12, which increases the nanoparticles' hydrophobicity. This drilling fluid showed adjustable rheology based on the pH of the formulation.

There are no studies yet in role of nanomaterials in stabilizing the biodiesel emulsion muds. Even literatures are also limited in terms of stabilizing SBDF emulsions. For example, Ghosen et al. studied olefin-based drilling fluids by adding 5 wt% of three modified types of commercial silica nanoparticles of various hydrophobicity (Ramy Ghosn, François Mihelic 2017). The stability of the emulsion increases with increasing hydrophobicity of the nanoparticles. The hydrophilic nano-silica showed underwhelming results. The particles' hydrophobicity depends on the pH or the salinity of the water phase. After ageing for 16 h at 177 °C and 500 psi, the nano-stabilized drilling fluids exhibited a better recovery for the rheological profile than surfactant-stabilized OBDF. Nano-stabilized OBDF also showed a shear-thinning behavior at 180 C and 75 bar while surfactant-stabilized OBDF failed to perform above 140 C. In a different study, Fan et al. investigated the stabilization of paraffin-based drilling fluids by adding 2 wt% of alkyl (methyl, octyl, and octadecyl) grafted silica nanoparticles without the presence of emulsifiers at low temperature (Fan et al. 2021). These nano-enhanced drilling fluids showed almost constant rheological performance at temperatures ranging from 5 to 75 C.

Furthermore, Zaboli et al. also studied hydrophobically modified silica with a contact angle of greater than 90° , which caused the nanoparticles to reside more in oil than in water (Dargahi-Zaboli, Sahraei, and Pourabbas 2017). They added 2 wt% of octadecyltrimethoxysilane (OTMOS)-modified silica nanoparticles as emulsifiers to stabilize the emulsion of polyalphaolefins drilling fluids as shown in Figure 12. This invert emulsion drilling fluids showed a good and stable rheological profile even after ageing at 120 °C. They found that the prepared nano-enhanced invert emulsion morphology meets the drilling fluid requirements, as shown in the Figure 13.

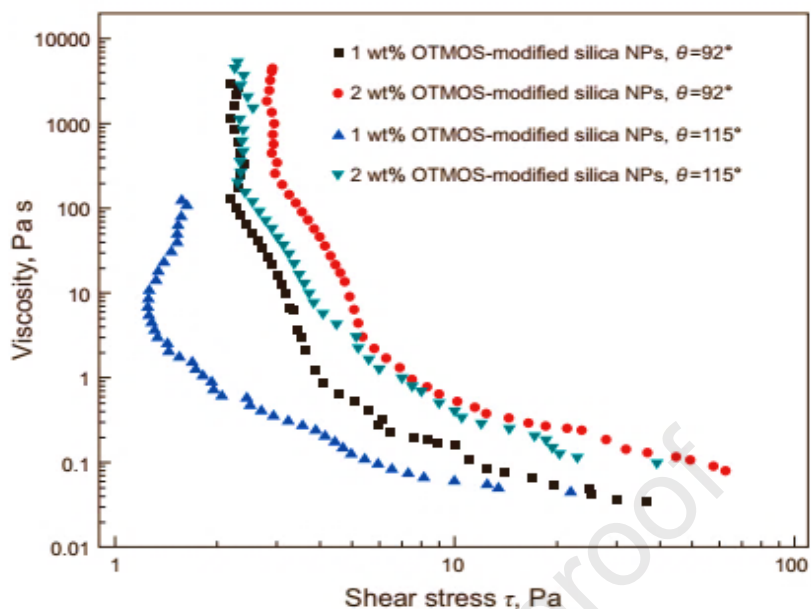


Figure 12: measured viscosity versus shear stress of invert emulsions containing 30 vol% water stabilized by OTMOS-modified silica NPs [reused with permission from Zaboli et al. (Dargahi-Zaboli, Sahraei, and Pourabbas 2017)]

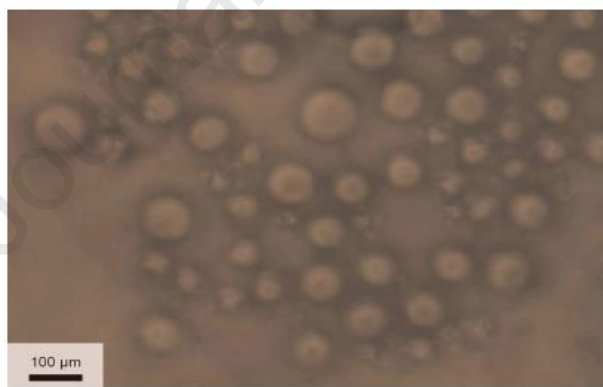


Figure 13: morphology of the invert emulsion stabilized by 2 wt% OTMOS-modified silica NPs ($\theta = 92^\circ$) after aging at 120 C [reused with permission from Zaboli et al. (Dargahi-Zaboli, Sahraei, and Pourabbas 2017)]

In addition to that, Agarwal et al. investigated a combination of hydrophobic nanoparticles (1 vol%) and organically modified nanoclay (2 wt%) that resulted in stable poly alpha olefin drilling fluid (Agarwal et al. 2013). The drilling fluid they formulated satisfies the rheological profile i.e., plastic viscosity but showed a decrease in yield point. However, they argue that it can be avoided

by adding weighting material such as barite or increasing the concentration of nano-silica. Moreover, they showed high-temperature stability in terms of rheology and proper emulsion morphology even after ageing at 225°C for 96 hours.

Also, Ahmed et al. stabilized the EBDF using aluminum oxide nanorods (hydrophilic nanoparticles) combined with commercial emulsifiers (oil-soluble emulsifiers). There was no separation in the emulsion by using 1 wt% of aluminium oxide nanorods as shown in figure 14. This can be explained due to the dispersion of particles and hence the electrostatic interaction between the colloidal particles (Ahmed et al. 2021; Katende et al. 2019).



Figure 14: Stability of the base EBDF (left) and nano-enhanced EBDF with 1 wt% NPs sample (right) after 16 hr of static aging at ambient conditions.

6. Designing a Proper Biodiesel Based Drilling Fluids

Based on the previous studies, the technical challenges for biodiesel-based drilling fluids can be resolved by three techniques (1) modification of biodiesel structure, (2) using additives and (3) blending biodiesel with other base oils.

6.1. Modification of Biodiesel Structure

The biodiesel structure is vital for its characteristics. For example, the biodiesels produced from linear alcohol, are prone to hydrolysis. While, the biodiesels that contain a branched chain, have

good hydrolytic stability. Also, biodiesel produced from branched alcohols, the hydrolytic/thermal stability and the flash point will significantly increase. At the same time, If the alcohol is highly branched, the pour point, low-temperature behavior, and biodegradability will decrease. The best candidate of alcohols moiety is a low branched alcohol as isoamyl or isobutanol alcohol. As, Staples found that the effects on the aqua environment for a C4 alcohol moiety (isobutanol) are less toxic than C8 alcohol (2-ethyl hexanol) due to the high solubility and volatility of C4 compared to C8 moieties [134]. Moreover, the best candidates of oil source, are rapeseed, soybean and sunflower due to their availability as base stook, low pour point, high flash point and proper kinematic viscosity.

Other studies suggested further chemical modification on the biodiesel structure (such as epoxidation and acylation) is required to find solutions for BBDFs. For instance, epoxidized biodiesel may enhance the pour point of biodiesel however, the compatibility of using epoxidized biodiesel with other drilling fluid components, has not been investigated.

6.2. Additives

Biodiesels or vegetable oil-based esters have poor pour point and excessive high viscosity compared to other base oils. This can be solved by adding pour point depressants as ethoxylate or acrylate-based compound by concentration ranging from 0.01 to 0.3 % (V/V). The pour point and viscosity can be adjusted by adding ethoxylate compounds.

Also, researchers have used anti-hydrolysis additives to overcome the hydrolysis issues related to esters. Some of these additives include, carbodiimides, polycarbodiimides, surface-modified macromolecules or polyalkylene glycols (PAG) (J P 2013; Stloukal et al. 2016; Tang et al. 1997)- (Greaves and Zaugg Hoozemans 2018). The formation of stable W/O emulsion can decrease the rate of hydrolysis. This can be achieved by using hydrophobic nanomaterials or a mixture of a low HLB emulsifier and hydrophilic nanomaterials. However, further investigation on the use of nanomaterials and the other additives mentioned above in BBDFs are needed.

6.3. Blending Biodiesel with Other Base Oils

Blends of biodiesels with other base oils, can enhance the rheological, filtration and low-temperature flowability properties of drilling fluids. For example, when ester (as POME) was

mixed with a low toxicity mineral oil or n-paraffin; the filtration and rheological properties were improved compared to the performance of the respective synthetic oils by its own (Amin et al. 2010). There are also reports of commercial esters that were mixed with other synthetic oil such as olefins (IO or LAO) or paraffins, and results demonstrated improvements in the overall drilling fluid performance (Kirsner J, Pober K 2004). The most proper oils to be mixed with biodiesel are synthetic ester, Fischer Tropsch paraffin and linear olefins based on the discussions in Sections 3.5, 3.6 and 4.3. That is to avoid the harmful impact on the environment and enhance the physiochemical properties of biodiesels (viscosity, high flash point and pour point) to meet the drilling fluid requirements. The pour point values can also be improved by mixing with olefins (Asadauskas and Erhan 1999). For example, Asadauskas & Erhan improved the pour point of soybean oil by adding polyalphaolefin (20%) to lower it to $-57\text{ }^{\circ}\text{C}$ (Asadauskas and Erhan 1999). The pour point value for the internal olefin is lower than the corresponding linear alpha-olefin; this is due to IO's presence in structural isomers (cis and trans), which causes the IO molecules to pack together uniformly upon cooling (Swaco 2006). Also, PAOs can be blended with biodiesel to enhance its pour point. The alkyl branches present on the PAO's backbone carbon atoms cause a fully amorphous structure due to the low packing density. That explains the reason for their excellent viscosity at extremely low temperatures as they remain viscous in liquid form and are not easily crystallized (Candler et al. 1995). Because of this branched nature, PAO also exhibits a low pour point. PAO's branching nature causes low biodegradation than linear alpha-olefins, internal olefins, and linear paraffin (Rabke, S. 1998).

Also, using multiple emulsion (W/O/W or O/W/O) such as a second oil phase (silicon based oil) could avoid the hydrolysis process (Daoust, Jennifer A., Lecomte, Jean-paul H., Liles, Donald Taylor, Liu, Yihan, Roggow II 2018). The application of such emulsions in BBDFs has not been investigated in detail, and requires more research.

7. Conclusions

This study discussed the use of biodiesels as base oil in drilling fluids. The relationship between the biodiesel molecular structures and their physiochemical properties, the performance of drilling fluids and their cytotoxicity were analyzed. The characteristics values of biodiesel properties were

summarized, and by comparing them with the values of practical base oils, engineers and researchers can choose a proper biodiesel and design a successful biodiesel-based drilling fluid. Biodiesel-based drilling fluids have demonstrated favourable results in terms of performance, biodegradability and cost. However, the use of biodiesel in drilling fluid, has faced some drawbacks due to technical challenges such as excessive viscosity, poor pour point, low hydrolytic stability and water-in-biodiesel emulsion stability. This review study discussed some of the proposed solutions to overcome these challenges including the modification of ester molecules, the use of additives (e.g., nanomaterials) and blends of oil base... The modification of biodiesel structure (as epoxidation) or the additives can increase the cost of drilling fluids, in addition they may not be compatible with other components of drilling fluids. Blending biodiesel with other base oils, showed promising solutions for most of biodiesel challenges such as high viscosity, poor pour point and hydrolytic stability, and the best environmentally-friendly choices to mix with the biodiesel, are synthetic esters, paraffins and linear alpha olefins. Additives such as nanoparticles can stabilize water-in-biodiesel emulsions in drilling fluids however there are limited understanding in this area and further investigations are required to achieve robust solutions for their use in biodiesel based drilling fluids.

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Highlights:

- The performance of drilling fluid depends on the type of used biodiesel.
- Modification of biodiesel structures improve the performance of drilling fluid.
- Branched chain biodiesels have high hydrolytic resistance and flash point.
- Nanomaterial may stabilize the emulsion of biodiesel based drilling fluid.

Declaration of interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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