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Enhancing the stability of local vegetable oils (esters) for high geothermal drilling applications

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Conventional drilling fluids such as diesel and mineral oil have posed some environmental and health challenges in their drilling applications but the introduction of synthetic-base fluids over the past two decades has considerably reduced such challenges. In some cases, a bottom hole temperature above 300°F (150°C) can cause significant instability in the rheological properties of these drilling fluids. Vegetable oils or pseudo oils are known to be environmentally friendly drilling fluids, but have not received much attention because of their instability in High Pressure Temperature High (HPHT) environments. The antioxidant potentials of Citric Acid (CA), Red Onion Skin Extract (ROSE) and Propyl Gallate (PG) on the oxidative stability of seven vegetable oils were examined. The additives (antioxidants) were able to protect the stability of the oils up to 250°C which is beyond the range for the 1st tier of HPHT environment (150 - 205°C). Though it was also observed that the peroxide values (PVs) of the oils as temperature increase also increase, it did not follow and defined pattern (no pattern was established). The applications of combined antioxidants improved the stability of the oil samples when compared with using individual antioxidants. The applications of appropriate antioxidants in local vegetables oils (esters) have revealed the potentials geothermal stability of the local esters to with stand the 1st tier of HPHT environments.

Key words: Drilling, high temperature, stability, antioxidant, vegetable oils (Esters), oil base muds.

INTRODUCTION

The introduction of synthetic base fluids in drilling mud formulations over the past two decades has considerably reduced the challenges usually associated with conventional drilling fluids (Growcock et al., 2011). Synthetic base fluids have now become the preferred type of based fluid for drilling through problem formations. However, the revolution in the use of non-aqueous drilling fluid (NADF), the synthetic base fluids technology, has its own challenges. As the complexity of drilling operations increased, environmental regulation issues have placed some restriction on the use of NADF in drilling mud formulation due to the toxicity of some of the base fluids (Growcock et al., 2011; Fadairo et al., 2012).

Conventional drilling fluid such as diesel and mineral oil pose some challenges such as initial high cost; health, safety and environmental (HSE) concerns; incompatibility

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with elastomers; high potential for lost circulation; high sensitivity to pressure and temperature; inability to detect gas kicks, and undesirable effects on some logging tools (Chen et al., 2003; Growcock et al., 2011). Every well drilled is subject to some element of temperature and pressure and these affect both drilling fluid and wellbore stability and even get more intense in HPHT environment where bottom hole temperature is above 300°F (150°C) (Oriji and Dosunmu, 2012).

Vegetable oils are known to be potential environmentally friendly drilling fluid but have not received much attention because of its high instability issues (Dosunmu and Ogunrinde, 2010). A sustainable environmentally friendly vegetable oil should be stable during usage under different operating conditions (Alves and Gomes de Oliveira, 2008). Malaysian palm oil and palm kernel oil have received worldwide approval for the preparing of environmentally friendly drilling fluids (Salleh and Tapavicza, 2004). The original Petro-free synthetic base fluid (SBF) system consisted of a mixture of five homologous fatty acid esters, of which the main component was 2-ethylhexyldodecanoate but later developed Petro-free formulations contain other SBF base chemicals such as Linear Alpha Olefins or Poly Alpha Olefins (Neff et al., 2000) for stability issues.

Vegetable oils challenges as drilling fluids

Most of the vegetable oils such as rapeseed, soya bean, groundnut, cotton, sunflower, coconut palm oils, though considered as prospect raw materials for oil based fluids, however require further refinery processes to obtain an API biodiesel standard (Okullo et al., 2012). It is investigated that the presence of OH group in ricinoleic acid in most of these vegetable oils tend to increase their viscosity significantly due to hydrogen bonding making most of these vegetable oils unsuitable to be used as biodiesel or uneconomical because of the need for trans-esterification processes like dilution, microemulsion, pyrolysis and catalytic cracking to reducing their viscosity (Okullo et al., 2012). At ambient temperatures, SBFs have base viscosity that are relatively 2 to 4 times higher than other oils based fluids but as temperature increases, the fluids then thin significantly more than other oils (Aluyor and Ori-Jesu, 2008; Baidu, 2014). Fats and oils deteriorate rapidly in the presence of oxygen and go rancid. The rate of oxidation by air varies and depends on the ease of hydrogen ion abstraction from the substrate molecule (Akaranta and Akaho, 2012).

There are basically three ways of improving the stability of the oil and these can be through:

1. Genetic modification through biotechnology to produce oils that have high saturated acid;
2. Chemical modifications through hydrogenation of the vegetable oil to alter the fatty acids;
3. The use of antioxidants (additives).

Antioxidants happen to be the most efficient and cost effective ways to improve the oxidative stability. Vegetable oils have some amount of natural antioxidants such as ascorbic acids, α-tocopherol, β-carotene, chlorogenic acids and flavanols but not so strong to withstand high temperatures (Aluyor and Ori-Jesu, 2008).

The antioxidants react with the fat radical to form a stable radical impending oxygen reaction. They function as hydrogen donors by either inhibiting the formation of free alkyl radicals in the initiation step or by interrupting the propagation of the free radical chain reacting with lipid free radicals to form stable and complex compounds. (Aluyor and Ori-Jesu, 2008) as shown in Figure 1.

Citric acid is commonly used in vegetable oils as a metal chelator; thus binds metal ions that contribute to rancidity as they catalyse free-radical oxidation of lipids (Akaranta and Akaho, 2012). According to Reda (2011), Propyl Gallate (PG)is one of the most effective antioxidant often used in the food industry. Red onion skin (Allium Cepa) has tannins in its protective layers polyhydroxyphenols of the flavonoid type (Akaranta and Akaho, 2012) which are good natural antioxidants. In most cases, mixtures of two or more antioxidants prove more effective than the effect of one. In such cases, one antioxidant reinforces the effect of the other for maximum efficiency (Akaranta and Akaho, 2012).

A good choice of antioxidant aims at the preservation of unsaturated fatty acids to increase the stability to thermal degradation, which usually happens between 150 and 220°C (Reda, 2011). The objective of this paper is to investigate the effect of antioxidants on the thermal stability of vegetable oils to be used as potential substitute of conventional oil drilling fluids which are environmentally unfriendly.

MATERIALS AND METHODS

An experimental design followed in carrying out the work is shown in Figure 2.

Oil samples collection

Seven vegetable oil samples were analysed in this work. The oils are coconut oil (CO), groundnut oil (GO), jatropha oil (JO), palm oil (PO), palm-kernel oil (PKO), soyabean oil (SO), and refined waste home-cooking oil (XB1000).

Thermal stability experiment of the oil samples were investigated to ascertain the viability of the oils for use as drilling oils. This was done through the increase in temperature of the oil samples and investigating the changes in peroxide values (PV). Peroxide value is a measure of the peroxides in a sample of fat, expressed as milli-equivalent of peroxide per 1 000 g of the material. It is one of the most important chemical parameters for appraising the degree of deterioration of oils (Ngassapaa et al., 2012).

Antioxidant selections and samples preparations

The antioxidants (Red Skin Onion Extract (ROSE), Propyl Gallate (PG)) were administered with citric acid in the ratio of 2:1. For each

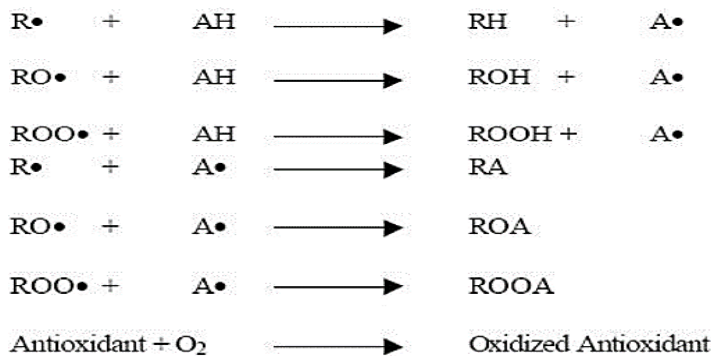

Figure 1. Reaction of antioxidants in vegetable oil stability (Aluyor and Ori-Jesu, 2008

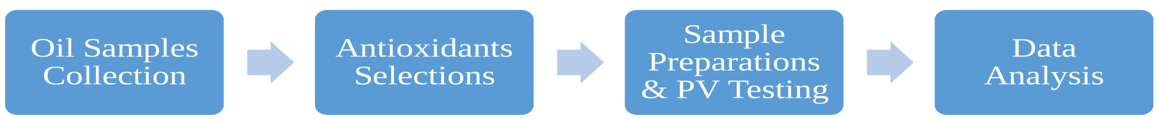

Figure 2. Experimental design.

100 g of oil sample, a total of 0.3 g of additives were added; citric acid inclusive (0.2 g main additive to 0.1 g citric acid). Though Akaranta and Akaho (2012), recommended a 1:1 ratio, but for prolong and higher temperature, 2:1 ratio was used. Sampling and subsequent analysis were carried out by the addition of antioxidants such as the ROSE, PG and a combination of ROSE and PG at the ratio of 1:1. ROSE antioxidant was extracted by the use of Acetone. The experiments were carried out at room temperature to a temperature of 482°F to monitor the changes in PV associated with the oxidation levels of the oils. The temperature ranges were 82, 212, 302, 392 and 482°F (28, 100, 150, 200, 250°C). The oil samples were heated at these temperatures at an average time of 200 s. The peroxide values were determined by titration method following the procedure of the American Oil Chemist Society (AOCS) (American Oil Chemists Society, 1960). Vegetable oil sample (3.0 g) was dissolved in a mixture of glacial acetic acid and chloroform (30 ml), by ratio of 3:2 v/v and saturated solution of potassium iodide (1 ml) was added. The solution was allowed to stand for one minute with occasional swirling and then 30 ml of water was added. The mixture obtained was titrated against 0.1 M solution of sodium thiosulphate to a (1 ml) starch indicator end point. A blanktitration (without oil sample) was also carried.

Data analysis

The peroxide values (PV) were then calculated as:

$$PV\ (Meq/Kg) = [M(S - B)/G] \times 100 \quad (1)$$

Where: G = weight of sample (g); S = titre of sample; B = titre of blank, and M = molarity of sodium thiosulphate.
A partial deviation mean stability test of the oil sample PV with antioxidant was analysed with the expression:

$$(x_i - \bar{x}) \quad (2)$$

A further stability test of the oil samples at various temperatures were determined by the method of applying standard deviation to the statistical data by the formula.

$$\sigma = \sqrt{\frac{\Sigma(x_i - \bar{x})^2}{n}} \quad (3)$$

Where: σ = the standard deviation; x_i = the PV at a particular temperature of an oil sample; $\bar{x}$ = the mean of the various PV of a particular oil sample, and n = the number of PV within an oil sample.

RESULTS AND DISCUSSION

The PVs of the various oil samples under various administrations of antioxidants are shown in Table 1.

PV analysis

The International Olive Oil Council (IOOC), the United Nations (UN) Codex Alimentarius Commission (often shortened to ‘Codex’) and other bodies have approved that the maximum PVs of most oils must not exceed 10 meq/kg for health and safety purposes as also reported by Ngassapa et al. (2012). The knowledge of thermal stability of antioxidants is very important in oil stability. The good choice of antioxidant aims at the preservation of unsaturated fatty acids to increase the stability to thermal degradation, which usually happens between 150 and 220°C. This was evidence in most of the oils sampled by their high corresponding PVs obtained and the fuming of the oils during their heating up (at such

Table 1. Summary of thermal stability of various oils with additives.

Temp. °C	CO	GO	JO	PO	PKO	SO	XB1000
100	ROSE + PG, ROSE	All Additive	With or without Additive	With or without Additive	ALL	All Additive	PG
150	ROSE + PG, ROSE	All Additive	No Additive, Rose + PG	With or without Additive	ROSE + PG, ROSE, PG	All Additive	PG
200	ROSE + PG, ROSE	ROSE + PG, PG	No Additive, Rose + PG	With or without Additive	ROSE + PG, PG	All Additive	PG, ROSE
250	ROSE + PG, ROSE	ROSE + PG	No additive, Rose + PG	With or without Additive	ROSE + PG, PG	All Additive	ROSE + PG, ROSE
All temperature	ROSE + PG	ROSE + PG	ROSE + PG	ROSE + PG	PG	PG, ROSE + PG	ROSE + PG

temperatures) especially without additives. Serjouie et al. (2010) stated that peroxides are unstable compounds towards at high temperatures, transforming them to carbonyl compounds. For the vegetable oils to pass the test to be used as HPHTSBFs, it must be stable at these critical temperatures. The results for the PVs tests are shown in Figures 3 to 6.

No Antioxidants

Generally, most of the oils without additives were stable to 302°F (150°C) due to some amount of antioxidants in their natural form such as ascorbic acids, α-tocopherole, β-carotene, chlorogenic acids and flavonoids as reported by Aluyor and Ori-Jesu (2008). Deterioration started at temperatures around 150°C and above, with the exception of JO and PO that remain relatively stable at all test temperature as shown in Figure 3. PO is highly saturated but has a strong enzyme activity which leads to hydrolytic rancidity that could make the oil unstable Anon. (2010). The high stability of the PO may likely be due to its fatty acids composition which contained nearly 40% of saturated fatty acids (SFA) as reported by Gharby et al. (2014). The heat treatment in processing of the oil inactivates these enzymes and this makes the oil quite resistant to oxidation. The stability of the two oils may be due to their acid profile which is less affluent to the most sensitive to oxidation unsaturated fatty acids. Oils such as SO is highly unsaturated, and its high linolenic acid content lead to oxidative deterioration as previously reported by Ngassapa et al. (2012) that linoleic and linolenic acids are the most readily oxidized components of oils. XB1000 by nature has a very high PV because it is a fatty acid methyl ester. The PVs ranged for nonaddition of antioxidant was from 3 to 37 meq/kg with an average value of 10.6 meq/kg. The wide variations in PVs confirm the instability of most of these oils in their natural state and therefore suggest the addition of antioxidants to the oils to make it suitable to be used as base oil for drilling HPHT conditions.

ROSE antioxidant

The addition of ROSE antioxidants to the oil samples helped to improve the stability for most of the oils over the entire temperature range of the test run than not adding any additive. GO and XB1000 did not improve much as the temperature was increased. The GO stated deteriorating after 150°C whiles that of XB1000 started after 100 °C as shown in Figures 3 and 4. GO is highly unsaturated and ROSE antioxidants begins to breakdown at around 150°C so the ROSE was unable to stabilize the highly unsaturated oils that much. More thermally stable antioxidants would therefore be needed. The PVs ranged for the addition of ROSE antioxidant was from 1 to 34 meq/kg with an average value of 7.2 meq/kg.

PG antioxidant

The addition of PG antioxidants to the oil samples help to improve the stability for most of the oils better than ROSE antioxidant with the exception of CO (Figure 5). CO started it deterioration after 150°C. CO is highly susceptible to hydrolytic rancidity, which imparts a soapy flavour to food and oils as shown in Figures 3 and 4. The rancin in CO is difficult to inhibit effectively by most antioxidants (Anon., 2010). The administration of PG to the oil samples recorded PVs ranges from 1 to 24 meq/kg with an average peroxide value of 7.76 meq/kg. Though most of the PVs of the oils fell within 2 to 10 meq/kg, they are not without fluctuations and would need an improvement to smoothen them for better stability.

Combined ROSE and PG antioxidants

The addition of blended Rose and PG antioxidants to the oil samples gave a better stability of the oils than the single effects of the applications of Rose and PG alone (Figure 6).

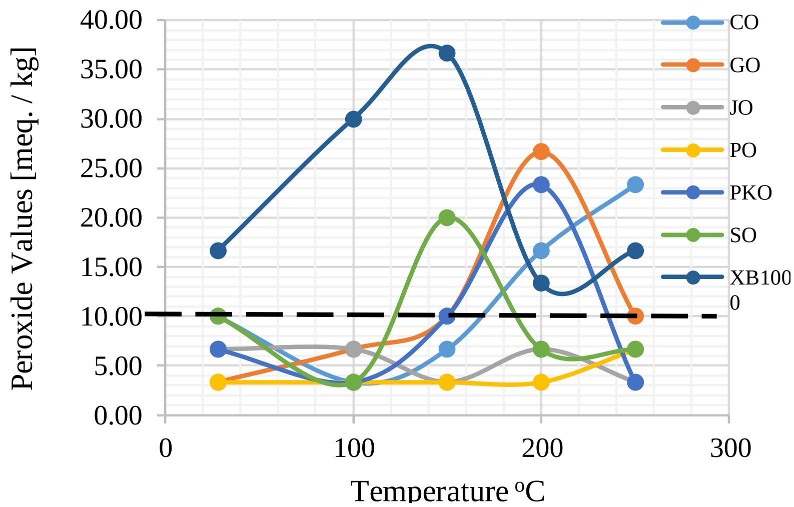

Figure 3. PVs of oil samples without additives.

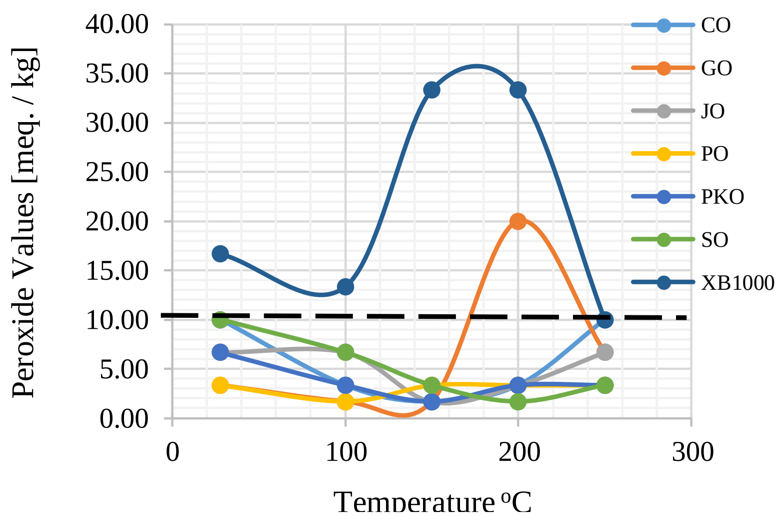

Figure 4. PVs of oil samples with ROSE additive.

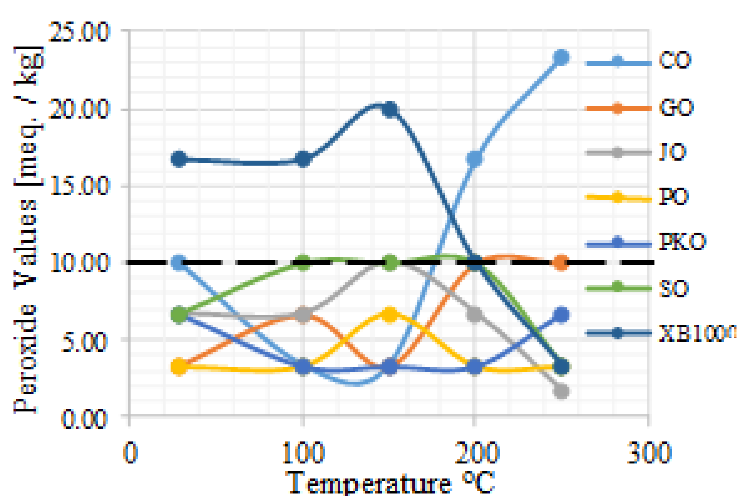

Figure 5. PVs of oil samples with PG additive.

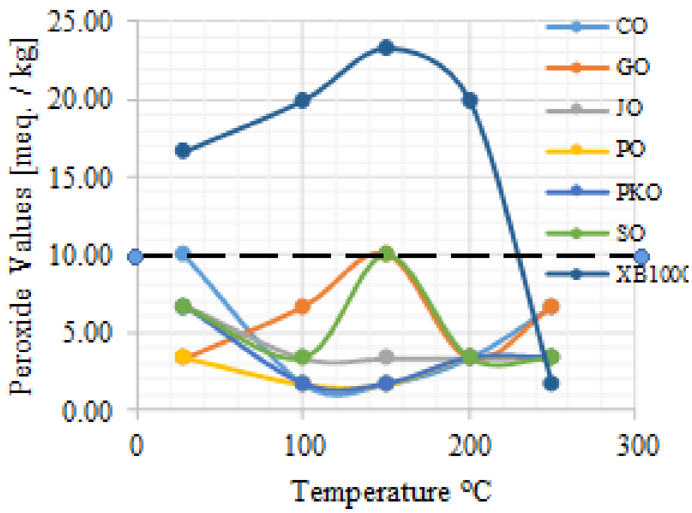

Figure 6. PVs of oil samples with ROSE and PG additives.

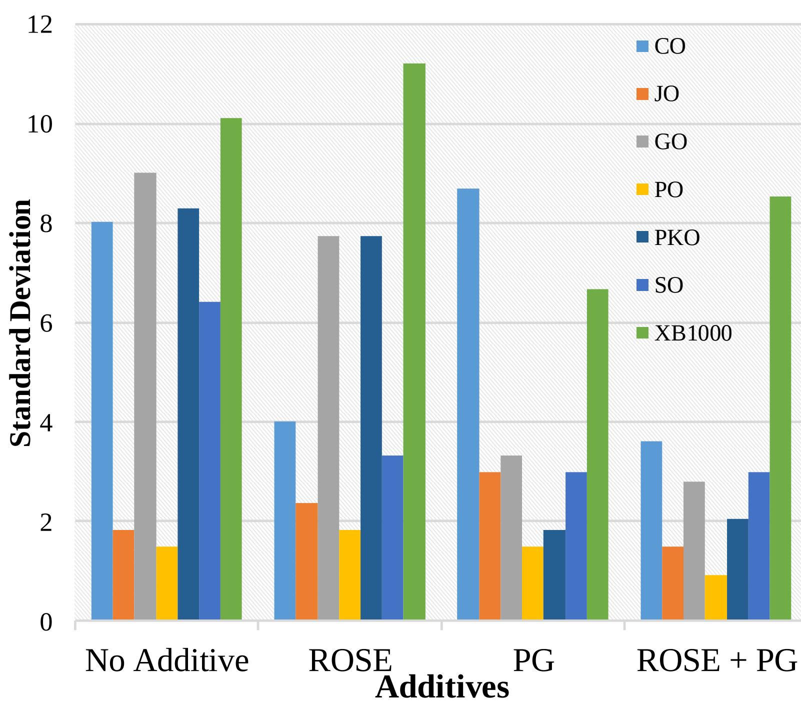

Figure 7. Standard deviation analysis of oils samples with antioxidants.

Though GO, SO and XB1000 increased in PVs from to 28 to 150°C, 100 to 150°C and 28 to 150°C respectively, the increases for GO and SO were within reasonable ranges. The PVs fell thereafter as shown in Figure 6. There was a significant improvement on the stability of the CO due to the combined effect of the additives than single antioxidant. This suggest that the combined effect of antioxidants are highly effective compared to the administration of single antioxidants as one antioxidant reinforces the effect of the other for maximum efficiency as reported by Akaranta and Akaho (2012). The PVs ranged for the combined ROSE and PG antioxidants was from 1 to 24 meq/kg with an average value of 6.1 meq/kg.

Statistical analysis

The stability analysis was done using standard deviation

Table 2. Standard deviation analysis of oils samples with antioxidants.

CO					JO				
Temp. °C	**No additive**	**ROSE**	**PG**	**ROSE + PG**	**Temp. °C**	**No Additive**	**ROSE**	**PG**	**ROSE + PG**
28	3.33	3.89	2.78	4.72	28	1.33	1.67	0.33	2.67
100	10	2.78	9.44	3.61	100	1.33	1.67	0.33	0.67
150	6.67	4.44	9.44	3.61	150	2	3.33	3.67	0.67
200	3.33	2.78	3.89	1.94	200	1.33	1.67	0.33	0.67
250	10	3.89	10.56	1.39	250	2	1.67	4.67	0.67
Mean	12	5.67	11.33	4.67	Mean	5.33	5	6.33	4
Variance	64.44	16.11	75.56	13.06	Variance	3.33	5	8.33	3.33
SD	8.03	4.01	8.69	3.61	SD	1.83	2.36	2.98	1.49
GO					**PO**				
Temp. °C	**No Additive**	**ROSE**	**PG**	**ROSE + PG**	**Temp. °C**	**No Additive**	**ROSE**	**PG**	**ROSE + PG**
28	10	5.28	3.33	2.78	28	0.67	3	0.67	0.67
100	6.67	6.94	0	0.56	100	0.67	0.33	0.67	1
150	3.33	6.94	3.33	3.89	150	0.67	2	2.67	1
200	13.33	11.39	3.33	2.78	200	0.67	0.33	0.67	0.67
250	11.33	1.94	3.33	0.56	250	2.67	0.33	0.67	0.67
Mean	11.33	6.67	6.67	6	Mean	4	3.67	4	2.67
Variance	81.11	59.72	11.11	7.78	Variance	2.22	3.33	2.22	0.83
SD	9.01	7.73	3.33	**2.79**	SD	1.49	1.83	1.49	**0.91**
PKO					**SO**				
Temp. °C	**No Additive**	**ROSE**	**PG**	**ROSE + PG**	**Temp. °C**	**No Additive**	**ROSE**	**PG**	**ROSE + PG**
28	2.67	3.33	2	3.33	28	0.67	5	1.33	1.33
100	6	5	1.33	1.67	100	6	1.67	2	2
150	0.67	5	1.33	1.67	150	10.67	1.67	2	4.67
200	14	13.33	1.33	0	200	2.67	3.33	2	2
250	6	0	2	0	250	2.67	1.67	4.67	2
Mean	9.33	6.67	4.67	3.33	Mean	9.33	5	8	5.33
Variance	68.89	59.72	3.33	4.17	Variance	41.11	11.11	8.89	8.89
SD	8.3	7.73	**1.83**	2.04	SD	6.41	3.33	**2.98**	**2.98**
XB1000									
Temp. °C	**No Additive**	**ROSE**	**PG**	**ROSE + PG**					
28	6	4.67	3.33	0.33					
100	7.33	8	3.33	3.67					
150	14	12	6.67	7					
200	9.33	12	3.33	3.67					
250	6	11.33	10	14.67					
Mean	22.67	21.33	13.33	16.33					
Variance	102.22	125.56	44.44	72.78					
SD	10.11	11.21	**6.67**	8.53					

(SD) techniques. This was used to analyse the internal stability trends of the various oil samples. It was used to select the most effective antioxidant. In order to find the most effective antioxidant to administer, the SD with the least value as shown in Figure 7 and Table 2 is selected. Table 1 shows the recommended thermal conditions for application of additives as well as the selected additives that are stable under all temperature ranges.

Conclusions

The vegetable oil samples exhibited thermal instability as temperature increases especially above 150°C without additives. With the addition of the additives, the vegetable oils exhibited some considerable measure of stability at temperature above 150°C. The most stable for all temperatures conditions under investigations for CO, GO, JO, PO and XB1000, is the combined ROSE and PG antioxidants, and for PKO is PG while for SO is PG and combined ROSE and PG.

The additives were able to protect the thermal stability of most oils up to 250°C. This is beyond the range for the 1st tier of HPHT environment (150 - 205°C). This reveals that with the addition of the right additives, vegetables oil esters become thermally stable and have the potential to be used as based fluid to drill to HPHT wells. The order of stability in ascending order based on the SD analysis and addition of additives are: XB1000 < CO < SO < GO < PKO < JO < PO. The applications of combined antioxidants improved the stability of the oil samples when compared with using individual antioxidants. The applications of appropriate antioxidants in local vegetables oils (esters) have revealed the potentials geothermal stability of the local esters to withstand the 1st tier of HPHT environments.

Conflict of Interest

The authors have not declared any conflict of interest.

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