Effective Separation and Hydrocarbon Accounting in Water-in-Oil Emulsion Through Demulsification Using Formulated Plant Extracts

[1]Tamunotonjo Obomanu, [2]Amieibibama Joseph and [3]Chukwu Chimarouke Godson

[1]Department of Petroleum Engineering Technology, Federal Polytechnic of Oil and Gas, Bonny, Rivers State, Nigeria
[2,3]Department of Petroleum and Gas Engineering, University of Port Harcourt, Rivers State, Nigeria

ABSTRACT

Background and Objective: Crude oil emulsions have been one of the major problems of the petroleum industry, as oil combines with water during production. This work focused on resolving water-in-oil emulsion using plant-based formulated demulsifiers made from cashew leaf (*Anacardium occidentale* B-1) and Moringa leaf extract (*Moringa oleifera*, C-1) and respective additives. **Materials and Methods:** The Soxhlet extraction technique with absolute ethanol as the solvent was employed as the extraction method to extract the solvents. Static bottle test and dynamic test were conducted to evaluate the effectiveness of these green demulsifiers at different concentration dosages and rpm and the results were compared with a chemical-based conventional demulsifier (pt6020). A single-factor ANOVA was performed at a significance level of 0.05. **Results:** It was observed that only B-1 was effective while C-1 performance was very poor in resolving the emulsion. At a dosage of 0.02 mL for static bottle and dynamic tests, the B-1 demulsifier competed favorably with the conventional demulsifier with a 0.1 mL volume of water separated and a percent relative error of 2%. In fact, at 30 min of the static bottle test, the B-1 demulsifier performed better than the conventional demulsifier, separating 4.8 mL of water compared to 4.6 mL by the conventional demulsifier. **Conclusion:** The B-1 shows a good prospect as a formidable demulsifier and could be used as an alternative to the chemical-based demulsifiers with 0.87 R^2 value and p-values of 1% for one-tail and two-tail tests; both less than $\alpha = 0.05$. The C-1 could be explored more to investigate its poor performance since it showed a propensity as a demulsifier.

INTRODUCTION

An emulsion is defined as a mixture of two immiscible liquids (e.g., oil and water) with one phase dispersed in a continuous phase formed when agitated in the presence of an emulsifier. Thus, for an emulsion to form, three basic conditions must prevail: The presence of two immiscible fluids, agitation, and a stabilizing agent[1]. The disperse phase, also called the sparse phase exists as globules while the continuous phase is the phase that produces the dispersion model wherein the droplets are suspended[2]. Three emulsions can be formed during oil production: Oil-in-water emulsion (O/W), water-in-oil emulsion (W/O), and Multiple-phase emulsion, respectively.

Oil field emulsions constitute one of the major operational challenges in the oil industry. The formation of emulsion is inevitable following the numerous inherent stabilizing agents and those induced not deliberately but, in an attempt, to resolve one or many operational challenges in the course of production such as corrosion inhibition, wax inhibition drag reduction etc[3]. Unfortunately, since no two hydrocarbon reservoirs are identical in all aspects, it becomes much more difficult to resolve emulsions as these stabilizing agents act as surfactants that bridge or decrease the interfacial tension between the two immiscible fluids[4]. The process of reversing an emulsion is what is termed demulsification. The industry is facing the challenge of selecting the most appropriate demulsifiers and as such resort to a trial-and-error process following the dynamic nature of surfactants and the timing. Thus, there are no universal demulsifiers and no demulsifier can resolve an emulsion throughout the life of a particular well[5]. The amount of mixing, the nature of the stabilizer, and the age of the emulsion strongly influence the stability of a given emulsion. The longer an emulsion is left unresolved, the more it becomes stable and difficult to reverse[6]. More challenging are the basic ingredients in formulating such demulsifiers as many of them if not all are not just expensive but also non-biodegradable, constituting an environmental menace once a given demulsification process is completed[7].

Demulsification is the process of breaking down or reversing emulsions into distinct phases. There are three basic demulsification techniques used in the industry and these include: physical, biological, and chemical methods of which the choice depends on the effectiveness of a given method in reducing the stability of a given emulsion[8,9]. The chemical method involves the use of chemical solvents called demulsifiers which are surfactants that reverse a given emulsion. The physical technique involves the use of heating, mechanical filtration, membrane separation, or electrical methods[10]. There is also the hybrid method called a thermochemical method that involves heating with the addition of a demulsifier[11]. There are also microwave and magnetic methods as well currently in use. The biological method, though not as popular as the chemical and physical method is an emerging technique that has shown prospects of conserving the environment. The biological method involves the use of bacteria to demulsify an emulsion with their cells or extracellular compounds[12].

However, chemical methods are the most popular and commonly preferred method used in the industry. To date, the selection process is by trial and error as no demuslifier can resolve an emulsion in the life of a well as emulsion properties change with changes in the stabilizing agent and the composition of the crude. Hence, in an attempt to curb this environmental challenge, plant-based extracts are now being formulated to investigate their potential to resolve emulsions reduce costs, and save the environment. In this work, the focus is on water-in-oil emulsion, which is also the most common in the industry.

MATERIALS AND METHODS

Study area: This study was conducted at the production laboratory of the Department of Petroleum and Gas Engineering, University of Port Harcourt, Rivers State, Nigeria between 29th October, 2023 to 5th March, 2024. The crude oil sample used was obtained from a flow station located in Niger Delta while the Moringa leaf (*Moringa oleifera*) and cashew leaf (*Anacardium occidentale*) were obtained from a farm in Choba, Rivers State.

Reagents and equipment: The Soxhlet extraction technique was used for the extraction of the leaves. The major materials used for the extraction and formulation of the demulsifiers are: Moringa (*Moringa oleifera*), Cashew leaf (*Anacardium occidentale*), Liquid soap, Cassava starch, Absolute ethanol (solvent), Paraffin wax, Camphor and Sodium hydroxide. Some of the equipment used are: AIBOTE Soxhlet extraction apparatus, Labkafe Water bath, Wincom Electronic weighing balance, Cannon BS/U-tube viscometer, Xiangtian Centrifuge machine, and NANBEI PH-1B digital pH meter.

Extraction of *Moringa oleifera* by Soxhlet method: The Moringa (*Moringa oleifera*) was locally sourced. After which they were rinsed and taken to the laboratory. They were properly hand-mashed before being placed in the Soxhlet extractor. Ethanol was used during this process. About 500 mL of ethanol was poured into a round bottom flask and was positioned into an electrothermal heater. The glass joints of the Soxhlet machine were properly lubricated before coupling. The condenser was then linked to the chiller and thereafter, the electrical source was switched on to commence the extraction. The 275 mL of the extract was obtained after the extraction.

Extraction of cashew leaf (*Anacardium occidentale*) by Soxhlet method: The cashew leaf (*Anacardium occidentale*) was locally sourced and washed properly to remove dirt. They were also hand mashed for easy solvent penetration, obtaining suitable extract before being placed in the Soxhlet extractor. The volume of the solvent (ethanol) used was 300 mL to achieve the desired concentration. The volume of *Anacardium occidentale* extract obtained after the extraction process was 130 mL.

Preparation of the emulsion used: The emulsion used for this study was a water-in-oil emulsion and it was prepared using brine and oil in the following proportions:

- The emulsion was prepared in a ratio of 60 to 40% for oil and water, oil is the continuous phase, and brine is the dispersed phase
- The brine used for the emulsion was at 90,000 ppm which is 90 g/L
- The emulsion was blended for 5 min to attain a homogeneous mixture

Preparation of the formulated demulsifiers: The materials and their functions used in preparing the demulsifiers are shown in Table 1.

Procedure for formulating cashew leaf (*Anacardium occidentale*, B-1) and Moringa leaf (*Moringa oleifera*, C-1) demulsifiers:

B-1 and C-1 were formulated using the following materials:

- *Anacardium occidentale* extract
- NaOH   Camphor
- Paraffin wax   Cassava starch
- Liquid soap   *Moringa oleifera* extract

- 45 g of distilled water was measured using the electronic weighing balance, after which 15 g of starch was also measured and dissolved in the distilled water
- 30 g of camphor is dissolved in 60 g of the plant extract at 60°C, stirring continuously until it was properly dissolved
- 15 g of paraffin wax was dissolved in the mixture of the camphorated extract
- The coagulated starch is then added to the mixture while stirring continuously
- 15 g of NaOH was dissolved in 45 g of distilled water and added to the mixture
- The mixture was then stirred continuously while 60 g of liquid soap was added to it
- The mixture was blended in a blender for 5 min to attain homogeneity of the mixture and thereafter, stored in a sealed bottle as shown in Fig. 1

Procedure in obtaining the rheology of the demulsifier B-1 and C-1:

- The viscosity of the samples was obtained with the aid of a Cannon BS/U-tube viscometer and the readings were taken at 300 and 600 rpm
- The gel strength of the samples was also measured at 10 sec and 10 min
- The pH of the samples was measured using the NANBEI PH-1B digital pH meter, which was done by immersing the electrode into the sample

Fig. 1: Samples of B-1 and C-1 formulated demulsifiers

Table 1: Functions of each material in the formulation

Materials	Functions
Cashew leaf	It contains polyphenols, which reduce interfacial tension
Moringa oleifera	It acts as a surface-active compound that reduces surface tension and disrupts interfacial film
Liquid soap	It serves as a binder of the formulation of locally sourced materials to bind the lipophilic and the hydrophilic end
Paraffin wax	It also serves as a water-repellent
Cassava starch	This forms the hydrophilic end of the locally sourced demulsifier because of its strong affinity for water
Camphor	It forms a lipophilic end of the local material sourced
Sodium hydroxide	It acts as a flocculant

Performance evaluation: The performance of the formulated demulsifiers was evaluated using the static bottle and dynamic test methods[8,9].

Static bottle test: A static bottle test was carried out to determine the efficiency of the demulsifiers at static conditions. The prepared emulsion was properly agitated before being used to achieve proper mixing of the crude and brine solution. During the test, the samples were placed in the water bath to reduce their viscous nature, thereafter, varying samples of the demulsifier were injected into a 12 mL labeled emulsion samples. The centrifuge tube was agitated properly, measurements were taken at 10, 20, 30, 40, 50, and 60 min to determine the amount of oil and water separated from each sample and recorded. The experiment was repeated using different dosages: 0.01, 0.02, 0.03, 0.04, and 0.05 mL, respectively.

Dynamic bottle test: The dynamic bottle test was carried out to measure the efficacy of the demulsifiers to break down the emulsion at dynamic conditions. The procedure entails adding varying quantities of the demulsifiers in the centrifuge tube with 12 mL of the emulsion prepared. Here, the centrifuge machine is connected to a power source, the process was carried out at 500, 1000, 1500, and 2000 rpm, respectively, and results were taken at 10 min intervals while measuring the quantities of separated brine and crude. The experiment was also repeated at different dosages of the demulsifiers: 0.01, 0.02, 0.03, 0.04, and 0.05 mL, respectively.

Statistical analysis: The analysis includes an ANOVA single-factor test performed at a 0.01 mL dosage with a significance level of $\alpha = 0.05$ to compare the means of three demulsifiers (pt6020, B-1, and C-1). Additionally, a t-test: Paired two sample for means was conducted at the same dosage to assess the performance of pt6020 and B-1, along with Pearson correlation analysis to determine their relationship. These tests were repeated for dosages of 0.02, 0.03, 0.04, and 0.05 mL.

RESULTS AND DISCUSSION

Table 2 shows the viscosity and density of the crude oil sample, the cashew leaf extract (*Anacardium occidentale*), and the Moringa leaf extract (*Moringa oleifera*) used for the experiment. From Table 2 it can be seen that the cashew leaf extract is most dense of all the samples while the crude oil is the most viscous compared to Moringa and Cashew leaf extracts.

The rheology of the formulated demulsifiers were shown in Table 3. From Table 3, it could be seen that B-1 was less viscous than C-1 as it attained a reading at 300 and 600 rpm which C-1 could not attain during the test. The viscosity at 300 and 600 rpm of C-1 exceeded the viscometer scale, depicting how viscous the demulsifier was. Both demulsifiers are alkaline in nature with pH readings above 13. The plastic viscosity is for B-1 is 90 rpm.

Figure 2 it could be seen that the best water separation was achieved with the conventional demulsifier at 0.02 mL dosage and this was closely followed by 0.03 mL dosage. The maximum quantity was obtained at 0.02 mL after 60 min of the static bottle test.

Figure 3 shows the performance of the formulated B-1 demulsifier using cashew leaf extract. As it can be seen in Fig. 2, 0.02 mL dosage of the B-1 demulsifier gave the best performance, and there was a progressive performance in breaking the emulsion as time progressed, achieving optimum of water separated after 30 min with 4.8 mL at 0.02 mL dosage. This shows a better performance compared to the conventional demulsifier (pt6020), which at the same time was able to separate only 4.6 mL of water and peaked at 4.9 mL after 60 min.

Table 2: Rheological properties of the extracts and the crude oil sample used in this study

Extracts	Viscosity (cp)	Density (g/cm^3)
Crude oil	12.00	0.886
Cashew leaf extract	02.30	0.953
Moringa extract	02.20	0.919

Table 3: Rheology of the demulsifier B-1 and C-1

	300 rpm	600 rpm	Gel at 10 sec	Gel at 10 min	PH
B-1	240	330	20	20	13.43
C-1	-	-	59	60	13.63

Cashew leaf (*Anacardium occidentale*): B-1 and Moringa leaf (*Moringa oleifera*): C-1 demulsifiers
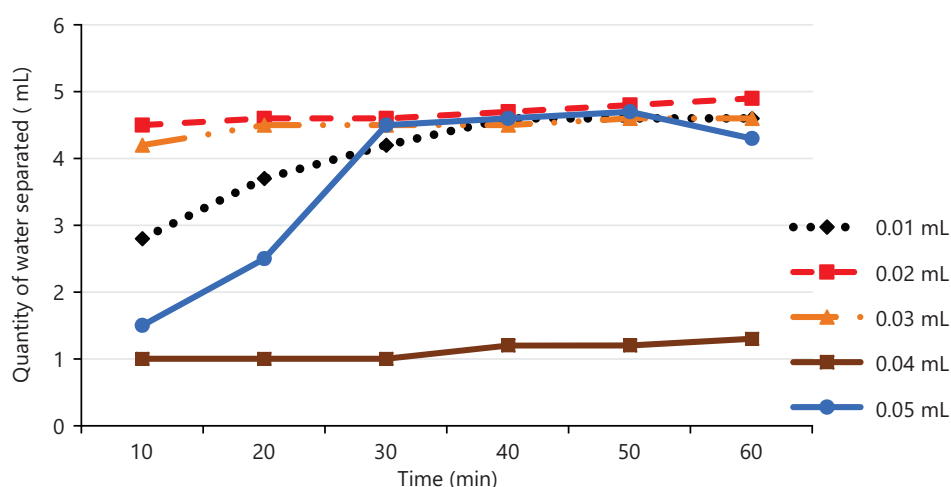

Fig. 2: Quantity of water separated per time using different dosages of the conventional demulsifier (pt6020) for static bottle test
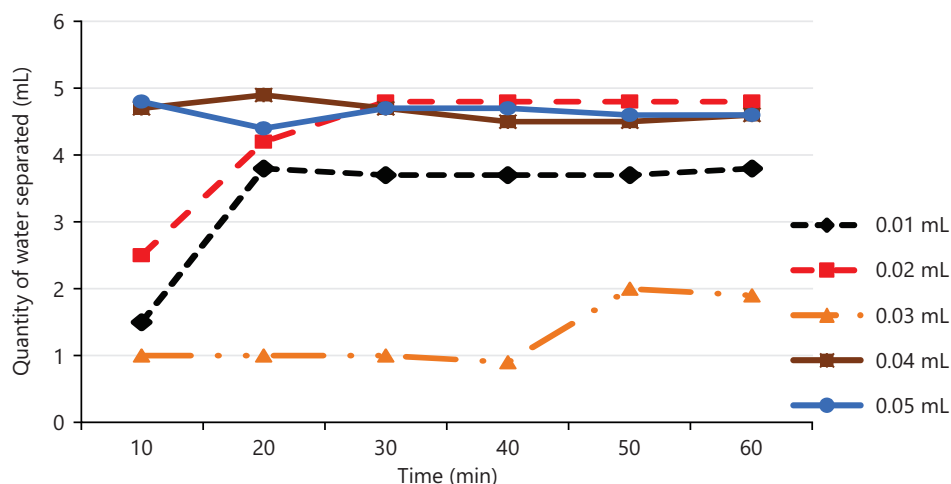
Fig. 3: Quantity of water separation per time using different dosages of B-1 demulsifier during the static test
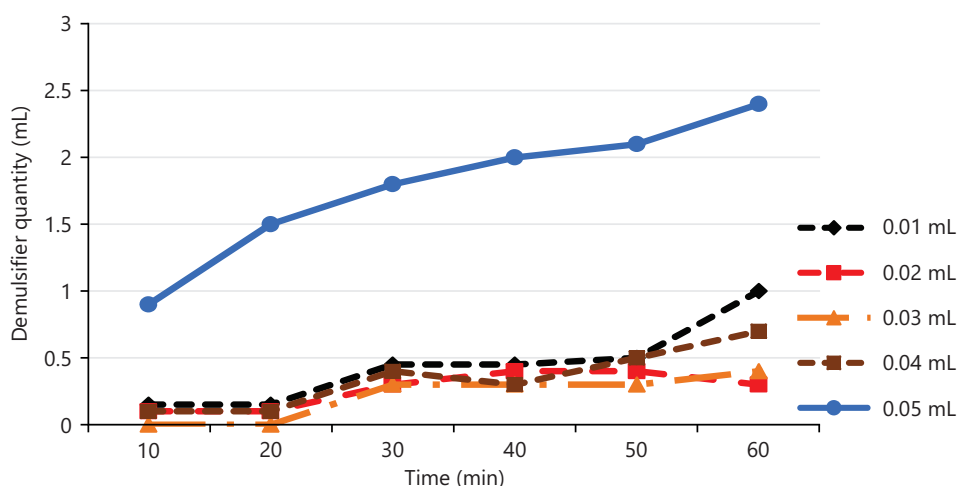

Fig. 4: Quantity of water separated per time using different dosages of C-1 as a demulsifier during the static test

The performance of the formulated C-1 demulsifier was very poor during the static bottle test compared to the B-1 and conventional (pt6020) demulsifiers as can be seen volume of water separated in Fig. 3. The best performance was seen at the highest dosage of 0.05 mL where 2.4 mL was only separated after 60 min of the bottle test. Moreover, the fact this was achieved at the highest dosage makes it less attractive as the lower the dosage might be better for a given demulsifier since demulsifiers are generally expensive and prevent demulsifier accumulation effects on the water drop-off[13].

In Fig. 4 and 5, increasing the dosage from 0.01 to 0.02 mL increased the volume of water separated for both pt6020 and B-1 demulsifiers[14]. However, increasing the dosage to 0.03 mL decreased the water separation compared to 0.02 mL. As the dosage increased to 0.04 and 0.05 mL, the performance of pt6020 dropped further while that of B-1 significantly improved as the residence time increased from 10-60 min with a minimum relative percent error of 2%. Again, it could be seen that as the residence time increases, the amount of water separated increases for both pt6020 and B-1 demulsifiers[15,16].

Unlike the static bottle test, the dynamic bottle test shows that the conventional demulsifier (pt6020) performed best at 1500 rpm and 0.01 mL dosage, which implies that the demulsifier 9 performs better at
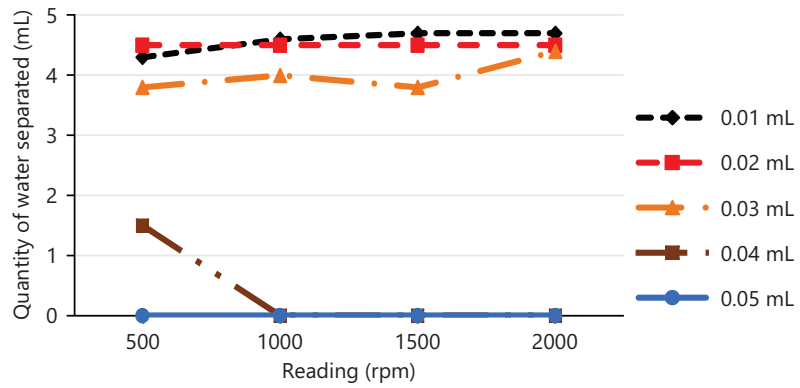
Fig. 5: Water separated at different dosages of pt6020 during the dynamic test
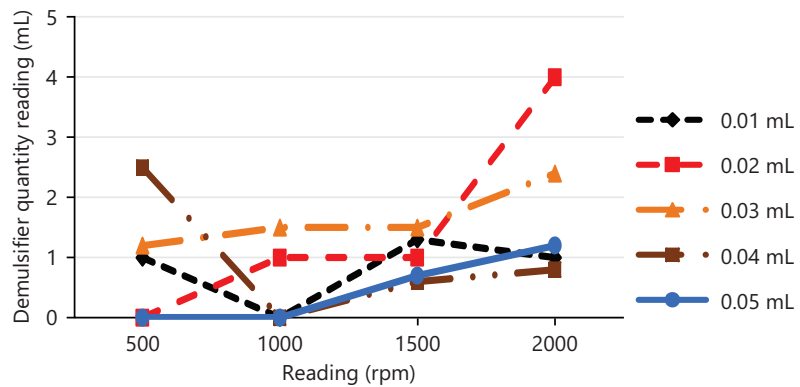

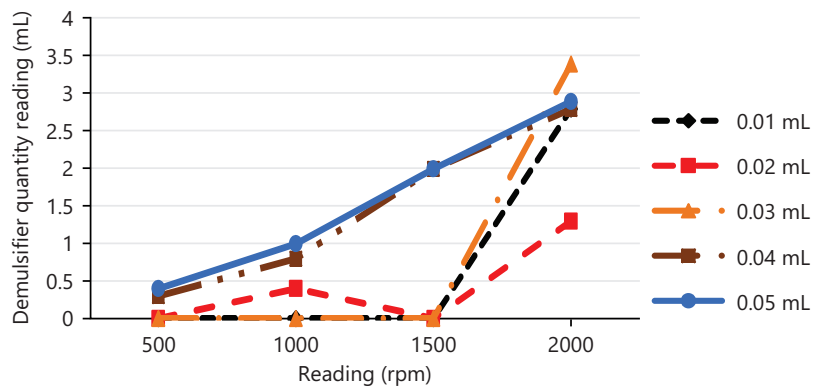Fig. 6: Water separated at different dosages of B-1 demulsifier during the dynamic test

Fig. 7: Water separated at different dosages of C-1 demulsifier during the dynamic test

higher resolutions and with lower concentration[17-19] as shown in Fig. 5. As the concentration increases, the efficiency of the demulsifier decreases[20].

As be seen in Fig. 6 and 7, the formulated B-1 and C-1 demulsifiers performed poorly compared to the conventional (pt6020) at dynamic conditions. The maximum water separation was 4 and 3.4 mL at 0.02 mL and 0.03 mL dosages for the B-1 and C-1 demulsifiers at 2000 rpm compared to the conventional demulsifier which separated 4.7 mL at 1500 rpm for a dosage of 0.01 mL.

Figure 8 shows the performance comparison of the different demulsifiers for the bottle test experiment at different demulsifier dosages. Figure 8(a) is for 0.01 mL, Fig. 8(b): 0.02 mL, Fig. 8(c): 0.03 mL,
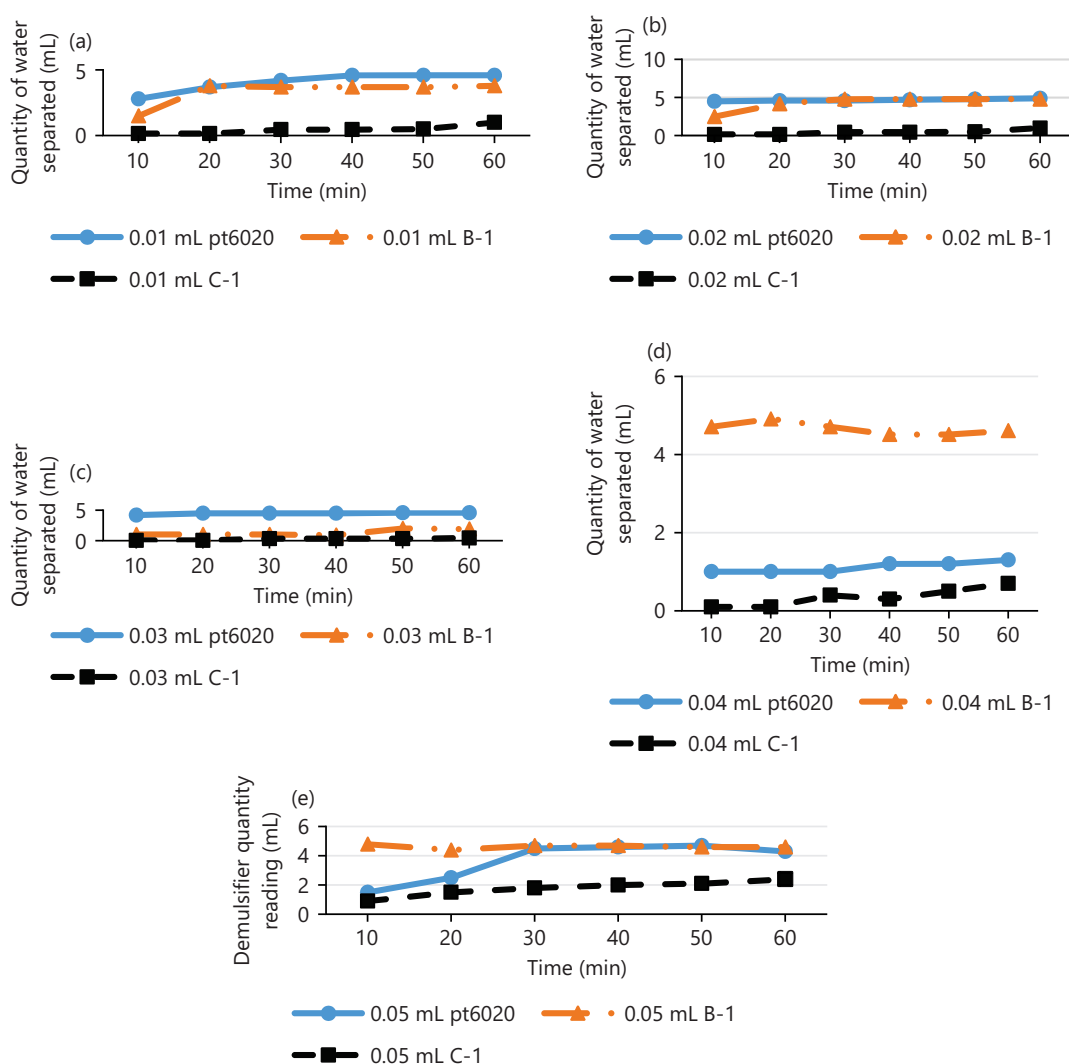
Fig. 8(a-e): Comparison of the performance of the conventional pt6020 demulsifier with the formulated B-1 and C-1 demulsifiers at different demulsifier dosages, (a) 0.01 mL, (b) 0.02 mL, (c) 0.03 mL, (d) 0.04 mL and (e) 0.05 mL

Fig. 8(d): 0.04 mL and Fig. 8(e): 0.05 mL, respectively. As it can be seen in Fig. 8, except for the 0.01 and 0.03 dosages, the B-1 performance compared very well with conventional demulsifier. The C-1 shows abysmal performance at all concentrations considered.

Statistical analysis findings: Table 4 shows an Anova single-factor analysis for the three demulsifiers at 0.01 mL dosage at $\alpha = 0.05$. From the results in Table 4, it can be seen that there is a reasonable difference in the sample means at 4.0833, 3.3667, and 0.45 for pt6020, B-1, and C-1, respectively. Moreover, because Abs(F)>F crit, and the $p<0.05$, we reject the null hypothesis. Again, the variance of C-1 is far higher compared to pt6020 and B-1, further analysis was focused on comparing performance between pt6020 and B-1.

Table 5 is a t-test, paired two sample for means at 0.01 mL dosage for pt6020 and *Anacardium occidentale* (B-1) demulsifiers. From Table 5, it can seen that the p-values for a one-tail and two-tail tests (approximately 1%) are less than 5%, implying that the null hypothesis is significant and thus rejected. The Pearson correlation coefficient of 0.85 also shows that a positive correlation exists between the two demulsifiers in resolving oil-in-water emulsions. A similar analysis was done for 0.02, 0.03, 0.04,

Table 4: Anova: Single factor for 0.01 mL dosage

Groups	Count	Sum	Average	Variance		
Summary						
pt6020	6	24.5	4.0833	0.5217		
B-1	6	20.2	3.3667	0.8387		
C-1	6	2.7	0.45	0.097		
ANOVA						
Source of variation	SS	df	MS	F	p-value	F-crit
Between groups	44.4433	2	22.2217	45.7445	4.12951E-07	3.682320344
Within groups	7.2867	15	0.4858			
Total	51.73	17				

SS: Sum of squares, df: Degrees of freedom, MS: Mean square and F: F-value

Table 5: t-Test: Paired two sample for means (0.01 mL dosage)

	pt6020	*Anacardium occidentale* (B-1)
Mean	4.0833	3.3667
Variance	0.5217	0.8387
Observations	6	6
Pearson correlation	0.8577	-
Hypothesized mean difference	0	-
df	5	-
t-stat	3.6954	-
P (T<=t) one-tail	0.0070	-
t-critical one-tail	2.0150	-
P (T<=t) two-tail	0.0140	-
t-critical two-tail	2.5705	-

and 0.05 mL, respectively. From these analysis, it was observed that except for the 0.05 mL dosage, all other dosages show p-values less than 5%:

- Null = Means are equal
- Alt = Means are not equal
- α = 0.05

CONCLUSION

Two plant-based demulsifiers were formulated using cashew extract (*Anarcadium occidentale*) B-1, and Moringa leaf extract (*Moringa oleifera*) C-1 with some additives to resolve water-in-oil emulsion. These green demulsifiers and a conventional chemical-based demulsifier (pt6020) were subjected to static bottle and dynamic tests to determine their effectiveness in resolving water-in-oil emulsion at different demulsifier dosages and rpm for a maximum of 60 min for the static bottle test and 10 min for the dynamic test. The B-1 shows a better prospect as a demuslfier compared to the C-1 and performed better in some cases than the conventional demulsifier (pt6020) considering the volume of water separated. More biodegradable plant extracts are recommended and further investigations should consider the additives that could enhance the performance of the B-1 demulsifier.

SIGNIFICANCE STATEMENT

Following the strong environmental protection and sustainability campaign, efforts are being made to explore green derivatives as substitutes for synthetic and chemical derivatives as oilfield chemicals to solve oilfield water-in-oil emulsion problems. One common oilfield problem that degrades crude oil quality and downgrades its market value is emulsion which is formed when oil combines with water in the presence of an emulsifier and agitation. Thus, this study explores using plant-based demulsifiers as substitutes for chemical demulsifiers. *Anacardium occidentale* and *Moringa oleifera* were explored following their unique properties. It was observed that *Anacardium occidentale* shows a better prospect than *Moringa oleifera* in resolving water-in-oil emulsion.

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