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Evaluation of modified cashew nutshell liquid as natural surfactants for chemical flooding in sandstone oil reservoirs

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Abstract

Bio-based oilfield chemicals are gaining attention due to their availability, cost-effectiveness, and non-toxic nature. Surfactants facilitate recovery of residual oil by reducing the interfacial tension between two immiscible fluids. Cashew nutshell liquid (CNSL) extracted from *Anacardium occidentale* waste shells was modified using triethanolamine and evaluated as a natural alternative to traditional surfactants. Phase behavior analysis, interfacial tension, critical micelle concentration (CMC) measurements, and core flooding analysis were performed to ascertain the compatibility and recoverability of the cashew nutshell liquid derivatives on sandstone reservoirs. Interfacial tension was reduced from 10.46 to 1.66 mN/m at CMC of 1 g/L. Additional recovery factor and displacement efficiency of 12% OOIP and 32.5%, respectively, was achieved at laboratory temperature. The effect of temperature on residual oil recovery was determined by subjecting the oil displacement experiment to reservoir temperature of 80°C, resulting in recovery factor and displacement efficiency of 9% OOIP and 25%, respectively.

Keywords: Enhanced oil recovery, Cashew nutshell liquid, Surfactant flooding, Biomass, Interfacial tension, Biobased surfactants

Introduction

During secondary and tertiary (enhanced oil recovery) phase of oil recovery, certain parameters such as crude oil composition and viscosity, formation type, reservoir temperature and pressure, permeability, reservoir depth, formation brine salinity, and concentration of additives are crucial for a successful oil production. These parameters will also determine the choice of the recovery method to be applied. For instance, if the oil is highly viscous, water flooding and some type of tertiary flooding will not be suitable because the oil will not flow at an economically viable rate [35].

Chemical flooding is a proven enhanced oil recovery method that involves injection of certain chemicals (alkaline, surfactants, and polymers) for the purpose of altering the wettability of the rock surface, decreasing the mobility ratio, reducing the interfacial tension between the displacing and displaced fluids, mobilizing the residual oil, and improving the sweep efficiencies. Surfactants are surface-active chemical additives that adsorb onto the surfaces or interfaces of oil–water system, altering the degree of surface

or interfacial-free energies, thereby reducing the capillary forces responsible for the restrictive movement of the oil and hence increasing oil recovery. They are unique based on their amphiphilic nature which makes them soluble in immiscible interfaces and/or multiphase system such as oil and water. The aim of designing a core flood system using surfactants as additives at optimal concentration is to modify surface properties of the fluid and rock formation and obtain a low interfacial tension at critical micelle concentration. A decrease in interfacial tension brought about by a reduction in the capillary pressures causes the residual oil in the pore space to flow. In addition, by forming emulsions of water and oil, larger droplets of oil turn into fine droplets which can freely move in the rock cavities [24].

The use of surfactants during chemical flooding spans over four decades [15, 19]. Several successful field applications have been recorded in various fields across the world such as Semonga field in Indonesia where injection of surfactants into the carbonate reservoir resulted in 8% reduction in water cut and increased oil recovery [33] and the Daqing oilfield (Chao 522 block) in China with a reported 30% increase in oil recovery from a single well after the injection of 1.0% nonionic surfactant [41]. Furthermore, Aslam et al. [4] reported an increase in oil recovery during surfactant flooding using 2.05% anionic surfactant-A127 on Tanjung Field in Indonesia. Despite the success rate of surfactant flooding, the technique has suffered some major setback over the years due to certain factors such as the high cost of surfactant chemicals and its non-biodegradable nature [27]. Recent studies on the use of natural-based surfactants as an alternative to synthetic surfactants is fast gaining global attention among researchers even as the global call for a greener environment increases. Nowrouzi et al. [25, 26] reported that natural surfactants are readily available, cheaper, environmentally friendly, and have excellent surface activities. The result of an experimental work conducted by Khayati et al. [23] using natural surfactant (saponin) showed that 5 g/L saponin reduced the interfacial tension and attained an additional oil recovery of 6–8% OOIP. Several researchers [25, 26, 28, 30, 40], reported the efficiency of some biomass based (agro-waste) chemicals as natural surfactant additives in displacing residual oil and increasing oil recovery between 19 and 23% OOIP.

Cashew nutshell is an abundant agro-industrial residue predominant in subtropical countries such as Brazil, Vietnam, India, Nigeria, and Burkina-Faso. Cashew nutshell liquid (CNSL) is a dark brown viscous and caustic extract readily obtainable from the shell of cashew nuts. It is a by-product of the cashew nut industry where the cashew kernel is the main interest and the shell is an agricultural waste turned into renewable resource because though not a food source, it has many technical uses. Natural (cold-extracted) cashew nutshell liquid (CNSL) consists of four structurally related alkyl/alkenyl phenols: anacardic acid (82%) (6-pentadecyl salicylic acid), cardol (13.8%) (5-pentadecyl resorcinol), cardanol (2.6%) (3-pentadecyl phenol), and 2-methyl cardol (1.6%) (2-methyl-5-pentadecyl resorcinol) [32, 38].

CNSL is a low cost, effective, renewable, and technologically significant raw material suitable as a replacement for synthetic phenols in various industries [18]. In the oil and gas industry, cashew nutshell liquid and its derivatives have been found to be good corrosion inhibitors and flow improvers for waxy and heavy crude oil [7, 12, 31]. Polyethylene glycol and diethanolamine esters of natural cashew nutshell liquid

as well as formaldehyde-novolac resins of distilled cashew nutshell liquid were effective as crude oil wax dispersants/inhibitors and pour point depressants [8, 9, 17, 18] reported the extensive use of cardanol and cardol and their derivatives as additives for the production of surfactants, resins, and polymers. However, no study has reported the application of CNSL derivatives in chemical EOR processes. In this work, natural cashew nutshell liquid which contains mainly anacardic acid was modified by esterification with triethanolamine to form di- and tri-esteramines. The rationale was to improve the surfactant properties of the major alkyl phenol in natural cashew nut liquid (6-pentadecyl salicylic acid) by chemically incorporating triethanolamine which is also a known surfactant. The acid group of the anacardic acid (6-pentadecyl salicylic acid) is reactive and easily esterified by the alcohol groups of triethanolamine to form the diester (TA-2) and triester (TA-3) derivatives, which consist of two and three anacardyl groups respectively connected to the amine by ester linkages. Thus, the nonionic surfactants produced possess ester and amine hydrophilic head groups and alkylated (pentadecyl) benzene groups as hydrophobic tail. As noted by Ogolo et al. [29], such chemical modifications yield more effective derivatives.

This paper presents novel application of CNSL-based surfactants (developed from natural cashew nutshell liquid by esterification with triethanolamine) in enhanced oil recovery process at elevated temperatures. It advances the concept from simply using raw biomass extracts as EOR agents or as additives in product formulations to executing systematic chemical modifications of biomass-based chemicals with the aim of enhancing the performance of the naturally occurring chemicals for the targeted application. Reservoir fluids and rock samples from an oilfield in the Niger Delta of Nigeria were used as case study in the work. Series of tests such as brine compatibility, surfactants interfacial tension modifications, salinity scan, and phase behavior tests as well as core flooding were conducted to investigate the efficiency and the prospect of using these biobased surfactants in enhanced oil recovery operation in the reservoir.

Methods

Materials

Natural agro-waste, cashew nutshell was locally sourced from the farmer's market at Ogbia town (4.6901 °N, 6.3213 °E) in Ogbia LGA, Bayelsa State. Deionized water and high-grade chemicals such as toluene, sodium chloride, calcium chloride, and magnesium chloride with their specifications outlined in Table 1 were purchased from local suppliers. Dead crude oil with an acidity of 0.9 mg KOH/g was obtained from a Niger Delta oil field, south-south of Nigeria.

Table 1 Properties of brine salts

Salt type	Molecular weight (g/mol)	Density (g/cm ³)	Water solubility(g/L)
NaCl	58.44	2.16	359
CaCl ₂	110.98	2.15	600 – 1520
MgCl ₂	95.21	2.32	520 – 720

Apparatus

Porcelain crucible, furnace, desiccator, sample cup, beaker, gas-chromatograph flame ionization detector (GC-FID), Fourier-transform infrared spectroscopy (FTIR), pH meter, water bath, thermometer, rotary evaporator, core flooding apparatus, capillary viscometer, magnetic stirrer, filter papers, core plug, funnels, beakers, glass tubes, pipettes, and Sigma 703D tensiometer.

Experimental procedures

The methodology adopted in the course of this study include the following: preparation of agro waste sample, extraction of cashew nutshell liquid (CNSL), characterization using FTIR, modification of cashew nutshell liquid, preparation of brine sample, characterization of crude oil, determination of physio-chemical properties of CNSL, interfacial tension analysis and CMC determination, phase behavior analysis, and finally coreflooding application of the modified surfactants as presented in the flowchart in Fig. 1.

Extraction of cashew nutshell liquid

Cashew nutshell liquid was extracted from waste cashew nutshell using acetone as described in literature [7]. The solvent was recovered in vacuo using a rotary evaporator. Extraction of CNSL using low-boiling solvents yields natural CNSL which consists of mainly (about 82%) anacardic acid [38].

Modification of cashew nutshell liquid

The CNSL was modified by mixing it with triethanolamine (TEA) at different ratio to produce two surfactants: TA-2 and TA-3. The TA-2 was produced with mixture of 38.1 ml TEA and 200 ml natural CNSL which is approximately 1:2 mol ratio in a three-necked flask. A catalytic quantity of sulfamic acid was added to the mixture. The mixture of CNSL and TEA was accompanied with the evolution of heat and the mixture was heated to 115–130 °C. The water of esterification was collected in a Dean-Stark trap, and

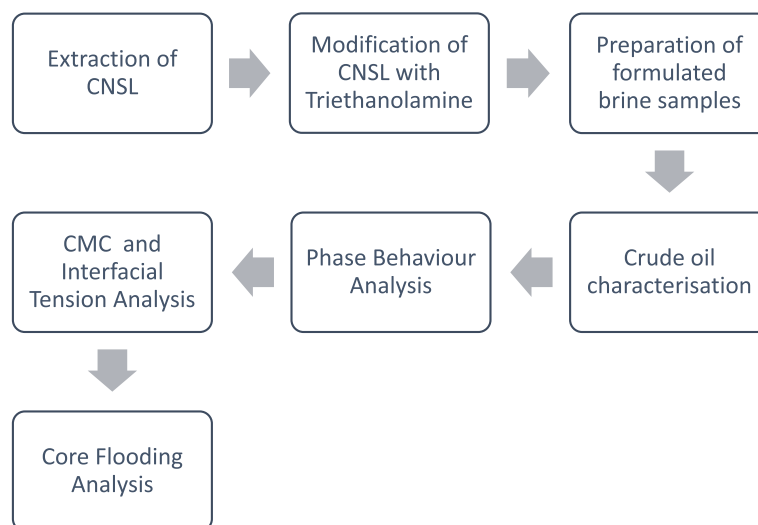


Fig. 1 Flowchart showing experimental procedure

the reaction was completed in 60 min upon cessation of water collection. A similar reaction was carried out using the same volume of TEA combined with 300 ml of natural CNSL equivalent to 1:3 mol ratio of reactants to produce the second surfactant, labeled TA-3. The spectra of each product and the reactants were recorded with a Fourier transform-infrared spectrophotometer.

Preparation of brine sample

Two types of formulated brines were prepared to replicate the formation brine using deionized water. The brine contained varying concentrations of sodium chloride, calcium, and magnesium chloride with a total dissolved solids (TDS) of 30,000 ppm with 5000 mg/l being total concentrations of calcium and magnesium chloride (divalent ions). The composition of the brines as presented in Table 2 was formulated based on the report of [5] which noted that the TDS of most formation water in Niger Delta reservoirs vary between 14,537 and 31,761 ppm with its composition in the order of Na > Ca > Mg > K. The varied concentrations were mixed in a beaker containing one litre of distilled water, and this was placed on a magnetic stirrer at a temperature of 27 °C for 10 min. The solution was allowed to settle and then filtered using a filter paper. The filtered solution was kept in a properly sealed vacuum flask and labeled FMB (formation brine). A similar process was performed using only sodium chloride at concentration of 30,000 ppm and labeled SFB (soft formation brine).

Characterisation of crude oil

The crude oil was characterised to determine its viscosity and specific gravity. Dynamic viscosity of the crude oil at 30.5 °C was determined using a rheometer. Specific gravity of the crude was also determined using a hydrometer with ASTM D1298-12b standard test method. The total acid number (TAN) of the crude oil was already determined.

Determination of physio-chemical properties

Varied concentrations of TA-2 and TA-3 in 100 ml deionised water was prepared at intervals into a glass beaker and properly stirred for 10 min, using a magnetic stirrer. Physio-chemical properties such as pH value and dynamic viscosity of the two natural surfactants were determined. The pH value was measured using a pH meter to determine the level of acidity or alkalinity of the surfactants while the dynamic viscosity at 30 °C (using a capillary viscometer to ascertain the viscous nature of the surfactants with respect to the crude oil).

Table 2 Composition of synthetic brine

FMB composition	Brine concentration(g/L)	SFB composition	Brine concentration (g/L)
NaCl	25.0	NaCl	30.0
CaCl ₂	3.0	-	-
MgCl ₂	2.0	-	-
TDS	30.0	TDS	30.0

Compatibility test

To analyze the fluid–fluid compatibility at different concentrations and temperatures, an aqueous stability test was conducted to determine the presence of any non-homogeneity such as phase separation, cloudiness, and precipitation in the aqueous phase. Sealed beakers containing 100 ml of brine with varying surfactant concentrations was observed at laboratory and increased temperature of 80 °C for about 48 h. This screening ensures a surfactant that is compatibility with the electrolyte concentration.

Interfacial tension analysis

The surface activities of TA-2 and TA-3 surfactants were investigated based on their interfacial tension (IFT) potential using the Du Nouy ring method with the aid of Sigma 703D tensiometer. The experiment was conducted in three phases using crude oil with deionised water and brines. In the first phase, the effect of varied surfactant concentrations on oil-deionized water was investigated. This was used to identify the critical micelle concentrations (CMC) beyond which surface activities of the surfactants do not change much with increase in their respective concentration.

Based on the results of the first phase, a fixed concentration of the surfactants with low IFT potential was identified and used in the second and third phase. The effect of varied brine composition on oil-brine interaction was investigated in the second phase. Brines of NaCl, CaCl₂, MgCl₂, and formation brine (FMB) with concentrations equivalent to their respective concentrations in the formation brine were used. This was aimed at identifying the effect of each salt on oil-brine IFT in the absence and presence of surfactants.

In the final phase, the effect of varied brine salinities on oil-brine IFT was investigated with and without surfactants. Three brine salinities used are formation brine (FMB), 50% formation brine (50D) and 10% formation brine (90D). This test helps us to see how the salinity level of brine influences its interaction with oil in the absence and presence of surfactants. A fixed concentration of 1 g/L of surfactants was used for all the tests conducted in phases two and three. All the experiments were conducted at ambient temperature of 25 °C.

Salinity scan test

Salinity scan test to determine the salinity tolerance of both brines (FMB & SFB) at varying electrolyte concentrations ranging from 0.1 to 1.0% carried out under ambient and increased temperature of 25 °C and 80°C, respectively. Surfactant concentration was kept constant, while brine salinity was changed. The experiment was conducted using a glass test tube each containing a total volume of 10 ml. The solution was mixed based on calculations of molarity as shown below:

$$C_1 V_1 = C_2 V_2 \quad (1.0)$$

where C_1 is initial concentration (%), C_2 is end concentration (%), V_1 is start volume (ml), and V_2 is total volume (ml). Pipette or phase separation test was conducted using pipettes containing equal volumes of aqueous pipette or phase separation test was conducted using pipettes containing equal volumes of aqueous solution (at

different concentrations) and crude oil, and the tightly sealed pipettes were carefully inverted to ensure a proper mix of the two phases at laboratory and reservoir temperature of 80 °C over an equilibration period of 21 days to ascertain the presence of microemulsion and the type formed.

Core flooding analysis

Core flooding experiment was conducted to determine the oil displacement efficiency and recovery factor of the modified natural surfactants using sandstone core plug. The absolute porosity and pore volume of the core sample was calculated. An AFS-300 Core flooding system was used in conducting the oil displacement experiment at reservoir conditions of 80 °C and 9000 psi. The surfactant at CMC value was used as surfactant concentration while brine concentration of 3% salinity was adopted. The injection rate was referenced from the works of [3]. The core flooding experiment followed a sequence from the initial brine saturation of the sandstone core, drainage, imbibition, and finally surfactant flooding. At the end of each experiment stage, the core was cleaned using toluene and deionized water.

Drainage Prior to the drainage experiment, the brine saturated sandstone core was placed in the core holder. Medium crude was injected into the core to displace brine until an initial drop of oil was seen. The displaced brine was collected into burettes and the original oil in place (OOIP), initial oil saturation, and irreducible water saturation were determined.

Imbibition This was done to imitate secondary oil recovery. Formulated brine containing divalent ions (FMB) was used as the displacing fluid at a constant flow rate of 1 ml/min to determine the effect of divalent ions (typically found on the rock surface) on the natural surfactant. Brine flooding up to 98% water cut was injected to completely displace oil to residual oil saturation.

Surfactant flooding The 2PV of surfactant-brine solution was used as the displacing fluid at a constant injection rate of 1 ml/min. This was injected into the core to enhance recovery of the residual oil at ambient and reservoir temperatures of 25 °C and 80 °C, respectively. The surfactant flooding continued until an oil cut of less than 1% was recorded and the recovery factors were determined.

Results and discussion

Crude oil characterization

The values obtained for certain properties of the crude oil sample are shown in Table 3. The resultant API gravity value falls within the medium crude which API gravity values range between 22.3 and 31.1°.

FTIR analyses

The FTIR spectra of natural CNSL and triethanolamine are shown in Fig. 2 while Fig. 3 presents the spectra of the CNSL-esteramines (TA-2 and TA-3). The broad band at

Table 3 Physical properties of crude oil sample

Physical property	Value
Specific gravity at 27°C	0.9 kg/m ³
API gravity	22.5°
Viscosity at 30.5°C	50.64 cP
Color	Dark brown
pH	6.2
Total acid number (TAN)	0.9 mgKOH/g

3306.1 cm⁻¹ in the spectrum of triethanolamine (TEA) is due to the O–H vibrations of alcohol group which is confirmed by C–O of primary alcohol at 1028 cm⁻¹. This intense band at 1028 cm⁻¹ is absent in the spectra of the derivatives (TA-2 and TA-3) which suggests that the alcohol groups are utilized in ester bond formation. This is corroborated by the higher transmittance value of the broad O–H band of alcohol in the derivatives relative to TEA, which increased from 63% in TEA to 85% in the derivatives, suggesting diminution of the alcohol groups (Figs. 2 and 3). The strong sharp band at 1699.7 cm⁻¹ in the spectrum of natural CNSL corresponds to the C=O stretching vibration of carboxylic acid attached to aromatic ring (confirmed by the C–O stretch vibrations of the acid 1211.4 cm⁻¹) which is characteristic of an anacardic acid rich liquid [6] (Fig. 2).

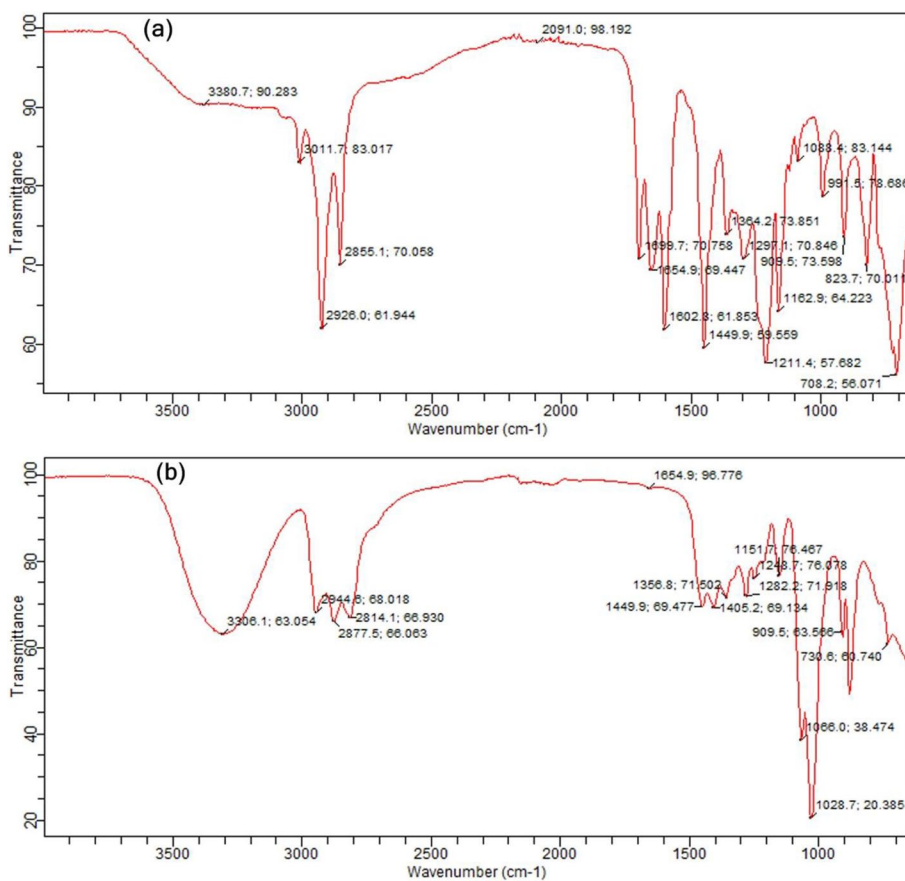


Fig. 2 FTIR spectra of natural cashew nutshell liquid (CNSL) and triethanolamine (TEA)

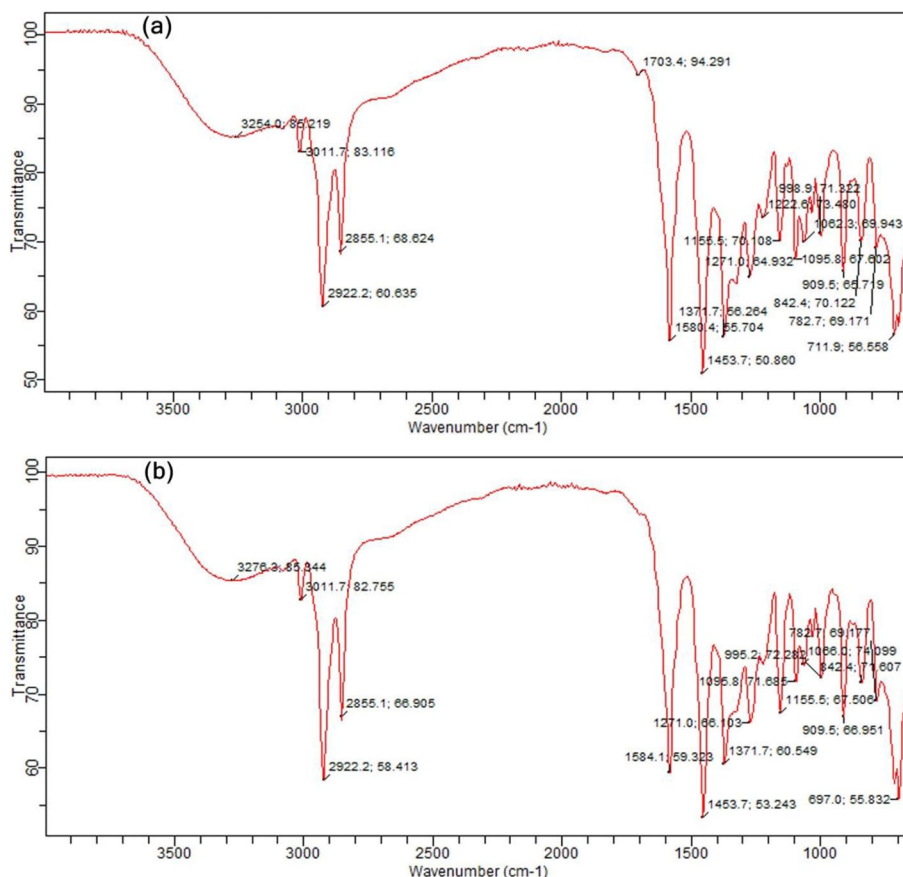


Fig. 3 FTIR spectra of cashew nutshell liquid-triethanolamine derivatives (esteramines)

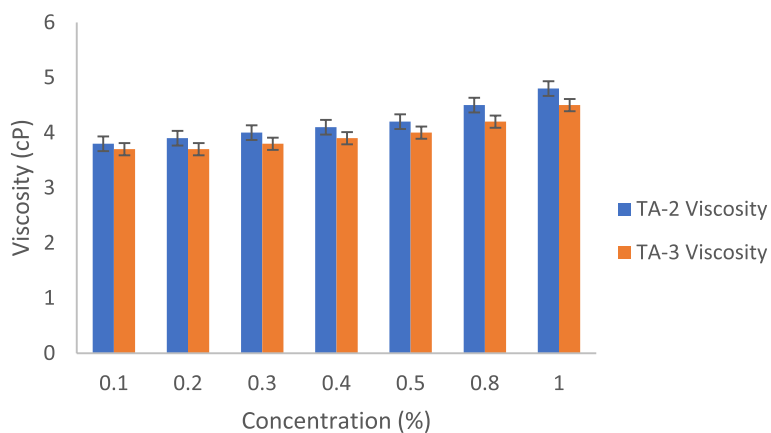


Fig. 4 Effect of varied surfactant concentration on viscosity

These bands are absent in the spectra of TA-2 and TA-3, indicating conversion of the carboxylic acid groups to ester groups. The new band appearing at 1271 cm⁻¹ in TA-2 and TA-3 corresponds to C–O of ester groups [11] as seen in Fig. 3. These observations

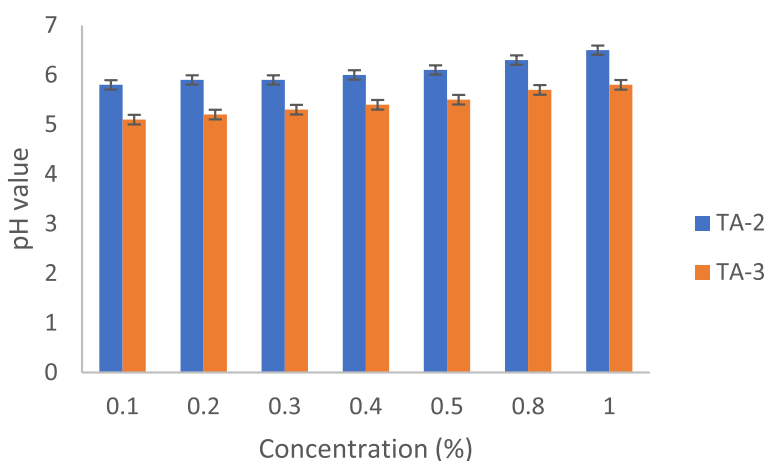


Fig. 5 Effect of varied surfactant concentration on pH value

indicate that ester linkages are formed between anacardic acid of CNSL and alcohol groups of TEA.

The band at 3011.7 cm^{-1} is the $=\text{C}-\text{H}$ stretching vibration of alkene originating from the 15-carbon aliphatic chains, while 2926.0 cm^{-1} and 2855.1 cm^{-1} are due to $\text{C}-\text{H}$ vibrations of methylene and methyl groups of the aliphatic chain [6]. Similar $\text{C}-\text{H}$ vibrations of the methylene groups in ethyl chain of TEA occurring at 2944.6 cm^{-1} , 2814.1 cm^{-1} , and 2877.5 cm^{-1} are subsumed within that of CNSL in the ester derivatives. The bands between 1602 and 1580 cm^{-1} in natural CNSL and the derivatives are due to $\text{C}=\text{C}$ stretch of the aromatic group. Furthermore, $\text{C}-\text{N}$ vibration of tertiary amine group contributed by TEA produced the bands at 1356.8 cm^{-1} and 1371.7 cm^{-1} [6, 11].

Physio-chemical properties of surfactants

The viscosity and pH of the two modified natural surfactants, TA-2 and TA-3, are outlined in Figs. 4 and 5. The results obtained indicate that viscosity and pH are functions of concentration, as evident in a corresponding increase in viscosity and pH as the surfactant concentration increases. The pH values obtained are in line with values of commonly used synthetic surfactant, e.g., SDS [36], natural CNSL [13], and certain natural surfactants [28].

The increase in viscosities as surfactant concentration increases reveals that these natural surfactants are non-Newtonian fluids. The dynamic viscosity values obtained for both surfactants fall within the range of $3.8\text{--}4.8\text{ cP}$ at $30\text{ }^\circ\text{C}$. These are higher than water viscosity (1.002 cP) and slightly higher than the viscosities of most synthetic surfactants which ranges between 1.4 and 3.1 cP as noted by Esteves et al. [10]. The viscosity of the crude oil (refer to Table 3) is 50.6 cP and is higher than that of the surfactant; however, viscosity is not an important parameter during surfactant flooding as the displacement mechanism of surfactants is to reduce interfacial tension.

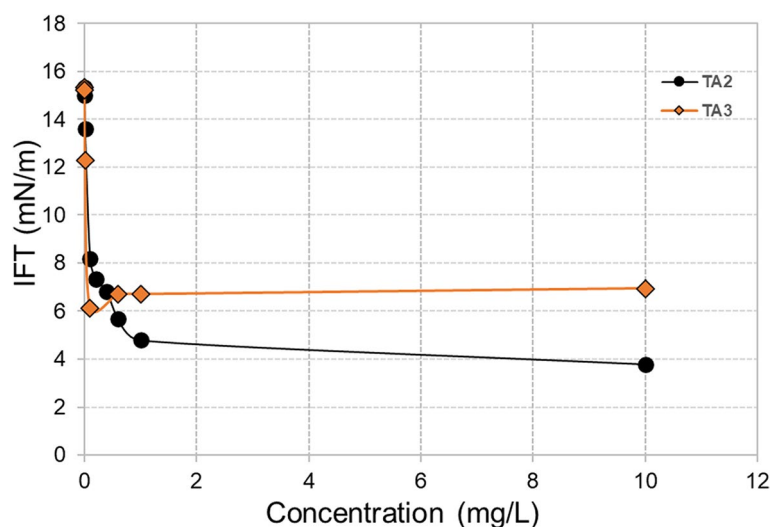


Fig. 6 Effect of surfactant concentrations on IFT reduction used for CMC determination

Compatibility test

Compatibility test conducted between the aqueous phase (TA-2 and FMB) produced solutions with slight precipitates as surfactant concentration increased (0.1–1.0%) under laboratory temperature. Sheng [35] had noted that the solubility of surfactants reduces as the concentration of the surfactants increases. These precipitates were found at the base, but when subjected to reservoir temperature of 80 °C, clear compatible solutions were produced at the various concentrations implying that surfactant solubility increased as the temperature increased. Thus, TA-2 shows high compatibility with formation brine at reservoir temperatures. Similar results were obtained for TA-3 and FMB where precipitates were seen to coagulate. At increased temperature of 80 °C, the precipitates were still visibly at concentrations above 0.7%. The compatibility effect of each salt ion on TA-2 and TA-3 was investigated to determine the cause of the precipitation, results showed that all except calcium ion (CaCl_2) produced precipitates. This implies that the calcium ions present in the formation brine has no effect on the modified CNSL (TA-2 & TA-3).

Critical Micelle concentration (CMC)

The critical micelle concentrations of TA-2 and TA-3 surfactants were determined by measuring their interfacial tensions at different concentrations. Figure 6 shows the results of the interfacial tensions of both surfactants at the varying concentrations. A decrease in interfacial tension was observed with increase in concentrations of the two surfactants, TA-2 however generated lower interfacial tension than TA-3. This observed decrease in IFT with increase in concentrations of surfactants is attributable to interfacial adsorption of the surfactants between the oil and water. At this interface, the molecules of the surfactant are oriented with the hydrophilic heads toward the water and hydrophobic tails toward the oil. It is observed that the initial rapid decrease in IFT with increase in the concentrations of surfactants faded out at higher concentrations due to aggregation of the surfactant molecules in the solution [25, 26, 34].

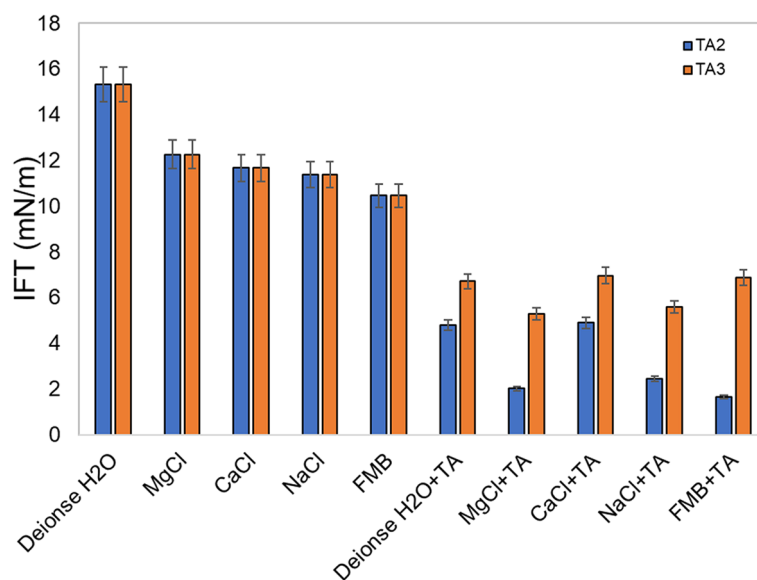


Fig. 7 Effect of varied brine composition on oil-brine IFT with and without surfactants

The concentration at which this aggregation formation is initiated is known as critical micelle concentration (CMC), and beyond this concentration, no significant IFT reduction is observed with increase in concentrations of the surfactants. For the TA-2 surfactant, the CMC was found to be 1 g/L beyond which no significant reduction in IFT was observed. At this concentration, an IFT of 4.79 mN/m was observed, while a slightly lower IFT of 3.79 mN/m was observed at 10 g/L. The difference in these concentrations (1 g/L and 10 g/L) is much more than the IFT reduction difference making it not economically viable to use the high concentration although it generated the lowest IFT. Hence, due to the minimal difference in IFT between surfactant concentrations at 1 g/L and 10 g/L, a more cost-effective option of 1 g/L was adopted for further investigations.

A similar test was conducted to determine the CMC of surfactant TA-3. The result shows that the interfacial tension reduces as the surfactant concentration increases up to 0.6 g/L. Beyond this point, further increase in the surfactant concentration resulted in increase in the IFT values as seen in Fig. 6. A CMC of 1.0 g/L with a corresponding IFT value of 6.71 mN/m was adopted for this surfactant and used for further investigations.

Interfacial tension measurements

The results of the effect of varied brine compositions on oil-brines IFT are presented in Fig. 7. The brine compositions considered are the individual salt compositions that are present in the formation brine (FMB) and the deionized water was used as the based solution for the analysis. These salts include monovalent salt (NaCl) and divalent salts (CaCl₂ and MgCl₂). The concentration of each salts used is equivalent to their respective concentration in the FMB. This enabled us to see the effect of each salt on IFT reduction and their respective contributions to FMB-oil interactions in the absence and presence of surfactants. Generally, higher IFT was observed for all the solutions in the absence of the surfactants. All the brines however had lower IFT

than deionised water, which suggest the possibility of interfacial influence of salt ions. The multicomponent FMB generated the lower IFT of 10.46 mN/m than the single salt solutions that have IFT of 12.27, 11.67, and 11.38 mN/m for MgCl₂, CaCl₂, and NaCl, respectively. The reduction observed with brines relative to the use of deionised water is consistent with previous study by [39] that showed that surface adsorption of salt ions lower interfacial energy.

The addition of the TA-2 and TA-3 surfactants to all the solutions resulted in reduction in their respective IFT relevant to the solutions alone without surfactants. The two surfactants however generated different effects in the same solutions. The observed reduction in IFT with the addition of the surfactants to the solutions is attributable to their interfacial adsorption driven by their molecular structure that allows them to simultaneously interact with oil and water [25, 26]. Better IFT reduction was observed with the TA-2 surfactant than TA-3 surfactant in the brines and deionized water. The lowest IFT (1.66 mN/m) was observed with the addition of TA-2 to multicomponent FMB, which shows that this surfactant is suitable for EOR application in reservoir brines of similar compositions and salinity to the FMB. A closer look at the effect of each single salt on the IFT modification of TA-2 showed that better IFT (2.01 mN/m) was generated with MgCl₂, followed by NaCl that has 2.11 mN/m and least with CaCl₂ that has 4.89 mN/m. Since better IFT was observed with FMB, it shows that this surfactant is more compatible with multicomponent brine than single brine. On the contrary, the TA-3 generated lowest IFT of 5.30 mN/m in MgCl₂, followed by NaCl (5.56 mN/m) and similar effect in deionized water (6.71 mN/m), CaCl₂ (6.59 mN/m), and FMB (6.86 mN/m). This surfactant seems not to be effective in saline environments and may not be efficient in EOR applications in brines with similar compositions to the brines used in this study.

The results of the effect of varied brine salinities on the interfacial tension in the presence and absence of TA-2 and TA-3 surfactants are shown in Fig. 8. Different dilutions

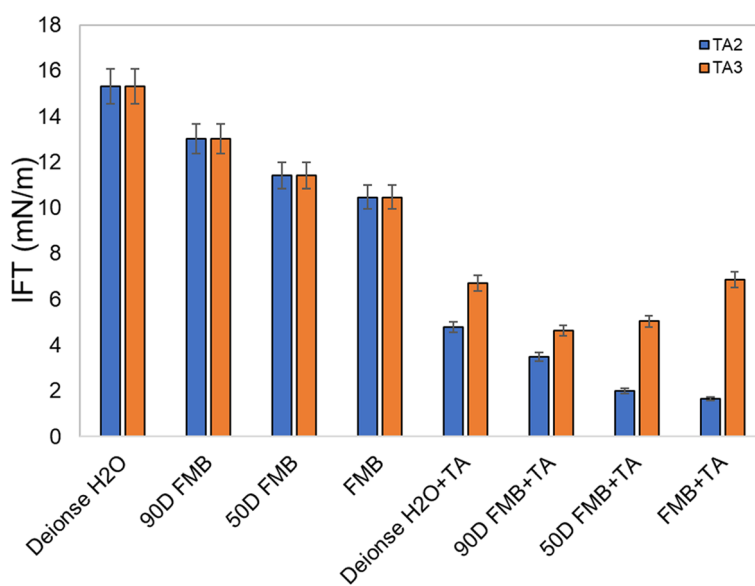


Fig. 8 Effect of varied brine salinity on oil-brine IFT with and without surfactants

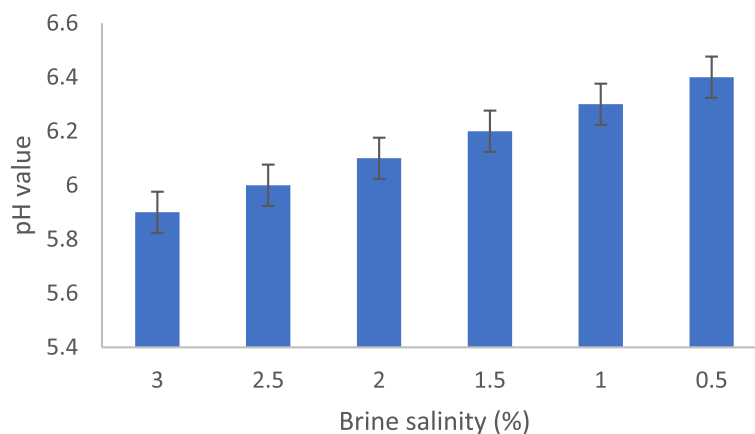


Fig. 9 Plot of salinity scan of TA-3 in SFB

(90% dilution [90D] and 50% dilution [50D]) of the multicomponent FMB were used in this investigation. Reduction in IFT was observed with the use of brines of different salinities relative to the deionised water in the absence of surfactant. This IFT reduction increases with increase in brine salinity, i.e., the lowest IFT 10.46 mN/m was observed with high salinity FMB. This further corroborates the interfacial adsorption of salt ions and the possibility of electrostatic effects brought about by the presence of salt contained in the brine solution which changes the distribution of the polar components at the oil-brine interface [22].

The addition of TA-2 and TA-3 surfactants to all the solutions generally resulted in IFT reduction of varied capacities from 10.46 to 1.66 mN/m and 6.86mN/m for TA-2 and TA-3, respectively. Better IFT reduction was achieved with the use of TA-2 surfactant in brines of varied salinities than TA-3 surfactant. Reduction in IFT with increase in brine salinity was observed with the use of TA-2, while increase in IFT with increase in salinity was observed with TA-3 indicating that there is a relationship between brine salinity and interfacial tension of the oleic-aqueous system, and it is dependent on the type of salt, amount of salt, type, and concentration of surfactant present [37]. This further showed the tolerance and efficiency of TA-2 surfactant in saline environments. The lowest IFT of 1.66 mN/m was observed with the application of TA-2 in FMB, which shows its compatibility with the reservoir formation brine without requiring any further brine treatment. Hence, this brine was used for the core flooding experiment.

Salinity scan analysis

Salinity scan test was performed on TA-2 and TA-3 using both brines; FMB and SFB at varying temperatures. The critical micelle concentration (CMC) of the surfactant obtained during IFT measurement was kept constant while varying the salinity of the brine. In terms of solubility, similar results were obtained for both surfactants in SFB showing clear, compatible, and highly soluble solutions for all salinities both at ambient and reservoir temperatures. On the contrary, TA-2 and TA-3 surfactants in FMB produced slight precipitates at salinities above 2% under laboratory temperature, at increased temperature of 80 °C, compatible solutions for all salinity range were observed. Sheng [35] noted that most petroleum sulfonates (anionic surfactants) which

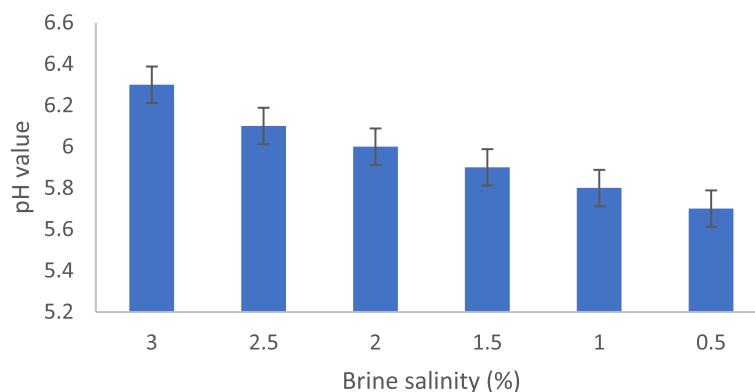


Fig. 10 Salinity scan plot of TA-2 in FMB

are commonly used in surfactant flooding, have limited solubility in brine in the presence of divalent ions. Solubility implies that surfactant has a strong bond with water. He stated that the solubility of the surfactants decreases with increasing salinity and temperature owing to the growing weaker bond in brine. This was observed for these natural surfactants which have similar structures with the nonionic surfactants [25, 26].

Figures 9 and 10 show the effect of brine salinity and divalent ions on pH value. Both surfactants, TA-2 and TA-3 in SFB (brine devoid of divalent ions) showed an increase in pH value of the solution as salinity decreased (as brine salinity increases, pH reduces) while in the presence of divalent ions using FMB, the pH value of the solution increased as the brine salinity increased as seen in Fig. 10 for TA-2.

Furthermore, aside from the solubility and compatibility analysis, the thermal stability of the surfactant chemical over a long period of time at reservoir temperature is crucial in designing a stable yet effective natural surfactant for EOR process. Hocine et al. [16] reported that in an oxygen free environment, sulfur-based anionic surfactants especially sulfonates remained chemically stable for a duration of 1 year at 100 °C temperature. Conversely, [20] in their evaluation of twenty-eight surfactants comprising of the four different types of surfactants, reported that only non-ionic and amphoteric surfactants showed good thermal stability at harsh reservoir conditions. The study on the thermal and chemical stability of these natural surfactants (TA-2 and TA-3) will form part of future studies.

Phase behaviour test

Phase separation test was performed on TA-2 and TA-3 with both brines and crude oil. Firstly, using equal volumes of the medium crude and clear aqueous solution of TA-2 in SFB (brine devoid of divalent ions) at salinities ranging from 0.5 to 3.0%, a type I or lower-phase microemulsion was formed under laboratory and reservoir temperature of 80 °C. A repeat test was conducted using the surfactant solution TA-2 in FMB at salinities ranging from 0.5 to 3.0%, for an equilibration period of 21 days, this also resulted in type I microemulsion, implying that the salinity of the aqueous solution is lower than optimal salinity, so the surfactants are concentrated in the water phase thereby reducing residual oil mobilization. A similar procedure was performed using TA-3 in both brines using the same salinity range

Table 4 Properties of sandstone core plug

Parameter	Value
Core length (cm)	5.80
Core diameter (cm)	3.40
Bulk volume (cm ³)	52.66
Dry sample mass (g)	128.54
Saturated sample mass (g)	142.97
Mass of brine (g)	14.43
Brine density (g/cm ³)	9.08
Pore volume (cm ³)	14.38
Cross sectional area (cm ²)	9.08
Porosity (%)	26.9

Table 5 Fluid properties at laboratory and reservoir temperatures

Fluid properties at laboratory temperature (25 °C)	Properties	Units	FMB brine	Crude oil	Surfactant (TA-2)
	Density	g/cm ³	1.0171	0.9174	1.0013
	API	API ^o		22.5	
	Viscosity	cP	0.97	50.61	4.11
Fluid properties at reservoir temperature (80 °C)					
	Density	g/cm ³	0.991	0.883	0.967
	API	API ^o		23.2	
	Viscosity	cP	0.75	6.50	1.04

Table 6 Initial water saturation and residual oil saturation

Parameter	Value
Pore volume (cm ³)	14.38
Irreducible water saturation at maximum oil saturation (cm ³)	3.38
Initial water saturation (%)	23.51
Volume of crude in the plug at S_{wi} (cm ³)	11
Initial oil saturation (%)	76.49
Volume of crude recovered after imbibition (cm ³)	7
Volume of residual crude oil (cm ³)	4
Residual oil saturation (S_{or})	0.3636
Residual oil saturation (S_{or}) (%)	36.36

under laboratory and reservoir temperature of 80 °C, and this also produced type I microemulsion. This result affirms the reports of [28, 35] which suggest that surfactant flooding alone is incapable of attaining an ultra-low interfacial tension due to its low salinity, noting that formation of microemulsion is largely dependent on brine salinity and IFT [2], thus, the need to readjust the composition of the injection brine [1] or the addition of alkali chemicals [14]. The addition of alkalis and/or cosurfactants to improve the salinity of the system is required, as the synergistic effect of both alkaline and surfactant chemicals will produce an ultra-low interfacial tension [35].

Table 7 Recovery factor after surfactant flooding at laboratory and reservoir temperature

Parameter	Laboratory temp. (22 °C)	Reservoir temp. (80 °C)
Pore volume (ml)	14.4	14.4
OOIP (ml)	11.0	11.0
Oil recovered during brine flooding (ml)	7.00	7.00
Recovery factor after secondary flooding (ml)	63.6	63.6
Residual oil volume (ml)	4.00	4.00
Oil recovered after surfactant flooding (ml)	1.30	1.00
Recovery factor after EOR (%)	11.8	9.10
Displacement efficiency (%)	32.5	25.0

Core flooding analysis

Oil displacement experiment was performed only on the natural surfactant, TA-2 at laboratory and reservoir conditions of 80 °C. The choice of TA-2 was due to its lower interfacial tension value as compared to TA-3. It also had a higher pH value and viscosity. The sandstone core plug which served as the porous medium was fully saturated with FMB prior to the experiment. Table 4 shows the properties of the sandstone core plug.

At the drainage phase, medium crude was used as the displacing fluid to displace brine from the fully saturated sandstone core. The physical properties of the various fluids at laboratory and reservoir temperatures are presented in Table 5.

The values of initial oil saturation, irreducible water saturation, and original oil in place (OOIP) were determined and presented in Table 6. Due to the absence of type III microemulsion, formulated brine (FMB) at 3% salinity was chosen as optimal concentration which was injected into the core plug to displace oil at an injection flow rate of 1.0 ml/min. An oil recovery of 63.3% was attained after secondary flooding with brine both at laboratory and reservoir temperatures indicating that temperature has no effect on the divalent ions. Surfactant flooding using the selected natural surfactant TA-2 at CMC of 1.0% was injected into the core to recover residual oil left behind after brine flooding due to capillary forces. 2PV of TA-2 was injected at a flow rate of 1.0 ml/min for about 2 h and 07 min. At an oil cut of less than 1% with negligible oil production, the experiment was terminated and the incremental oil recovery. At laboratory temperature of 25 °C, an additional recovery factor of 12% OOIP and a displacement efficiency of 32.5% (see Table 7) resulting in a total oil recovery of 75.3%. The effect of temperature on residual oil recovery using the natural surfactant was determined by subjecting the oil displacement experiment to reservoir temperature of 80 °C. This resulted in a recovery factor and displacement efficiency of 9% OOIP and 25%. This implies that increased temperature has a minimal effect on the surfactant and oil recoverability. Karnanda et al. [21] had reported that a direct relationship exists between IFT and temperature, stating that IFT increases with temperature. An increase in IFT invariably reduces the amount of oil recovered. The principal mechanism of surfactant flooding is IFT reduction, however, [35] noted that the addition of an alkaline chemical will result in an ultra-low IFT. This implies that if an alkaline chemical is added to the surfactant slug, the IFT will further reduce, and a higher recovery factor can be achieved even under reservoir temperature. Finally, based on the experimental results obtained, the use of biobased

surfactant TA-2 showed a significant effect in enhancing oil recovery at reservoir temperature and reducing water–oil interfacial tension.

Conclusions

Derivatives of natural cashew nutshell liquid, TA-2 and TA-3, were prepared by esterification with triethanolamine and evaluated as surfactants for chemical EOR. Phase behaviour analysis, interfacial tension, CMC measurement, and core flooding analysis were performed to ascertain the compatibility and recoverability of these biobased surfactants in sandstone reservoirs. Results obtained showed that TA-2 had higher viscosity, higher pH value and lower interfacial tension value compared to TA-3. It was observed that TA-2 can reduce the water–oil IFT to 1.66 mN/m at critical micelle concentration of 1 g/L making it more economically viable compared to TA-3. Oil displacement efficiency using TA-2 at laboratory temperature resulted in additional recovery of 12% OOIP and a displacement efficiency of 32.5%. The effect of reservoir temperature (80 °C) on residual oil recovery was determined, resulting in a recovery factor and displacement efficiency of 9% OOIP and 25%, respectively, indicating that reservoir temperature affected the performance of the natural surfactant to enhance oil recovery. This novel study showed the chemically modified cashew nutshell liquid is an effective biomass-based product for chemical enhanced oil recovery. The derivative (TA-2) is a potential low-cost green alternative to conventional surfactants for chemical EOR. This work demonstrates the feasibility of developing a variety of novel CNSL derivatives and the potential of biobased green formulations from agro-waste materials in oil and gas industry applications. A comprehensive study on the thermal and chemical stability of the natural surfactant over time is suggested for future studies. Furthermore, the addition of a natural alkaline chemical is suggested for future studies to further enhance the phase behaviour, achieve optimum salinity, and attain an ultra-low IFT.

Abbreviations

OOIP	Original oil in place
GC-FID	Gas-chromatograph flame ionization detector
FTIR	Fourier-transform infrared spectroscopy
CNSL	Cashew nutshell liquid
CMC	Critical Micelle concentration
FMB	Formation brine
SFB	Soft formation brine
TEA	Triethanolamine
TAN	Total acid number
TDS	Total dissolved solids
IFT	Interfacial tension
EOR	Enhanced oil recovery

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Authors' contributions

OAA provided the conceptualization, bench work, data curation, draft manuscript preparation, and data analysis. EW provided the data analysis and interpretation. TU provided the data analysis, curation, and interpretation. The authors read and approved the final manuscript.

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Availability of data and materials

All datasets generated and/or analyzed during this study are available from the corresponding author on reasonable request.

Declarations

Competing interests

The authors declare that they have no competing interest.

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