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Experimental Investigation of the Suitability of Surfactants Extracted from Plant Leaves for Enhanced Oil Recovery

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Abstract - **The application of natural surfactants in enhanced oil recovery has recently gained popularity due to their environmentally friendly nature and low cost of production. Several natural surfactants have been extracted from plant leaves and tested for interfacial tension reduction and increased oil recovery. However, it is important to evaluate the oil recovery potential of natural surfactants extracted from plant leaves that are abundant and available in each region for easy production. This study evaluates the recovery potential of surfactants produced from Citrus sinensis leaves, Carica papaya leaves, and Garcinia kola leaves due to their saponin contents for enhanced recovery while reducing the cost of conventional surfactants. The plant leaves (Citrus sinensis, Carica papaya, Garcinia kola) were washed, dried for one week, ground, and measured in quantities of 2 grams and 4 grams. The measured quantities were added to 30 ml of hot water, stirred to mix uniformly, and then brine was added to fill up to 1000 ml in the cylinder. The solution was filtered to obtain the surfactant extract. Core samples were saturated with low salinity brine (10,000 ppm) and flooded with crude oil until irreducible water saturation was achieved. The breakthrough time and volume of water and oil recovered were measured. The brine was replaced with the extracted surfactants at concentrations of 2000 ppm and 4000 ppm, and used for flooding. The volume of oil recovered for each surfactant was measured. The results show that the Citrus sinensis surfactant had an early breakthrough at 28 seconds with 73.68% oil recovery, compared to the 4000 ppm dosage, which had a breakthrough at 34 seconds with 77.5% oil recovery. The Carica papaya surfactant at a 2000 ppm dosage had a breakthrough at 33 seconds with an oil recovery of 85%, and at 39 seconds with an 80.95% recovery for the 4000 ppm dosage. The Garcinia kola surfactant had a late breakthrough at 44 seconds with 76.08% oil recovery for the 2000 ppm dosage and 80.00% recovery at 40 seconds breakthrough for the 4000 ppm dosage. The Carica papaya surfactant provided the highest oil recovery at the lowest dosage of 2000 ppm.**
Keywords: **Natural Surfactants, Oil Recovery, Plant Leaves, Interfacial Tension, Saponin Contents**

I. INTRODUCTION

Flooding with surfactants is one of the most efficient methods for enhancing oil recovery through chemical injection. The need to improve the recovery factor to above 50% to 60% of the original oil in place has arisen, driven by chemical-enhanced methods [1]. This approach displaces crude oil in the void spaces to the producing wells [2], [3]. The primary factors determining the selection and success of flooding with surfactants include the structure, salinity level, surfactant pH value, temperature, rock properties, and adsorption behavior.

Surfactants come in various forms proposed for enhanced oil recovery (EOR), including cationic, anionic, nonionic, and natural surfactants. However, the application has focused on natural surfactants as active agents for enhanced oil recovery due to their effectiveness. Surfactant injection into mature oil reservoirs as a chemical approach for EOR has proven promising and effective [4], [5]. Surfactants have been shown to decrease interfacial tension, modify fluid-to-fluid and rock-to-fluid interactions, and increase production [6], [7]. Many natural surfactants have been extracted from various plant parts such as flowers, roots, leaves, seeds, and oils, and have demonstrated the ability to reduce interfacial tension and improve recovery [8], [9], [10].

Additionally, animal and plant oils such as sesame oil, coconut oil, and linseed oil have been processed into natural surfactants for enhanced oil recovery and have shown good performance [11], [12], [13]. For instance, A.B. Chhetri *et al.,* [14] extracted surfactants from the shell pericarp of Mukurossi sapindus fruit to decrease the interfacial tension (IFT) between oil and water, achieving over a 30% reduction. Similarly, natural surfactants derived from Seidlitzia rosea and Rosemary plants were used to decrease interfacial tension and increase oil recovery, resulting in an improved recovery factor [15]. Surfactants extracted from Ziziphus spina-christi plant leaves were utilized for EOR flooding in a carbonate reservoir, recording a significant increase in oil recovery [16].

Likewise, S. Kumar *et al.,* [17] investigated the suitability of Glycyrrhizin glabra and Mulberry leaf surfactant extracts in carbonate reservoirs, achieving an improved oil recovery of 17.8% and 34% of the original oil in place. Cedar-derived surfactants produced from mulberry tree leaves were used to alter wettability in carbonate and sandstone rocks, reducing the interfacial tension between water and kerosene from 44 to 17.9 mN/m [18]. Surfactant extracts from Matricaria chamomilla plants were tested for interfacial tension reduction with kerosene as the oil phase and distilled water, showing a decrease from 30.6 to 12.67 mN/m [19]. A. Khorram Ghahfarokhi *et al.,* [20] evaluated surfactants extracted from Prosopis leaves to reduce the interfacial tension between distilled water and kerosene, achieving a reduction from about 36.5 to 15 mN/m.

C. Ojukwu *et al.,* [21] used lecithin, a phospholipid, with soybean surfactant extracts to form surfactants for Surfactant-Alkaline-Polymer (SAP) flooding, demonstrating improved oil recovery. The surfactant in the test reduced the interfacial tension to drive more oil out of the sand pores, recovering 23.3% of the original oil in place. Omo and Keysoap detergent surfactants yielded the highest recovery of 80%, compared to industrial surfactants (sodium hydroxide and potassium hydroxide), which faced emulsion problems for improved recovery [22]. Lignin surfactants extracted from black liquor, obtained through oil palm empty fruit bunch pulping with sodium hydroxide, showed good recovery potential in surfactant flooding when tested with standard lignin [23].

Recently, the application of plant surfactants in chemical flooding has increased due to their environmental friendliness and lower cost compared to synthetic surfactants. Plants with saponin content are found to be effective surfactants with potential for enhanced recovery. Saponins are surface-active agents with foaming and emulsification properties, including detergent and wetting capabilities. Saponin extracted from Ziziphus spina-christi tree leaves showed significant potential for enhancing recovery, reducing interfacial tension from 48 to 9 mN/m [24]. In this study, plant leaves (Carica papaya, Garcinia kola, Citrus sinensis) were chosen for surfactant extraction due to their saponin content of 2.00%, 1.29%, and 0.98% [25], [26]. These plant leaves are common, readily available, and can be extracted on a commercial scale if found effective as surfactants for enhanced oil recovery.

II. MATERIALS AND METHODS

A. Materials and Equipment

The materials and equipment used are unconsolidated sand samples, crude oil, natural surfactants derived from Citrus sinensis leaf, Garcinia kola leaf, and Carica papaya leaf, distilled water, sodium chloride, clay, EOR apparatus, stopwatch, weighing balance, pycnometer, separating funnel, retort stand, clamp, syringe, spatula, aluminum foil, vernier caliper, mesh screen, hydrometer, oven, plunger, thermometer, electric mixer, beaker, conical flasks, measuring cylinder, and tubes.

B. Method

Niger Delta field crude oil was used for this experiment. Encapsulated (mounted) cores from unconsolidated sand were prepared, saturated with brine, and their porosities were determined using the saturation method. The sand pack oil displacement flood test was conducted to evaluate the effectiveness of the plant leaf extracts for enhanced oil recovery using surfactant flooding.

1. Brine Preparation

A brine concentration of 10,000 ppm was prepared using 10 grams of industrial salt measured with an electronic weighing balance. The steps are as follows:

a. Ten grams of industrial salt were measured with the weighing balance.
b. The measured salt was mixed with 30 mL of distilled water and stirred until it was completely dissolved.
c. The salt solution was poured into a 1000 mL graduated cylinder.
d. Distilled water was then added to bring the volume up to the 1000 mL mark on the cylinder.

2. Surfactant Preparation

The concentrations of surfactants used were 2000 ppm and 4000 ppm. The procedure for preparing the surfactants with different plant leaves is as follows:

a. Fresh Carica papaya leaves were washed and dried at room temperature for one week.
b. The dried leaves of Carica papaya were crushed into small pieces.
c. Two grams and four grams of Carica papaya leaves were measured and weighed.
d. The leaves were added to a beaker with 30 mL of hot water and stirred. The filtered solution was then transferred into a clean container and labeled accordingly.

The same steps were repeated for Citrus sinensis and Garcinia kola leaves. The prepared surfactants are presented in Fig. 1.

Fig. 1 Prepared Surfactant from Carica papaya (pawpaw leave), Citrus sinensis (orange leave) and Garcinia kola (bitter kola leave)

3. Flooding Procedure

a. The core sample was saturated with low salinity brine at 10,000 ppm.

b. The saturated core was placed in the core holder and clamped with a retort stand.
c. The flow line was attached to the top of the core holder in a horizontal position, with a beaker placed under the retort stand.
d. The flow line was connected from the bottle containing crude oil to the other end of the stem.
e. Crude oil was used to flood the core until irreducible water saturation was attained, and the breakthrough time and volume of water recovered were recorded.
f. The flow line for crude oil was removed and replaced with that for brine, and the beaker was replaced.
g. The core was flooded with brine until little or no oil was recovered, and the volume of oil recovered was recorded.
h. The flow line for brine was removed and replaced with that for the surfactant.
i. The core was soaked and flooded with the surfactant, and the volume of oil recovered was recorded.

4. Surfactant Sample Identification

Sample B1 represents the surfactant derived from Citrus sinensis leaf.
Sample B2 represents the surfactant derived from Carica papaya leaf.
Sample B3 represents the surfactant derived from Garcinia kola leaf.

III. RESULTS AND DISCUSSION

A. Oil Volume Recovered for 2000ppm Surfactants Concentration

The oil recovered from brine flooding (secondary), surfactant flooding (Citrus sinensis leaf, Carica papaya leaf, Garcinia kola leaf), and the residual oil saturation is presented in Fig. 2.

When a 2000 ppm concentration of the surfactants was injected, the *Citrus sinensis* surfactant exhibited an earlier breakthrough of 28 seconds with 73.68% oil recovery, the *Carica papaya* surfactant had a breakthrough at 33 seconds with 85% oil recovery, and the *Garcinia kola* surfactant showed a breakthrough at 44 seconds with 76.08% oil recovery. The *Carica papaya* surfactant provided the highest oil recovery at the 2000 ppm concentration.

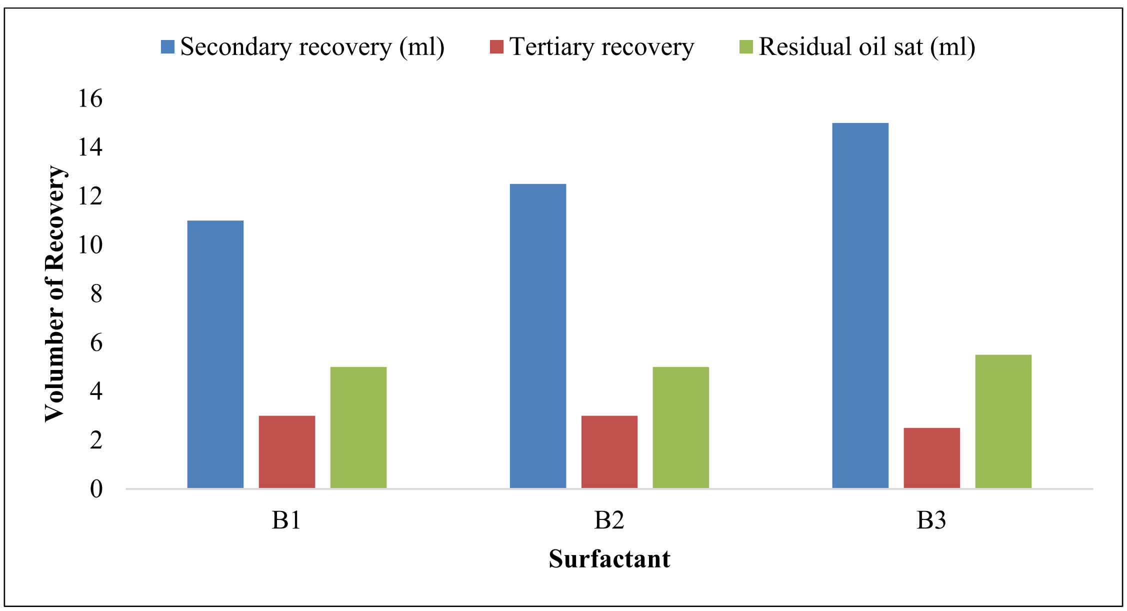

Fig. 2 Surfactants with 2000ppm concentration for B1 to B3

B. Oil Volume Recovered for 4000ppm Surfactant Concentration

The oil recovered from brine flooding (secondary), surfactant flooding (Citrus sinensis, *Carica papaya* leaves, *Garcinia kola* leaves) at a 4000 ppm concentration, and the residual oil saturation are presented in Fig. 3. When a 4000 ppm concentration of the surfactants was injected, the *Citrus sinensis* surfactant exhibited an earlier breakthrough of 34 seconds with 77.50% oil recovery, the *Carica papaya* surfactant achieved a breakthrough at 34 seconds with 80.95% oil recovery, and the *Garcinia kola* surfactant had a breakthrough at 40 seconds with 80% oil recovery. The *Carica papaya* surfactant provided the highest oil recovery at the 4000 ppm concentration. The results show that increasing the surfactant concentration increases the breakthrough time and oil recovery for *Citrus sinensis* and *Garcinia kola* surfactants but decreases the oil recovery for *Carica papaya* surfactant.

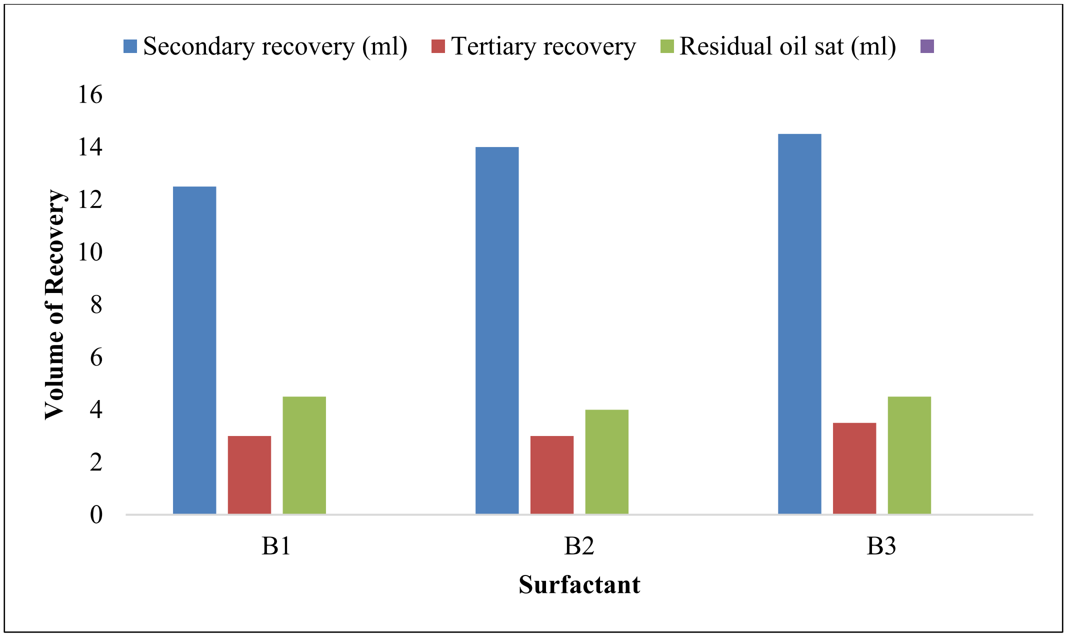

Fig. 3 Surfactants with 4000ppm concentration for B1 to B3

C. Cumulative Recovery for 2000ppm Surfactant Concentration

Fig. 4 shows the cumulative oil recovered for the surfactants at 2000 ppm. The *Carica papaya* surfactant achieved the highest cumulative oil recovery, followed by the *Garcinia kola* surfactant, with the *Citrus sinensis* surfactant showing the least cumulative recovery, as presented in Fig. 4.

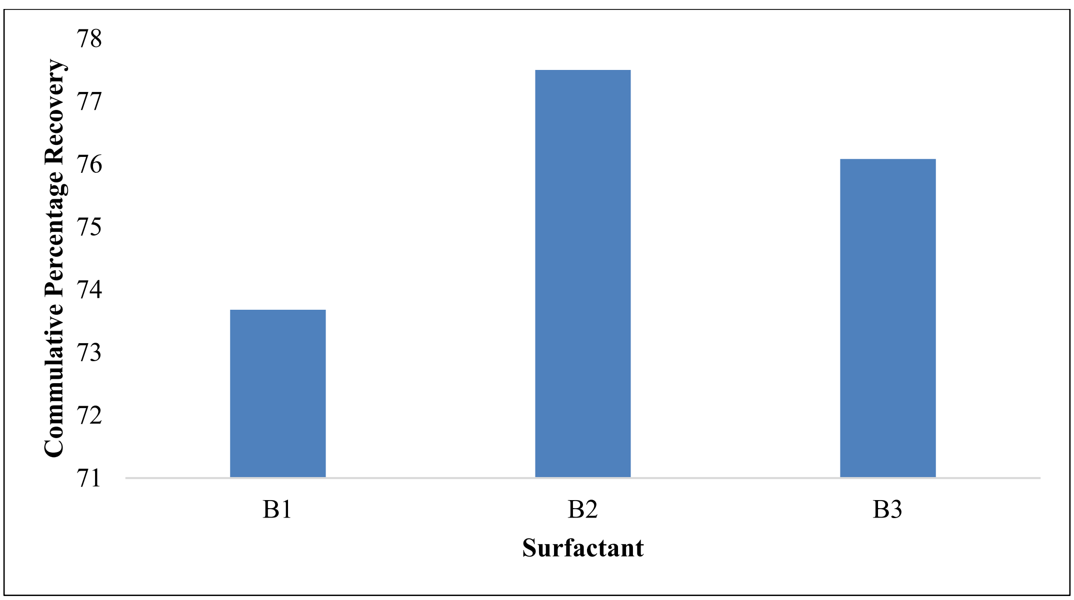

Fig. 4 Cumulative percentage recovery of Surfactant with 2000ppm concentration for B1 to B3

D. Cumulative Recovery Form Surfactants for 4000 ppm Concentration

The cumulative oil recovered for the surfactants at 4000 ppm is presented in Fig. 5. Similarly, the *Carica papaya* surfactant achieved the highest cumulative oil recovery, followed by the *Garcinia kola* surfactant, with the *Citrus sinensis* surfactant showing the least cumulative recovery, as shown in Fig. 5.

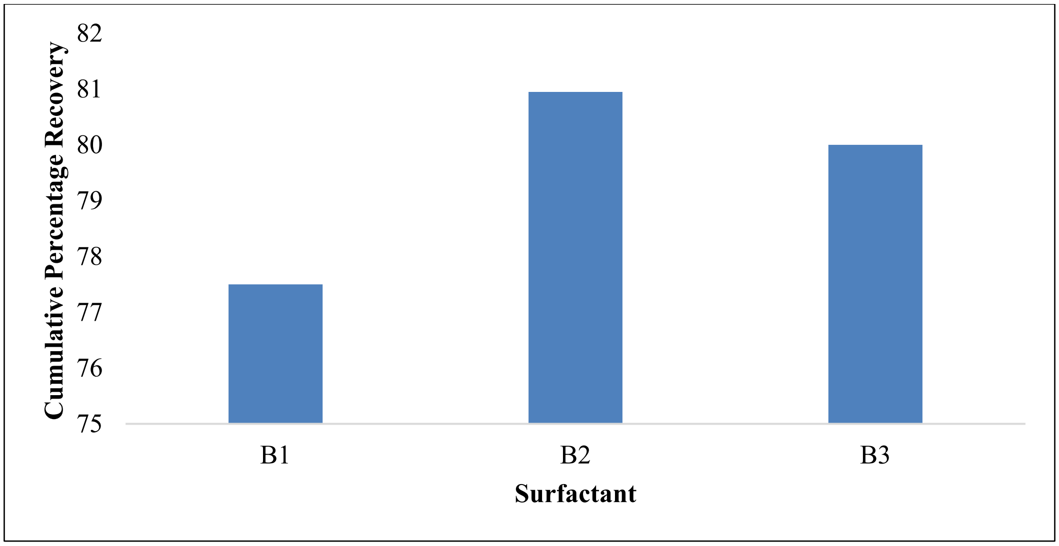

Fig. 5 Cumulative percentage recovery of Surfactant with 4000ppm concentration for B1 to B3

IV. CONCLUSION

This study examined the effectiveness of surfactants derived from the leaves of *Carica papaya*, *Citrus sinensis*, and *Garcinia kola* for enhanced oil recovery. The findings revealed that the selected plant leaves are rich in saponins, making them suitable as surfactants. Among the three, the surfactant from *Carica papaya* leaves demonstrated the highest oil recovery across all concentrations, while the surfactant from *Citrus sinensis* leaves resulted in the lowest recovery. Additionally, the study showed that higher concentrations of surfactants led to increased oil recovery.

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