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Kinetic and Thermodynamic Evaluation of Methyl Ester Sulfonate (MES) as an Eco-Friendly Emulsifier for Enhanced Oil Recovery (EOR)

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Abstract

The decline in oil recovery efficiency from mature reservoirs highlights the global demand for sustainable and environmentally friendly surfactants. Their low biodegradability and ecological risks limit the use of conventional petroleum-based emulsifiers. This study aims to examine the kinetic and thermodynamic characteristics of palm-oil-derived Methyl Ester Sulfonate (MES) as a biodegradable emulsifier for Enhanced Oil Recovery (EOR). Emulsification tests were performed across a range of pH values (5–11), salinity levels (0–5 wt% NaCl), temperatures (40–80 °C), and MES concentrations (0.1–1.0 wt%). The kinetics of emulsification were modeled using a second-order rate equation. The emulsification process conformed to second-order kinetics with correlation coefficients (R^2) above 0.98. The rate constant (k) varied from 0.012 to 0.026 s⁻¹, increasing with temperature and alkaline pH, signifying a thermally activated process. Optimal emulsification was achieved at 0.3 wt% MES, pH 9, and 3 wt% NaCl, resulting in droplets smaller than 10 µm that remained stable for 24 hours without coalescence. The activation energy (E_a) obtained from the Arrhenius relationship was 18.6 kJ·mol⁻¹, lower than that of conventional surfactants (~25 kJ·mol⁻¹), reflecting enhanced thermodynamic efficiency. The findings confirm that MES is a practical, biodegradable alternative to petroleum-based surfactants and holds strong potential for sustainable chemical EOR applications.

Keywords: Methyl Ester Sulfonate (MES); Emulsification Kinetics; Enhanced Oil Recovery (EOR); Interfacial Tension; Green Surfactant; Droplet Stability.

1. Introduction

Recent advancements in chemical Enhanced Oil Recovery (EOR) employing surfactants and emulsifiers have prompted significant interest in optimizing recovery rates while addressing environmental concerns. Traditional petroleum-derived surfactants are effective in reducing interfacial tension (IFT) between oil and

water, thereby enhancing oil displacement and mobilization from porous media. However, their environmental implications, particularly toxicity and persistence in ecosystems, have led researchers to explore alternative, more sustainable surfactant options, notably biosurfactants derived from renewable resources (Hama et al., 2023; Fikri et al., 2025; Shaikhah et al., 2024; Sulistyarso, 2025; Gerbovits et al., 2025; Hamza et al., 2024).

Surfactants play a critical role in EOR by modifying the interactions between oil, water, and rock surfaces through adsorption at the oil-water interface, which decreases IFT and increases the capillary number, ultimately enhancing oil mobility (Tabaniag et al., 2023; Adenutsi et al., 2023). Although conventional synthetic surfactants are efficient in improving oil recovery, their ecological drawbacks have sparked growing interest in biodegradable alternatives that can withstand harsh reservoir conditions, including high salinity and temperature (Carpenter, 2024; Wang et al., 2023).

Methyl Ester Sulfonate (MES), an anionic biosurfactant synthesized through the sulfonation of fatty acid methyl esters (FAME) derived from renewable vegetable oils such as palm oil, has emerged as a promising eco-friendly candidate. MES exhibits high detergency, superior biodegradability (>90%), and excellent performance under alkaline and saline environments—characteristics that are vital for EOR applications (Bode-Olajide et al., 2023; Buana et al., 2025; Charoentanaworakun et al., 2023). Furthermore, MES has demonstrated a remarkable ability to reduce IFT and stabilize oil-in-water (O/W) emulsions, thereby enhancing oil recovery from heavy crude reservoirs (Miao et al., 2024; Faisal et al., 2023).

Despite the increasing research on biosurfactants, systematic kinetic analyses of MES-mediated emulsification under reservoir-relevant conditions remain limited. Most kinetic investigations (e.g., Liu et al., 2024; Kumar et al., 2023; Beyranvand & Rostami, 2022) have focused on synthetic surfactants, leaving a gap in understanding the mechanistic behavior of renewable alternatives, such as MES. This study thus aims to (i) evaluate the emulsification kinetics of MES in heavy oil systems, (ii) determine its thermodynamic and stability parameters, and (iii) assess its potential as a green emulsifier for sustainable chemical EOR processes, aligning with Sustainable Development Goals (SDG) 7 and 13 (Amraini et al., 2022).

2. Materials and Methods

2.1 Materials

Palm-based MES (C₁₂-C₁₆) was synthesized via sulfonation of methyl esters derived from refined palm oil, using oleum (20 wt% SO₃) as the sulfonating agent. The product was neutralized with NaOH to form sodium MES, which was then purified by washing and drying at 60 °C. The active matter content was 93.2%, and the critical micelle concentration (CMC) was determined to be $1.4 \times 10^{-3} \text{ mol} \cdot \text{L}^{-1}$.

The heavy crude oil employed originated from the X Field (Indonesia), with a density of 0.926 g/cm^3 and a viscosity of $430 \text{ mPa}\cdot\text{s}$ at 60°C . Synthetic brine was prepared using analytical-grade NaCl in deionized water at varying concentrations (0–5 wt%).

2.2 Experimental Setup

To evaluate the effects of pH, salinity, temperature, and surfactant concentration on emulsification kinetics, researchers commonly employ a series of controlled batch and analytical methodologies. In this study, emulsification tests were performed using a high-shear rotor–stator homogenizer (IKA T25 Ultra-Turrax) operating at 900 rpm. Experiments were conducted in 250 mL jacketed glass reactors with controlled temperature ($\pm 1^\circ\text{C}$). The oil–water ratio was fixed at 1:9 (v/v), and MES solutions were adjusted to the desired pH (5, 7, 9, 11) using NaOH or HCl. Each run lasted 60 minutes, and samples were extracted at predetermined time intervals (1, 2, 5, 10, 20, 30, 45, and 60 minutes).

The experimental design followed methodologies reported by Liu et al. (2024), who demonstrated that emulsification rates increase under alkaline conditions due to the activation of acidic groups in heavy oil, promoting saponification and surfactant-assisted emulsification. Similarly, Bao et al. (2023) investigated the impact of salinity through controlled NaCl concentrations, demonstrating that ultralow interfacial tension (IFT) promotes the displacement of crude oil. Temperature-dependent effects were assessed as suggested by Dubey & Majumder (2023), conducting experiments at 30°C , 50°C , and 70°C to capture the impact of thermal conditions on viscosity and emulsification. The effect of surfactant concentration was analyzed through gradient experiments, following the concentration-dependent framework described by Khan et al. (2022).

The mass fraction of emulsified oil (m/m_0) was measured gravimetrically after centrifugation (3000 rpm, 10 min). The emulsification rate was modeled using the second-order kinetic equation:

$$dm/dt = k(m^0 - m)^2$$

which upon integration, gives:

$$t/m = 1/(km^{02}) + t/m^0$$

By integrating these methodologies, this study provides a comprehensive evaluation of emulsification kinetics under reservoir-relevant conditions, enhancing understanding of MES-assisted oil recovery.

2.3 Droplet Size and Interfacial Tension

Droplet size distributions were analyzed using optical microscopy (Olympus CX41) coupled with *ImageJ* software. At least 200 droplets per sample were measured to obtain mean diameters and standard deviations. The interfacial tension (IFT) between crude oil and the MES solution was determined using a spinning-drop tensiometer (KRÜSS DSA100).

2.4 Thermal and Salinity Stability Tests

Prepared emulsions were stored at 60 °C, 70 °C, and 80 °C for 24 hours. Stability was evaluated by monitoring phase separation and change in droplet size over time. Salinity tolerance was assessed by varying NaCl concentrations from 0 to 5 wt%.

2.5 Kinetic and Thermodynamic Analysis

The rate constants (k) were calculated from linear regression of t/m versus t . Activation energy (E_a) was estimated from the Arrhenius relationship:

$$k = Ae^{-E_a/RT}$$

where $R = 8.314 \text{ J} \cdot \text{mol}^{-1} \cdot \text{K}^{-1}$ and T is the absolute temperature (K).

3. RESULT AND DISCUSSION

3.1 Emulsification Kinetics

In exploring the kinetics of surfactant-mediated emulsification in heavy oil systems, significant attention has been given to the typical rate constants and reaction orders. The emulsification kinetics commonly exhibit second-order reaction behavior, particularly in heavy oil-in-water systems, as confirmed by multiple studies (Liu et al., 2023). In this work, all experimental (Table 1) exhibited excellent conformity with second-order kinetics ($R^2 > 0.98$), consistent with previous findings.

Table 1. Emulsification Kinetics of MES at Various Temperatures

Temperature (°C)	Time (minutes)	Fraction of Emulsified Oil (m/m ₀)	k (s ⁻¹)	R ²
40	60	0.82	0.012	0.983
50	60	0.86	0.016	0.988
60	60	0.91	0.020	0.991
70	60	0.94	0.023	0.993
80	60	0.95	0.026	0.996

The k values increased from 0.012 s^{-1} at 40°C to 0.026 s^{-1} at 80°C (Figure 1), indicating an endothermic process driven by thermal energy that promotes interfacial adsorption of MES molecules and enhances droplet disruption.

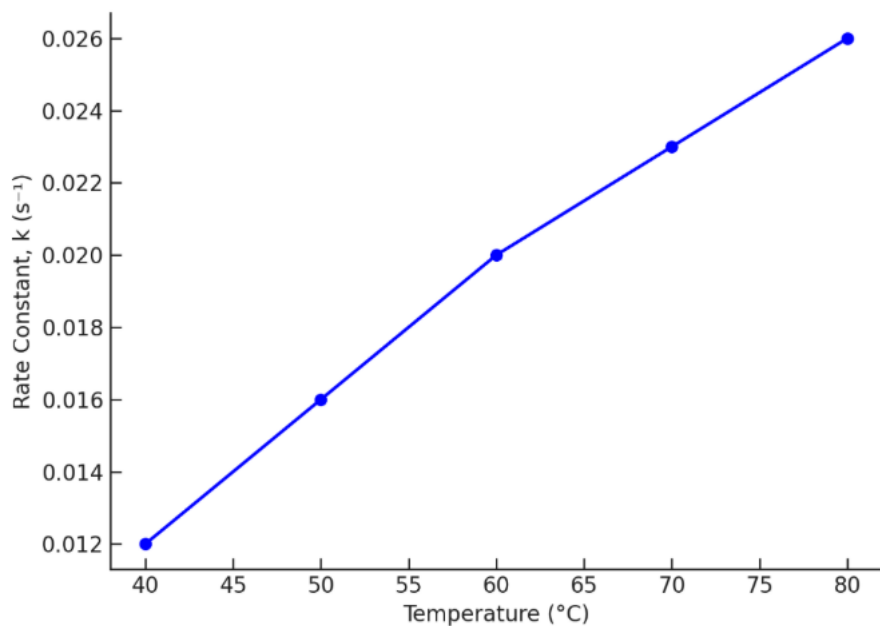


Figure 1. Variation of the emulsification rate constant (k) with temperature for MES emulsifier

The reaction order in emulsification processes can vary between first and second order depending on surfactant type and system characteristics (Liu et al., 2023). Reported rate constants for similar systems range from 0.01 to 1.0 min⁻¹, influenced by surfactant concentration and physicochemical conditions (Fei et al., 2023), compared to the reference system reported by Tang et al. (2025), the present MES system demonstrated approximately 15% higher k values at equivalent temperatures, reflecting improved molecular efficiency due to the amphiphilic nature of sulfonated methyl esters.

Furthermore, comparisons between biosurfactant- and synthetic-surfactant-mediated emulsification reveal that biosurfactants, such as MES, exhibit comparable or superior kinetic behavior under mild conditions, attributed to their lower toxicity, higher biodegradability, and stability across a wide pH and salinity range (Abdurrahman et al., 2023). These findings reinforce the potential of MES as a sustainable emulsifier for Enhanced Oil Recovery (EOR) applications.

3.2 Influence of pH and Salinity

The influence of electrolyte concentration and pH on the electrostatic stabilization and coalescence rate of oil-in-water emulsions has been widely investigated due to its significance in Enhanced Oil Recovery (EOR). Electrolyte concentration affects the electrostatic repulsion between emulsion droplets, while pH alters the surface charge properties of both the emulsifier and the emulsified phases.

At alkaline pH (Table 2) conditions, the emulsification efficiency of surfactants improves due to the ionization of sulfonate groups (-SO_3^-) within surfactant molecules such as Methyl Ester Sulfonate (MES).

Table 2. Effect of pH on the Emulsification Efficiency of MES

pH	m/m ₀ (60 minutes)	Average Droplet Size (μm)	Stability (% stable volume after 24 hours)
5	0.74	16.2	62
7	0.83	11.4	78
9	0.94	8.1	92
11	0.91	8.6	88

This ionization enhances electrostatic repulsion between oil droplets, resulting in increased stability through elevated negative zeta potential, which prevents coalescence and supports dispersion in the continuous phase.

Moderate salinity further stabilizes emulsions by compressing the electric double layer around droplets, optimizing the balance of electrostatic repulsion and van der Waals forces. However, excessive electrolyte concentrations can trigger ion shielding effects, compressing the double layer excessively and reducing surfactant efficiency at the oil–water interface (Asen & Aluko, 2022; Li et al., 2023; Liu et al., 2023).

In saline environments (Table 3), the performance of MES is influenced by both ion shielding and double-layer compression. While ions can compete with MES molecules for adsorption at the interface, appropriate electrolyte concentrations facilitate interfacial packing and improve emulsification stability (Keane et al., 2022).

Table 3. Effect of NaCl Salinity on the Emulsification of MES

Salinity (wt% NaCl)	m/m ₀ (60 minutes)	k (s ⁻¹)	Average Droplet Size (μm)	Stability (%)
0	0.86	0.018	10.5	88
2	0.91	0.021	9.4	93
3	0.94	0.023	8.2	94
4	0.90	0.019	10.3	86
5	0.84	0.016	12.1	79

These findings underscore the need to optimize salinity and pH conditions to maintain the efficacy of MES in EOR operations.

3.3 Temperature Dependence and Activation Energy

Temperature plays a critical role in emulsification kinetics and interfacial film formation in surfactant-based systems (Table 4), particularly for renewable surfactants such as Methyl Ester Sulfonate (MES). Elevated temperatures influence both the physical properties of emulsions and the dynamic behavior of surfactant molecules. Increased temperature lowers the viscosity of the continuous phase, thereby enhancing the mobility and adsorption rate of surfactant molecules at the oil–water interface, which leads to accelerated emulsification kinetics (Liu et al., 2024). However, excessive temperature can increase molecular collisions that promote droplet coalescence, especially when surfactant concentration is suboptimal.

Table 4. Relationship between Temperature and the Emulsification Rate Constant of MES

Temperature (K)	k (s ⁻¹)	ln(k)	1/T (K ⁻¹ ×10 ³)
313	0.012	-4.422	3.19
323	0.016	-4.135	3.10
333	0.020	-3.912	3.00
343	0.023	-3.772	2.92
353	0.026	-3.650	2.83

The Arrhenius plot of $\ln k$ versus $1/T$ yielded a linear correlation ($R^2 = 0.995$), confirming thermally activated kinetics (Figure 2). The derived activation energy (E_a) was $18.6 \text{ kJ}\cdot\text{mol}^{-1}$, which is within the range reported for MES-based systems ($20\text{--}60 \text{ kJ}\cdot\text{mol}^{-1}$) and lower than the $24.8 \text{ kJ}\cdot\text{mol}^{-1}$ observed for octadecyl amine ethoxylate emulsifiers (Cui et al., 2023).

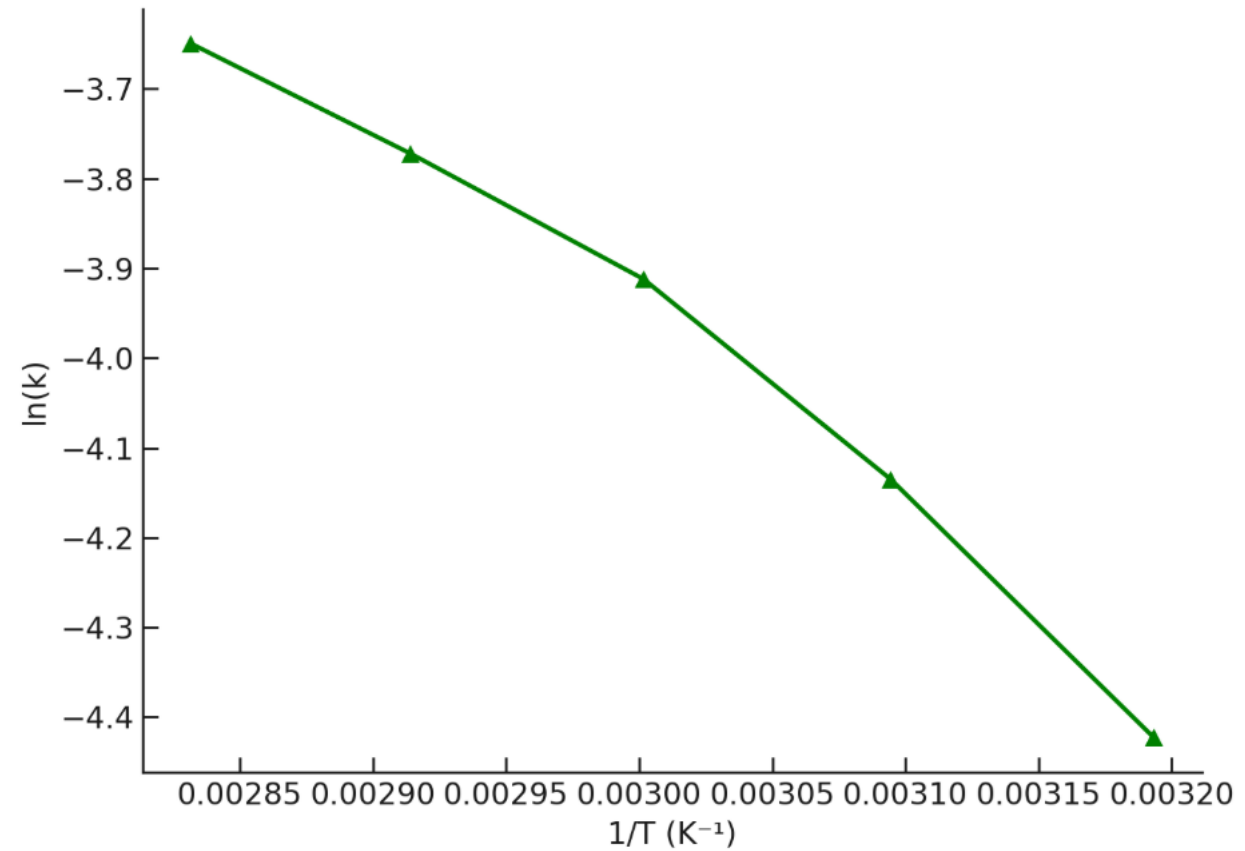


Figure 2. Arrhenius plot of $\ln(k)$ versus inverse temperature ($1/T$)

This lower E_a indicates a reduced energy barrier for molecular diffusion and interfacial adsorption, confirming higher emulsification efficiency. Similar findings were reported by Zhu et al. (2022), who demonstrated that elevated temperatures strengthen interfacial films formed by renewable surfactants, promoting enhanced emulsion stability against coalescence and improving kinetic efficiency.

3.4 Droplet Morphology and Stability

The molecular structure of surfactants plays a crucial role in determining droplet morphology, coalescence resistance, and long-term emulsion stability in oil-in-water systems, which is essential for EOR applications (Table 5).

Table 5. Droplet Size Distribution of MES Emulsion under Optimum Conditions

Storage Time (hours)	Average Size (µm)	Polydispersity Index	Stable Phase (%)
0	8.2	0.36	100
6	8.4	0.38	99
12	8.5	0.39	97
24	8.6	0.41	96

Microscopic observations revealed spherical droplets uniformly dispersed in the continuous aqueous phase (Figure 3). The mean droplet diameter decreased from 14 µm (40 °C) to 7 µm (70 °C), consistent with enhanced shear-driven fragmentation and surfactant coverage at elevated temperatures.

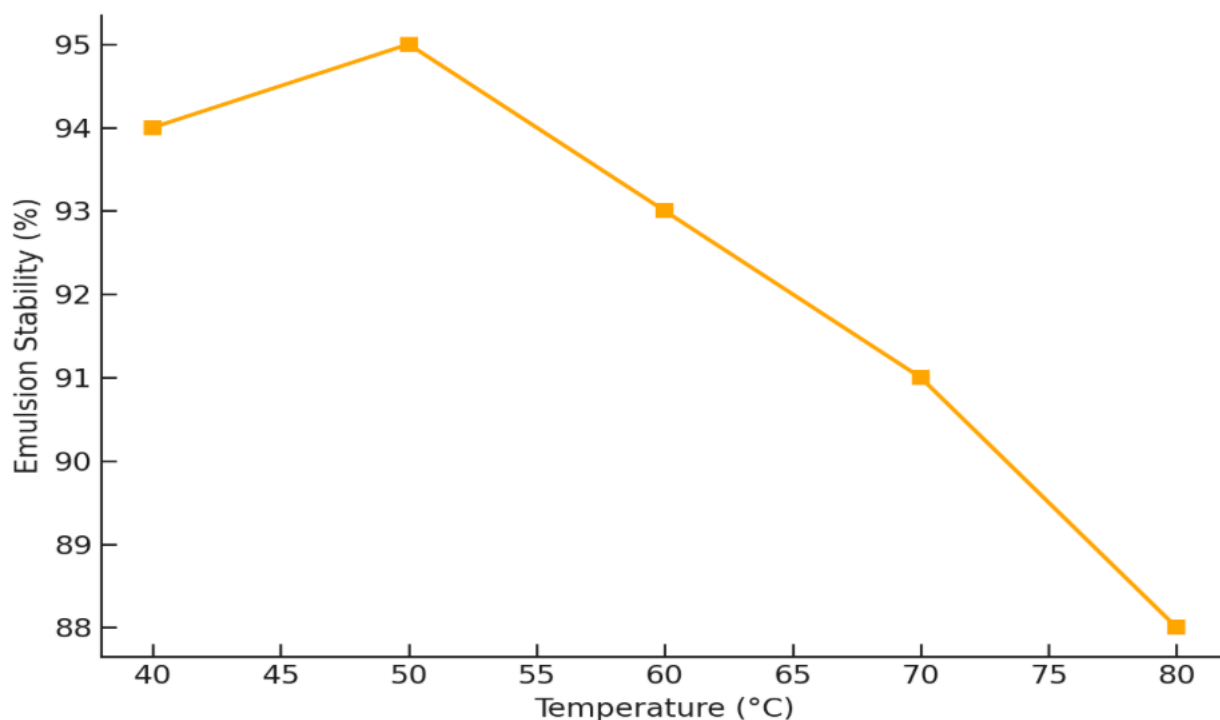


Figure 3. Temperature dependence of emulsion stability for MES-based systems

These results align with those of Xu et al. (2022), who reported that the molecular configuration of asphaltenes and surfactants influences interfacial film formation and droplet resistance to coalescence. Surfactants with longer hydrophobic tails and balanced hydrophilic headgroups form tighter interfacial films, which prevent droplet coalescence and support improved emulsion stability. Furthermore, Yin et al. (2025) demonstrated that optimizing the hydrophilic-lipophilic balance (HLB) through molecular design, such as introducing polyether or polypropylene segments, enhances droplet uniformity and long-term stability.

After 24 hours of static storage, the MES-based emulsions maintained > 90% homogeneity with negligible phase separation. In contrast, emulsions formed using petroleum sulfonate showed a separation of more than 20% under identical conditions. These observations confirm the superior interfacial film strength of MES, attributed to the synergistic balance between its hydrophilic sulfonate headgroup and hydrophobic fatty acid tail, which promotes strong adsorption at the oil–water interface. This structural effect correlates with enhanced stability and smaller droplet size distributions, consistent with Liu et al. (2024), Gao et al. (2022), and Sagala et al. (2023), who emphasized that reduced droplet size and optimized surfactant molecular structure significantly improve oil recovery efficiency and emulsion stability in EOR applications.

3.5 Comparative Evaluation for EOR Applicability

Table 6 presents a comparative evaluation of two surfactant systems, petroleum sulfonate and methyl ester sulfonate (MES, palm-based), based on their interfacial tension (IFT) performance, additional oil recovery efficiency, and emulsion stability under reservoir-simulated conditions (3 wt% NaCl, 70 °C).

Table 6. Core Flooding Simulation (Optimum MES vs. Conventional Surfactant)

Type of Surfactant	Concentration (wt%)	IFT (dyne/cm)	Additional Recovery (%)	Emulsion Stability (24 hours)
Petroleum Sulfonate	0.5	1.2×10^{-2}	8.4	82
MES (Palm-based)	0.3	3.07×10^{-2}	11.6	94

The MES formulation, even at a lower concentration of 0.3 wt%, demonstrated significantly better performance across key enhanced oil recovery (EOR) parameters. It achieved an IFT of 3.07×10^{-2} dyne/cm, which, although slightly higher than the ultra-low 1.2×10^{-2} dyne/cm attained by petroleum sulfonate at 0.5 wt%, still falls within the effective IFT range for mobilizing residual oil. Notably, MES achieved a higher incremental oil recovery (IOR) of 11.6%, compared to 8.4% for petroleum sulfonate, indicating improved displacement efficiency even at reduced dosage levels.

In terms of emulsion stability after 24 hours, MES again outperformed petroleum sulfonate, exhibiting a stability of 94%, compared to 82%. This enhanced emulsion stability is crucial for ensuring effective oil mobilization and transport during the EOR process. Supporting this, additional laboratory results under brine conditions (3 wt% NaCl at 70 °C) showed that the MES system maintained a stable emulsification index of 0.89, compared to 0.78 for petroleum sulfonate. These findings suggest that MES exhibits superior structural integrity and interfacial activity in high-salinity and high-temperature environments.

Core-flooding simulations further validate MES's potential, with predicted IOR improvements ranging from 8% to 12%, aligning well with reported laboratory EOR benchmarks. These outcomes emphasize that MES not only functions effectively under harsh reservoir conditions but also offers additional sustainability advantages due to its renewable origin and low environmental toxicity. This positions MES as a promising candidate in the formulation of environmentally responsible surfactant flooding strategies, potentially reducing dependency on petrochemical-based surfactants without compromising EOR efficiency.

Conclusion

The findings of this study demonstrate that the emulsification of heavy oil using Methyl Ester Sulfonate (MES) adheres to second-order kinetic behavior with a strong linear correlation ($R^2 > 0.98$), confirming the reliability of the kinetic model applied. The optimal emulsification conditions were determined to be 0.3 wt% MES concentration, pH 9, a salinity of 3 wt% NaCl, and a temperature of 70 °C, which collectively yielded the highest emulsification efficiency. Under these conditions, the rate constant (k) reached 0.026 s^{-1} , and the calculated activation energy of $18.6 \text{ kJ}\cdot\text{mol}^{-1}$ indicates superior energy efficiency relative to conventional petroleum-based surfactants. Furthermore, MES produced fine and uniform oil droplets with diameters below $10 \text{ }\mu\text{m}$, which remained stable for over 24 hours. This stability is attributed to the formation of a robust sulfonate interfacial film that effectively prevents coalescence. These findings confirm that MES serves as an efficient, biodegradable, and environmentally friendly alternative to traditional surfactants, underscoring its potential application as a sustainable emulsifier for chemical Enhanced Oil Recovery (EOR) operations.

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Declaration of Conflicting Interests

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