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Review Article

A Review on Biosurfactant Applications in the Petroleum Industry

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The inadequacy of worldwide fossil fuel resources, combined with increasing energy demands, encourages global attention to either using alternative energy resources or improving the recovery factor and produce larger quantities from present reservoirs. Among all enhanced oil recovery (EOR) methods, surfactant injection is a well-known technique that reduces the interfacial tension (IFT) between oil and water and increases oil production. Despite numerous advantages of using surfactants, there are also a few obstacles like environmental impacts, high cost, effect on humans and other organisms due to toxicological potential, and availability from nonrenewable resources. Biosurfactants are microbial surface-active agents that decrease the surface tension (ST) of a liquid phase and the IFT of two diverse phases. They are biotechnological products of high value owing to their widespread applications, low toxicity, relatively easy preparation, and specific performance, applied in different industries like organic chemicals and fertilizers, agrochemicals, metallurgy and mining, cosmetics, foods, medical and pharmaceuticals, beverages, environmental management, and petroleum and petrochemical applications in emulsifying and demulsifying wetting agents, detergent spreading and foaming agents, and functional food ingredients. Biosurfactants are synthesized by microbes; therefore, various genetic diversities of microorganisms provide the considerable capability to produce new types of biosurfactants, which can develop EOR technology. Biosurfactants are classified into ex situ and in situ MEOR processes. The genera *Pseudomonas*, *Bacillus*, *Sphingomonas*, and *Actinobacteria* are the foremost biosurfactant-producing bacteria. This paper reviews relevant reports and results from various presented papers by researchers and companies on applications of microorganisms and biosurfactant technology with specific emphasis on EOR and MEOR processes, based on recently published articles since 2010 until now.

1. Introduction

These days, human life is highly dependent on fossil fuel and its related products such as kerosene, gas, petrol, and diesel [1, 2]. Among the various fossil fuel types, crude oil plays an essential role in the industrial revolution since the beginning of civilizations to provide global energy supplies [1, 3]. Statistics of global oil resources consumption exhibit an increasing trend from the last century until 2019. As a primary energy source, oil consumption was about 36390.5 Mbbl in 2019, and much increase has been assumed for the future, which would increase oil prices [4]. Figure 1 shows the global crude oil consumption trend from 1980 until now and predicts growing consumption demand in the future. The graph experienced a reduction in 2020 due to the worldwide coronavirus pandemic and widespread shutdowns; however, an increase in worldwide energy demand is expected in the approaching decades [5]. Rising universal energy demands, in addition to limited fossil fuel resources across the world, require a robust response to energy supply. Development and improvement of the oil recovery technique yield from the existing reservoirs; furthermore, identifying alternative resources is the best way to reduce dependence on fossil fuels in the future [6–9].

Various alternatives have been developed and suggested, including wind energy, solar energy, nuclear energy, and different biomass-converted products such as firewood, biogas, fuel pellets, biodiesel, bioethanol, bio-oil, and

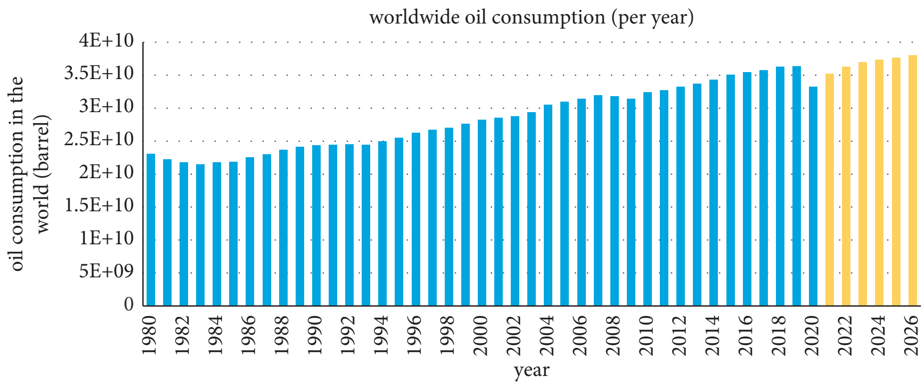

Figure 1: Worldwide consumption of oil resources (barrel per year) [5].

biohydrogen [10–14]. Despite the availability and advantages of renewable resources, their technology and investigations are in the initial stages, also requiring fossil fuel-based infrastructure changes [15, 16]. Some cases are not cost-effective in comparison with fossil fuels; therefore, fossil fuels remain the essential energy resource, applied for household and industrial purposes, power generation, and transportation [17–20].

There are a number of considered techniques for enhancing oil recovery from depleted reserves [21]. The procedure of recovering oil from reservoirs includes primary, secondary, and tertiary recovery phases [22–24]. In the first recovery phase, oil production is driven by the reservoir's initial pressure energy, which can produce just 10–20% of the present oil in the reservoir [25, 26]. In the second recovery phase, either gas or water will be injected to the reservoir to maintain both reservoir and oil pressures during production, and the final oil recovery phase increases 20–40% [21, 26–28]. To determine the amount of relative oil and water saturation, choosing appropriate injection fluid, chemical factors, and reservoir conditions such as pressure, temperature, brine characterization, pollution levels, rock type and sizes, and distribution of pores structure information is essential [29–31]. If the injected fluid's viscosity is lower than the displacing fluid, the injected compound may flow faster than the initial compound's flow across the porous media [32, 33]. Depending on oil properties, geographic difficulties, and reservoir conditions, initial and secondary oil production methods are capable of producing 40–60% of the original oil in place (OOIP), while residual oil saturation (S_{or}) is a target for EOR [21, 26, 28, 34]. The most economically viable time to apply EOR methods in a field seems to be at the early stages after primary recovery; however, the best time usually depends on the economic factors of the production process [34, 35]. Figure 2 represents an overview of the field development program and three production stages.

High IFT between oil and water combined with high capillary force and electrostatic charge causes a large amount of unrecovered oil to be trapped in the pores [3, 36, 37]. To decrease residual oil value and improve oil recovery yields, improved and upgraded technologies such as enhanced oil recovery (EOR) have been employed in secondary or tertiary stages [26, 37]. A significant difference between EOR and conventional reservoir recovery techniques like water and gas injection is that EOR mobilizes the remaining oil in the porous media for more economical outcomes, further extending the lifetime of reservoirs and prolonging crude oil production [38–41]. IFT of the injecting fluid has a crucial role in EOR. The high value of IFT between injected water and the oil prevents oil displacement, resulting in a high amount of unrecovered oil in reservoirs. Meanwhile, decrease in IFT caused by the EOR fluid reduces and mobilizes the unrecovered oil value and enhances microscopic sweep efficiency [32, 33, 42–44]. The second reason for low oil recovery and oil remaining in the pores is capillary forces, which can be overcome by using the EOR methods [33, 45].

To demonstrate the effect of capillary forces, the nondimensional capillary number has been considered as the ratio of fluid viscosity (μ) to velocity (V), IFT (σ) between oil and water, and contact angle (θ) [46–48]. As this ratio increases by four or five orders in the magnitude of an EOR process, the residual oil volume will reduce, which requires either an IFT decline or adjusting the contact angles for a moderate wettability, which is nearly 90 degree of interphase (θ) [46, 48].

2. A Review of EOR and MEOR Methods

The EOR methods mainly include injecting a particular fluid with chemical, thermal (in situ combustion, injecting steam), or microbial characteristics into the reservoir [21, 23, 37, 49–53]. Thomas provided a detailed classification of EOR methods [54]. MEOR (microbial enhanced oil recovery) is an EOR technique that utilizes microorganisms and their metabolic products, which can produce nearly 30% of the remaining oil in the reservoir [55]. A considerable number of EOR techniques employ chemical methods due to their easy applicability and easy access to various chemical compounds. Meanwhile, the MEOR methods continue to gain attention because of their improved applicability, environmental friendly nature, and competitive prices [21, 56–58]. The existence of various microorganisms with different metabolite productions and growing properties affects the used recovery method. There are three feasible mechanisms to use biosurfactants in the MEOR processes:

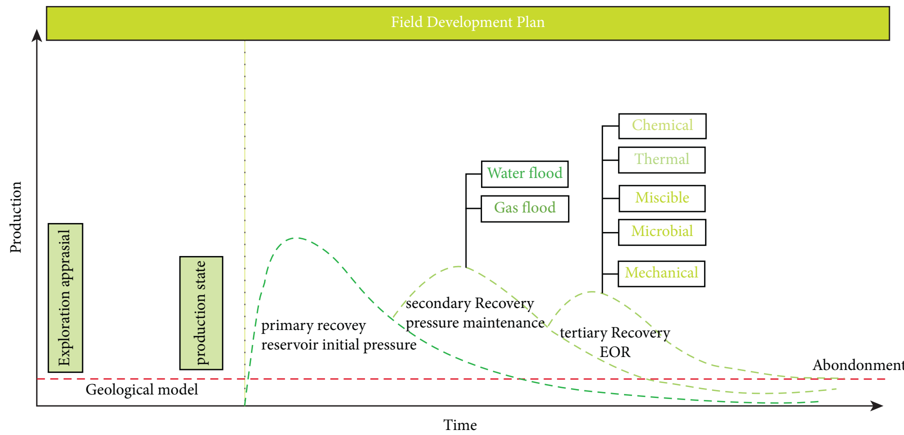

FIGURE 2: Various stages of petroleum production (inspired from [1]).

(1) Injecting biosurfactant-producing microorganisms from wells toward the reservoir and consequent in situ diffusion by the reservoir rock
(2) Injecting appropriate nutrients inside the reservoir to stimulate the reservoir's biosurfactant-producing endemic microorganisms to grow
(3) Producing biosurfactants in ex situ bioreactors and injecting them subsequently into the reservoir [59, 60]

Chemical EOR, a highly effective EOR method, includes surfactant injection, polymer injection, acid and alkaline flooding, and injecting other suitable chemical compounds within the reservoir to alter the interactions between crude oil/brine/rock (COBR) and properties and enhance oil recovery [53, 61, 62]. In polymer injection for enhanced oil recovery, polymers are dispersed into water to increase water viscosity and then injected into the reservoir in order to reach a greater capillary number, higher vertical sweep power, and also increased upward-moving control [63, 64]. Alkaline flooding is another powerful EOR method to recover heavy oil, which has been used in many projects since 1970 [42, 65, 66]. Notwithstanding other methods, this method has no limitation about depths or formation thicknesses and needs no high-cost surface equipment. The reaction between alkali and the acidic compound of heavy oil forms surface-active materials, which reduces IFT by ionization at the water-oil interface by several orders of magnitude [67].

Various experiments have been performed in other branches of chemical EOR, using alkaline surfactants (ASs), alkaline polymers (APs), and alkaline surfactant polymers (ASPs), which has attracted interest as one of the most effective techniques [47, 68]. Delshad et al. [69] simulated Chinese onshore oil reservoir characteristics using a specific method to measure the amount of recovered oil. Surfactant polymer, alkaline, and ASP flooding methods were examined where the alkaline surfactant polymer flooding method increased 24% of OOIP further recovery in comparison with using only water and produced the least value for residual oil. Synthetic chemicals are generally derived from fossil fuels, have high prices, and cause inappropriate environmental influences. Surfactant flooding includes injecting either chemical surfactants or biosurfactants and natural surfactants into the reservoir [9]. Both biosurfactants and chemical surfactants decrease IFT at the interface and surface tension at the surface by accumulating between the liquid phases [70, 71]. Chemical surfactants and polymers are generally expensive, hazardous, and subtend several obstacles, such as undesirable residues that are hard to dispose, environmental impacts, and linking with fossil fuels. The natural surfactants are classified in the same way as chemical surfactants into amphoteric, nonionic, cationic, and anionic types, and they have a nature-based source, for example, saponin derived from *Zizyphus spina-christi* leaves, nonionic surfactants derived from *Glycyrrhiza glabra*, and cationic surfactants from olive, *Prosopis*, spistan, and *Seidlitzia rosmarinus* [70–72]. Several articles have investigated the application of chemical surfactants and their effect on the water and oil IFT in EOR projects, due to relatively lower costs. Biosurfactants have been reviewed for their potential to be used in petroleum production, especially to achieve eco-friendly biodegradable surfactants.

3. Surfactants

The use of surfactants dates back 2,800 years in soap production [73]. The global production of surfactants has grown to over 13 million tons every year as one of the most widely used industrial chemicals, half of which is used as laundry and household detergents [74, 75]. Surfactants include both hydrophilic and hydrophobic domains as a group of amphiphilic chemical compounds, which makes it an essential component in most modern industries like agriculture (i.e., organic chemicals and fertilizers), agrochemicals, metallurgy, mining, cosmetics, foods, paper, public health and pharmaceuticals, beverages, environmental management, textiles, petroleum, petrochemicals, and bioremediation [72, 74, 76, 77].

Taking advantage of petrochemical and oleochemical resources, organochemical synthesis produces most of the surfactants used in the industry today. Hence, most of the surfactants used today are petroleum-derived, which is problematic due to environmental incongruity and toxic effects on humans and the environment, leading to major ecological problems since they inevitably enter the environment after use [74]. Use of surfactants is an increasing concern due to their biodegradability, toxicity, bioaccumulation, and phosphate releases [78]. The environmental community should make efforts to decrease detergent loads and biodegradable use since detergents release phosphates. The extensive diversity and large volumes of applications have been a significant drive to move forwards in producing surfactants from natural and renewable feedstock, which are biosurfactants. An extended genetic variety of microorganisms provides the substantial capability of forming new sorts of biosurfactants from the natural fermentation process instead of organochemical-synthesized surfactants. The following section discusses microbially produced biosurfactants and effective parameters in the EOR process.

4. Biosurfactants

Biosurfactants are a group of amphiphilic compounds with antiviral, hemolytic, insecticidal, and antimicrobial biological activities. They are applicable in numerous industries, including foods, cleaning products, pharmacology (drug solvents), cosmetics, pesticides, textiles, fungicides on different organic surfaces, medicine, and oil and gas fields as essential biotechnology products [7, 79]. The growing interest in biosurfactants is because of their low environmental impacts, low toxicity, and biodegradability [80]. The global market for biosurfactants reached 1.5 billion USD until 2019, which is assumed to experience over a 5.5% CAGR by 2026, of which over half of it belongs to Europe [7]. Household detergents, cosmetics, personal care, and the food industry account for the highest application market [7, 81].

By 1960, the first biosurfactants were synthesized via microbes throughout hydrocarbon fermenting in the form of extracellular compounds [82]. They are generated on living surfaces and can reduce ST as well as IFT. The best production grounds for them are extracellular hydrophilic and hydrophobic moieties or surfaces of microbial cells [83]. Due to growing awareness of the adverse effects of chemical surfactants on the environment, interests have shifted toward environment-friendly surfactants [80, 84]. Affective factors in producing biosurfactants are carbon content, pH, the type of the nitrogen source, temperature, aeration, and carbon-to-nitrogen Ratio [74, 76]. Evonik, Jeneil Biotech, Biotensidon, and Ecover are the major biosurfactant-producing companies; also, genera *Pseudomonas*, *Bacillus*, *Sphingomonas*, and *Actinobacteria* include the foremost biosurfactant-producing bacteria. Desai and Banat [80] provided a detailed review of biosurfactants, and we will only focus on biosurfactants that may be useful for improving oil recovery such as glycolipids, rhamnolipids, lipopeptides, and sophorolipids.

Özdemir et al. [83] compared the interfacial tension reduction in two pure rhamnolipid solutions. They found that molecules of rhamnolipids have strong intermolecular interactions, thus resulting in excellent foam-formation properties even at a narrow air flow rate. However, other yeast biosurfactants, including sophorolipids and glycolipids, had moderate foaming abilities.

Joshi-Navare et al. [85] produced and investigated sophorolipids (SLs) from nonedible Jatropha oil and realized their effectiveness in removing stains. They optimized fermentation parameters to maximize Jatropha oil-derived SL yield (SLJO), which demonstrated a significant ability to decrease the ST in distilled water, coupled with antibacterial and stain-removing capabilities, in addition to good emulsion stability under pH stress and temperature. Reduced immersion time during the washing process accompanying their antibacterial activity, biodegradability, and skin-friendly properties make them ideal alternatives for synthetic surfactants in household detergents to reduce their adverse effects.

Sajna et al. [86] studied a *Pseudozyma* sp. NII08165 biosurfactant containing a combination of some unknown glycolipid and three mannosylerythritol lipid (MEL) isomers. Their stability over the alkaline pH range and high temperatures make them suitable as laundry detergent additives to remove stains efficiently. Figure 3 compares the yield of various biosurfactant-producing microorganisms. Some biosurfactant yields are not exactly presented in reviewed articles, and we did not mention them in the figure and used accurate data.

5. Application of Biosurfactants in EOR Methods

The foaming property of biosurfactants allows use of them in various sectors, such as reducing oil viscosity and cleaning crude oil storage tanks as detergents. Several researchers have been investigating the usage of biosurfactants in EOR operations. The cost of biosurfactants is generally higher than chemical surfactants, making EOR less viable commercially, but they perform better than their chemical counterparts due to higher environmental compatibility, reduced toxicity, and ability to be generated from renewable sources [87]. Utilizing cheaper raw materials, optimizing media components, hyperproduction, or using fermentation extracts that contain significant amounts of biosurfactants can reduce the cost [2, 57]. Biosurfactant use for EOR can be divided into ex situ and in situ MEOR procedures, called biosurfactant-mediated MEOR (BS-MEOR) [59]. In the former, laboratory-produced biosurfactants are injected into the reservoir, while the in situ process identifies proper microorganisms present in the reservoir and supports their growth to synthesize needed metabolites, for instance, surfactants or polymers that provide favorable factors for EOR [88]. Following this phase, wells are shut in and monitored for microbial activity and metabolite production. There has been a successful substantial increase in oil production resulting from MEOR field-scale in situ research projects.

Lal [89] constructed a combined microbial consortium consisting of three hyperthermophilic, acidogenic, and barophilic anaerobic strains to improve oil recovery from oil reservoirs under 70°C to 90°C temperature, resulting in various metabolic outputs like alcohols, biosurfactants,

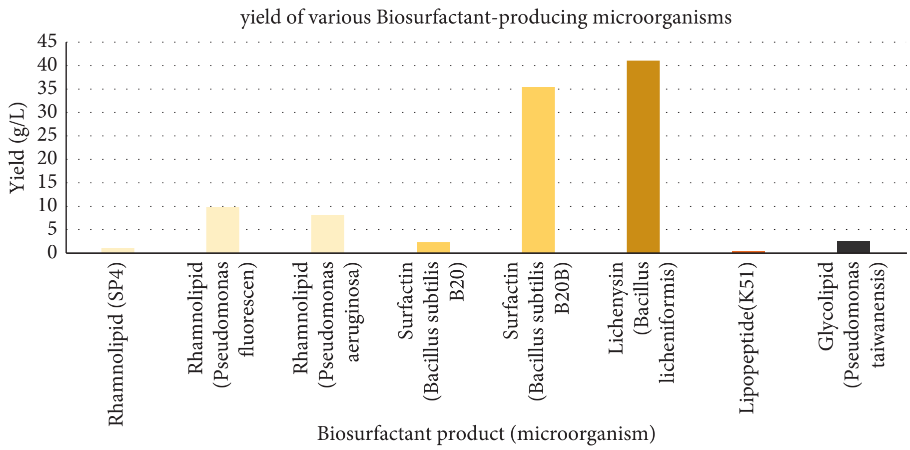

Figure 3: Yield of various biosurfactant-producing microorganisms (data extracted from [82]).

methane, volatile fatty acids, and CO_2 under the specially designed nutrient medium. In situ application of the microbial consortium provided an oil-improving recovery process and increased the efficiency of sweeping crude oil from oil-bearing poles of rock formations. Studies of the MEOR applications mainly focus on two types of biosurfactants: glycolipids and lipopeptides [90]. Lipopeptides show greater efficacy in reducing IFT and ST [90, 91].

Cooper et al. [92] studied surfactin production on a large scale by B*acillus subtilis*. They discovered that surfactin was recoverable from collapsed foam through acid precipitation. Glucose substrate fermentation by sequential product removal by foam fractionation resulted in a good yield, which also increased via the application of iron or manganese salts. *B. subtilis* completely stopped producing surfactin when hydrocarbon was added to the medium, which regularly improves biosurfactant production. By reducing the pH to 2.0, the surfactin precipitates from *Bacillus subtilis* culture spent media, and after extracting dichloromethane and reprecipitating it with acid, it can be better purified.

Biochemical investigation on one of the JF-2 proteins exhibited a similar amino acid composition and infrared spectrum to surfactin but the JF-2 acid precipitate has other components that may be necessary for interactions between all components and JF-2 as full activity surfactant. Following this, required solvents have been developed for complete activity and interaction among these compounds. Among four present components in the crude extract, chloroform, dichloromethane, and methanol could extract sequentially 1, 2, and 4 components with 23%, 56%, and 94% values of activity recovery, respectively. Reconstituted surfactant preparation was performed more actively than preparation of individual extracted components in experiments [92, 93].

Cooper and Goldenberg [94] obtained that the monoglyceride biosurfactant produced by *B. cereus* increased the polyhexosamine emulsifier activity by this organism.

Investigations of McInerney et al. [93] and Jenneman et al. [95] on the JF-2 surfactant revealed promising properties for enhanced oil recovery. Those concentrations where NaC1 was at least 5% (w/v) provided the lowest IFT; thus, the JF-2 biosurfactant is more effective when the NaC1 concentration is high. Oil reservoirs have high levels of salt, which are the ideal conditions for the organism to grow. Very low CMC of JF-2 makes it an effective biosurfactant in dilute concentrations; the IFT was obtained less than 0.1 mN/m. High biosurfactant concentrations or cosurfactants may result in lower IFT.

McInerney et al. [93] studied anaerobic biosurfactant production from *Bacillus licheniformis* strain JF-2 growing in a medium consisting of glucose-mineral salts, $NaNO_3$, and yeast extract. By decreasing the pH to 2.0, an anionic biosurfactant precipitated from the spent medium, which resulted in a substantial decline of the ST in the medium from 70 to 74 mN/m to as low as 28 mN/m. Both JF-2 and surfactin biosurfactants reduced water surface tension by 27 mN/m, the lowest determined surface tension for biosurfactants. *B. subtilis* completely stopped producing surfactin when hydrocarbon was added to the mechanism, which normally boosts biosurfactant production. By reducing the pH to 2.0, the surfactin precipitates from *Bacillus subtilis* culture spent media, which can be further purified by dichloromethane extraction and reprecipitation with acid.

Arima et al. [96] characterized surfactin, an efficient biosurfactant synthesized from *Bacillus* strains, as it possesses high surface activity and a low critical micelle concentration (CMC) comparable to synthetic surfactants. Lipopeptide surfactin inhibits fibrin clot formation and induces lowered surface tension and reduced interfacial tension against a hexadecane concentration of less than 1 mN/m. It is stable under high pH conditions, high salinity conditions, and high temperatures. Furthermore, low CMC characteristics may be advantageous for EOR applications since they impact the economy of EOR development [97, 98].

McInerney et al. [93] grew and synthesized biosurfactants using *Bacillus mojavensis* JF-2 anaerobically along with other competing organisms in a sand environment. *Bacillus mojavensis* JF-2 grew fastest on glucose; besides, monosaccharides tended to be the preferred sources of carbon generally. The highest growing yield was observed with fructose. Under anaerobic conditions, the presence of peptone #2 (PP2) in the medium enhanced the production of biosurfactants.

Table 1: Utilized microorganisms and their products.

Microorganisms	Growth requirement	Microbial products
Clostridium sp.	Anaerobic	Acids, surfactants, gases, alcohols
Bacillus licheniformis	Facultative	Acids, surfactants
Bacillus sp.	Facultative	Acids, surfactants
Gram-negative rods	Facultative	Acids, gases

The microbial formulation consists of four microorganisms (NIPER Bac 1) that have been injected into four wells of the Delaware–Childers field in Oklahoma, producing primarily surfactants, alcohols, and acids, followed by regular injections of molasses used as a nutrient [99]. Applied microorganisms and their products are classified separately in Table 1. After core waterflooding, laboratory microbial tests recovered 28% of the residual oil.

Bryant et al. [99] performed micromodels to reveal if oil could be mobilized via the microbial formulation in the porous environment and related oil mobilization with oil recovery efficiency in Berea sandstone cores. After micromodel waterflooding, about 60% of the oil has been mobilized. When the micromodel was incubating, gas bubbles were observed, as well as crude oil emulsification. NIPER Bac 1 mobilizes oil primarily through gas production, surfactants, and solvents. The initial results showed a 13% sustainable increase in oil recovery along with decreased surface tension of produced brine, revealing the effectivity of in situ microbial biosurfactant production in recovering additional petroleum. As a definite record that strain JF-2 can grow and manufacture its surfactants all over an oil reservoir, strain JF-2 was detached from the produced brine 30 weeks after injection.

In another study on micromodels, Bryant and Douglas [100] noticed that microorganisms that have a high recovery efficiency in a core flood also perform better in mobilizing crude oil. As microorganisms grow and produce CO and chemicals, they can significantly enhance oil recovery in reservoirs under suitable salinity and temperature conditions. A microbial study of Berea sandstone cores revealed that ability of bacteria in recovering remaining oil after waterflooding varies widely, ranging from 7.5 to 71%; the type of core encapsulation had no effect on the recovered oil amount. Studies using a wide permeability range (134–1,920) showed that some microorganisms might be more efficient at recovering oil in lower-permeability cores. As a result, some surfactant-producing bacteria enhance oil recovery in ways that no gas-producer microorganisms can do; as a result, gas production is not the only affecting factor. It is also vital to consider the kind of surfactant; two microorganisms can produce surfactants that differ in their recovery capacity. Improving the areal sweep efficiency (EA) of microorganisms appears to be a beneficial mechanism in microbially mobilizing oil. Comparing two heavy oil and medium-to-light-gravity oil recovery yields showed that the MEOR method might be appropriate for both light and heavy oils.

The work of McInerney et al. [93, 101] and other papers [102, 103] indicated lower than 0.1 mN/m interfacial tension between water and oil while using lipopeptides. Lipopeptide surfactants have a poor yield and high manufacturing costs when produced through fermentation; therefore, they are rarely applied in field-scale EOR projects, and utilizing them is limited to industries like paper, health care, pulp, and foods [104].

Long et al. [105] conducted a study on the pH-regulated emulsifying activity of surfactin and its possible application in separating oil in EOR. The results demonstrated that surfactin is able to effectively stabilize emulsions over pH 7.4, which allows easy oil separation by adjusting pH when using surfactin as an emulsifier. The surfactin-based EOR technique has great application potential since it can be reused multiple times and sustains its activity after demulsification.

6. Conclusions

In this study, applied microorganisms on oil fields were explored. Although biosurfactants tend to be more expensive than chemical surfactants, they are preferred due to their environmental compatibility, reduced toxicity, and ability to be produced from renewable resources. Utilizing cheaper raw materials, optimizing media components, hyperproduction, or using fermentation extracts that contain significant amounts of biosurfactants can reduce the cost. Oil recovery can be improved using both in situ and ex situ microorganisms. Utilizing in situ microbial EOR-produced metabolic products such as biosurfactants, alcohols, methane, volatile fatty acids, and CO_2 improved oil recovery and sweep efficiency. The effect of different parameters, including pH, concentration, and components, on the behavior of different kinds of *Bacillus* microorganisms was investigated. An efficient biosurfactant synthesized with low CMC, surfactin, was examined to achieve a low interfacial tension. By optimizing microbial mobilization in the porous environment, oil recovery efficiency was maximized.

Abbreviations

AS:	Alkaline surfactant
ASP:	Alkaline surfactant polymer
AP:	Alkaline polymer
BS-MEOR:	Biosurfactant-mediated microbial enhanced oil recovery
B. subtilis:	*Bacillus subtilis*
CMC:	Critical micelle concentration
COBR:	Crude oil/brine/rock
EA:	Areal sweep efficiency
EOR:	Enhanced oil recovery
IFT:	Interfacial tension
OOIP:	Original oil in place
MELs:	Mannosylerythritol lipids

MEOR: Microbial enhanced oil recovery
PP2: Proteose peptone #2
ST: Surface tension
S_{or}: Residual oil saturation
SLs: Sophorolipids
SLJOs: Jatropha oil-derived sophorolipids.

Data Availability

The data used to support the findings of this study are included within the article.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

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