Role of Biosurfactants in Marine Sediment Remediation of Organic Pollutants

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1 Introduction

Petroleum crude oil and its hydrocarbons constitute a significant concern among the numerous environmental organic pollutants that adversely impact marine ecosystems and their function (Jamal 2022; Wang et al. 2022; Wei et al. 2020). Oil spills in the marine environment often lead to immediate and long-term ecological damage. In several crude oil-producing regions of the world, marine sediments have been significantly contaminated with organic pollutants with a total petroleum hydrocarbon concentration (TPH) of 44,600 mg per kg of dry soil (Feng et al. 2021). The sources of organic hydrocarbon pollutants in the marine environment include industrial zones, commercial ports, touristic cities, aquacultural/agricultural practices, oil/gas exploitation, megacities, and other anthropogenic activities (Dai et al. 2022; Kumar et al. 2021). When crude oil is spilled into the marine environment, it persists in the sediment, enters the marine food web, and exerts detrimental effects on humans and other organisms (Biswas et al. 2019; Gayathiri et al. 2022; Mgbechidinma et al. 2022a; Shuai et al. 2019). These oils consist of aliphatic and aromatic compounds that greatly hinder remediation technology while negatively impacting the surrounding environment due to their toxicity, complexity, persistence, bioaccumulation tendency, and susceptibility to long-range atmospheric transport.

Although many studies have documented different treatment processes for cleaning up organic pollutants in the environment, biological methods using microbes and plants remain a promising alternative green method (Dai et al. 2022; Kariyawasam et al. 2022; Kumar et al. 2021; Lal et al. 2018; Nayak et al. 2020;

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Sonowal et al. 2022). Biological remediation of oil-contaminated environments is highly recommended and widely practiced because of its efficiency, cost-effectiveness, and environmentally friendly nature than the mechanical or chemical methods (da Silva et al. 2021; Muneeswari et al. 2022; Pete et al. 2021). While developmental approaches to biological remediation are growing, the major strategies include phytoremediation (using plants), bioremediation (using nutrients—bio-stimulation and using microbes—bio-augmentation), and bio-electroremediation (Fdez-Sanromán et al. 2021; Laothamteep et al. 2022; Mapelli et al. 2017). Recent studies on crude oil biological remediation focus on constructing effective microbial consortiums (mono and mixed cultures), inoculating co-metabolic substrates, re-inoculating contaminated sites with indigenous microorganisms, and genetic manipulation of microbes/plants (Feng et al. 2021; Gayathiri et al. 2022; Yan et al. 2020).

Despite the advances in improving crude oil biological cleanup methods, the degradation of petroleum hydrocarbon is limited mainly by the bioavailability and toxicity of the pollutants, the spatial distribution of microbes/plants, and their metabolic capability (Huang et al. 2020; Jamal 2022; Zhou et al. 2021). In marine sediments, the bioavailability mechanism of hydrocarbon pollutants involves desorption from the soil matrix, transport, and uptake/absorption by plants or microorganisms (Feng et al. 2021). Also, hydrocarbon moieties of crude oil are highly hydrophobic, leading to their absorption unto sediments, thereby limiting the pollutant mass transfer rate. Thus, most hydrocarbon pollutants in marine sediments are not readily available to the plant or microbes as nutrient sources, hindering their degradation. The primary factor limiting crude oil bioremediation in contaminated sediments is the slow desorption of these hydrocarbons from the solid phase (sediment) to the aqueous phase, causing low bio-accessibility (Dai et al. 2022; Feng et al. 2021; Guo et al. 2022; Kumar et al. 2021). In an attempt to advert these limitations in the biological remediation of organic pollutants in the marine environment, advances have been made toward incorporating surfactants to lower the surface tension between the solid and aqueous phases (Dhanya 2021; Hentati et al. 2021).

Surfactant-enhanced bioremediation is a promising technique for improving the bio-accessibility of organic hydrocarbon pollutants (da Silva et al. 2021; Gidudu and Chirwa 2021; Liduino et al. 2018). However, as surfactants increase organic pollutants desorption and free transport into the aqueous phase to intensify remediation, petroleum-derived surfactants are not eco-friendly because of their high toxicity and low biodegradability. Hence the increasing interest in biosurfactants as a green, non-toxic alternative to their chemical counterpart. Biosurfactants are active surface secondary metabolites synthesized by microbes that can utilize substrates like simple sugars and oils as nutrient sources (Durval et al. 2020; Femina et al. 2021). They have different structures and are amphiphilic compounds with polar and nonpolar moieties. These secondary metabolites are grouped based on the type of producer, the substrate used, and their chemical composition as low molecular weight (glycolipids, phospholipids, lipopeptides, fatty acids) and high molecular weight (polysaccharide–protein complexes) biosurfactants (Sarubbo et al. 2022). Following a systematic approach, published articles that report the application of biosurfactants in sediment remediation were reviewed and quantitatively evaluated using comprehensive databases such as Web of Science, Scopus, Google scholar, and Pub Med between 2017 and May 2022 (Fig. 1).

Figure 1 shows the rise in research incorporating biosurfactants in sediment remediation to remove and degrade pollutants. However, biosurfactants have a wide range of applications in several industries, including food, cosmetics, agricultural, and pharmaceutical (Ashitha et al. 2020; da Silva et al. 2021; Pandey et al. 2022; Xu et al. 2020). The dramatic increase in biosurfactant application in the marine environment can be attributed to its numerous environmental compatibility properties. These include reducing surface and interface tension, emulsion formation, foaming capability, oil displacement, biodegradability, nontoxic, and stability over varying environmental conditions like temperature, pH, and salinity (Dell'Anno et al. 2018; Gidudu and Chirwa 2021; Ram et al. 2019; Wei et al. 2020). These properties make biosurfactants an environmentally compatible biomolecule of the twenty-first century. Biosurfactants are produced within cells or secreted extracellularly to form thin films for cellular communications that regulate several physiological activities (Sharma et al. 2021). Many recent studies have reported successes in applying biosurfactant producer as remediation agents. However, there are advances in applying biosurfactants during pollutant remediation by employing nanotechnology, immobilization, dose-supplementation, and direct use of crude extracts (Mandal et al. 2018; Rong et al. 2021).

Following the unpredictable flow of organic pollutants such as hydrocarbons (aliphatic and aromatic), pesticides, and chlorinated solvents in the marine environment and their hazardous impact on the exposed populations, ecosystem productivity, and global economic growth, this chapter is focused on revealing the role of biosurfactants in the marine sediment remediation of organic hydrocarbon pollutants. The sediment remediation trend in biosurfactant application, as shown in Fig. 1, also emphasizes the scope of this chapter. The environmental compatibility properties of biosurfactants and the factors that influence their production are discussed. Considering the anoxic conditions of the marine environment, we reviewed biosurfactant production under aerobic/anaerobic conditions. Moreover, application strategies of biosurfactants and the interaction mechanism underlying biosurfactant–pollutants' complexation during remediation are discussed. We further revealed recent advances in biosurfactant-mediated remediations while providing future outlooks for developing efficient and eco-sustainable biosurfactant-based strategies in marine sediments remediation in view of large-scale applications. This chapter will provide relevant insight into the possible achievement of environmental sustainability of marine sediments in beaches, wetlands, marshes, offshores, and intertidal zones.

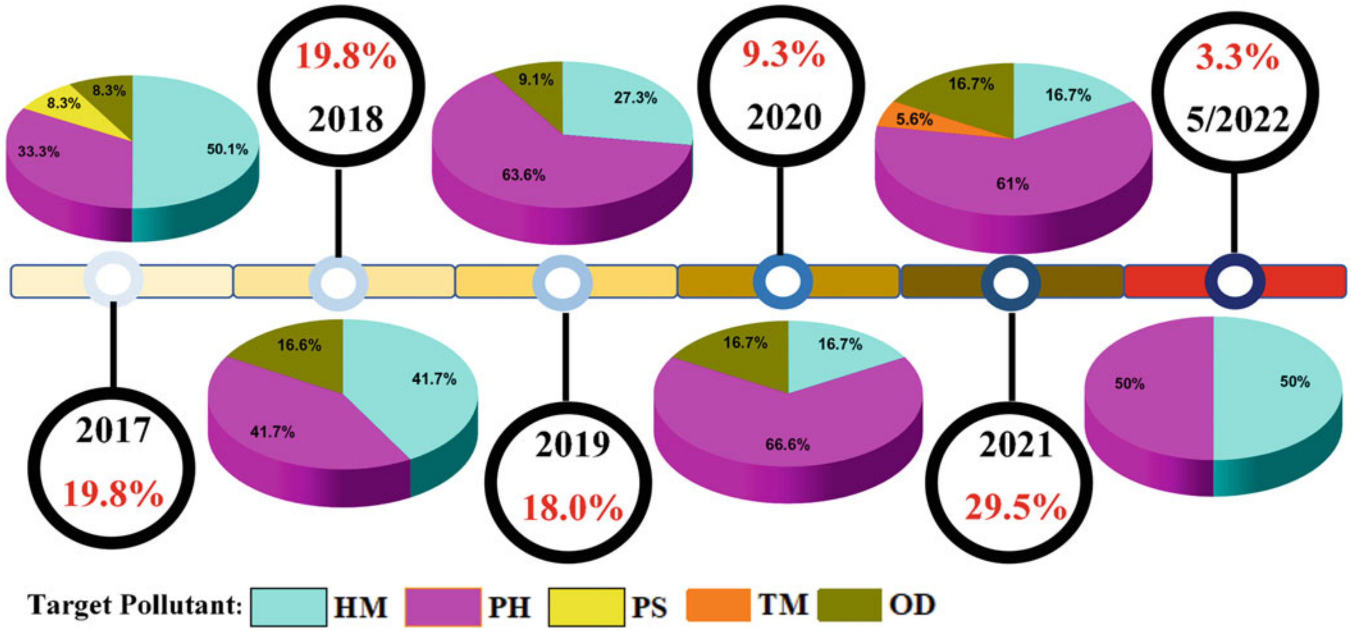

Fig. 1 The systematic trend in published articles on biosurfactant application in sediment remediation as indexed in several databases between 2017 and May 2022. The target pollutants observed were heavy metals (HM), petroleum hydrocarbons (PH), pesticides (PS), transition metals (TM), and others (OD). Others represent combined pollutants. This process was performed using limited searching terms: biosurfactant AND sediment AND remediation. The outcome measurement technique was restricted to specific criteria focusing on the title, abstract, and keywords. Duplicate search results were removed, and studies on petroleum surfactants and aqueous environmental remediation were removed

2 Biosurfactants: Production, Environmental Influence, and Remediation Properties

Intrinsic and extrinsic factors involved in biosurfactant production affect their activity. The effectiveness of these factors is commonly evaluated under conditions of different oxygen levels, temperatures, pH, salinity, and microorganism used.

2.1 Biosurfactants and Their Main Microbial Producers

Biosurfactants are amphiphilic molecules presenting hydrophobic features (consisting of saturated and unsaturated long-chain fatty acids, hydroxy-fatty acids, or α-alkyl-β-hydroxyl fatty acids or amphiphilic/hydrophobic peptides) and hydrophilic features (composed of anionic or cationic amino acids, peptides, monosaccharides, disaccharides, polysaccharides, phosphate, carboxylic acid, or alcohol). Biosurfactants are mainly anionic or neutral, but some cationic forms have amine groups. They are classified based on their chemical composition, molecular weight, physicochemical properties, mode of action, and microbial origin into several classes like simple fatty acids, glycolipids, lipopeptides, lipopolysaccharides, phospholipids, polymeric and particulate compounds (Ashitha et al. 2020; Gayathiri et al. 2022).

Several biosurfactants produced by different microbial taxa have been isolated from marine environments and mostly reported are rhamnolipid, cellobiolipids, trehalose lipids, sophorolipids, mannosylerythriol lipids, and surfactin from microbes in genera *Pseudomonas*, *Bacillus*, *Actinobacteria*, *Ustilago*, *Rhodococcus*, *Arthrobacter*, *Candida*, and *Pseudozyma* (Gayathiri et al. 2022). Biosurfactants regulate quorum sensing and significant microbial roles like motility, antagonism, and virulence (Sharma et al. 2021). These roles form the basis of most biosurfactant interactions with microbes in terrestrial and aquatic ecosystems during pollutant degradation (Dhanya 2021; Wei et al. 2020). Table 1 shows microbial biosurfactant producers and their fermentation conditions.

2.2 Environmental Factors That Influence Biosurfactant Production

At the late exponential and stationary growth phase of microbes during fermentation, several environmental factors significantly influence metabolite production (surfactants) (Uddin et al. 2021). These factors account for the differences in biosurfactant composition, structure, and properties that affect their applicability (Filho et al. 2021). Some of the intrinsic factors include the nutrient source and the fermentation mode. Most literature now reports on the use of renewable waste as substrates for

Table 1 Biosurfactant producers and their relevant environmental conditions

Biosurfactant producers	Isolation site	Fermentation conditions	Produced biosurfactant (Yield)	References
P. aeruginosa CH1	Zhoushan island, China	Anaerobic cultivation in 200 mL medium (containing 0.5 mg/L resazurin) in 250 mL serum bottle aerated with oxygen-free N_2 gas at 30 °C, 180 rpm for 5 d	Rhamnolipid	Jiang et al. (2022)
Pseudomonas mendocina ADY2b	Chennai harbor	Aerobic, 28 °C, pH 7.2 at 150 rpm	Rhamnolipid	Balakrishnan et al. (2022)
Enterobacter hormaechei	Tamil Nadu, India	Aerobic, 35 °C, 150 rpm for 10 d	Lipopeptide	Muneeswari et al. (2022)
Aeromonas hydrophila RP1	Himachal Pradesh, India	Aerobic, 27 °C, pH 7 for 5 d	Glycolipopepetide	Pandey et al. (2022)
B. subtilis AnPL-1	Xinjiang, China	Anaerobic cultivation in 100 ml serum bottles containing 80 mL medium at 39 °C, 80 rpm for 10 d	Surfactin	Zhao et al. (2021)
Vibrio sp. LQ2	The South China Sea	Aerobic, 30 °C, pH 7 at 180 rpm	Phospholipid	Zhou et al. (2021)
B. subtilis AS2, *B. licheniformis* AS3 and *B. velezensis* AS4	Tamil Nadu, India	Aerobic, 40 °C, pH 7 at 150 rpm	Lipopeptide	Prakash et al. (2021)
Staphylococcus sp. CO100	Sfax, Tunisia, Mediterranean Sea	Aerobic, 37 °C, pH 7.6, 100 g/L NaCl at 180 rpm	Lipopeptide	Hentati et al. (2021)
P. aeruginosa ASW-4	Zhoushan island, China	Aerobic, 25 °C, pH 7 at 150 rpm	Rhamnolipid	Chen et al. (2021)
P. cepacia CCT 6659	São Paulo state, Brazil	Aerobic, 28 °C, pH 7, 250 rpm for 60 h	Rhamnolipid	da Silva et al. (2021)
Bacillus cereus UCP 1615	Pernambuco, Brazil	Aerobic, 28 °C, pH 7 at 200 rpm	Lipopeptide (4.6 g/L)	Durval et al. (2020)
P. aeruginosa CH1	Zhoushan island, China	Aerobic, 30 °C at 180 rpm for 8 d	Rhamnolipid	Huang et al. (2020)
B. licheniformis LRK1	Bhavnagar, India	Aerobic, 35 °C, pH 7, 3% salt concentration at 150 rpm	Lipopeptide	Nayak et al. (2020)
Paracoccus sp. MJ9	Jiaozhou Bay, China	Aerobic, 30 °C, pH 7.2 at 130 rpm	Rhamnolipid	Xu et al. (2020)
Acinetobacter sp. Y2	Xinjiang Uygur, China	Aerobic, 30 °C, pH 6.5–7.0 for 3 d	Lipopeptide	Zhou et al. (2020)

(continued)

Table 1 (continued)

Biosurfactant producers	Isolation site	Fermentation conditions	Produced biosurfactant (Yield)	References
Bacillus sp. SGD-AC-13	Chorao Island, Goa, India	Aerobic, 30 °C, pH 7.6 at 150 rpm	Novel thermosta-ble biosurfactant with fatty alkene	Ram et al. (2019)
P. aeruginosa 709	Xinjiang oil reservoir, China.	Anaerobic cultivation in 250 mL serum bottles sealed with butyl rubber stoppers at 39 °C for 10-d.	ND (422.8 ± 16.23 mg/L)	Zhao et al. (2017)
P. stutzeri DQ1	Heilongjiang Province, China	Anaerobic fermenta-tion medium-boiled under a stream of oxygen-free nitrogen and incubated at 40 °C, pH 7.2 for 36 h.	Lipopeptide	Liang et al. (2017)

ND not detected

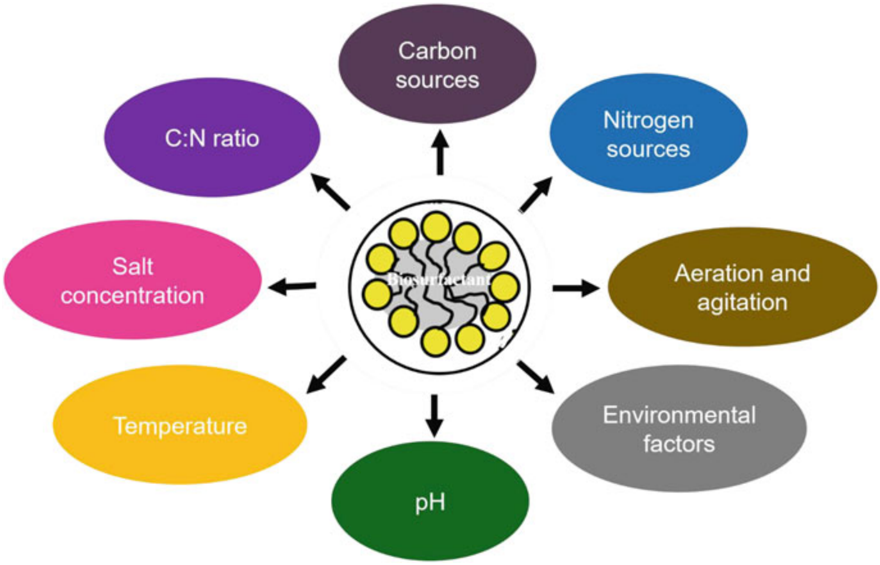

Fig. 2 Factors that influence biosurfactant production

biosurfactant production, mainly agro-industrial wastes (molasses, fruit peels, wheat straw, rice straw, cassava flour, and sugarcane bagasse), animal oil/fats (fish waste, fish peptones, and crude fish oil), dairy/distillery by-products, petroleum refining wastes (marine leachates), and food processing by-products (Das and Kumar 2018; Femina et al. 2021; Gaur et al. 2022; Mgbechidinma et al. 2022b). The major factors affecting biosurfactant production are shown in Fig. 2.

Biosurfactant production relies on the feeding methods employed in a shake flask or bioreactor fermentation. There are three modes of fermentation commonly used in microbial surfactant research. In the batch mode, the media and inoculum are added simultaneously to the bioreactor, and the product is recovered at the end of the fermentation process (Sarubbo et al. 2022). Fed-batch mode entails adding new media regularly without removing the product. In contrast, the continuous mode is run through unceasing substrate streaming and product collection once the maximum product concentration is reached (Gayathiri et al. 2022). During fermentation, biosurfactant producers grow and function in a wide range of environmental conditions such as pH, temperature, salinity, and oxygen availability (aeration and agitation speed).

According to Gayathiri et al. (2022), some ecosystem-dependent biosurfactants include trehalose lipids in cold environments, rhamnolipids in thermophilic environments, lipoproteins in acidophilic/alkaliphilic environments, and glycolipids in saline/hypersaline environments. These environmental factors form the basis for determining the fermentation parameters used during biosurfactant production. Statistical methods can optimize these parameters to investigate their variable interactions and ensure maximum biosurfactant yield at the lowest possible costs (Christopher et al. 2021; Mandal et al. 2018; Mgbechidinma et al. 2022a; Muneeswari et al. 2022; Uddin et al. 2021; Vaishnavi et al. 2021). The commonly reported fermentation conditions are pH 6–8, temperature 28–37 °C, NaCl concentration up to 10%, and aeration modulation at 150–200 rpm agitation speed (Dierickx et al. 2022; Gaur et al. 2022). Although agitation and aeration cause foam formation during biosurfactant production (Domingues et al. 2017), the presence or absence of oxygen transfer remains essential. Although previous studies on biosurfactant production have emphasized the effect of varying temperature, pH, and salinity as relevant environmental conditions, oxygen availability is addressed in this section, considering the anoxic nature of marine sediment below the surface.

2.2.1 Aerobic Biosurfactant Production

Oxygen availability in microbial cultures implies an aerobic condition, whereby molecular oxygen (>30%) is the electron acceptor and limiting factor (Domingues et al. 2017). In field experiments, bulking agents are used to save costs; however, most laboratory studies on biosurfactant production are conducted in aerated bioreactors equipped with agitators. As a result, aerobic biosurfactant production is more widely explored (Balakrishnan et al. 2022; Chen et al. 2021; Hentati et al. 2021; Muneeswari et al. 2022; Pandey et al. 2022; Prakash et al. 2021; Zhou et al. 2021). The presence or absence of oxygen affects genes that regulate biosurfactant production. Under aerobic or anaerobic conditions, the rhl genes expression in *P. aeruginosa* is altered, leading to rhamnolipid production with different yields, homologs, structural composition, and physicochemical properties (Jiang et al. 2022; Zhao et al. 2021).

2.2.2 Anaerobic Biosurfactant Production

The surface marine sediment might allow the aerobic production of biosurfactants by microbes; however, locations in the deep underground areas of the marine environment are anaerobic with high pressure and high salinity (Liang et al. 2017; Zhao et al. 2017). Studies show that aerobic biosurfactants-producing microbes exhibit weaker metabolic activity and lower oil displacement efficiency in anaerobic marine environments (Zhao et al. 2018). The unique biosphere of marine sediments modifies the microbial communities to adapt to diverse metabolic functions using nitrate, iron, bicarbonate, nitrous oxide, and sulfate as electron acceptors. Although some anaerobic biosurfactant syntheses are nutrient dependent, there are currently only a few microbes capable of such processes. According to Zhao et al. (2021), *Bacillus subtilis* AnPL-1 anaerobically produces surfactin (150 mg/L) with emulsification and viscosity reduction effects on crude oil at 20–50 °C, 6–9 pH, and 0–7% of NaCl. The surfactin had a mixture of C13-, C14-, and C15-surfactin congeners with 28.5 mN/m ST, 30 mg/L CMC, 70.5% emulsification index, and 40.6% viscosity reduction against crude oil. Jiang et al. (2022) also revealed that *Pseudomonas* sp. CH1 anaerobically produces rhamnolipids with lower CMC (40 mg/L) than 100 mg/L in an aerobic condition. The biosurfactant from CH1 had six homologs with 87.83% mono-rhamnolipids capable of enhancing PAHs solubilization in water from 1.29 mg/L to 193.14 mg/L with over 90% viscosity reduction.

2.3 Characteristic Remediation Properties of Biosurfactant

Although biosurfactants are ecologically safe, they can self-assemble and form micelles that define their morphological structures and specificity, like synthetic surfactants. The three main micelles forms are spherical, rod-like, and wormlike micelles (Fig. 3).

The self-assemblage and micelle formation account for several favorable biosurfactant properties that can be affected by changes in the congener molecular structure (Sarubbo et al. 2022). These properties are the basis for the observable physiochemical methods for developing several rapid techniques for isolating and screening biosurfactant-producing microbes.

2.3.1 Microbial Cellular Communication

Biosurfactants mediate a myriad of cellular communication in microorganisms while allowing for physiological processes such as motility, antagonism, virulence, quorum sensing (detect and modulate cell population density), and biofilm formation/dispersion (Sharma et al. 2021). These cellular communication features can be

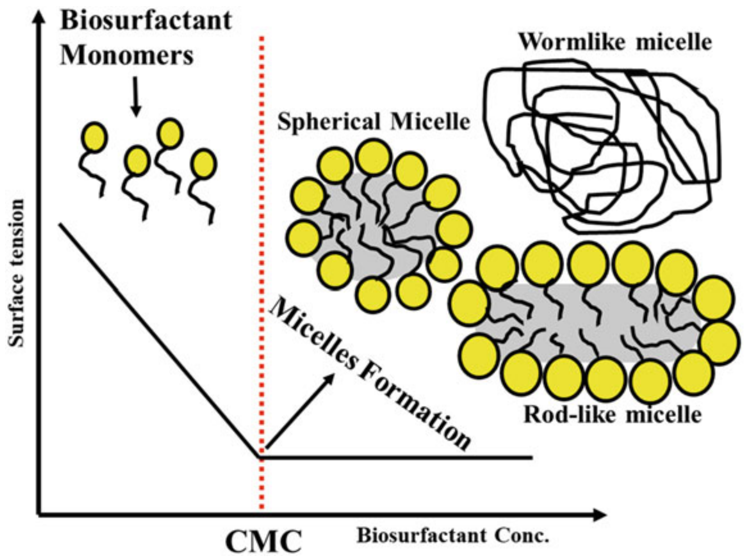

Fig. 3 Biosurfactant micelle structures

explored as an alternative approach for sustainable and economic biosurfactant production.

2.3.2 Surface and Interfacial Tension Reduction

Biosurfactants are known to reduce surface tension (ST) of water (72 mN/m) and interfacial tension (IT) between oil/water interfaces (10–40 times) more than synthetic surfactants owing to their lower CMC values (Femina et al. 2021). This implies that less biosurfactant concentration is required for maximum ST/IT reduction than synthetic surfactants. The IT measures the cohesive energy present at the interface between liquid and liquid or gas and liquid. However, ST and IT are determined similarly in mN/m units by measuring the fermentation liquids or purified biosurfactant extracts using capillary rise, Du Nouy, Wilhelmy plate, and release/drop-weight methods. The principles of these techniques include measuring (i) the counterbalance gravity force and weight of the liquid in the Capillary rise method (Das and Kumar 2018), (ii) the force required to remove a platinum-iridium ring placed on a surface or interface by the Du Nouy method (Balan et al. 2019; Gayathiri et al. 2022; Hentati et al. 2019), (iii) the direct force imposed on a platinum plate at the interface using Wilhelmy plate method (Christopher et al. 2021; Lee et al. 2018; Pandey et al. 2022; Zhao et al. 2018), (iv) the force required to remove a wire ring from a liquid surface (release method) and the droplet weight from a pipe (drop-weight method) (Gidudu and Chirwa 2021; Ram et al. 2019; Uddin et al. 2021).

Corresponding to the decrease in ST and IT, most biosurfactants have CMC values less than 2000 mg/L depending on their molecular structure.

2.3.3 Enhanced Solubilization, Mass Transfer, and Bioavailability

Biosurfactants can increase the solubility of hydrophobic organic compounds, thereby enhancing their mass transfer and bioavailability by reducing ST at the interfacial phase (Femina et al. 2021). This leads to emulsions that dispense solute inside the core of hydrophobic micelles, thus altering the CMC, size, and shape of the biosurfactant. Moreover, biosurfactant solubilization is based on ST reduction and micelle formation, which modifies the hydrophobicity of the cell surface and increases cell-substrate affinity (Zhou et al. 2021). At standard micelle formation, a most compatible phase with the hydrophobic pollutant solubilization, the biosurfactant hydrophobic ends are connected inside. In contrast, the hydrophilic ends are connected to the aqueous phase (Xu et al. 2020). However, the hydrocarbon concentration of any organic pollutant is the limiting factor for effective solubilization (Femina et al. 2021).

2.3.4 Environmental Tolerance and Ionic Strength

The surface activity of most biosurfactants is unaffected by environmental conditions like pH (3–12), temperature (up to 120 °C), and salt concentration (up to 10% w/v). Meanwhile, synthetic surfactants are inactivated at extreme pH, temperature, and salt concentrations greater than 2% (Sarubbo et al. 2022). Therefore, the biosurfactant produced by microorganisms has high foaming and emulsifying activities with stability at extreme temperatures, pH, and salt concentrations (Kumar et al. 2021).

2.3.5 Biodegradability and Low Toxicity

Biosurfactants are not persistent molecules, and their intermittent or end-products are not hazardous. They are highly eco-friendly and safe compared to synthetic surfactants, with a minimal report on their harmful effects when used without purification. Although sucrose-stearate, a synthetic surfactant, has an identical homolog to microbially produced glycolipid, the latter has faster degradability (Gayathiri et al. 2022). Assays that are commonly used to determine biosurfactant toxicity include cytotoxicity activity (using cell lines or animal samples) (Balan et al. 2019; Dierickx et al. 2022) and phytotoxicity activity (using seed or plant samples) (Wei et al. 2020). Biosurfactants are of considerable interest in the food, cosmetic, and pharmaceutical industries, emphasizing the importance of using safe producers like lactic acid bacteria due to their detoxifying and antimicrobial properties (Ashitha et al. 2020; Mgbechidinma et al. 2020). In addition, biosurfactants in hydrocarbon-contaminated environments enhance biodegradability by increasing pollutant solubilization (Feng et al. 2021; Xu et al. 2020).

2.3.6 Emulsion Forming and Breaking

The emulsifying potential of biosurfactants is independent of their ST reduction capability. Although high molecular mass biosurfactants are excellent emulsifiers, not all bio-emulsifiers can significantly reduce surface/interfacial tension. Emulsifiers (polymeric biosurfactants) are in high market demand in the food, cosmetics, and pharmaceutical industries. Also, biosurfactants are used for the demulsification of industrial waste emulsions. While a common mechanism of biosurfactant emulsion formation is polymerization, the breaking mechanism occurs through creaming, flocculation, coagulation, and coalescence (Femina et al. 2021).

3 The Fate of Organic Pollutants and Biosurfactant Mechanism of Action

Marine pollution with petroleum hydrocarbons has detrimental effects on the ecosystems and possible economic resources (Pete et al. 2021). Crude oil is one of the major globally needed natural resources, although only produced significantly by countries like Russia, Iran, Qatar, the United States, Turkmenistan, Saudi Arabia, China, United Arab Emirate, Nigeria, and Venezuela, following the recent report by the US Energy Information Administration (https://www.eia.gov/). As such, the global distribution of oil in line with meeting the high market demand results in crude oil spills and seepage, thus, impacting the lives of the terrestrial and aquatic communities (Pete et al. 2021). Significantly, the marine sediments are the vulnerable sink for these crude oil hydrocarbons consisting of heavy metals, volatile organic compounds (VOCs), aliphatic hydrocarbons, acidic aerosols, hydrogen sulfide, PAHs, and particulates (Gayathiri et al. 2022; Mishra et al. 2021; Wei et al. 2020).

When crude oil reaches the surface, the composition and properties of the oil change almost immediately by processes like evaporation, oxidation, emulsification, sedimentation, biodegradation, and dispersion (Fig. 4). The presence of biosurfactant increases hydrophobicity that improves the interaction of the surface-active agents with the pollutant leading to desorption (Ram et al. 2019). Pollutant solubilization is affected by the charge of the hydrophilic group and the chain lengths that determine the micelles' orientation (Dierickx et al. 2022).

In contaminated marine sediments, the hydrophobic moiety entraps the pollutant hydrocarbon, thereby increasing its adsorption by microbes. Bioremediation efficiency depends on pollutant bioavailability, microbial growth, and degradation capacity. The mechanism of action of biosurfactants in marine sediment remediation

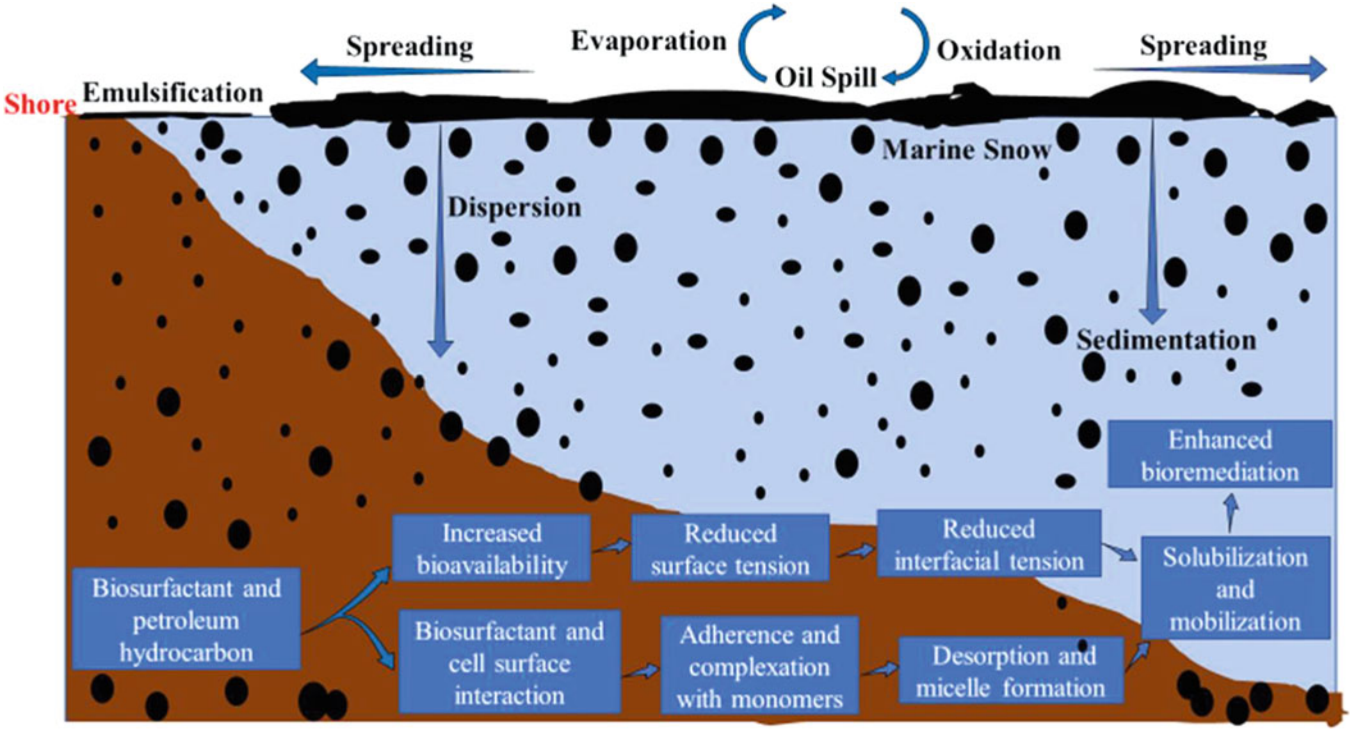

Fig. 4 Biosurfactant-mediated bioremediation. The fate of organic pollutants includes processes in the upper part of the figure, such as spreading, evaporation, oxidation, dispersion, and sedimentation. On the other hand, biosurfactant mechanisms of action are processes in the lower part of the figure, including bioavailability, surface tension, interfacial tension, hydrophobicity, micelle formation, solubilization, and mobilization

of organic pollutants is shown in Fig. 4. The mechanism of hydrocarbon removal by biosurfactant to increase substrate bioavailability for microorganisms and increases cell hydrophobicity can be described based on their molecular mass and concentration. Low molecular mass biosurfactant below CMC allows for mobilization by reducing the surface and interfacial tension between air/water and soil/water systems. Thus, the contact angle increases as the capillary force reduce. Meanwhile, solubilization is favored above the CMC for low molecular mass biosurfactant through the micelle formation with the hydrophobic ends connected inward.

In contrast, the hydrophilic ends are exposed to the exterior aqueous phase (Christopher et al. 2021). Also, a high molecular mass biosurfactant enhances emulsification, a process that forms emulsion (droplet of oil suspended in a fluid).

Interestingly, microbial surface-active agents solubilize crude petroleum oil in aqueous and solid media. Using biosurfactants from marine bacteria (*Bacillus licheniformis* MTCC 5514 producing surfactin) in comparison to synthetic surfactants, Kavitha et al. (2014) described the solubilization of crude oil in soil matrix of different types (sandy, fine sand soil, clay, and clay loam). It was observed that complete solubilization could be achieved at 2% concentration of crude oil, with the biosurfactant having $>25\%$ removal rate than the synthetic surfactants. Compared to other soil types, clay absorbs more crude oil, accounting for its least solubilization efficiency (Kavitha et al. 2014). In a dose-dependent manner, as reported by Saimmai et al. (2013), biosurfactant solubilization of PAHs such as anthracene, fluoranthene, or pyrene (15–20 times higher compared to control) is higher compared to fluorene, naphthalene, or phenanthrene (about 3–5 times compared to control). Biosurfactants increase petroleum hydrocarbon solubilization by multiple folds compared to water (Feng et al. 2021), with efficiency ranging from fivefold solubility (for cell-free supernatant containing the biosurfactant), threefold solubility (for crude biosurfactant) and between 1.6 and 2.8 fold solubility (for synthetic surfactants) (Hentati et al. 2019). This suggests the applicability of biosurfactants for microbial-enhanced oil recovery and environmental bioremediation (Mapelli et al. 2017).

Notable, non-homogenous solubilization is called "pseudo-solubilization," the incorporation of hydrophobic pollutants into the micelle is called "solubilization," and a high amount of hydrocarbon act as a limiting factor during solubilization in different ecosystems. The two significant solubilization mechanisms during bioremediation are the increased bioavailability of microbial substrates and the increasing surface hydrophobicity to allow hydrophobic substrates easily associate with bacterial cells (Xu et al. 2020). Although, further investigation is required to enable model predictions for ex situ sediment remediation mediated by biosurfactant solubilization.

4 Environmental Application of Biosurfactant During Pollutant Remediation

The recent advances in the environmental application of biosurfactants are shown in Table 2.

4.1 Direct Supplementation of Biosurfactant and Dose Effect

Direct addition of biosurfactant during remediation is usually conducted at a known CMC concentration. According to Sarubbo et al. (2022), the biosurfactant mechanism of action during sand washing can occur in 2 ways. Firstly, below CMC allows surfactant accumulation at the soil–pollutant interface, resulting in a change in the system's affinity for water and increasing repulsion force. Meanwhile, the second mode of action occurs above the CMC level when micelles are formed, favoring the partitioning of pollutants into the aqueous phase, increasing solubilization, recovery, demulsification, and possibly recycling to reduce remediation costs (Huang et al. 2020).

In simulated field remediation, Rong et al. (2021) demonstrated bacteria isolation and surfactant toxicity matching as a new sediment remediation technology. It was reported that adding 500 mg/kg rhamnolipid to a bacteria-enriched soil can remove 80.24% of aged total petroleum hydrocarbons (TPHs) within 30 d and significantly increase the specie richness of indigenous petroleum-degrading bacteria (such as *Massilia* and *Streptomyces*) (Rong et al. 2021). This suggests that biosurfactant addition to polluted marine sediment improves microbial community interaction, enhancing organic pollutants degradation.

Also, Huang et al. (2020) showed that glycolipid with 80 mg/L CMC has high stability over a wide range of temperatures (0–120 °C), pH (4–12), and salinity (0–16%, w/v) could be used in dose-effect to enhance organic pollutant degradation. The microbial growth and activity improved at sub-CMC (40 mg/L concentration of glycolipid) with upregulation of the expression levels of degradation-related genes and effectively promoted the biodegradation of n-alkanes (reduction from 272.21 to 56.93 mg/L) and PAHs (reduction from 61.6 to 16.36 g/L) in 7 d. The results by Huang et al. (2020) suggest that the feasibility of applying biosurfactant at known dose enhances remediation and contributes significantly to the optimization of surfactant-facilitated bioremediation strategies. Therefore, knowing the optimal dose of biosurfactant improve the remediation processes and decreases operational cost.

Table 2 Recent advances in the environmental application of biosurfactant

Biosurfactant application	Biosurfactant	Application strategy	Pollutant	Remarks	References
Direct supplementation	Rhamnolipid	500 mg/kg concentration	Petroleum hydrocarbon	80.24% TPH degradation after 30 d and increased species richness of indigenous petroleum-degrading bacteria	Rong et al. (2021)
	Rhamnolipid	5x CMC concentration	Crude oil	Hydrocarbon removal efficiency of 72.50 ± 0.11%	da Silva et al. (2021)
	Glycolipid	Sub-CMC concentration (40 mg/L)	Hydraulic fracturing flow back of petroleum hydrocarbon	Upregulation of degradation-related genes and effective biodegradation of n-alkanes from 272.21 to 56.93 mg/L and PAHs from 61.6 to 16.36 g/L in 7 d	Huang et al. (2020)
	Rhamnolipid	50 mg/L CMC concentration	PAHs	Enhanced the degradation of PAHs from 35.3% to 54.4% within 7 d	Ammami et al. (2015)
	Rhamnolipid	10 mg/L of purified rhamnolipid	Crude oil	Enhanced degradation of TPH from 63.4% to 82.1%	Antoniou et al. (2015)
Supplementation of biosurfactant-producing microbes	Lipopeptide	Biosurfactant-producing *Enterobacter hormaechei* with high biocatalysts	Crude oil	85% petroleum hydrocarbons degradation within 10 d following a pseudo-second-order kinetics with rate constant k^2 0.2775 and R^2 0.9923	Muneeswari et al. (2022)
	Glycolipopepetide	Biosurfactant-producing *Aeromonas hydrophila* RP1	Diesel and n-hexadecane	77.33% and 55.98% percentage degradation efficiency of diesel and n-hexadecane after 7 d	Pandey et al. (2022)
	Lipopeptide	Biosurfactant-producing halotolerant marine bacterium *Bacillus licheniformis* LRK1	Engine oil	Degrades 24.23% of engine oil after 21-d at neutral pH, 35 ± 2 °C temperature and 3% w/v NaCl concentration	Nayak et al. (2020)

	Lipopeptide	Indigenous biosurfactant-producing bacteria *Acinetobacter* sp. Y2	Hydraulic fracturing flowback of petroleum hydrocarbon	Increased microbial activity and growth. Significant decrease in chemical oxygen demand from 6646.7 mg/L to 1546.7 mg/L. Degradation of n-alkanes from 2635.4 mg/L to 159.7 mg/L and PAHs from 918.6 μg/L to 209.6 μg/L in 7 d	Zhou et al. (2020)
	Rhamnolipid	Biosurfactant-producing *Paracoccus* sp. MJ9	Diesel oil	81% degradation efficiency within 5 d	Xu et al. (2020)
	Lipopeptide	Biosurfactant-producing *Bacillus cereus* UCP 1615	Motor oil	Organic pollutant degradation with little or no detrimental effect on fish *Poecilia vivipara* (higher than 90% survival rate)	Durval et al. (2020)
	Lipopeptide	Biosurfactant-producing halotolerant marine bacterium *Bacillus licheniformis* LRK1	Engine oil	Degrades 24.23% of engine oil after 21-d at neutral pH, 35 ± 2 °C temperature and 3% w/v NaCl concentration	Nayak et al. (2020)
	Lipopeptide	Biosurfactant-producing marine bacterium, *Bacillus stratosphericus* FLU5	Petroleum hydrocarbon	Hydrocarbonoclastic effect with 50 mg/L CMC value and 28 mN/m ST	Hentati et al. (2019)
Immobilization of biosurfactant or its producers	ND	Zeolite immobilization	Crude oil	80.67% crude oil bioremediation efficiency within 21 d compared to bio-stimulation and natural attenuation	Laothamteep et al. (2022)
	ND	Immobilization on novel carrier material from coated puffed rhubarb rice with calcium alginate membrane	Diesel oil	The immobilized consortium exhibited floatability, and slow nutrient release properties while degrading 86% of diesel oil	Luo et al. (2022)
	Phospholipid	Biochar immobilization prepared from corn straw biomass	Diesel oil	Upregulation of degradation-related genes (alkB and CYP450)	Zhou et al. (2021)

(continued)

Table 2 (continued)

Biosurfactant application	Biosurfactant	Application strategy	Pollutant	Remarks	References
				and 94.7% effective removal of diesel oil corresponding to 169.2 mg to 8.91 mg in 7 d compared to the 54.4% degradation by free-cell culture of LQ2	
	Lipopeptide	Immobilization on functionalized nanoporous activated bio-carbon	Petroleum hydrocarbon	Bioremediation of municipal landfill leachate through lignin biosequestration mechanism	Uddin et al. (2021)
	Lipopeptide	Immobilization on activated functionalized carbon (AFC) matrix prepared from rice husk	Petroleum hydrocarbon	61.8% TPH reduction and improving seed germination of cowpea *Vigna unguiculate* after 28 d of treatment	Christopher et al. (2021)
	ND	Size-optimized coconut fibers cellulosic bio-carrier	n-hexadecane	Increased microbial biomass in contact with the pollutant and the degradation rate enhanced to 95.7% in solid phase of soil at 27 °C after 60 d	Hajieghrari and Hejazi (2020)
Nanotechnological practices	ND	Biosurfactant and ZnO nanoparticles	Benzo[a]pyrene	Degradation kinetics was a first-order reaction with a maximum rate of 82.67 ± 0.01 (%) at 130 rpm, 30 °C, pH 7.0 after 6 d	Mandal et al. (2018)
	Rhamnolipid	Biosurfactant and nanoparticles (α-Fe_2O_3 and $Zn_5(OH)_8C_{12}$)	Crude oil	Improved degradation rate with 80% emulsification index and 36 mN/m ST	El-Sheshtawy et al. (2014)
	Rhamnolipid	Biosurfactant layered double hydroxides	Naphthalene	Removal of hydrophobic organic pollutants with 1.3 efficiency higher than SDS, a synthetic surfactant	Chuang et al. (2010)

ND not detected

4.2 Supplementation of Biosurfactant-Producing Microbes and Synergistic Effect

Biosurfactant-producing microorganisms are widely used in organic pollutant remediation either as mono- or mixed culture in synergistic interactions with non-producers have been extensively detailed in previous studies (Feng et al. 2021; Shuai et al. 2019; Xu et al. 2020). The biosurfactants are secreted either intercellularly or extracellularly in the form of biofilm, which interacts with an interface and alters the surface features such as wettability and other properties.

Indigenous biosurfactant-producing bacteria *Acinetobacter* sp. Y2 was reported to increase microbial activity and growth while significantly ($P < 0.05$) removing chemical oxygen demand (from 6646.7 mg/L to 1546.7 mg/L) and increasing the degradation rate of n-alkanes (from 2635.4 mg/L to 159.7 mg/L) and PAHs (from 918.6 μg/L to 209.6 μg/L) in 7 d (Zhou et al. 2020). Lipopeptide-producing halotolerant marine bacterium *B. licheniformis* LRK1 degrades 24.23% of engine oil after 21 d of incubation at neutral pH, 35 ± 2 °C temperature, and 3% w/v NaCl concentration (Nayak et al. 2020). The lipopeptide had a 70% Emulsification Index (EI24) and 31.43% ST reduction (Nayak et al. 2020). Similarly, Hentati et al. (2019) showed that lipopeptide-producing marine bacterium *B. stratosphericus* FLU5 is hydrocarbonoclastic and, at 50 mg/L CMC value reduces the ST of water from 72 to 28 mN/m. In Jiaozhou Bay, China, *Paracoccus* sp. MJ9 can produce rhamnolipid that enhances the bioavailability of recalcitrant hydrocarbon for easy degradation of diesel oil (81%) by microorganisms within 5 d (Xu et al. 2020).

Recently, Muneeswari et al. (2022) revealed that the successful remediation of marine crude oil spills depends on biosurfactant production and biocatalysts by the native hydrocarbon-degrading microbes. The report showed that halotolerant bacterium, *Enterobacter hormaechei,* capable of producing anionic, high molecular weight (48 kDa) lipopeptide can reduce ST of water to 35 mN/m, with an emulsification index of 46.34%, and biocatalysts of extracellular enzymes [lipase (160 U/mL) and laccase (38 U/mL)] and intracellular enzymes [alkane hydroxylase (48 U/mL), alcohol dehydrogenase (86 U/mL), and esterase (102 U/mL)]. The petroleum hydrocarbon degradation was 85% within 10 d following a pseudo-second-order kinetics with rate constant k^2 0.2775 and R^2 0.9923. This suggests that aside from biosurfactant production, the synergetic activity of the biocatalysts secreted by *E. hormaechei* enhances bioremediation.

Pandey et al. (2022) reported that biosurfactant-mediated biodegradation of 77.33% of diesel and 55.98% of n-hexadecane is greater than 26.68% of n-hexadecane and 48.36% of diesel degradation by *Aeromonas hydrophila* RP1 alone after 7 d. The biosurfactant used was glycolipopepetide consisting of pentadecanoic and octadecenoic fatty acids and having an ST reduction value of 27.4 mN/m at 123 mg/L CMC concentration. The glycolipopepetide displayed high stability over several environmental conditions (4–100 °C temperature, 2–10 pH, and 5–150 g/L NaCl concentration) with a greater than 50% emulsification index against hydrocarbons like aviation turbine fuel, n-hexadecane, hexane, paraffin oil, and xylene.

Durval et al. (2020) reported using *B. cereus* UCP 1615 biosurfactant "lipopeptide" to stimulate the growth of autochthonous microorganisms independently of the presence of motor oil in the marine environment. The nutrient sources for the anionic lipopeptide production were 2% frying oil and 0.12% peptone. The toxicity assay revealed that aside from the lipopeptide being a good candidate for remediating polluted marine environment through enhancing pollutant solubilization and mobilization, the biosurfactant had little or no detrimental effect on fish *Poecilia vivipara* (higher than 90% survival rate).

4.3 Immobilization of Biosurfactant or Biosurfactant Producers

In nature, the violent fluctuation and complex constituents of the marine environment significantly influence the activity of free microbial cells while reducing pollutant degradability (Zhou et al. 2021). As such, immobilization of microbes using inert carriers is practiced to stabilize the interaction between the degraders and pollutants by minimizing biomass loss even under adverse environmental conditions (Hajieghrari and Hejazi 2020). Thus, ensuring biosurfactant sustainability during remediation reduces the regeneration cost, increasing cell density and reusability associated with immobilization technology (Luo et al. 2022). Immobilized biosurfactants, especially the producers, promote pollutant degradation rate with stability in varying environmental conditions. Commonly used immobilization substances include zeolites, carbonaceous materials, alginate, biosilica, and biochar, following methods like entrapment, encapsulation, adhesion/adsorption, covalent bonding, cross-linking, and combined techniques (Lapponi et al. 2022). The immobilization substances can increase cell biomass, substrate–microbe interaction, and pollutant absorption to facilitate bioremediation. Biochar is cost-effective, readily available, and has an excellent physiochemical substance with large surface area/porosity. Waste materials in immobilization matrix formation (such as biochar) enhance renewable resource recycling and reduce pollution.

Recently, Zhou et al. (2021) reported the enhanced bioremediation of diesel oil-contaminated seawater by cell immobilization technology. The biochar used was corn straw biomass, and the cell organism was a phospholipid-producing marine *Vibrio* sp. LQ2 isolated from cold-seep sediment. The biochar-immobilized phospholipid producer removed 94.7% of diesel oil, corresponding to 169.2 mg–8.91 mg in 7 d, compared to the 54.4% degradation percentage by the free-cell culture of LQ2. Besides the improvement in LQ2 biomass and activity in the immobilized matrix, the degradation-related genes (alkB and CYP450) also increased by 3.8 and 15.2 folds to the cell-free LQ2. The findings by Zhou et al. (2021) indicate that using immobilized biosurfactant-producing microbes in treating organic pollutants in the marine environment can be feasible. Previously, Hajieghrari and Hejazi (2020) showed that immobilized biosurfactant-producing *P. aeruginosa* on size-optimized coconut fibers, a cellulosic bio-carrier, enhances biodegradation n-hexadecane (95.7%) in solid phase of soil at 27 °C after 60 d. The study revealed that optimizing the size of fibers used for immobilization influences the amount of microbial biomass in contact with the pollutant and the degradation rate.

Christopher et al. (2021) showed that lipopeptide biosurfactant (5.0 g) from *Bacillus Malacitensis* could be immobilized on Activated Functionalized Carbon (AFC) matrix (30.0 g) prepared from rice husk at 80 rpm agitation, room temperature, and pH 7 in 30 min. The immobilization process increased the amino acids (71.22% polar and 28.78% hydrophobic) content of the lipopeptide, thereby significantly enhancing the removal of recalcitrant organic pollutants through increased hydrophobicity, solubilization, micelle formation, and pollutant adsorption. The TPH (2642.5 ± 131 mg/kg) of the contaminated sediment (50 cm depth) from an industrial area used in the study was reduced by 61.8% while also improving the seed germination of cowpea *Vigna unguiculate* after 28 d of treatment. Therefore, suggesting that toxic organic pollutants affect plant metabolic rate as the soil property and microbial community are negatively impacted. Following an advanced approach, the lignin biosequestration mechanism by immobilized cationic lipoprotein biosurfactant was proposed for the bioremediation of municipal landfill leachate (Uddin et al. 2021). Optimal bioremediation was achieved using functionalized nanoporous activated bio-carbon as the immobilization material.

Studies have demonstrated that petroleum hydrocarbon consortiums containing biosurfactant producers can be immobilized to improve their functionality during bioremediation. In enhancing the bioremediation of crude oil-polluted marine sandy soil microcosms, Laothamteep et al. (2022) showed biosurfactant-producing *Mycolicibacterium* sp. PO2 can be zeolite-immobilized with other PAH degraders. It was revealed that the synergistic interactions between the bacteria strains increased the biodegradation of recalcitrant high molecular weight hydrocarbon since their genomes harbor degradative genes that allow for a meta-cleavage pathway. The zeolite-immobilized consortium has a broad activity range of pH (5.0–9.0), temperatures (30–40 °C), and salinities (20–60‰), highly increasing the indigenous hydrocarbon-degrading bacteria. The bioremediation efficiency of the crude oil (10,000 mg/kg) contaminated sandy soil was 80.67% within 21 d compared to bio-stimulation and natural attenuation. Similarly, Luo et al. (2022) reported that biosurfactant-producing *Gordonia* sp., in addition to other bacteria strains in a consortium, can be immobilized with a novel carrier material from coated puffed rhubarb rice (PRR) with calcium alginate (CA) membrane. The immobilized consortium exhibited floattability and slow nutrient release properties while degrading 86% of diesel oil.

4.4 Nanotechnological Practices

Nanomaterials are sized from 1 to 100 nm with a large surface area-to-volume ratio spatial confinement, possessing optical, magnetic, catalytic, and electronic properties (Pete et al. 2021). These properties make nanoparticles (either organic or inorganic) have good adsorbing characteristics that increase their potent preparation with biosurfactants to form biocompatible conjugates with synergistic interaction to improve organic pollutant remediation (Biswas et al. 2019). Therefore, nano-biosurfactants are a promising green technology material due to their surface activity, aggregative, stabilization, ion exchange, affinity, adsorption, and molecular sieving properties.

Biosurfactant addition to nanoparticles increases the electronegativity between the materials and their interaction, thereby acting as a dispersant, foaming agent, and flocculant to favor the removal of organic pollutants (Kumari and Singh 2016). The first report on the statistical optimization of benzo[a]pyrene degradation by response surface methodology using yeast consortium in the presence of 2 g/L ZnO nanoparticles and 3% biosurfactant was reported by Mandal et al. (2018). The pollutant degradation followed a first-order reaction with a maximum 82.67 ± 0.01 (%) rate at 130 rpm, 30 °C, pH 7.0 after 6 d. El-Sheshtawy et al. (2014) showed that maximum bioremediation of crude oil pollution at Gemsa Bay could be achieved with the use of biosurfactants from *Pseudomonas* species and nanoparticles (α-Fe_2O_3 and $Zn_5(OH)_8C_{12}$) after 7 d with an emulsification index of 80% and 36 mN/m ST. The application of nano-biosurfactant in pollutant remediation from the environment depends on their classification (high- or low molecular weight surface-active substance) and chemical (anionic, cationic, or neutral) nature (Kumari and Singh 2016; Płaza et al. 2014). The commonly synthesized nano-biosurfactants are metallic nanoparticles using rhamnolipids, lipopeptides, and sophorolipids through water in oil microemulsion, reverse micelle, and borohydrate reduction methods (Pete et al. 2021; Płaza et al. 2014).

Chuang et al. (2010) reported that rhamnolipid produced from *P. aeruginosa* could synthesize biosurfactant layered double hydroxides (LDHs) to remove hydrophobic organic pollutants such as naphthalene. The removal efficiency was 1.3 times higher than when SDS, a synthetic surfactant, was used, suggesting that rhamnolipid-LDH has a good adsorption capability. Biosurfactant nanotechnological practices employed for remediation have great potential, but some shortcomings like the high cost of biosurfactant production and expensive downstream processing need to be addressed.

5 Role of Biosurfactants in Marine Sediment Remediation

One of the problems with remediating contaminated marine sediments with organic hydrocarbons is their absorption into the sediment matrix, low solubility into the aqueous phase, and limited bioavailability for degradation (Dell'Anno et al. 2018). Therefore, several efforts have been taken to incorporate biosurfactants and their producers in the remediation of organic pollutants from marine sediments, as illustrated in Table 3.

5.1 *Biosurfactant-Mediated Microbial Remediation*

This concept entails enhancing the bioavailability of organic pollutants for easy accessibility by microbes. As more petroleum reservoirs are discovered and explored, oil spill into the marine environment is inevitable. These oil spills cause many severe consequences, such as heavily impacting the marine planktonic ecosystem (organisms unable to swim against water currents), bottom-dwellers, and other higher tropic organisms in the marine sediments. Bioremediation is up to date widely accepted in the remediation of contaminated marine sediments as a more sustainable approach. However, the integration of biosurfactants and bioremediation has been a significant advancement in the biological remediation section.

Feng et al. (2021) demonstrated sophorolipid-assisted bioremediation of petroleum hydrocarbon-contaminated soil with an isolated indigenous bacterial consortium. The biodegradation efficiency of TPH increased from 12.2% in the contaminated soil (control) to 44.5% and 57.7% in the isolated consortium and isolated consortium +1.5 g sophorolipid per kg dry soil, respectively. The half-life of TPH degradation also decreased from 32.5 d to 20.4 d in the treatment having sophorolipid compared with only the bacteria consortium. The sophorolipid mechanism of action includes TPH desorption from the solid matrix to the aqueous solution, increased solubilization, and improved hydrocarbon bioavailability. The stimulated microbial growth and activity observed suggest that sophorolipid also served as carbon for the bacterial community co-metabolism in the degradation system.

Although bioremediation studies on varying sediment types, including marine stones with porous spacing, are scarce, da Silva et al. (2021) recently described the removal of hydrophobic contaminant adsorbed in marine stones. The stones resulting from wave fragmentation of coral reefs had an average pore size between 230 μm and 520 μm and a porosity of 72.0%. The biosurfactant was prepared at different concentrations of ½xCMC, CMC, 2xCMC, and 5xCMC; however, the highest hydrocarbon removal efficiency (72.50%) was observed in treatment with 5xCMC (3000 mg/L). This suggests that hydrocarbon mobilization on porous sediment surfaces increases with increasing biosurfactant concentration. Although the changes in hydrocarbon removal efficiency from CMC to 5xCMC are not apparent (71.30 ± 0.19 to 72.50 ± 0.11%), the biosurfactant enhanced the oil viscosity reduction and formation of an oil-in-water emulsion.

Das and Kumar (2018) studied an indigenous glycolipid-producing *Pseudomonas azotoformans* AJ15 strain for remediation of petroleum-contaminated soil under hypersaline conditions. The biosurfactant substrate was agro-industrial wastes of bagasse and potato peels produced under submerged fermentation. The glycolipid class was identified as rhamnolipid following several chemical analyses, and the product had high stability against environmental stress (90 °C, 6% NaCl concentration, and varying pH). It was observed that the rhamnolipid effectively enhanced the removal of about 36.56% of trapped petroleum hydrocarbon in soil matrix under saline conditions.

In isolating indigenous microbes from Taean beach sediment, Lee et al. (2018) showed that employing biosurfactant-producing and hydrocarbon-utilizing indigenous bacteria enhances the effectiveness of crude oil bioremediation. The hydrocarbon bioavailability was increased by the biosurfactant-producing bacteria in the genus *Bacillus*, *Rhodococcus*, *Isoptericola*, and *Pseudoalteromonas* during the degradation. The biosurfactant produced was rhamnolipid with a reduced ST of 33.9–41.3 mN/m, high oil spreading (1.2–2.4 cm), and hydrocarbon emulsification (up to 65%), justifying the hydrocarbon degradation performance observed in the marine sediment tested.

5.2 *Biosurfactant-Mediated Phytoremediation*

Phytoremediation is an eco-friendly approach for repairing and restoring contaminated lands, and with recent advances in the biotechnology field, its application potential is widening and has opened up new possibilities in the reclamation of the degraded sediments in marine ecosystems (Sonowal et al. 2022). This green technological approach uses site adaptive or endemic plants (Lal et al. 2018) to stabilize, extract, accumulate, degrade, or transform organic pollutants in marine sediments into less toxic molecules (Sonowal et al. 2022). While several promising assisted phytoremediation methods such as genetic engineering, nanoparticle-assisted, microbial-assisted, and electrokinetic-assisted approaches are gaining increasing attention (Yan et al. 2020), studies on the incorporation of biosurfactants are limited. As such, we focus on revealing biosurfactant-assisted phytoremediation considering the limitations highlighted by Moradi et al. (2021).

Moradi et al. (2021) reported the physiological responses and phytoremediation capability of *Avicennia marina* to PAHs contamination sediment in the vulnerable coastal ecosystems of the Persian Gulf area. The *A. marina* phytoremediation mechanism involved allocating more biomass to the root than shoot regions and activating the antioxidative enzymatic/non-enzymatic reactions (activities of peroxidase, ascorbate peroxidase, and polyphenol oxidase). The decreasing pattern of PAHs in the polluted sediments with *A. marina* rhizosphere was 37 ± 0.4, 21.84 ± 0.27, 12.78 ± 0.11, and 14.74 ± 0.03%, corresponding to 2.5, 5.0, 7.5,

Table 3 Role of biosurfactants in marine sediment remediation

Biosurfactant-assisted remediation	Organic pollutants	Soil/sediment	Biosurfactant used	Biosurfactant mechanism of action	Observed effects	References
Bioremediation	Petroleum hydrocarbon	Contaminated soils (5–15 cm depth) were obtained from an abandoned plant located at the Yangtze River Delta, China	Sophorolipid	TPH desorption from solid matrix to the aqueous solution, increased solubilization, and improved hydrocarbon bioavailability. Stimulated microbial growth, activity, and co-metabolism of the bacterial community	Decreased TPH biodegradation half-life and increased removal efficiency from 12.2% in the contaminated soil (control) to 57.7% in isolated consortium +1.5 g sophorolipid per kg dry soil	Feng et al. (2021)
	Crude oil	Marine stones were collected at Suape beach, Ipojuca city—Pernambuco, close to the coral reefs	Rhamnolipid	Enhanced oil viscosity reduction and formation of an oil-in-water emulsion	Hydrocarbon removal efficiency of 71.30 ± 0.19 to 72.50 ± 0.11% from CMC to 5xCMC	da Silva et al. (2021)
	Crude oil and PAHs	Heavily contaminated Taean beach sediment on coast of Taean, Korea	Rhamnolipid	ST reduction (33.9–41.3 mN/m), high oil spreading, and hydrocarbon emulsification (up to 65%)	Hydrocarbon degradation in the marine sediment	Lee et al. (2018)
Phytoremediation	Petroleum hydrocarbons	Soil samples contaminated with oily	Rhamnolipid	No significant effect ($p < 0.05$)	Over a 90-d, 4 mg/kg rhamnolipid	Liduino et al. (2018)

(continued)

Table 3 (continued)

Biosurfactant-assisted remediation	Organic pollutants	Soil/sediment	Biosurfactant used	Biosurfactant mechanism of action	Observed effects	References
		residues were collected from petrochemical facility in Sao Paulo State, Brazil		on the structure of the dominant bacterial community in the contaminated industrial soil sample analyzed	results in 58% TPH and 48% PAH concentrations reduction. The metal removal percentage was Ni (41%), Cr (30%), Pb (29%), and Zn (20%)	
	Dichlorodiphenyltrichloroethane (DDT)	The DDT-contaminated soil was collected from a long-term cotton field in Henan province, China	Direct use of biosurfactant-producing *Pseudomonas* sp. SB	Increased pollutant bioavailability and microbial community	The removal efficiency of tall fescue in treatment containing biosurfactant-producing *Pseudomonas* sp. SB increased from 40.3 to 59.4%	Wang et al. (2017)
	Crude oil	Spiked soil (5000 mg/kg, TPH) collected from the upper layer (0–20 cm) of a farm in Guangzhou higher education mega center	Rhamnolipid and soybean lecithin	Enhanced the soil microbial population, which correlates with the crude oil degradation efficiency	The saturated hydrocarbons were greatly reduced while the recalcitrant PAH degradation was enhanced in treatment having rhamnolipid in the	Liao et al. (2016)

					plant root compared to the leaf	
	Gasoline	Spiked soil	Biosurfactant from *Serratia marcescens*	Increased bacterial richness at the root surface and stimulated bio-decomposition of pollutants through enhanced rhizodegradation. The biosurfactant also acts as biocatalysts to increase the performance of phytoremediation by *Ludwigia octovalvis*	The biosurfactant removed a significant amount (up to 93.5%) of TPH compared to other additives that removed only 85.4% (*Serratia marcescens*), 70.3% (*Serratia marcescens* culture supernatant) and 86.3% (sodium dodecyl sulfate)	Almansoory et al. (2015)
Bio-electrokinetic remediation	Diesel oil	Spiked soil located in Serkkadu (Tamil Nadu, India)	Rhamnolipid from *Staphylococcus epidermidis* EVR4	Synergistic role of biosurfactant and catabolic enzymes	With a maximum degradation efficiency of 96% within 4 d	Vaishnavi et al. (2021)
	Petroleum hydrocarbon	Spiked soil from Pretoria (South Africa)	Rhamnolipid (7 mono-rhamnolipid and 4 di-rhamnolipid homologues) from *P. aeruginosa*	Biosurfactant and electrode with different currents.	Microbial survival and biosurfactant yield decreased 36.25 ± 3.75 mg/mL, 22.5 ± 5 mg/mL, 6.25 ± 1.25 with increasing currents of 0.5 A, 1 A, and 1.5 A	Gidudu and Chirwa (2021)

(continued)

Table 3 (continued)

Biosurfactant-assisted remediation	Organic pollutants	Soil/sediment	Biosurfactant used	Biosurfactant mechanism of action	Observed effects	References
	Crude oil	Spiked soil collected from Thiruvalluvar university campus, Serkadu, Vellore district (India)	Lipopeptide	Improved solubilization. Increased pollutant bioavailability and microbial accessibility of hydrocarbons as carbon and energy source. The microbes were *B. subtilis* AS2, *B. licheniformis* AS3, and *B. velezensis* AS4	Degraded higher molecular weight hydrocarbons (C8–C28) with about 92% crude oil which is more than the conventional electrokinetic method (60%) in 2 d operation	Prakash et al. (2021)
	Engine oil	Spiked soil from Pretoria (South Africa)	Rhamnolipid from *P. aeruginosa* PA1	Biosurfactant and lower electrode spacings enhanced desorption and demulsification	Oil recovery was improved to 75.15% in 240 h	Gidudu and Chirwa (2020)
	PAHs	Polluted dredged sediment was collected from the disposal site of a French harbor in Normandy	Rhamnolipid viscosin-like biosurfactant from *P. fluorescens* PfA7B	Biosurfactants and periodic voltage gradient	Removal of 16 PAHs	Ammami et al. (2015)

and 10% crude oil contamination, respectively. The highest percentage rate of PAH fraction removal was observed for fluoranthene (71.18 ± 0.56) in 2.5% crude oil contamination sediment and anthracene (69.45 ± 6.33, 55.66 ± 4.38, and 35.97 ± 0.22) in 5.0, 7.5 and 10% crude oil-contaminated sediments. The findings by Moradi et al. (2021) indicate that *A. marina* is an excellent phytoremediation candidate for small-scale oil pollution and can only remove some PAH fractions from contaminated marine sediments. Therefore, incorporating biosurfactant treatment into the studies could have enhanced the activity of *A. marina* in removing more PAHs fractions easily.

The previous studies on phytoremediation are more focused on addressing the removal of heavy metals. However, Liduino et al. (2018) reported the multi-decontamination of organic hydrocarbon pollutants and heavy metals through biosurfactant-assisted phytoremediation. The study revealed the mixed functionality of commercial biosurfactant (rhamnolipid) and sunflower (*Helianthus annuus* L.) cultivation. Over a 90-d examination, the best phytoremediation efficiency occurred in the treatment with 4 mg/kg rhamnolipid, reducing 58% TPH and 48% PAH concentrations. The metal removal percentage was Ni (41%), Cr (30%), Pb (29%), and Zn (20%). Nevertheless, rhamnolipid addition and phytoremediation activity of *Helianthus annuus* L. had no significant effect ($p < 0.05$) on the structure of the dominant bacterial community. Still, increased pollutant degradation in the soil samples analyzed.

Wang et al. (2017) revealed that biosurfactant-producing microorganisms could promote the phytoremediation of organic pollutants in soil media. The biosurfactants produced by *Pseudomonas* sp. SB increased the bioavailability of organic pollutants and enhanced their microbial degradation. In synergetic response, the plants (tall fescue and perennial ryegrass) improved the rhizosphere environment for *Pseudomonas* sp. SB proliferation, thus promoting an integrated phyto- and bioremediation system. The removal efficiency of the different treatments for the pot experiment conducted: T0 = fertilizer (control), T1 = fertilizer + tall fescue, T2 = fertilizer + tall fescue + *Pseudomonas* sp. SB, T3 = fertilizer + perennial ryegrass, and T4 = fertilizer + perennial ryegrass + *Pseudomonas* sp. SB were 40.3, 59.4, 65.6, 69.0, and 65.9%. The result observed suggests that while biosurfactant addition has a significant effect on pollutant removal, the plant type influenced the rate of remediation and soil bacterial community, with Proteobacteria, Acidobacteria, and Actinobacteria being the three most dominant phyla in all groups.

Liao et al. (2016) demonstrated that biosurfactant-enhanced phytoremediation of organic pollutants as a green technology for treating contaminated soil. Microbially synthesized (rhamnolipid and soybean lecithin) and chemically synthesized (Tween 80) surfactants were employed in the phytoremediation of crude oil-contaminated soil using maize (*Zea mays*. L). It was observed that the addition of surfactants inhibited the chlorophyll fluorescence of the maize leaf but had no significant effect on the maize biomass production at $p < 0.05$. Compared to Tween 80, rhamnolipid and soybean lecithin enhanced the soil microbial population, which correlates with the crude oil degradation efficiency observed. Among the crude oil constituents, the saturated hydrocarbons were greatly reduced (100%), while the recalcitrant PAH degradation occurred more at the plant root region than the leaf.

Almansoory et al. (2015) showed the potential application of a biosurfactant extracted from hydrocarbon-degrading bacteria *Serratia marcescens* in improving phytoremediation technology using *Ludwigia octovalvis*. Biosurfactant addition (10%) resulted in up to 93.5% TPH removal compared to the other treatments that removed only 85.4% (*S. marcescens*), 70.3% (*S. marcescens* culture supernatant), and 86.3% (synthetic surfactant). The biosurfactant-assisted phytoremediation of the gasoline-contaminated soil was pseudo-second-order kinetics with 0.9318 coefficient of determination (R^2) and 0.0032 second-order rate constant (k^2) (g TPH/kg plant *d*). It was observed that the biosurfactant increased the bacterial richness at the root surface and stimulated the bio-decomposition of pollutants through enhanced rhizodegradation. Hence, *S. marcescens* secreted biosurfactant is an effective biocatalyst for the phytoremediation of polluted sediment.

5.3 Biosurfactant-Mediated Bio-electrokinetic Remediation

Electrokinetics (EK) is used to remove organic pollutants in solid sediments based on charges using limited direct current (DC) applied to electrodes. The outcome effectively separates the hydrocarbon molecules based on electroosmosis, electromigration, and electrophoresis (Prakash et al. 2021). Electrochemical factors such as the slow bio-oxidation process, low microbial biomass, and pH fluctuations greatly impact the rate of pollutant degradation. However, coupling bioremediation and electrokinetic remediation methods known as the bio-electrokinetic technique are highly advantageous for simultaneously increasing the degradation rates and cell yield in the polluted marine environment (Fdez-Sanromán et al. 2021).

The major reason for integrating bioremediation and electrokinetic remediation is the long degradation period required to clean up contaminated marine sediments compared to the aqueous media (Ammami et al. 2015). This reason can be attributed to several factors such as microbial types, nutrient availability, pollutant composition, and level of environmental parameters. Using electrochemical techniques can increase the temperature inside the soil system, thereby improving nutrient supply and possible biosurfactant production by inherent microbes (Ammami et al. 2015). These processes enhance pollutant solubilization and mobilization (electromigration) through the soil matrix toward the electrode chambers, thus improving biodegradation. Besides temperature increase, the electrokinetic technique using anode and cathode in the same soil compartment can modulate pH (polarity reversal approach to maintain soil acidity or alkalinity) and pollutant concentration (nutrients distribution across the soil), which correlates with the remediation rate (Fdez-Sanromán et al. 2021). These mechanisms can increase marine sediment permeability and functionality of biosurfactant producers.

According to Prakash et al. (2021), bacterial biosurfactant “lipopeptide” is applied in a system containing *B. subtilis* AS2, *B. licheniformis* AS3, and

B. velezensis A enhances the remediation efficiency of crude oil in contaminated soil from 60% to 90% after 2 d. The mechanism of bio-electrokinetic involves increasing the solubility of the organic pollutant, which leads to faster electromigration of hydrocarbon constituents to the anodic compartment, which then confirms the decrease in the total organic content. Similarly, Vaishnavi et al. (2021) show that biosurfactant secretion by *Staphylococcus epidermidis* EVR4 can improve diesel degradation in a bio-electrokinetic remediation system. The remediation efficiency observed was 100% degradation of nonane (C9) to tricosane (C23) hydrocarbons, while pentacosane and octacosane were degraded at 85%, and 47%, respectively. The improvement in the degradation of TPHs 96% (liquid system) and 84% (soil system) was recorded within 4 d. This can be attributed to the synergistic role of biosurfactant and catabolic enzymes (dehydrogenase, catalase, and cytochrome C). Thus, making biosurfactant-assisted bio-electrokinetic remediation a potential method for the in situ removal of organic pollutants in marine sediment.

Gidudu and Chirwa (2020) reported that integrating high voltage, low electrode spacing, and biosurfactants enhances bio-electrokinetic remediation. The experiment was conducted at a varying voltage (30 V and 10 V), electrode spacing (335 mm and 185 mm), and rhamnolipid from *P. aeruginosa* PA1 with ST 30.35 mN/m at 156 mg/L CMC concentration. The result revealed that the bio-electrokinetic remediation run at 30 V and 185 mm with biosurfactant had the highest petroleum hydrocarbon recovery efficiency in the soil system. The high voltage allowed higher electroosmosis and electrophoresis in favor of electron transfer within the soil. Meanwhile, the biosurfactant and 185 mm electrode spacing enhanced the hydrocarbon remediation rate by decreasing energy expenditure and increasing desorption and demulsification. Although the change in voltage had no significant detrimental effect ($p < 0.05$) on the cells except close to the electrode (pH extremes), higher microbial proliferation was recorded in the compartment with 185 mm electrode spacing. When field scale (in situ) environmental conditions were further evaluated, it was observed that microbial survival and biosurfactant yield decreased 36.25 ± 3.75 mg/mL, 22.5 ± 5 mg/mL, 6.25 ± 1.25 of the organic pollutants with increasing currents of 0.5 A, 1 A and 1.5 A (Gidudu and Chirwa 2021).

6 Remediation Evaluation Techniques of Contaminated Marine Sediment

While developing appropriate remediation methods is required to mitigate the possible risk of organic pollutants in the marine environment, determining the suitable extraction and qualification assay is important. According to Zhang et al. (2021), the major criteria for evaluating a sediment remediation technology include the organic pollutant type, duration, residue, costs, safety, technological readiness level, efficacy/monitoring, reliability/maintenance, and preliminary investigations, auto-sustainability, acceptability. The analytical flow for evaluating biosurfactant-mediated remediation of organic pollutants in marine sediments is extensively discussed and illustrated in Fig. 5.

6.1 Organic Pollutant Extraction

Due to the low aqueous solubility of hydrophobic pollutants, they tend to adsorb tightly to the organic matter in soils and sediments, thereby making the pollutants less extractable and difficult to recover if the sample size is small or the contaminants are at trace levels (Ammami et al. 2015; Chen et al. 2021; Dell'Anno et al. 2018; Lee et al. 2018). The regularly used techniques to extract organic pollutants from sediments are soxhlet, ultrasonic, liquid–liquid, and solid-phase extractions, although the need for large solvent volumes, longer period, more purification steps, limited efficiency, and analyte loss limit their application. However, selecting the most efficient and sustainable technique plays a major role in minimizing organic solvent wastage and human exposure. More rapid, simplified, safe, eco-friendly, and cost-effective techniques are employed in modern studies, including supercritical and subcritical fluid extraction, microwave-assisted solvent extraction, plant oil-assisted extraction, and microextraction methods (Kariyawasam et al. 2022).

6.2 Organic Pollutant Quantification

Quantification of the organic hydrocarbons extracted from contaminated marine sediment matrices is essential to understanding the extent of remediation. The most common analytic techniques with high sensitivity and low detection limit are gas chromatography (GC) and high-performance liquid chromatography (HPLC). The coupled detection systems include mass detectors (MS), flame ionization detectors (FID), fluorescence detectors (FLD), diode array detectors (DAD), and ultraviolet detectors (UV). Fourier transforms mass spectrometry (FT-IR) has been reported to reveal non-target hydrocarbons after multidimensional ionization (Kariyawasam et al. 2022).

7 Some Relevant Enterprises Working on the Massive Biosurfactant Production

Some companies, organizations, and research groups working on the massive production of biosurfactants for possible application in sediment remediation of organic pollutants in the environment are listed below:

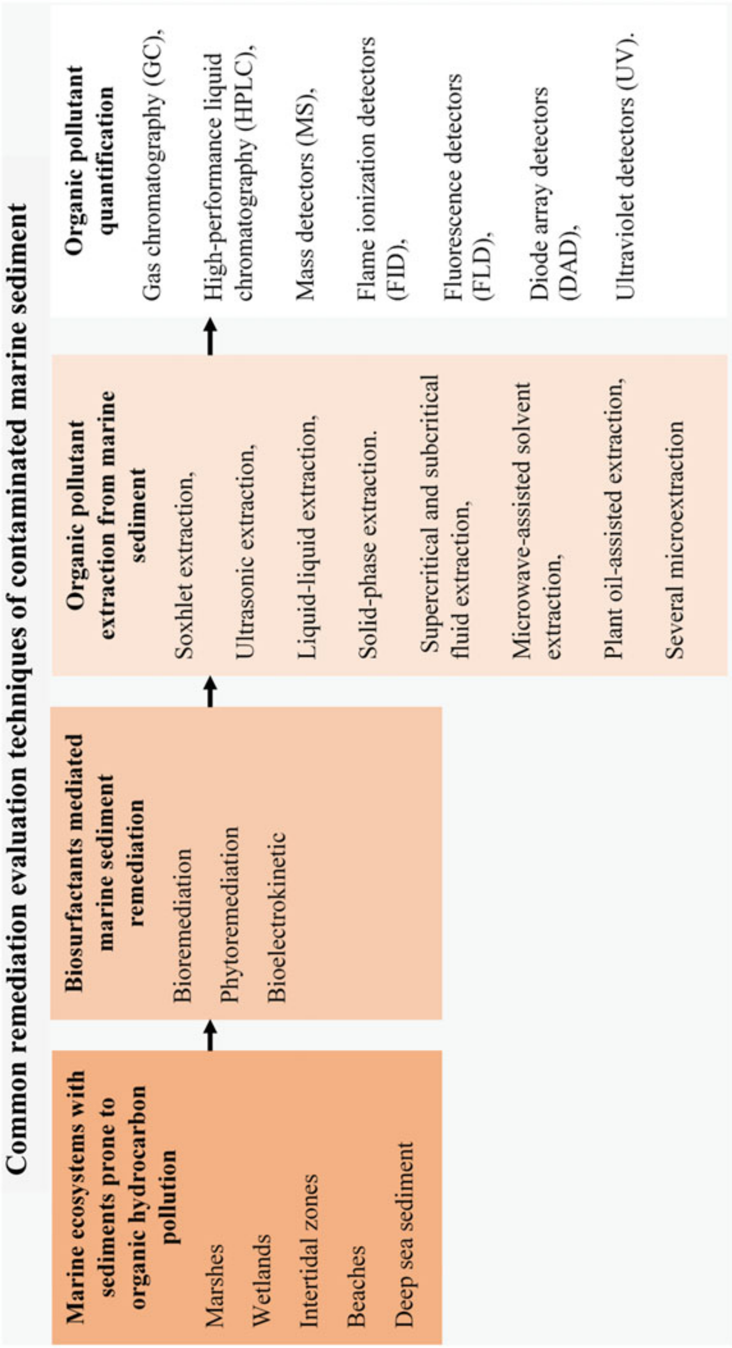

Fig. 5 The analytical flow for evaluating biosurfactant-mediated organic pollutant remediation in marine sediments

(i). Holiferm, a private company located in the United Kingdom and founded in 2018—https://holiferm.com/
(ii). TeeGene Biotech, a private company located in the United Kingdom and founded in 2014—https://www.teegene.co.uk/
(iii). Rhamnolipid, Inc., a company located in the USA, uses artificial intelligence and machine learning to determine rhamnolipid application—https://www.rhamnolipids.com/
(iv). AGAE Technologies, a private company located in the USA and founded in 2010—http://www.agaetech.com
(v). Logos Technologies LLC, a company located in the USA and known for the production of sulfate and phosphate-free biosurfactants—https://www.logostech.net/?s=Biosurfactant
(vi). National Science Foundation, sponsored Columbia University research on biosurfactant production by anaerobes and their cleansing/environmental remediation performance—https://www.nsf.gov/awardsearch/showAward?AWD_ID=0942962&HistoricalAwards=false
(vii). KAM Biotechnology Ltd. is a leading international company located in British Columbia, Canada, and established in 1981. http://www.kambiotechnology.com/
(viii). G&C Ambientpetrol V, Inc. is a site remediation company located in Florida, USA, and is known for producing biodegradable and ecological products. https://gcambientpetrol.com/
(ix). Micro-Bac International, Inc., an environmental biotechnology research company based in the USA. https://www.micro-bac.com/
(x). MCF Environmental Services, Inc., founded in 1989, USA. https://mcfenvironmental.com/
(xi). Hull's Environmental Services, Inc. was founded in 1983. https://www.hullsenvironmental.com/services/environmental-remediation-services/
(xii). Professor Zhang Chunfang research group, Microbiology laboratory, Institute of Marine Biology and Pharmacology, Ocean College, Zhejiang University, Zhejiang Province, China. https://person.zju.edu.cn/en/zhang_cf#673570

8 Future Prospects

Future research should be devoted to:

- Understand the trophic transfer and impacts on human health of different petroleum hydrocarbons accumulated in marine sediments.
- Investigate the mechanism of pollutants solubilization by biosurfactants is required to enable model predictions as information regarding biosurfactant-pollutant interactions based on their structure, texture, complexity, and geochemical characteristics is scarce.

- Optimize factors that influence biosurfactant-mediated remediation in marine sediments focusing on site characteristics, surrounding environment parameters (oxygen availability, temperature, pH, and salinity), indigenous microorganisms, hydrocarbon components, and remediation cost. This will allow for a comparative analysis of different biosurfactants for assisting pollutant remediation and the scaling-up of this practice for in situ applications.
- Incorporate new micro-nano methods for applying biosurfactant in a biosurfactant-mediated remediation system while developing on the existing methods like immobilization and use of bubbles. Also, culture-independent techniques need to be developed to reduce the cost of isolating and screening biosurfactant producers.
- Develop monitoring tools and technologies to study biosurfactant-mediated remediation's efficacy in marine sediments and understand the persistence of petroleum hydrocarbon. These techniques need to be advanced, smart, sustainable, cost-effective, and energy efficient.
- Determine the effect of different sediment texture types on biosurfactant-assisted remediation. Sediment properties are often not considered during experimental design. However, the knowledge can create a link and cross-talk between remediation method, sediment texture, biosurfactant application, and microbial community structure/diversity.

It is known that intense human activities around and within marine ecosystems lead to serious pollutant accumulation in sediments; therefore, in addition to biosurfactant-assisted remediation as a sustainable approach, governmental interventions, and policy implementation is required.

9 Conclusion

Several technologies have emerged over the years for petroleum hydrocarbon remediation in marine sediments to transform these pollutants into less and non-toxic forms at a minimum environmental cost. As cost-effective remediation technologies are being developed at a slow pace, biosurfactant-assisted systems are being employed to enhance remediation performance in marine sediment reclamation. Thus, this chapter reveals the sustainable strategies for applying biosurfactants in organic pollutant remediation of marine environments and the possibility for improvement.

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