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REVIEW ARTICLE

# Review of the application of surfactants in microemulsion systems for remediation of petroleum contaminated soil and sediments

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**Abstract**
Microemulsions are important for soil and sediment remediation technology. The characteristics of the surfactants that make up these microemulsions include low sorption into soil or sediments, low surface and interfacial tension, the ability to penetrate tiny pores, and good solubilization of contaminants. This review revealed that microemulsions formulated with nonionic and anionic surfactants have higher recovery efficiencies for hydrophobic contaminants than cationic ones, as evidenced by the surveyed studies reporting effective remediation of soils and sediments using on microemulsions. These microemulsified systems have been found to remove petroleum and its derivatives from soil or sediments at percentages ranging from 40 to 100%. As such, this review can aid with the choice of surfactants used in microemulsions for remediation, such as those with plant-based components, which are promising solutions for the remediation of contaminated soils due to their contaminant extraction efficiency and biodegradability.

**Keywords** Microemulsions · Removal of organic contaminants · Soil and sediment washing · Anionic and nonionic surfactants · Remediation · Petroleum

## Introduction

As optimized surfactant systems, microemulsions can be considered a chemical tool for the remediation of oil-contaminated environments, including soil and sediments (Yuan et al. 2020). Their use requires an understanding of the surface contamination process, which in turn relies on knowledge of the contaminant substance and the nature of the clean and contaminated surfaces.

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Microemulsions are thermodynamically stable, optically transparent, isotropic dispersions that contain oil, water, and surfactants (Schulman et al. 1959). A substance called a co-surfactant may also be present. Soils or sediments contaminated with oil can be decontaminated by mixing them with microemulsions formulated using biodegradable components (Haegel et al. 1999).

In this review, we considered soils and sediments as different entities. According to Schiffer (1987), sediments are a collection of mineral particles that can undergo the action of time at their source site and can be re-deposited, while soil is made up of mineral and organic materials characterized by physically and chemically altered deposits in situ. As such, the washing of soils and sediments can take different forms, primarily dependent on how the microemulsion in these surfaces is classified.

The remediation processes in soils and sediments differ, and it is essential to take into account the vegetal origin, biodegradability, and low foam generation of surfactants (Yuan et al. 2020), since the choice of an inappropriate surfactant can lead to serious environmental problems. However, the use of surfactants is widely recognized as mandatory to achieve adequate remediation yields (Badr et al. 2004) due to its strong adsorption into sediments and the low aqueous solubility of petroleum, which is characterized as a component of nonpolar composition.

According to Ritoré et al. (2022), soils with higher clay content have lower hydrocarbon removal efficiency and a greater number of small particles. Thus, soil washing with microemulsions and surfactants initially removes organic contaminants from the coarser fraction (e.g., sand), while the finer fraction adsorbs oil more strongly (Fox 1996; Ossai et al. 2020).

In comparison, Wang et al. (2019) investigated the influence of the concentration of surfactant sodium dodecyl benzene sulfonate (SDBS) in microemulsions on the crude oil removal efficiency in different contaminated soils. They concluded that the organic matter content in the soil increases while the clay content decreases and that the microemulsion treatment has little influence on the physical and chemical properties of the soil.

Remediation of beach sand-type sediments differs from that of sediments present in mangroves. Oil spilled in coastal regions, transported by the action of tides and waves, tends to adsorb into, precipitate from, and adhere to sediments. Depending on the nature of the oil and the beach sediment, the oil can penetrate through the sediment, normally to an average depth of 15–25 cm (Miller and Mudge 1997), making it difficult to clean sediment-oil mixtures. The granulometry of sandy sediments found on beaches reduces the interaction between oil and sediment. In contrast, in mangrove regions, naturally dispersed oil droplets can easily aggregate with suspended particles (clay minerals or organic matter) to form aggregates (Gong et al. 2014). Mangroves are characterized by low-energy environments with predominant deposition of fine-grained sediments and high concentrations of organic matter (Cheriyan et al. 2022).

Washing of soils contaminated with oil using surfactant solutions is an efficient method for remediation. However, its disadvantage is the potential formation of unstable macroemulsions in porous media (Oliveira et al. 2004) (Fig. 1a), which can be highly viscous and contain relatively large oil droplets that can cause pore clogging (Zhou and Rhue 2000; Karthick et al. 2019). In turn, microemulsions (Fig. 1b) are advantageous because they are characterized by a high stability and solubilization capacity, small size distribution of the dispersed phase, and low viscosity (Mahboob et al. 2022). Thus, microemulsions have favorable characteristics for soil remediation compared with their respective surfactant solutions (Zheng et al. 2011; Wang et al. 2019).

The low aqueous solubility of oil and its components, characterized as nonpolar, as well as their strong adsorption into soil or sediments makes the use of surfactant-type technologies essential to reducing contaminant levels back to safe levels compatible with human health (Badr et al. 2004). In this regard, the unique properties of microemulsions, such as ultra-low interfacial tension, a large interfacial area, thermodynamic stability, and the ability to solubilize immiscible liquids (Malik et al. 2012), make them an attractive tool for remediation.

However, to the best of our knowledge, few literature reviews (Klier et al. 2000; Karthick et al. 2019; Cheng et al. 2017; Gradzielski et al. 2021) specifically focused on detailing the composition and application of microemulsions for

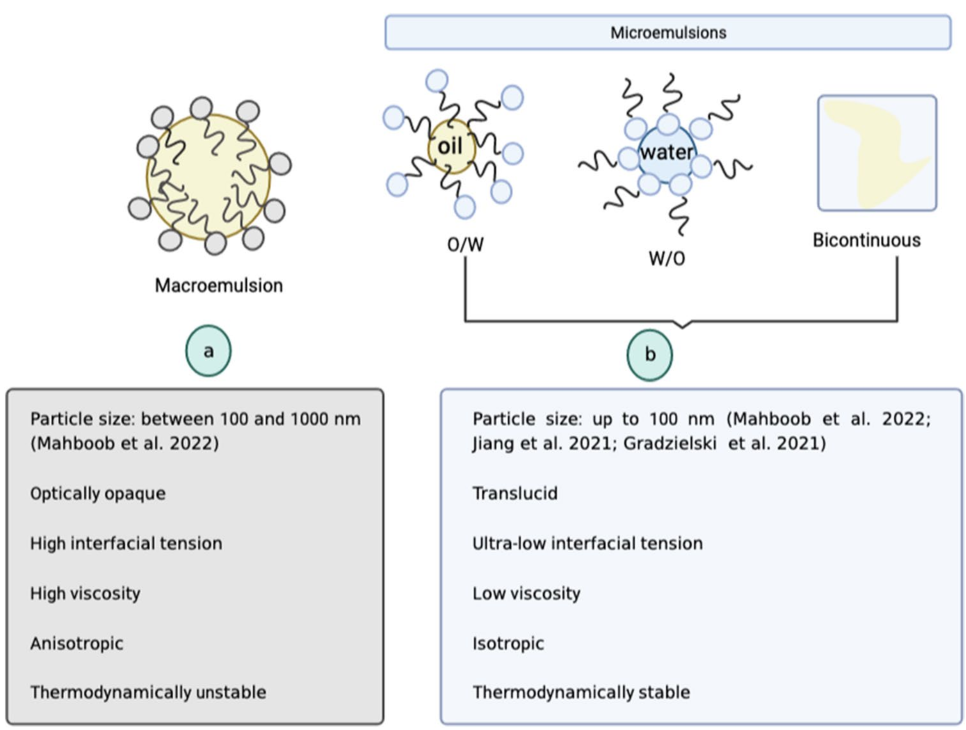



**Fig. 1** Illustration of the differences between macroemulsions and microemulsions: **a** the use of surfactants with the formation of macroemulsions in a system composed of immiscible liquids intercalated in a more or less stable form, and **b** the use of oil/water (O/W), water/oil (W/O), and bicontinuous microemulsions. The behavior of a macroemulsion or microemulsion in oil-contaminated soil will depend on the type of emulsion or microemulsion, the properties of the oil, and the soil

the remediation of oil-contaminated sediments. Karthick et al. (2019) discussed the removal mechanisms of petroleum oil contaminants from soil using chemical surfactant systems, such as surfactant solutions, surfactant foams, and nanoparticle-stabilized surfactant foams. Cheng et al. (2017) reviewed the application of the nonionic surfactant Tween-80 for the remediation of soils contaminated with hydrophobic organic compounds. Klier et al. (2000) and Gradzielski et al. (2021) described various applications of microemulsions, including the decontamination of soils and other surfaces, emphasizing the importance of the surfactant properties. Furthermore, Rosen (1989) pointed out the potential of microemulsions for soil remediation owing to their high solubilization capacity compared to micellar surfactant solutions. The authors Dierkes et al. (1998), Ouyang et al. (2002), and Bragato et al. (2002) studied microemulsions for remediation of soils contaminated with hydrophobic organic compounds, proving that microemulsions can be used in in situ or ex situ processes.

This review investigated studies on the implementation of microemulsions for removal of petroleum contaminants from sediments and soils. A detailed discussion highlights the types of surfactants used in the compositions formulated for this type of remediation. The presence of surfactants can increase the desorption of hydrophobic compounds by solubilization and, therefore, influence the rate of degradation when soil washing is associated with bioremediation (Singh et al. 2007). Additionally, the types of surfactants used in the composition of microemulsion systems and their performance in soil and sediment remediation are presented. Considering the lack of publications that elucidate the application of microemulsions for oil remediation of soil or sediment-type environments, this review includes an evaluation of the choice of surfactant type in microemulsion systems from the perspective of empirical work. Lastly, prospects for the application of microemulsions for the remediation of contaminated sediments, especially sand, are discussed based on surfactant properties, specifically biodegradability and toxicity.

## Composition of microemulsions used in the remediation of petroleum-contaminated soils and sediments

Microemulsions behave as mixtures that have the power to decrease the interfacial tension to below 50 mN $m^{-1}$, approximately the value of the interface between pure water and oil (Kahlweit et al. 1987). Their composition is complex because of the nature of the surfactants that compose them, the properties of which may vary widely (Gradzielski 2008).

The application of microemulsions in the remediation of contaminated soils or sediments has received little attention despite their potential for being promising alternatives for the remediation of sediments contaminated with hydrocarbons (Zhao et al. 2005). Microemulsified systems are composed of a mixture of a polar phase (aqueous), nonpolar phase (oil), surfactants, and co-surfactants; the latter often optionally used to decrease interfacial tension. Microemulsified systems with ionic surfactants, however, require co-surfactants to decrease the interfacial tension to below the limit of the emulsified system (Moulik and Paul 1998).

Alcohols are generally used in microemulsions for soil and sediment remediation (Valiente and Alvarez 2001), but microemulsions with fatty acids can also be used as co-surfactants since they are biodegradable and have low volatility (Lohateeraparp et al. 2003). Co-surfactants aid surfactants in establishing the interfacial membrane stability. They are characterized as nonionic molecules that decrease the repulsion forces between the hydrophilic parts of surfactant molecules (Dantas et al. 2007). A hydrophilic head provides surfactants with an affinity for polar materials, and a hydrophobic tail with an affinity for nonpolar materials. The apolar region is a hydrocarbon chain, whereas the polar region can be ionic (anionic or cationic), nonionic, or amphoteric. Thus, surfactants that stabilize microemulsion systems can be nonionic, amphoteric, or ionic (Trellu et al. 2016, 2021). Their potential to enhance the desorption of pollutants from soils or sediments during decontamination treatments has drawn considerable attention (Jafvert and Heath 1991).

## Oil removal in sediment and soil remediation

In the remediation of solid surfaces, the critical micellar concentration (cmc) of the surfactants composing the microemulsions is highly important, as the use of this parameter is recommended for the washing method (Sabatini et al. 2000). At values above the cmc, the micellar concentration and, thus, the solubility of the organic contaminant is increased.

In general, in oil-in-water micelles, the application of surfactants increases the solubility of petroleum constituents, with hydrocarbons located in the core of the micelles, while weakly polar compounds, such as fatty acids, alcohols, and esters, are on the outside of the micelles (Porter 1993; Mulligan et al. 2001). The polar exterior makes the micelles water soluble, and the nonpolar interior results in a higher affinity for oil. However, Kim and Lee (2002) showed that the contaminant removal rate was reduced when the surfactant concentration was too high, perhaps because of surfactant aggregation or emulsion formation.

The main processes of remediation of soils and sediments contaminated by oil and its derivatives involve dissolution and mobilization. While surfactants are monomeric units with both hydrophobic and hydrophilic parts, micelles aggregate at the interface between the solvent and surfactant as the concentration of surfactants increases (Ali et al. 2020). Microemulsions form swollen micelles capable of penetrating pores in soil and sediments (Hernandez et al. 2019a, b; Dela Fonte et al. 2021), and the simultaneous solubilization capacity of hydrophobic and hydrophilic substances in microemulsions is much greater than that of micelles (Fig. 2).

Oil-type solutes, which are hydrophobic contaminants, can solubilize in both the oil phase fraction and the fraction corresponding to the interfacial layer of the microemulsion (Testard and Zemb 1998), demonstrating the influence of the surfactant, oil phase, and co-surfactant on the solubilization ability of the microemulsified system.

The co-surfactant, defined as an amphiphilic co-solvent with low molecular mass and high water solubility, is frequently used for the hydrophilic-lipophilic balance of a system and can further aid with stabilizing the microemulsion over a wide range of surfactant concentrations (Zheng et al. 2012) and preventing the precipitation of the surfactant.

Factors such as soil properties, the type of oil or organic contaminant, and the process' characteristics affect the scrubbing process' efficiency. One type of remediation that can be performed on soils or sediments contaminated with organic, petroleum-type pollutants consists of solubilization of the contaminants in a microemulsion, resulting in improved wettability of the soil particles as a result of very low interfacial tensions (Schwuger et al. 1995). Tolmacheva et al. (2017) reported that a microemulsion formulation composed of the anionic surfactant sodium dodecyl sulfate acted as a good extractant of different organic contaminants, with a recovery of more than 90%, and that it depended little on the soil type.

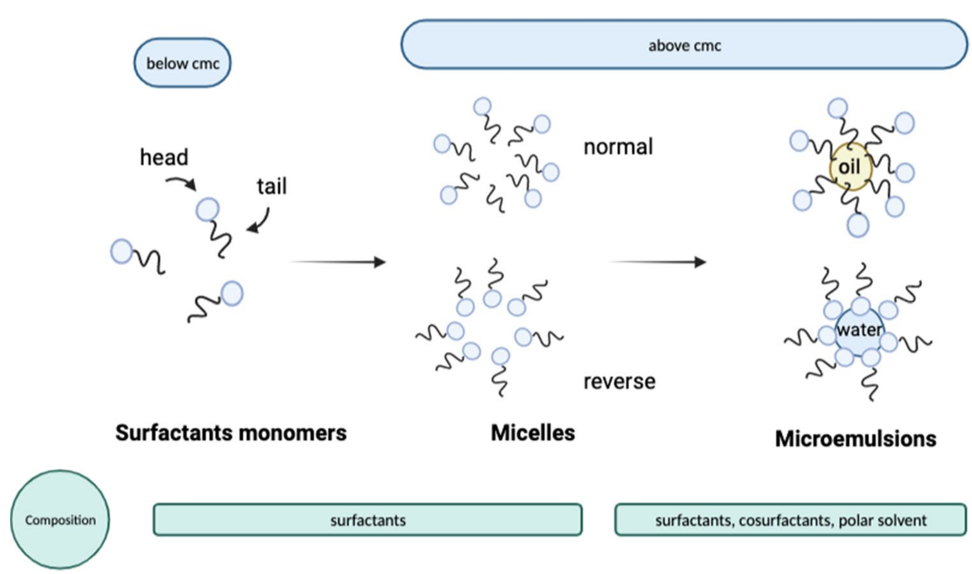
The tendency of contaminants to adsorb into the soil matrix, their low water solubility, and limited rate of mass transfer between phases for biodegradation, among other factors, causes contamination by organic complexes. Surfactants can increase the bioavailability of hydrophobic compounds, such as petroleum, by solubilization, thus increasing the degradation rate (Haigh 1996).

## Characteristics of surfactants in microemulsion formulations for soil and sediment remediation

It is essential to understand the characteristics of each soil or sediment, the chemical composition of the contaminant, and how these can interfere with the remediation process. Anionic and nonionic surfactants are selected over cationic surfactants because of their reduced degree of adsorption into soil (Fardin et al. 2021; Haigh 1996) owing to their interesting adsorption characteristics, where low levels may be present in pores and have the potential to interact with hydrophobic organic compounds (Haigh 1996). Additionally, the adsorption of nonionic surfactants is mainly influenced by the soil's organic carbon content, and the solubility of apolar compounds in soil is increased by the chain length of the surfactant (Yang and Robb 2005).

The performance of anionic and nonionic surfactants is effective but depends on the texture, size, and composition of the soil, as well as the chemical structure of the hydrocarbons present at a contaminated site. When deciding on the type of surfactant, factors such as soil organic matter content, clay fraction, and particle size should be considered (Ritoré et al. 2022.) Anionic and nonionic surfactants may be mixed to obtain low surface tension, improved solubilization, and better results compared to that of pure surfactants (Paria et al. 2005).

**Fig. 2** Differences in the performance of surfactants, micelles, and microemulsions



Using sand-type sediments and their chemical composition as an example, the sorption of surfactants can be explained. Sand is composed of calcite, quartz, and aragonite (Taqvi et al. 2007), and the negative sites in their structure in the form of carbonates and oxides have an affinity for positively charged cationic surfactants. Organic contaminants, such as petroleum hydrocarbons, tend to be adsorbed due to the hydrophobic forces in the soil or sediment matrix (Fig. 3).

Paul and Moulik (2001) considered possible mechanisms for using surfactants for remediation with microemulsions. One was the co-solution of polar and apolar compounds, providing an advantage similar to that of micellar solubilization (Romero et al. 1990), and the other was mobilization through reducing interfacial tension and increasing the ability to penetrate pores. Remediation efficiency is higher when the interfacial tension is lower and, under this condition, the free hydrocarbon phase can flow spontaneously (Ganeshalingam et al. 1994).

Concerning sediments and soil, microemulsions have further composition-dependent advantages associated with their biodegradability at the contamination site occurring within a reasonable timespan, leading to nontoxic residues (Gradzielski et al. 2021). Characteristics such as solubility and bioavailability of hydrophobic organic pollutants are attributed to the surfactants that make up microemulsions, which can aid in bioremediation processes (Mulligan et al. 2001).

Table 1 summarizes the proposed uses of microemulsions for remediation of areas affected by petroleum (crude oil or its derivatives). It reports the source of the sediment or contaminated site, the composition of the microemulsion, the types of surfactants used for the composition of the microemulsions, the effectiveness of the remediation, and observations of the parameters used by the respective empirical papers.

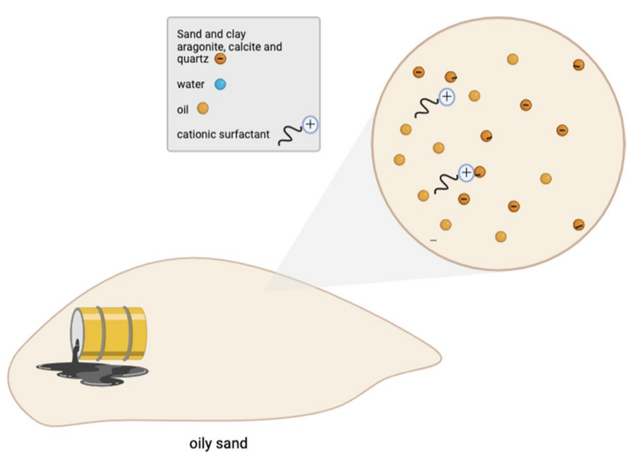



**Fig. 3** The chemical composition of cationic surfactants and oil-contaminated sand. The sorption of this type of surfactant into sand decreases the remediation efficiency

Most of the studies surveyed in this review mention that the use of nonionic and anionic surfactants in the composition of the microemulsion improved the solubilization and availability of contaminants, resulting in high remediation effectiveness. The results in Table 1 agree with the predicted theory of remediation of soils impacted by hydrophobic components, where the order of surfactants is nonionic > anionic > cationic (Boulakradeche et al. 2015; Mao et al. 2015; Lau et al. 2014; Dierkes et al. 1998). Microemulsions composed of mixtures rich in anionic and cationic or ionic and nonionic surfactants can also be effective for environmental remediation (Doan et al. 2003); however, these synergies need to be studied further.

Yuan et al. (2020) investigated the removal of polycyclic aromatic hydrocarbons (PAHs) by microemulsion washing, which was the highest (89.7%) compared to that by the other washing agents studied (Tween-80, TX-100, TX-100: Tween-80 = 1:1, and biodiesel). This study evaluated the effects of the surfactant composition and ratio in the microemulsion, the effect of the co-surfactant, its C/S ratio, pH, and the temperature.

Hernandez et al. (2019a, b) demonstrated improved efficiency of removing oil residues from sand using a microemulsion with a d-limonene solvent over an equivalent surfactant. The advantages of using microemulsions to remove crude oil residues from solid surfaces, specifically the mechanisms of softening and dispersion, were highlighted. The amount of removal depended strongly on the concentration of the microemulsion, solvent type and content, surfactant type, and immersion time. The type of surfactant used in the microemulsion affected oil removal, where anionic surfactants outperformed cationic surfactants.

Gu et al. (2020) used a microemulsion with 2% by weight anionic surfactant sodium dodecyl benzene sulfonate, 4.05% by weight n-butanol, 0.40% sodium chloride salt solution, and 15% diesel oil as the oil phase for the remediation of diesel oil-contaminated sand. Increasing the surfactant concentration increased the solubilization capacity of the microemulsion in oil and water, confirming that surfactants can alter the hydrophilic-lipophilic equilibrium at the interface and reduce the interfacial tension.

As listed in Table 1, Dantas et al. (2007), Zhao et al. (2005), Bragato et al. (2002), Bragato and El Seoud (2003), and Haegel et al. (1999) used vegetable oils in the composition of microemulsions for remediation of contaminated soils. Vegetable oils are used in the oil phase or as surfactants when saponified into microemulsions. The vegetable oils mentioned in this review include coconut, palm, rapeseed, and castor oil.

Most of the studies listed in Table 1 used medium-chain alcohols as co-surfactant, such as n-butanol. Owing to its

**Table 1** Summary of the types of microemulsions used for soil and sediment remediation after contamination with petroleum and its derivatives

| Source of sediment/soil/contaminated sites | Microemulsion | Type of surfactant | Comments | Correction effectiveness | Refs |
|---|---|---|---|---|---|
| Sand contaminated with diesel oil | 15% diesel oil and a formulation of 2.00% by weight SDBS, 4.05% by weight n-butanol, and 0.40% by weight sodium chloride | Anionic | With increasing salinity, the microemulsion (μE) phase changed from Winsor I (O/W) to Winsor II (W/O) to Winsor III (bicontinuous). The effects of inorganic salts, alcohol, and temperature on the oil removal rate of diesel-contaminated sand were investigated. Phase behavior and solubilization ability of the microemulsion system were affected by surfactant, alcohol, salt, temperature, and other factors. The SDBS surfactant anionic microemulsion system was selected to study the effects of the amount of n-butanol and NaCl on the upper phase oil yield of a Windsor I system through a static oil removal experiment. Inorganic salts and co-surfactants had little effect on the nonionic surfactant microemulsion system solubilization and phase behavior, even though temperature had a significant effect | 82.84% | Gu et al. 2020 |
| Soil contaminated by polycyclic aromatic hydrocarbons (PAHs) | 7.7% Tween-80 and 7.7% TX-100 (surfactants), 15.4% n-butanol (co-surfactant), 20.5% biodiesel (oil phase), and water (aqueous phase) | Nonionic | pH = 7. The ideal conditions for microemulsion remediation were a liquid–solid ratio of 4:1 and an agitation time of 8 h. The microemulsion prepared with a surfactant-biodiesel mixture ratio of 6:4 had the best dirt washing and PHA removal effect | 89.70% | Yuan et al. 2020 |

**Table 1** (continued)

| Source of sediment/soil/contaminated sites | Microemulsion | Type of surfactant | Comments | Correction effectiveness | Refs |
|---|---|---|---|---|---|
| Sand contaminated with crude oil residues | 23% ethoxylated surfactant ($C_{12\text{-}15}EO_7$), 23% isopropyl alcohol, 39% fresh water, and 15% solvent (containing d-limonene, β-pinene, α-pinene, xylene, nopol, and α-terpineol) | Nonionic | The amount of removal strongly depended on the crude oil composition, microemulsion (μE) concentration, and amount and type of solvent contained in the microemulsion. 5 g of the coated sand were treated with 10 g of diluted surfactant or microemulsion solution and shaken for 10 min. The supernatant was analyzed using UV–vis spectrometry at 400 nm. Increasing the μE concentration from 0.2 to 2% by volume resulted in an increase in removal efficiency from 10 to 40%<br>However, increasing the surfactant concentration up to 2% by volume triggered a minimal increase (less than 2%). This demonstrated the need for solvents within μE concentrations to effectively remove hard asphaltene deposits, even when high surfactant concentrations are used. All formulations were diluted to 0.2% by volume in 2% KCl, which contained 0.46% by weight surfactant and 0.3% by weight solvent | 40% | Hernandez et al. 2019a |

**Table 1** (continued)

| Source of sediment/soil/contaminated sites | Microemulsion | Type of surfactant | Comments | Correction effectiveness | Refs |
|---|---|---|---|---|---|
| Sand contaminated with asphalt waste | 23% by weight ethoxylated surfactant ($C_{12-15}EO_7$), 23% by weight isopropyl alcohol (co-surfactant), 39% by weight fresh water, and 15% by weight d- limonene. Two cationic surfactants (dodecyl bromide trimethyl ammonium [DTAB] and coconut bis [hydroxyethyl] benzyl ammonium chloride [CBAC]) and two anionic surfactants (sodium lauryl ether sulfate [SLES] and sodium dodecylbenzene sulfonate [SDBS]) were used. A small amount of co-surfactant was replaced by 1-octanol (0.3% by weight) | Anionic, cationic, and nonionic | The removal amount strongly depended on the microemulsion concentration, solvent and content within the microemulsion, surfactant type, and immersion time | Surfactants were compared, and the anionic one (SDBS) showed better efficacy (13–49%) | Hernandez et al. 2019b |
| Soil contaminated with PHAs | 3% sodium dodecyl sulfate, 0.8% benzene, 0.6% n-butanol, and 90.2% water | Anionic | At an extraction time of 10 min and a microemulsion volume of 30 mL per 2 g of soil, the recovery rate reached its maximum | 90–105%[a] | Tolmacheva et al. 2017 |
| Soil contaminated with petroleum | 1% w/v surfactants, such as sodium dodecyl sulfate (SDS) and sodium dodecyl benzene sulfonate (SDBS), kerosene, and a 1:1 mixture of 1-pentanol and 1-butanol | Anionic | The surface tension in the wash water ranged from 48.0 ± 0.2 to 72.5 ± 0.2 dynes/cm. Equal volumes of aqueous and oil phases (W/O = 1.0) were used, and the optimal salinity conditions were 4.9% w/v for SDS and 2.2% w/v for SDBS when contact time was 4 h; 250.0 mg of the solid target substrate were placed in contact with 2.00 mL of each microemulsion system | 95.43% | Pérez et al. 2012 |

**Table 1** (continued)

| Source of sediment/soil/contaminated sites | Microemulsion | Type of surfactant | Comments | Correction effectiveness | Refs |
|---|---|---|---|---|---|
| Sandy soil contaminated with diesel oil | Surfactant (saponified coconut oil and butan-1-ol), an aqueous phase (distilled water with or without NaCl), and an oil phase (diesel oil) | Anionic | The co-surfactant/surfactant ratio (C/S) ratio was either 0.5, 1.0, or 2.0. The parameters tested that influenced microemulsion formation included: the nature of the co-surfactant, the C/S, and presence or absence of an electrolyte in the aqueous phase (NaCl). An increase in the carbonic chain length of the co-surfactant led to a decrease in the solubility of the active material in the aqueous phase. This effect is demonstrated where the microemulsion region (μE) formed by the butan-1-ol system was larger than that formed by the pentan-1-ol system. Therefore, butan-1-ol with a C/S ratio of 0.5 was the co-surfactant chosen for soil remediation experiments | 75% | Dantas et al. 2007 |
| Soil impregnated with phenanthrene | Castor oil, commercial castor sodium sulfate (SCOS), and water | Nonionic | 500 mg/L of surfactant solutions were used for 1:10 soil–water systems. The results showed that SCOS could be a potential agent in the ex situ washing of soils contaminated with phenanthrene | 69.90% | Zhao et al. 2005 |

**Table 1** (continued)

| Source of sediment/soil/contaminated sites | Microemulsion | Type of surfactant | Comments | Correction effectiveness | Refs |
|---|---|---|---|---|---|
| Sand contaminated with asphaltene residue | 15% polyethoxylene-10 ($C_{12}E_{10}$) lauryl ether, isopropyl, butyl (15%) and isoamyl co-surfactants, sodium hydrotropes p-toluenesulfonate, and sodium cumenesulfonate and triethylene glycol ether. The oil phase (60%) was a mixture of decane, toluene, and cyclohexane (DTC), and the aqueous phase was 10% (water and different aqueous solutions of hydrotropes) | Nonionic | Three C/S ratios (0.5, 1.0, and 1.5) using butyl alcohol were investigated. Butyl alcohol promoted a monophasic region at C/S = 1. An extraction time of 4 h was selected to analyze the effects of a microemulsion composition of 60% DTC, 15% $C_{12}E_{10}$, 15% butyl alcohol, and 10% aqueous phase. The action of the DTC mixture was compared to the actions of terpene, orange oil, and pine oil, all considered safe for the environment. While pine oil did not show good oil removal efficiency, the other natural solvents showed a solubilization capacity very close to that of DTC. Microemulsions containing 30% orange oil exhibited high removal (93%) of residual crude oil from the sand | 92–93% | Oliveira et al. 2004 |
| Soil contaminated by PHAs | Coconut oil methyl esters (CME), the saturated palm oil fraction (SPME), and surfactants of the type Synperonic 91/4 ($REO_4$) and Synperonic 91/5 ($REO_5$), where R = $CH_3$ $(CH_2)_{8/10}$ and EO refer to one unit of oxyethylene and water | Nonionic | Interfacial tension: $10^{-6}$ mN $m^{-1}$. Contact time: 15 min (μE and contaminated soil). Increasing temperature (for nonionic surfactants) caused the surfactant solution to become more hydrophobic and, thus, to segregate further towards the oil–water interface, thereby reducing surfactant film curvature and interfacial tension | 88% | Bragato, and El Seoud 2003 |

**Table 1** (continued)

| Source of sediment/soil/contaminated sites | Microemulsion | Type of surfactant | Comments | Correction effectiveness | Refs |
|---|---|---|---|---|---|
| Soil contaminated by PHAs | Coconut and palm oils, alkyl linear polyethoxylates: Synperonic 91/4, $CH_3\ (CH_2)_{8–10}\ (OCH_2CH_2)\ _4OH$, Synperonic 91/5, $CH_3\ (CH_2)\ _{8–10}\ (OCH_2CH_2)\ _5OH$ (ICI, Middlesborough, UK), and Marlipal 24/50, $CH_3\ (CH_2)_{11–13}\ (OCH_2)$, and $(CH_2)\ _5OH$ (Hüls, Marl, Germany) | Nonionic | The concentration of all components of the microemulsion was given by weight. In these experiments, the variables were microemulsion composition, temperature, µE/soil ratio, and contact time with an interfacial tension of $10^{-6}$ mN $m^{-1}$ and a contact time of 3 h. The best µE/soil ratio was 6 (extraction efficiency). At 37.5 °C, the efficiency of the microemulsion was greater than that of toluene. Microemulsion extraction based on the shorter CME and PME oils was more efficient than that based on RME (rapeseed oil methyl ester) whose extraction value is around 105% (T = 43 °C; µE/soil = 6). The results showed that oils from the CME and PME chains form µE close to room temperature | > 100%[b] | Bragato et al. 2002 |
| Soil contaminated with PHAs or biphenyls polychlorinated compounds (PCBs) | Alkyl polyethoxylates, ethoxylated castor oil, rapeseed oil methyl ester (oil), and water | Nonionic-anionic blend | 3.3 g of earth and 10 g of microemulsion were used | 89–106%[c] | Haegel et al. 1999 |
| Soil contaminated with gasoline and organic lead | Sodium lauryl sulfate as a surfactant, n-pentanol as a co-surfactant, and water | Anionic | Interfacial tension: 0.03 N $m^{-1}$. The surfactant/co-surfactant/water (S/CoS/W) solution was made by mixing 4.3 g sodium lauryl sulfate, 8.7 g n-pentanol, and 87 g deionized water. The solution thus contained 4.3% and 8.7% surfactant and co-surfactant, respectively | 95% gasoline and 90% organolead | Ouyang et al. 1996 |

**Table 1** (continued)

| Source of sediment/soil/contaminated sites | Microemulsion | Type of surfactant | Comments | Correction effectiveness | Refs |
|---|---|---|---|---|---|
| Bitumen-impregnated sand | 2-Butoxyethanol (co-surfactant), water, toluene, and various surfactants | Cationic and anionic | The co-surfactant/surfactant molar ratio was 10:1. The active mixture was $H_2O$:co-surfactant:surfactant in a molar ratio of 0.800:0.182:0.018 | 40–60% depending on the surfactant. Dowfax 2A1 (anionic) showed a recovery rate of 100% at 25 °C | Sarbar et al. 1984 |

[a]The degree of extraction of PHAs from soil samples was determined by a standard addition method, with additions of 100 μg/kg and 20 μg/kg of each compound.

[b]Extraction efficiency is given in % relative to the amount extracted from the soil using hot toluene (Soxhlet, 6 h). According to this criterion, efficiencies > 100% can be achieved.

[c]Toluene extraction after washing with water determined the PCB remaining in the soil (3–7%). Adding the amounts from the batch extraction, the amounts in the soil were found to be more than 100% of the total PCB.

characteristics, alcohol partially enters the aqueous and oil phases, changing their polarity and promoting the phase transition of the system (Jiang et al. 2021; Wang et al. 2019). For the composition of microemulsion systems, studies have indicated the optimization of the process to obtain the maximum formulation region of the microemulsion by evaluating the nature of the surfactant and co-surfactant, the influence of the C/S ratio, and the effect of the oil phase.

The reviewed studies evaluated the influence of temperature, pH, surfactant ratio, co-surfactant/surfactant ratio, salinity, and contact time on the remediation effectiveness. The amount of crude oil or its derivatives removed from soils and sediments was strongly dependent on the microemulsion concentration, solvent type, and content within the microemulsion, immersion time, and surfactant type.

Nonionic surfactants are preferred over anionic surfactants because, in soils or sediments rich in bivalent cations, the complexation of these cations with anionic surfactants makes remediation ineffective. Cationic surfactants have a higher affinity for the negative charges of soils and sediments (Gu et al. 2020) considering similar hydrocarbon chain length, pH levels, and use of the same set of sediments or soils (Brownawell et al. 1997; Paria 2008).

The use of surfactants for soil washing is a widespread, promising technology. Only surfactants were evaluated for their cleaning abilities. Many studies have considered the higher solubilization capacity of nonionic surfactants, which are more economical and biodegradable (Gharibzadeh et al. 2016; Takeuchi et al. 2014). In contrast, Hernandez et al. (2019a, b) pointed out the feasibility of using anionic surfactants in the composition of microemulsions for cleaning asphaltene-contaminated sand. However, the soil washing process is affected by several factors, and when using microemulsions, other components of their composition must be considered.

Due to the negatively charged particles present in soils, compared with anionic and nonionic surfactants, cationic surfactants have a greater tendency to adsorb into the soil, justifying the greater consumption of this type of surfactant. This also explains the large number of studies using anionic and nonionic surfactants for soil washing.

Although interfacial tension is essential data related to the performance of microemulsions, most articles did not discuss this. The works of Ouyang et al. (1996), Bragato et al. (2002), and Bragato and El Seoud (2003) specifically reported low interfacial tension values, and other works mentioned them, leading us to conclude what is described in the literature (Gradzielski et al. 2021; Ganeshalingam et al. 1994).

As for biodegradability, the works of Bragato et al. (2002) and Bragato and El Seoud (2003) mention the biodegradability of the oil phase of the microemulsion. For environmental purposes, the microemulsion components should be biodegradable and, as such, vegetable oils were used (Bragato and El Seoud 2003).

Surfactants are toxic to humans, animals, and plants, owing to the absence or low biodegradability of these compounds in soil and sediment (Mao et al. 2015). Decomposition and degradation of surfactants in soils and sediments occur via microorganisms, usually fungi and bacteria, and biodegradation can be low, especially that of synthetic surfactants, due to precipitation and sorption phenomena. Some studies have indicated that nonionic surfactants have greater biodegradability than cationic and anionic surfactants (Bolan et al. 2022; Zhang et al. 2021; Gharibzadeh et al. 2016).

Most studies have investigated the removal efficiency of contaminants in soil or sediments. Further studies on the influence of the type of co-surfactant used, contact time of the microemulsion with the soil, surfactant concentration, biodegradability of the components, and hydrophilic-lipophilic balance (Lau et al. 2014) are required, as these factors may be jointly responsible for remediation effectiveness.

## Prospects of microemulsions

Given the potential of microemulsions as a remediation technology for soil and sediment cleanup (Dela Fonte et al. 2021), new formulations or combinations of surfactants for microemulsion compositions should be realized. Lower cost, more environmentally "friendly" (i.e., biodegradable) oil removal techniques with efficiency equal to or greater than those presented in the literature are desirable (Zhao et al. 2005).

Biodegradability is still a challenge considering that mass degradation is required for decontamination purposes, which is toxic, albeit to a lesser degree than the initial toxicity (Gradzielski et al. 2021). Therefore, the use of surfactants of plant origin needs to be further explored for the remediation of sand-type sediments. Our literature review showed that the number of studies on the use of vegetable oils is incipient.

Bioremediation is considered one of the most promising techniques for soil and sediment remediation (Guerin 1999). However, the time factor may be disadvantageous. In this regard, microemulsions deserve further study as a remediation technique associated with bioremediation, since soil pollutants are mainly degraded in solutions, becoming more readily available to microbial actions (Harms and Bosma 1997).

Regarding the cost of microemulsion remediation processes (Lucas et al. 2020), further studies are needed to consider separating the oil-rich phase containing dissolved oil contaminants and the surfactant-rich phase so that it can be recycled for other cleanups. The application of microemulsions as a remediation technique aimed at contaminant reuse and microemulsion recycling should be considered. One possible method using nonionic surfactants is to promote phase separation of microemulsion systems by increasing the temperature. Additionally, microemulsions can be reused in new soils or sediment masses.

The use of strategies such as microemulsions or their combination with others can help reduce the toxicity of surfactants before they are released into the environment. Fatty acid methyl esters from vegetable oils are gaining ground in the choice of microemulsion components because of their low toxicity and rapid biodegradability in soils and sediments (Kumar et al. 2021; Zhang et al. 2021; Klossek et al. 2012).

## Conclusion

This review addresses the literature on the application of microemulsions for the remediation of soils and sediments contaminated with petroleum or its derivatives. The findings can be summarized as follows:

- Anionic and nonionic surfactants have been used in most studies and are effective, although their relative performances depend on the type of soil or sediment. Cationic surfactants can sorb into the soil owing to their positive charges, and nonionic surfactants are selected due to their higher solubilization capabilities and them being more economical than cationic and anionic surfactants.
- To explore the technical feasibility of decontaminating oil-contaminated soils and sediments using microemulsions, it is essential to understand the contribution of different components in microemulsion formation and their effects on contaminant solubilization. This involves choosing surfactants that increase the solubility of oil components or decrease interfacial tension to increase oil mobility. Desirable properties for soil and sediment remediation include increased solubility of hydrophobic contaminants, reduced surface tension, critical microemulsion concentrations, and wettability.
- The choice of surfactant is essential for the efficient remediation of oil-contaminated sediments and biodegradability. This literature review indicated that microemulsions can be formulated with vegetable oils, alcohols, and surfactants such as saponified vegetable oil. It is critical to take into account the vegetable origin, biodegradability, and non-toxicity of the microemulsion components for soil and sediment remediation because selecting an inappropriate surfactant can result in serious environmental problems.
- The remediation of soils and sediments is a matter of socioeconomic interest because of its impacts on humans, animals, and plants. In coastal environments,

such as beaches and mangroves, populations depend on natural resources for food and the development of economic activities. In addition, the aesthetic aspects of such environments relate to touristic, economic, and environmental issues of ecosystems.

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**Data availability** All relevant data and material are presented in the main paper.

## Declarations

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