Removal of Oil Pollutants in Soil

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Oil-contaminated soil is one of the most concerning problems due to its potential damage to humans, animals, and the environment. In recent years, surfactant foam and nanoparticles have shown high removal of oil pollutants from contaminated soil. Researchers provide an overview on the remediation of oil pollutants in soil using nanoparticles, surfactant foams, and nanoparticle-stabilized surfactant foams. In particular, the fate and transport of oil compounds in the soil, the interaction of nanoparticles and surfactant foam, the removal mechanisms of nanoparticles and various surfactant foams, the effect of some factors (e.g., soil characteristics and amount, nanoparticle properties, surfactant concentration) on remediation efficiency, and some advantages and disadvantages of these methods are evaluated.

Keywords: soil remediation ; oil pollutants ; surfactant ; biosurfactant ; surfactant foam ; nanoparticles ; contaminated soil

1. Introduction

Oil compounds have been widely utilized as an energy source in human life and industry for a long time. In nature, they can be found in deposits or deep sediment as a result of the decomposition of dead plants and animals over many years. They mostly consist of aliphatics (e.g., alkanes, alkenes), aromatics (e.g., polycyclic aromatic hydrocarbons—PAHs), and non-hydrocarbon compounds (e.g., sulfides, pyridine, metals) ^[1].

Oil pollutants can be formed by exploration, production, and transformation processes ^[2]. After entering the soil, the interaction of oil pollutants with soil components and microorganisms may alter their properties and transport ^{[3][4]}. They continue in the soil for a long time due to the attachment or adsorption to soil components, which can harm the soil, ecosystem, or animals ^[5].

In recent years, the use of oil-related products has increased as a result of economic development and population growth. According to a British Petroleum report ^[6], global oil consumption was 5.3 million barrels per day in 2021, whereas 1.5 million barrels per day were consumed by the United States. A small amount of oil pollutants may cause serious problems for animal and human health, such as teratogenicity, cardiotoxicity, cancer, and fetus malformation ^{[5][7]}. Moreover, they are also listed in the priority pollutants category by the United States Environmental Protection Agency (USEPA) as an origin of cancer for humans ^[8]. The toxicity of oil pollutants to humans is strongly dependent on their specific composition, features, and contact time and level ^{[9][10]}. In addition, the presence of oil pollutants in the soil can decrease the resistance to diseases and stunted growth of plants or limit the development of soil microbes and the aquatic environment ^{[7][11][12]}.

2. Remediation Methods of Oil Pollutants in Soil

Different approaches have been utilized to remediate oil pollutants in soil. Some common techniques are physicochemical (e.g., surface capping, pump and treat, soil washing, soil vapor extraction, soil extraction) $^{[13][14][15][16]}$, chemical (e.g., stabilization, oxidation–reduction, adsorption, supercritical fluid extraction and oxidation, encapsulation) $^{[15][17][18]}$, biological (bioremediation, bioattenuation, biodegradation, bioventing, biosparging, biotransformation, composting) $^{[19][20]}$ $^{[21][22]}$, thermal (e.g., incineration, pyrolysis) $^{[23][24]}$, and phytoremediation (phytostabilization, phytovolatilization, phytotransformation) methods $^{[15][25]}$. Many criteria should be considered to select the optimal treatment method, such as site characteristics, oil pollutant features, soil composition and properties, remediation time and cost $^{[15]}$. Generally, these common methods have many disadvantages that limit their wide application, e.g., they are not effective for removing oil pollutants adsorbed on clay-size particles (soil washing) $^{[26][27]}$ or high oil content (soil vapor extraction) $^{[25]}$. There is also the possibility of the formation of by-products (chemical oxidation–reduction) $^{[12]}$. In addition, they have high operation costs, further treatment demand for off-gases and combustion residuals (thermal treatment) $^{[24]}$, and long treatment time (phytoremediation) $^{[15]}$. Thus, it is critical to research and develop new oil-contaminated soil remediation approaches.

2.1. Application of Nanoparticles for Remediating Oil Pollutants in Soil

Nanoparticles are particles with a size of less than 100 nm (or 10^{-9} m). Due to their unique characteristics, for example, small size or high specific surface area, they can be transported to complex target zones at contaminated sites ^[28]. Together with their simple and uniform operating conditions ^[29], they have been widely used for soil remediation. In contrast, long treatment times and possible formation of toxic by-products are some disadvantages of using nanoparticles for soil remediation ^[30].

2.1.1. Effect of Nanoparticles on Soil Properties

The presence of nanoparticles decreases the soil pH, organic carbon, activity of dehydrogenase enzyme, microbial biomass transformation rate, soil bacteria, and amount of fungal colonies in the soil, reducing the soil microbial diversity ^[31]. Due to their magnetic attraction, nanoparticles tend to aggregate to form larger particles, lowering soil mobility and reactivity ^[32].

Meanwhile, the addition of nanoparticles enhances the available phosphorus in the soil. In another study, adding ZnO nanoparticles (10 mg/kg soil) reduced the soil pH after seven days and decreased the $eqCO_2$ value in soil or the conversion rate of carbon sources into biomass. However, the presence of ZnO nanoparticles also enhanced the development of some bacteria in the soil, which improved the soil microbial diversity ^[33].

2.1.2. Removal Mechanisms

Nanoparticles have been used to remediate contaminated soil under different conditions for a long time. Due to their high solvent affinity and large specific surface area, nanoparticles can easily contact oil compounds and improve their solubility, leading to a high removal rate ^{[34][35]}. The interaction of nanoparticles and other counterparts strongly depends on their types, amount, and properties ^[36]. Their main treatment mechanisms are adsorption (e.g., nZVI, carbon nanotubes), oxidation (e.g., manganese nanoparticles, cobalt nanoparticles), and photocatalysis (e.g., bismuth nanocomposite, BiPO₄-based photocatalysts) applications ^{[37][38]}. Oil pollutants can be removed from contaminated soil by adsorption on the nanoparticle's surface via π - π and van der Waals interactions ^{[18][39][40]}. Nonetheless, the potential aggregation of nanoparticles, which can decrease the surface area and active sites of nanoparticles and reduce the treatment efficiency, is one of the most significant disadvantages of this method.

In the oxidation method, the oil pollutants can be reduced into less toxic or non-toxic compounds, such as CO_2 and H_2O , by Fenton-like reactions ^{[35][41][42]}. This method involves the degradation of oil pollutants by reactive oxygen species (ROS), which are formed via the reaction of iron oxides with H_2O_2 , UV light, or under ultrasound ^[43]. In particular, the generation of ROS such as hydroxyl radicals (HO·) or hydroperoxyl (HO₂·), may degrade oil pollutants to form final products, such as CO_2 and H_2O , as follows ^[44]:

$$\begin{aligned} &\mathsf{Fe}^{2^{+}} + \mathsf{H}_2\mathsf{O}_2 \to \mathsf{Fe}^{3^{+}} + \mathsf{OH}^- + \mathsf{HO} \ (1) \\ &\mathsf{Fe}^{3^{+}} + \mathsf{H}_2\mathsf{O}_2 \to \mathsf{Fe}(\mathsf{OOH})^{2^{+}} + \mathsf{H}^+ \ (2) \\ &\mathsf{Fe}(\mathsf{OOH})^{2^{+}} \to \mathsf{Fe}^{2^{+}} + \mathsf{HO}_2 \ (3) \\ &\mathsf{H}_2\mathsf{O}_2 + \mathsf{HO}^{\cdot} \to \mathsf{HO}_2^{\cdot} + \mathsf{H}_2\mathsf{O} \ (4) \\ &\mathsf{Fe}^{3^{+}} + \mathsf{HO}_2^{\cdot} \to \mathsf{Fe}^{2^{+}} + \mathsf{H}^+ + \mathsf{O}_2 \ (5) \\ &\mathsf{ROS} + \mathsf{oil} \ \mathsf{pollutants} \to \mathsf{CO}_2 + \mathsf{H}_2\mathsf{O} \ (6) \end{aligned}$$

This method is simple, cheap, effective for various organic pollutants in soil, and safe for the environment and human health. However, the potential toxicity of intermediates and slow treatment efficiency are some disadvantages that should be considered for this approach. Moreover, its treatment rate is influenced by concentrations of H_2O_2 , the dosage of iron oxides, power and time of UV light and ultrasound, pH, and temperature ^[44].

In the photocatalysis method, oil pollutants are degraded into mostly CO_2 and H_2O by reactive oxygen species, such as HO^{\cdot} or superoxide anions (O_2^{\cdot}), formed under the activation of light and semiconductors such as TiO_2 or $ZnO^{[45]}$. In particular, under the illumination of a light source, such as ultraviolet (UV) or sunlight, electrons from the valence band in TiO_2 nanoparticles will be activated and jump to the conduction band, leaving behind some holes— h^+ (**Figure 1**). These h^+ and e^- may react with H_2O and O_2 in the atmosphere to generate ROS, which will degrade oil pollutants into less toxic or non-toxic products. The advantages of photocatalytic techniques include high treatment efficiency, clean technology,

high stability, no formation of toxic by-products, and low toxicity. Meanwhile, the high energy cost, quick recombination rate of ROS, and unavailability of pollutants deep in the soil are some disadvantages of this method. The treatment efficiency by photocatalysis methods is strongly dependent on various factors, such as temperature, soil particle size and type, soil thickness, humic acid, light source and time, and characteristics of oil compounds in soil ^[46].

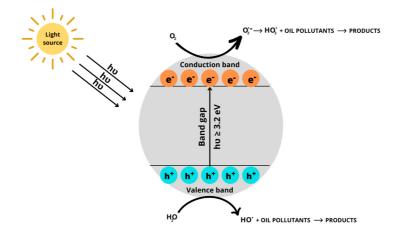


Figure 1. Treatment of oil pollutants by photocatalytic activity of TiO₂ nanoparticles.

2.1.3. Treatment of Oil Pollutants in Soil by Nanoparticles

Various nanoparticle types have been successfully utilized to remove different oil pollutants from contaminated soil. Carbon nanotubes (CNTs) were effectively utilized for adsorbing PAHs ^[47] or dichloro-diphenyl-trichloroethane (DDT) in the natural soil, whereas 56% of DDT was degraded by nZVI after 7-day treatment ^[48]. Bentonite clay combined with nZVI removed PCBs from soil-sorbed PCBs 10 times more than only nZVI ^[49]. Furthermore, the addition of ethanol increased PCB desorption and enhanced the contact between PCBs and nZVI, leading to 50% higher treatment efficiency.

Iron nano-oxide particles removed 99% pyrene in contaminated soils via a Fenton oxidation reaction with hydrogen peroxide (H₂O₂) ^[50]. Karam et al. ^[51] showed a high degradation rate of anthracene using nano-TiO₂-photocatalysts. Furthermore, PAHs were productively treated by different nanoparticles, such as gold nanoparticles ^[52], iron hexacyanoferrate nanoparticles ^[53], ZrO₂ nanoparticles ^[54], nano Fe³⁺-montmorillonite ^[55], nano anatase TiO₂, ^[56], ZnO nanoparticles ^[57], Ti/ZnO-Cr₂O₃ nanocomposite ^[58], Fe₃O₄ nanoparticles ^[59], TiO₂-graphene nanocomposites ^[45], Fe/Cu bimetallic nanoparticles ^{[38][60]}. More oil-contaminated soil treatment methods using nanoparticles are shown in **Table 1**.

Nanoparticle Name	Pollutant Name	Treatment Time, Day	Treatment Efficiency, %	Reference
MWCNTs ¹	Phenanthrene	21	54.2	[61]
MWCNTs	PAHs ²	5	79	[62]
nZVI ³	Phenol	12 h	9	[63]
nZVI/BFN ⁴	Phenol	7 h	98.5	[63]
Iron nanoparticles	PCBs ⁵	6 h	95	[64]
nZVI	PAHs	1 h	70	[<u>65</u>]
APU nanoparticles ⁶	PAHs	5	67	<u>[66]</u>
nZVI/biosurfactant	Oil compounds	1 h	83	[<u>40]</u>
nZVI/biosurfactant foam	Oil compounds	30 min	67	[<u>34]</u>
nZVI	PCBs	15	42	[67]
nZVI-Pd	PCBs	15	64	[67]
nFe ₃ O ₄	PCBs	15	68	[<u>67]</u>
Fe-Cu/biochar/geopolymer	Naphthalene	2 h	68	[<u>68]</u>
nZVI/bioattenuation	Diesel fuel	75	41.0	[21]

Table 1. Use of selected nanoparticle for remediation of oil pollutants from soil.

Nanoparticle Name	Pollutant Name	Treatment Time, Day	Treatment Efficiency, %	Reference
nZVI/biostimulation	Diesel fuel	45	64.6	[21]
nZVI/bioaugmentation	Diesel fuel	15–30	85.3	[21]
nZVI/biostimulation + bioaugmentation	Diesel fuel	30–60	89.5	[21]
Iron oxide nanoparticles	Crude oil	1	N/A	[42]
Nano rutile TiO ₂	Pyrene	25 h	52.2	<u>[69]</u>
Nano rutile TiO ₂	Phenanthrene	25 h	38.9	[<u>69]</u>
Iron oxide nanoparticles	PAHs	5	70	[<u>70</u>]
Akaganeite nano-rods	PAHs	1	65	<u>[71]</u>
Iron oxide nanoparticles	Anthracene	10	99	<u>[41]</u>
Graphene oxide	PAHs	7 min	~100	[72]
Fe-doped TiO ₂ nanocatalyst	PAHs	35 min	80	[<u>73</u>]
TiO ₂ -based ZnHCF nanocomposite	PAHs	1	86	[74]
C ₃ N ₄ /Fe ₃ O ₄ nanocomposite	Phenanthrene	2 h	92.3	[<u>75</u>]
Cu ₂ OPLA composite nanofiber	Fluoranthene	8 h	67.6	[<u>76</u>]

In other research articles, nanoparticles have been combined with microorganisms to remove oil pollutants in soil. The presence of nanoparticles might have improved microbial metabolism and microbial enzymes, which increased the treatment efficiency of toxic organic contaminants ^[72]. The combination of iron magnetic nanoparticles and *Bacillus* spp. degraded up to 89.7% atrazine in soil ^[78]. Bebić et al. ^[79] indicated the degradation of lindane up to 68.3% using silica nanoparticles and *Myceliophthora thermophila* at pH 5.0 in 40 min.

3. Remediation of Oil Pollutants in Soil by Surfactant Foam/Nanoparticle Mixture

3.1. Interaction of Surfactant Foam and Nanoparticles

The existence of contaminants may decrease the spreading velocity of aqueous foam in soil layers [80]. In this case, the foam interaction with soil contaminants can be represented by the dimensionless Lamela number [81]. Hence, nanoparticles can stabilize the surfactant foam (Figure 5), enhancing their movement in the unsaturated soil zone [82]. The foam stabilization by nanoparticles involves the agglomeration of nanoparticles at the oil-water interface to create a thick layer that may hinder foam aggregation [83]. In particular, the generation of nanoparticle monolayers or adjacent nanoparticle bilayers may cause the stabilization of liquid films in the foam [84][85]. The attachment of colloidal nanoparticles at the gas-liquid or liquid-liquid interfaces of foam may decrease bubble breakage, contributing to foam stabilization [86][87]. For example, the foam formation by silica nanoparticles and SDS surfactant was 10 times more stable than that by only SDS surfactant due to the attachment of SDS molecules, which lowered the silica nanoparticle surface charge [87]. Li and Prigiobbe [88] showed a similar result, where high foam quality was formed by cationic surfactant and silica nanoparticles under N2-gas. The generation mechanism of anionic surfactant foam in porous media with or without nanoparticles is similar [89]. In another study, the mixture of hydrophobic fine particles and surfactant was also proven to improve the bubble combination efficiency and reduce the foam stability ^[90]. The reduction in nanoparticle retention due to the decrease of surface tension at the liquid-gas interfaces may influence foam stability [91][92]. The gas used for foam generation may also influence the transport of surfactant foam-nanoparticle mixture through the soil, affecting the remediation efficiency of oil pollutants [93].

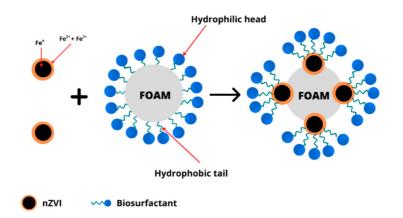


Figure 2. Interaction of surfactant foam and nZVIs.

Nanoparticles can also stabilize and enhance foam transport in the unsaturated zone of the soil, leading to higher soil treatment effectiveness ^[93]. The accumulation of nanoparticles at the oil–water interface may generate particle monolayers or bilayers, which limit bubble breakage and stabilize the surfactant foam ^[83]. In addition, the adsorption of colloidal nanoparticles at gas–liquid or liquid–liquid interfaces may also contribute to foam stability ^[86]. The foam produced by silica nanoparticles and SDS surfactant is ten times more stable than by only SDS surfactant. This is due to the adsorption of SDS surfactant foam on the nanoparticle surface lowering the surface charge, which promotes the capacity to adsorb more uncharged nanoparticles on the foam surface ^[87]. The foam stability produced by hydrophobic fine particles and surfactant is much lower than by surfactant only due to the development of bubble coalescence ^[90].

3.2. Use of Surfactant Foam-Nanoparticle Mixture in Soil Remediation

The effect of colloidal particles on foam formation, stability, and prevention has been studied for a long time ^[94]. Due to their ability to enhance foam stability, nanoparticles have been effectively used for oil recovery ^[95] or soil remediation ^[38] $\frac{[40][96]}{[96]}$. The viscoelastic layer was enhanced by attaching 50% silica nanoparticles to the interface, which hindered the collapse of the bubble and improved the foam stability up to 23 h ^[97]. In another study, the half-life of SDBS surfactant foam after adding silica nanoparticles was double that of only SDBS surfactant foam due to the development of foam stability ^[98]. The presence of silica nanoparticles in a sand column also decreased the hydraulic conductivity of CTAB surfactant foam, which enhanced the foam stability after 17 days and led to higher isolation efficiency of the contaminant in soil ^[99].

The transport of nanoparticles in soil was improved by combining them with surfactant foam. Surfactant foam may stabilize the nanoparticle suspension and prevent them from aggregating in an aqueous solution, which decreases the nanoparticle retention on the soil surface and enhances the movement of nanoparticles in the soil. For example, 1% SLES surfactant foam delivered 100% nZVI in the soil vadose zone, leading to higher removal efficiency of the soil contaminants ^[100]. Shen et al. ^[101] reported that the transport of nZVI in the soil subsurface was significantly improved with foam generated by different surfactants, such as SDS, TW20, TW80, TX100, leading to higher remediation efficiency in the vadose zone. The film breakage of foam was reduced with the addition of nZVI, which enhanced the microsphere transport in soil and led to better treatment effectiveness. The same results were reported in other papers, where the remediation rate of oil contaminants was 78–99% by foam-stabilized nanoparticles under various environmental conditions ^{[102][103]}.

Using a surfactant foam–nanoparticle combination for soil remediation is less common than surfactant solutions only or surfactant foams. However, due to the synergistic effect of nanoparticles and surfactant foam, the soil remediation efficiency by this mixture is surpassed by only surfactant or surfactant foam at the same concentration ^[103]. With other advantages, such as simplicity and effectiveness for various soil contaminants, applying a surfactant foam–nanoparticle mixture can become a productive method for soil remediation in the future.

A surfactant foam–nanoparticle mixture has been employed for soil remediation in the field. According to Quinn et al. ^[104], the treatment performance of TCE from contaminated soil was remarkably enhanced (up to 100%) after five months. In another article, Zhao et al. ^[105] pointed out that the degradation rate of chlorinated volatile organic compounds (CVOCs) and perchloroethylene (PCE) in field sites using a surfactant–corn oil–nZVI mixture after 2.5 years was 86% and 93%, respectively. He et al. ^[106] showed that 88% TCE was removed using carboxymethyl cellulose and Fe/Pd nanoparticle mixture after 596 days. Moreover, the presence of hydrogen improved the remediation performance. Bennett et al. ^[107] pointed out that the chlorinated ethenes at an aerospace facility were rapidly degraded by applying a carboxymethyl

cellulose and Fe/Pd nanoparticle combination. The reduction in oil concentration from contaminated soil using biosurfactant foam–nanoparticle mixture has been shown by gas chromatography (**Figure 3**).

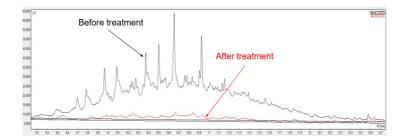


Figure 3. Peaks of oil pollutants before (black) and after (red) treatment by rhamnolipid biosurfactant foam and nZVI.

3.3. Effect of Some Factors on Soil Remediation Performance by Surfactant Foam-Nanoparticles

3.3.1. Effect of Environmental Conditions

The environmental conditions (or weather) are one of the most vital factors affecting the remediation efficiency of surfactant foam–nanoparticles. The weather may change the toxicity and biodegradability of surfactant foam, which alters the properties of the surfactant foam–nanoparticle mixture ^[108]. Toxicity and biodegradability are possible adverse effects of surfactant foam on the soil and the potential influence of soil microorganisms on the surfactant foam, respectively ^[109]. Due to the low biodegradability of most chemical surfactant foams used for soil remediation, combining these surfactant foams with nanoparticles may adsorb on the soil surface and harm the soil properties and soil microorganisms ^[110].

The adsorption of these surfactant foams, especially nonionic surfactant foam, on the soil surface can generate aggregation, which will alter the soil hydrophobicity, reduce soil retention, and cause toxicity to the soil ^[111]. The toxicity to the soil is more serious due to the potential absorption of surfactant foam into the plant roots, which can decrease crop growth and yield. In addition, the surfactant foam can break the cellular membrane, interact with lipids and proteins, and harm the soil microorganisms. The potential toxicity of some surfactants on soil microbes has been indicated in previous studies ^{[112][113]}. Therefore, biosurfactant foam, biologically produced from the microbial population, is suggested for soil remediation ^{[34][114]}.

3.3.2. Effect of Soil Characteristics

Soil type and particle size can affect the remediation rate. The smaller the soil particle size, the higher porosity and stronger bonds with oil pollutants, which will decrease the soil wettability and lead to lower treatment effectiveness. The remediation of motor oil from clay soil (soil porosity of 68.7%) was found to be lower than from desert soil and coastal soil (soil porosity of 42.5% and 37.5%) ^[115]. In another study, the removal performance of PCBs in clay soil was lower than in sandy soil due to the smaller desorption of PCBs in clay soil ^[17].

The presence of organic matter in soil components creates more competitive factors with oil pollutants in the mixture, which will inhibit and limit the removal efficiency of oil pollutants $^{[116]}$. The bond of organic matter molecules to the nanoparticle surface may generate a film that prevents the mass and electron transfer rate, resulting in a lower remediation percentage $^{[16]}$. Soil type also changes the activation energy connecting oil pollutants and soil surface, thus influencing the oil remediation efficiency $^{[117]}$. In particular, the binding of adsorbed oil compounds and soil particles may cause clogging and block the available pores, limiting the transport of flow through the pores and decreasing the removal effectiveness $^{[118]}$. A reduction in treatment efficiency was also observed by adding some salts representing the ionic strength in soil components $^{[40]}$. Consequently, treatment of oil compounds from contaminated soil greatly relies on the soil characteristics.

Soil pH also plays a critical role in the treatment efficiency of oil pollutants. Mańko et al. showed a change in CMC value and micelle generation because of the pH effect on the surface and interfacial tension of surfactant molecules, which will alter the oil treatment performance ^{[98][119][120][121]}. At low pH, more H⁺ ions are present, which makes the soil surface more positively charged, leading to a higher reduction of oil pollutants from the soil ^[122].

3.3.3. Effect of Nanoparticle Properties

The surface area of nanoparticles plays a vital role in the treatment effectiveness of surfactant foam-nanoparticle mixtures. The higher the surface area of nanoparticles, the greater the remediation rate. The larger surface area will lead to more interaction between nanoparticles and oil pollutants. In other words, more oil compounds may be adsorbed,

complexed, or reduced on the nanoparticle surface, resulting in higher treatment efficiency ^[123]. The high specific area also increases the agglomeration of nanoparticles due to their magnetic attraction, which may reduce their reactivity and mobility in soil, subsequently leading to lower remediation efficiency. However, the presence of surfactant foam may act as a stabilizer and inhibit nanoparticle aggregation ^[34]. The use of 20 nm Fe/Cu nanoparticles showed higher treatment efficiency of oil pollutants in soil than 200 nm Fe/Cu nanoparticles ^[40].

The interaction of nanoparticles and hydrophilic components of surfactant foam may prevent surfactant foam collapse and enhance foam stability and quality [10][40][124]. The repulsive electrostatic force between nanoparticles and surrounding liquid is improved due to the adsorption of surfactant molecules on the solid–liquid interface, which will lower the surface tension of the mixture, resulting in a change in remediation rate [125]. If the number of nanoparticles exceeds the threshold value, more surfactant molecules will be attracted to the solid–liquid interface. Therefore, fewer surfactant molecules appear at the gas–liquid interface, reducing the cohesive force between surfactant molecules. Consequently, the surfactant foam will collapse, and treatment efficiency will decrease [124]. The increase in nanoparticle quantity may also improve the attractive van der Waals force, decreasing the interfacial tension of surfactant and oil pollutants in soil is shown in **Figure 4**, where the oil treatment efficiency by biosurfactant foam/nanoparticle mixture is higher than only biosurfactant foam and only nanoparticles.

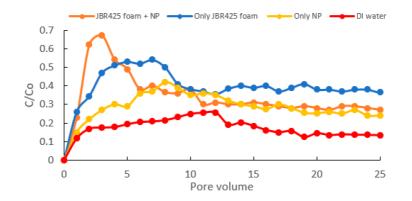


Figure 4. Oil treatment rate (C/Co) in column experiments using JBR425 rhamnolipid biosurfactant foam and Fe/Cu nanoparticles (NP) under various conditions.

3.3.4. Effect of Surfactant Concentration

The presence of surfactant foam may prevent nanoparticle aggregation, which increases the delivery and transport of nanoparticles in soil, leading to higher treatment efficiency ^{[34][127]}. Surfactant concentration changes the CMC value of the mixture, which may alter and surface and interfacial tension of oil pollutants, resulting in a change in oil treatment efficiency. Moreover, foam quality and stability greatly depend on the surfactant concentration. At pH 7, the combination of 2 vol% rhamnolipid and 2 wt% Fe/Cu nanoparticles displayed high foam stability and quality, leading to a better remediation rate of oil pollutants in soil ^[34]. Therefore, a suitable amount of surfactant can generate high foam quality, improving the aggregation of nanoparticles at the interface and increasing the interaction of a surfactant foam– nanoparticle mixture with oil pollutants, resulting in high effectiveness.

3.4. Limitations

Nanoparticles can penetrate organisms through ingestion or inhalation and cause some negative effects. The toxicity of nanoparticles to humans, animals, and soil microorganisms has raised some public concerns ^{[128][129]}. Some adverse effects are cell membrane damage, respiration interference, and DNA oxidative damage ^{[130][131]}. When nanoparticles enter the cell, they may concentrate at the cell membrane and increase their concentration on the cell surface. Some nanoparticles, such as nZVI or nano-iron oxide, can react with hydrogen peroxide on the cell surface to generate ROS, damaging the cell membrane ^[132]. In addition, nanoparticles may precipitate on the cell surface through the interaction with lipoteichoic acids in the cell wall, which will block the pores on the outer cell membrane, prevent nutrient transformation, and lead to the death of the cell ^[133].

However, the potential toxicity of nanoparticles is still controversial and needs more research. Vanzetto and Thome ^[134] found that nZVI caused no negative effect on the development of bacteria (*Bacillus* and *P. aeruginosa*) in pentachlorophenol-contaminated soil during the nanoremediation process. No major change in temperature, electrical conductivity, pH, and humidity of soil was observed after 90 days. Fajardo et al. ^[135] found no cytotoxicity on *Klebsiella planticola* bacteria in soil by the high concentration of nZVI. Nanoparticles have little or no adverse influence on the

growth of different fungi, such as *Trametes versicolor* and *Aspergillus versicolor*. In other studies, nanoparticles caused no significant toxicity on different bacteria, such as *P. stutzeri*, *Klebsiella oxytoca*, *P. putida*, or *Escherichia coli* under various incubation conditions ^{[132][136][137][138]}. The resistance mechanisms of bacteria or fungi are mainly due to the limitation of nanoparticle adsorption into the cell by some cell wall components, such as intracellular antioxidants, which decreases the adverse effects of nanoparticles ^{[125][126]}. Chitin cell walls also play a critical role in the low adsorption of nanoparticles, leading to their high resistance to nanoparticles ^{[121][131]}.

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