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Invited review

A review on remediation of spilled oil-contaminated soil from a pore-scale perspective

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Abstract:
Many innovative decontamination techniques, such as pulsed pumping and surfactant flushing, have been proposed to enhance the remediation performance of oil-contaminated soils. Their practical application is dependent on injection and extraction well. Therefore, these techniques can be viewed as an enhanced version of pump-and-treat technology. Since macroscopic flow phenomena are determined by microscopic fluid flow behaviors, conducting pore-scale studies on soil remediation will contribute to a deeper understanding of the remediation mechanisms associated with different pumping methods. This study examines the application of microfluidic experiments and pore-scale numerical simulations to the fluid dynamics of immiscible fluid displacement processes. The main application scenarios are reservoir development and CO_2 geological sequestration. Additionally, the primary distinction between soil remediation studies and the aforementioned scenarios is pointed out, i.e., the unsaturated initial fluid distribution. Finally, future research directions in soil remediation are discussed, emphasizing the fluid dynamic effects of initial contaminant distribution.

1. Introduction

Pump-and-Treat (P&T) technology is the most extensively utilized remediation technology for oil-contaminated soils (Fetter et al., 2017; Kahler and Kabala, 2019). However, its performance is suboptimal due to several factors. For example, preferential flow channels will form in the subsurface as a result of soil heterogeneity. This phenomenon triggers the formation of immobilized contaminant ganglia within the soil, thereby decreasing the remediation efficiency (Lin et al., 2022). The adsorption and dissociation of contaminants on soil grain surfaces can lead to a significant increase in the remediation time. Numerous measures have been proposed to improve the decontamination capacity of P&T technology. Pulsed pumping, periodically altering the pumping velocity during the remediation, was proposed by Kahler and Kabala (2016). They indicated that both deep sweeping with increased pumping velocity and vortex injection with reduced pumping velocity can accelerate the remediation. Suk et al. (2021) found that chaotic advection under pulsed pumping leads to a larger cumulative swept area than the pumping with a constant velocity. Similarly, adaptive pumping, exchanging the location of injection and extraction wells during the remediation process, can also effectively increase the swept area of the injected fluid, thus enhancing the remediation

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efficiency (He et al., 2022). However, achieving the cleanup target for contaminated soils is often challenging when using the aforementioned measures for P&T technologies (Obiri-Nyarko et al., 2014).

Many other decontamination methods, including permeable reactive barrier, chemically enhanced flushing technology, enhanced bioremediation, in-situ thermal treatment, and in-situ chemical oxidation, were developed to address the low remediation efficiency of P&T technology. Most of the above methods require injection and extraction well systems for practical application. The effectiveness of enhanced bioremediation technology is closely related to the mixing degree of injected nutrients and contaminants (Kao et al., 2016). The destruction and transformation of pollutants by chemical oxidizers is also controlled by the mixing degree (Tsitonaki et al., 2010). Numerous studies have been conducted on enhancing subsurface mixing due to the importance of the mixing degree (Cho et al., 2019). Previous studies have shown that adaptive pumping can effectively increase the interfacial area, thus facilitate enhanced the mixing degree of injected fluid and contaminants (He et al., 2023). Moreover, a larger interfacial area can promote the heat and mass transfer between the fluid (Basirat et al., 2017). In this sense, combining adaptive and pulsed pumping with remediation measures, such as hot water and steam flooding, can further improve the remediation performance. Since previous related studies are mostly based on field scales, they are unable to quantify the mixing degree of the injected fluid and contaminants. This limitation severely restricts the development of combined remediation technologies. Pore-scale studies can capture the flow details within soil pores, helping to fill the gap of field-scale studies. Undoubtedly, conducting pore-scale studies on oil-contaminated soil remediation can help to reveal remediation mechanisms of various pumping methods, thereby advancing the design and development of decontamination techniques.

Although pore-scale studies are relatively rare in soil remediation, they have been widely used in immiscible fluid displacement researches, such as enhanced oil recovery (Liu et al., 2022a; Zhou et al., 2024) and CO_2 geological sequestration (Chaturvedi et al., 2021; Zhu et al., 2024). The decontamination process also can be considered as an immiscible fluid displacement process. These subsurface applications have the same goal as soil remediation, i.e., achieving higher displacement efficiency. Therefore, previous studies on immiscible fluid displacement can provide references for the remediation of contaminated soil. The remainder of this study is structured as follows. Section 2 describes the direct visualization of fluid distribution within porous media based on microfluidic experiments. Section 3 focuses on the application of pore-scale numerical simulations for immiscible fluid displacement. Section 4 reviews pore-scale studies on the remediation of oil-contaminated soil. Finally, future developments in soil remediation research are discussed.

2. Direct visualization for fluid distribution

Pore-scale flow within porous media can be visualized by using optically transparent microfluidic models and microscopic imaging technology (Browne et al., 2020; Guo et al., 2022). Liu et al. (2024a) probed the effect of pore throat ratio, coordination number and tortuosity on the imbibition process in a polydimethylsiloxane microfluidic device. They noted that the imbibition velocity in regular channels is typically higher than that in irregular channels. Wu et al. (2021) conducted air-silicone oil displacement experiments in a rectangular microchannel, focusing on the transport and retention behavior of polyethylene particles within the silicone oil during the displacement process. They identified three transport patterns of particles, influenced by liquid withdrawal velocity and particle volume fraction, i.e., no deposition, particle entrainment, and particle layering within liquid films. In addition to these channels with relatively simple structures, researchers have also explored porous media flow in more complex structures using direct visualization methods. Ju et al. (2022) investigated the effect of pore topology skeleton on flow regimes and preferential flow paths using a 3D printed transparent microfluidic model. They observed a positive correlation between displacing fluid saturation and pore connectivity at breakthrough under imbibition condition, and a negative correlation under drainage condition. Lv et al. (2020) explored the transport mechanism of foams in porous media using a microfluidic model produced by polydimethylsiloxane. The foam transport process, along with the effect of permeability on bubble morphology and gas trapping in both homogeneous and heterogeneous models, was observed by direct visualization. They indicated that the bubble regeneration mechanism for trapped foam is snap-off and lamella division, while for flowing foam is mainly lamella division, as shown in Fig. 1(a). Compared to the high permeability model, the trapped bubbles in the low permeability model exhibit smaller diameters but a higher global gas saturation.

Etched microfluidic models were also widely used in pore-scale studies (Anbari et al., 2018). Jafari et al. (2017a) probed the evolutionary behavior of the fluid interface under the interaction of matrix and fracture, using a fracture microfluidic model obtained by etching techniques. They found that higher injection velocities, greater matrix porosity, and the presence of lateral fractures all contribute to an increase in ultimate oil recovery. Song et al. (2020) analyzed the effect of circular and square grains on the CO_2 displacement process using brine based on a glass-etched pore throat micromodel. The wetting phase primarily exists as films, bridges, and pools. Besides, the microfluidic model with square grains has a better sequestration capacity for CO_2. Hu et al. (2017) also observed the trapping phenomenon of Supercritical CO_2 ($scCO_2$) under different wettability, as shown in Fig. 1(b). The dynamic oil displacement process of polymers was visualized by Buchgraber et al. (2011). The results show that the higher viscosity of the polymer can change the displacement regime. It can inhibit the fingering phenomenon and delay the breakthrough time, thus enhancing the oil displacement efficiency. By combining microfluidic experiments with Micro-PIV technology, Roman et al. (2019) obtained the velocity field distribution of the captured fluid at the end of displacement and analyzed the momentum transfer between fluids.

Numerous similar studies for direct visualization of mul-

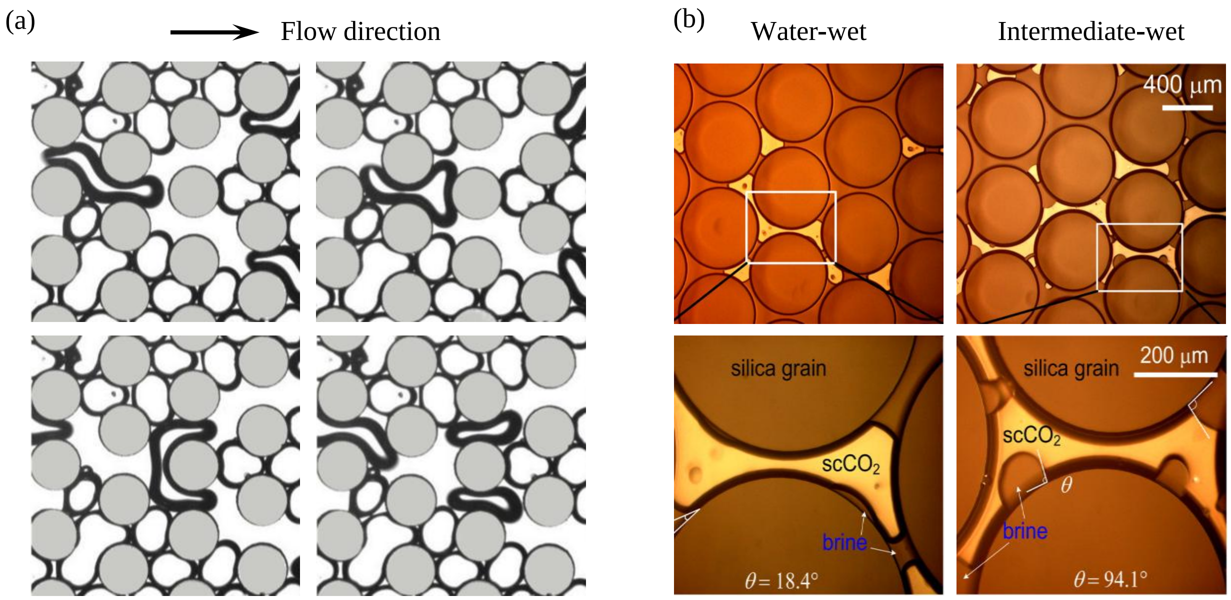

Fig. 1. Visualization experiment of microfluidic model. (a) Bubble regeneration process based on lamella division (Lv et al., 2020) and (b) $scCO_2$ capillary trapping phenomenon (Hu et al., 2017).

Table 1. Summary of studies on direct visualization of multiphase flows in porous media.

Fluid types	Grain shape	Research emphasis	References
Water, oil	Square	Pore aspect ratio, aspect ratio, flow rates	Kim et al. (2023)
Water, air	Irregular	Mixed wettability	AlOmier et al. (2024)
0.01% HCl	Round	Mineral dissolution	Musabbir Rahman et al. (2025)
Water, oil, N_2	Irregular	N_2 injection after water-flooding	Su et al. (2022)
Water, oil, CO_2	Irregular	CO_2 injection after water-flooding	Zhang et al. (2022)
Water, fluorinert	Round	Effects of flow rates on fluid connectivity	Dastjerdi et al. (2023)
Brine, $scCO_2$	Irregular	Mixed wettability and flow rates	Chang et al. (2020)
Brine, decane	Irregular	Wettability and flow rates	Zou et al. (2024)
Water, oil, polystyrene particles	Round	Particle transport and aggregation	Wu et al. (2025)

tiphase flows within porous media have been conducted, as shown in Table 1. Researchers have used direct visualization experiments to investigate the effects of various factors, including flow rates, wettability, fluid type, and channel structure, on the fluid displacement process within porous media. These experiments have deepened our understanding of the immiscible fluid displacement process. However, the microfluidic chips adopted in these studies typically have a uniform depth along the vertical direction. This geometric confinement induces a neglect of capillary pressure variation in the depth direction, and thus affects the flow instability (Liu et al., 2021). Lei et al. (2023) proposed an optimized sequential lithography and successfully fabricated a microfluidic chip with three distinct depths. Their experimental results show that unstable interfacial phenomena within uniform depth porous media are inhibited compared to those in depth-variable microfluidic porous media. To better model the real porous media, the three-dimensional structure effects of pores, such as different depths and pore connectivity, should be fully considered in the future fabrication of microfluidic chips.

3. Numerical simulations for fluid displacement

Compared to microfluidic model displacement experiments, pore-scale simulation can provide more abundant flow field information, including velocity field and pressure field distribution, enabling quantitative analysis of immiscible fluid displacement mechanisms. Pore-scale simulation methods are typically classified into Pore Network Model (PNM) and Direct Numerical Simulation (DNS) (Qin et al., 2024). PNM approximates real pores as an interconnected network of regular geometries, such as spheres, cubes, and pipes. The calculation process of multiphase flow is performed based on the predetermined capillary entry pressure and algorithmic, resulting in significantly lower computational cost compared

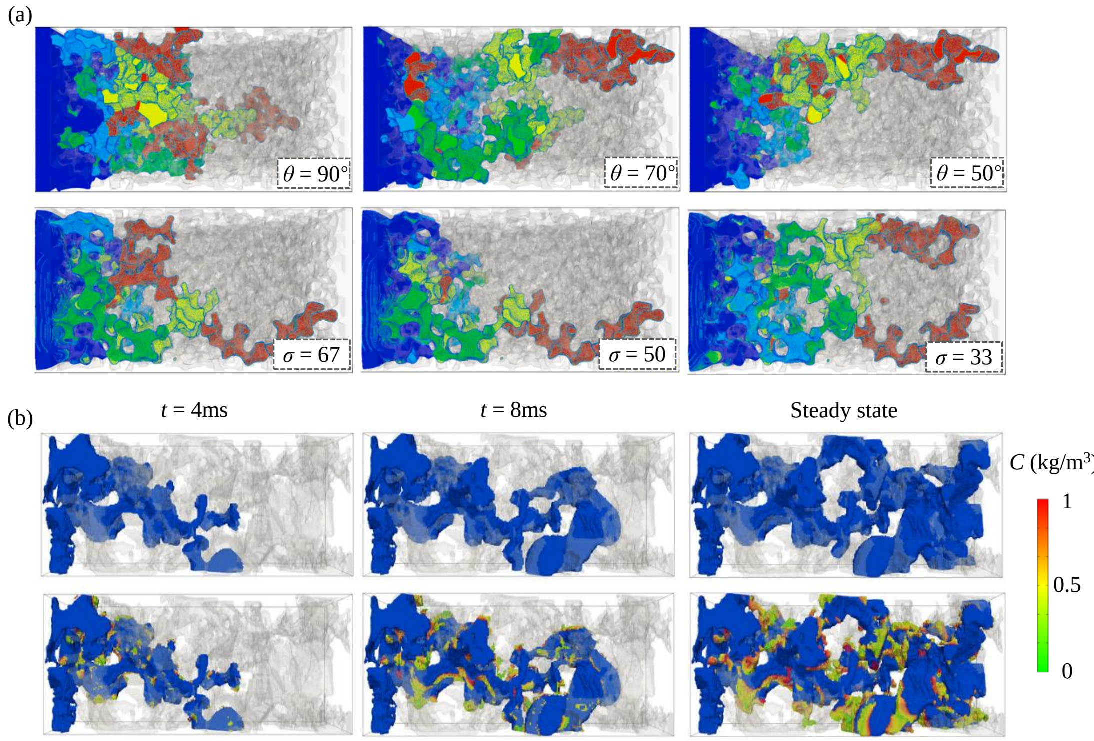

Fig. 2. (a) Effects of contact angle and interfacial tension on the water-oil displacement process (Liu et al., 2024b) and (b) CO_2 geological sequestration process (Yang et al., 2023).

to DNS. However, this simplification leads to the inability of PNM to obtain detailed ganglion dynamics within the pore space (Berg et al., 2016). DNS is modeled based on the realistic fluid flow space. Therefore, it can capture pore-scale phenomena, such as interfacial rupture and coalescence. DNS methods are classified into two categories: the lattice Boltzmann method and methods based on the Navier-Stokes equations, including the Volume-of-Fluid (VOF), the Phase-Field Method (PFM), and the level-set method (Liang et al., 2023; Cai et al., 2024). The most widely applied scenarios for these methods in immiscible fluid displacement are reservoir development and CO_2 geological sequestration.

3.1 Reservoir development

The process in which a wetting fluid displaces a non-wetting fluid is referred to as imbibition, whereas the reverse process is known as drainage (Ju et al., 2019). The reservoir development process is generally dominated by imbibition, but drainage also plays a role. For example, Kong et al. (2020) obtained the evolution of the interface within porous media during gas-assisted gravity drainage based on PFM. They considered the effects of gas injection velocity, viscosity ratio, density difference, and pore diameter. The results showed that the oil recovery factor is negatively correlated with the injection velocity and viscosity ratio, and positively correlated with the density difference and pore diameter. The imbibition process is classified into co-current and counter-current types, depending on the flow directions of the two fluids (Gu et al., 2019).

The oil-water displacement is the most common imbibition process (Cai et al., 2025). Liu et al. (2024b) probed the effect of capillary number on the dynamic displacement process based on the lattice Boltzmann method, as shown in Fig. 2(a). The simulation results show that the displacement efficiency increases as the capillary number increases. At high capillary numbers, the flow is dominated by ganglion dynamics. The snap-off phenomenon of the injected fluid results in a large number of residual ganglia at low capillary numbers. (Yang et al., 2021) obtained the evolution of the fluid interface during water-oil displacement in a three-dimensional pore based on the VOF method. Recirculation in the trapped residual oil within the pore corners was observed. This phenomenon is classified into two forms, namely the co-current driven pattern and lid-cavity driven pattern, based on differences in the circulation direction. The form of the recirculation phenomenon will change when the wettability of the matrix is altered.

The counter-current imbibition is the fundamental displacement mechanism in low-permeability fractured reservoirs. Rokhforouz and Akhlaghi Amiri (2017) constructed a two-dimensional model using circular particles with diameters ranging from 0.6 to 1.15 mm. They explored the counter-current spontaneous imbibition process under the effects of wettability, interfacial tension, and viscosity ratio using the PFM. Using the same model, Jafari et al. (2017b) probed the effects of injection velocity, grain shape, and fracture diameter on the dynamic spontaneous imbibition process. They observed pore-scale events, such as snap-off, oil film thinning, interface coalescence, and water film bridging, and found that the square-grain model yielded a 12% higher oil recovery than the circular-grain model. Zhu et al. (2021) obtained the oil-water interface evolution within a porous medium, considering the influence of tree-shaped fracture morphology (tortuosity, angle, and width) using the level-set method. They found that the flow involves a combination of co-current and counter-current imbibition at the initial displacement stage, with counter-current imbibition gradually dominating as displacement proceeds. Several similar studies have explored the effects of heterogeneity (Liu et al., 2021), fracture spacing (Gu et al., 2019), and surface roughness (Liu et al., 2022b) on the spontaneous imbibition process.

3.2 CO_2 geological sequestration

Different from reservoir development, CO_2 is the displacing fluid in geological sequestration. Basirat et al. (2017) analyzed the evolution pattern of interfacial morphology during CO_2 and brine displacement based on PFM. They quantitatively evaluated the effect of wettability on macroscopic parameters, such as residual brine saturation, capillary pressure, relative permeability, and specific surface area. The results showed that both brine saturation and interfacial area within the porous media decreased with decreasing contact angle at the end of the displacement, while the CO_2 breakthrough time was less affected. Based on the same method, Liu et al. (2020) captured the dynamic drainage process of CO_2 under different wettability, interfacial tension, and injection velocity. They found that the ultimate CO_2 saturation demonstrates a positive correlation with the injection velocity and contact angle, while exhibiting a negative correlation with the interfacial tension. Zhou et al. (2023) modeled the displacement process of CO_2 and oil in a 2D heterogeneous porous media. Similar to Liu et al. (2020), they also focused on the two key factors: wettability and injection velocity. The simulation results demonstrated a non-monotonic relationship between CO_2 saturation and contact angle.

In addition to two-phase flow, researchers have begun to focus on the interphase mass transfer (Wang et al., 2023; Tariq et al., 2024) and mineral reaction processes (An et al., 2021) in geological sequestration. Based on VOF and continuous species transfer method, Yang et al. (2023) probed the flow and dissolution processes of $scCO_2$ at the pore-scale, as shown in Fig. 2(b). They indicated that the dissolution of $scCO_2$ is highly dependent on the distribution of flow paths. Li et al. (2024) obtained the effect of CO_2 concentration on interfacial tension, viscosity, and contact angle using molecular dynamics simulations. These effects were then upscaled to the pore scale based on lattice Boltzmann simulations. A phase diagram was proposed to evaluate the relationship between residual oil saturation, initial oil saturation, and CO_2 concentration in the water-oil-CO_2 system. Zhu et al. (2024) modeled the CO_2-oil miscible flooding process, considering the viscosity reduction and swelling effects induced by mixing. They indicated that the displacement pattern before breakthrough is determined by both the molecular and convective diffusion, whereas after breakthrough, it is primarily controlled by molecular diffusion. For accurate modeling the process of CO_2 geological sequestration, future studies should consider the interactions of convection, diffusion, and reactive transport mechanisms.

4. Pore-scale studies on soil remediation

Pore-scale experiments and numerical simulations have been widely employed to investigate the mechanisms related to enhanced oil recovery and CO_2 geological sequestration. However, their application is relatively less in the remediation of oil-contaminated soil. Wang et al. (2022) visualized the degradation process of trichloroethylene by permanganate using microfluidic experiments. They evaluated the effects of injection velocity, oxidant concentration, and stabilization supplement on the remediation performance. The mobilization and solubilization of contaminants by four surfactants were also quantified. The results showed that sodium dodecyl sulfate exhibited the strongest mobilization of contaminants, while sorbitan monooleate (Tween 80) demonstrated the highest solubilization capacity (Wang et al., 2023). Jung et al. (2016) explored the displacement process of polyacrylamide solutions for decane. The experimental results showed that the viscosity of polyacrylamide solutions is positively correlated with the solution concentration. The economic concentration in soil remediation is about 5 g/L. Aitkhozha et al. (2025) explored the remediation mechanism of aqueous xanthan gum solution on oil-contaminated soil in glass micromodels. The effects of polymer concentration, injection pressure, and wettability on residual oil saturation were quantified. They found that the blocking effect of xanthan gum on high-conductance flow paths resulted in a higher pollutant removal efficiency compared to water flooding.

Pore-scale numerical simulations were also employed to investigate the remediation mechanism of oil-contaminated soil. He et al. (2023) captured the evolution of the oil-water interface within the porous media under adaptive pumping through PFM. They revealed the effects of boundary exchange time and pore topography on remediation performance and indicated that adaptive pumping can obtain a larger interfacial area than positive pumping (He et al., 2022). Pu et al. (2025) explored the factors that affect the performance of surfactant flushing technology, including the viscosity of the flushing agent, flushing velocity, interfacial tension, and soil wettability. Zhang et al. (2024) modelled the mobilization of captured pollutant ganglia within a biconical pore structure and indicated that the asymmetric jump mechanism facilitates their mobilization. Yang et al. (2024) proposed a remediation reagent, colloidal biliquid aphron (CBLA), and compared its performance in remediating contaminated soil to that of water using pore-scale simulation. They found that the shear thinning property of CBLA significantly enhanced the contaminant removal efficiency.

The aforementioned studies can help to enhance the understanding of the remediation mechanisms associated with various decontamination technologies. However, these studies generally assume that the initial state of the pore space is oil-saturated. Due to the heterogeneity of the soil and the presence of dead-end pores, the spilled oil is impossible to

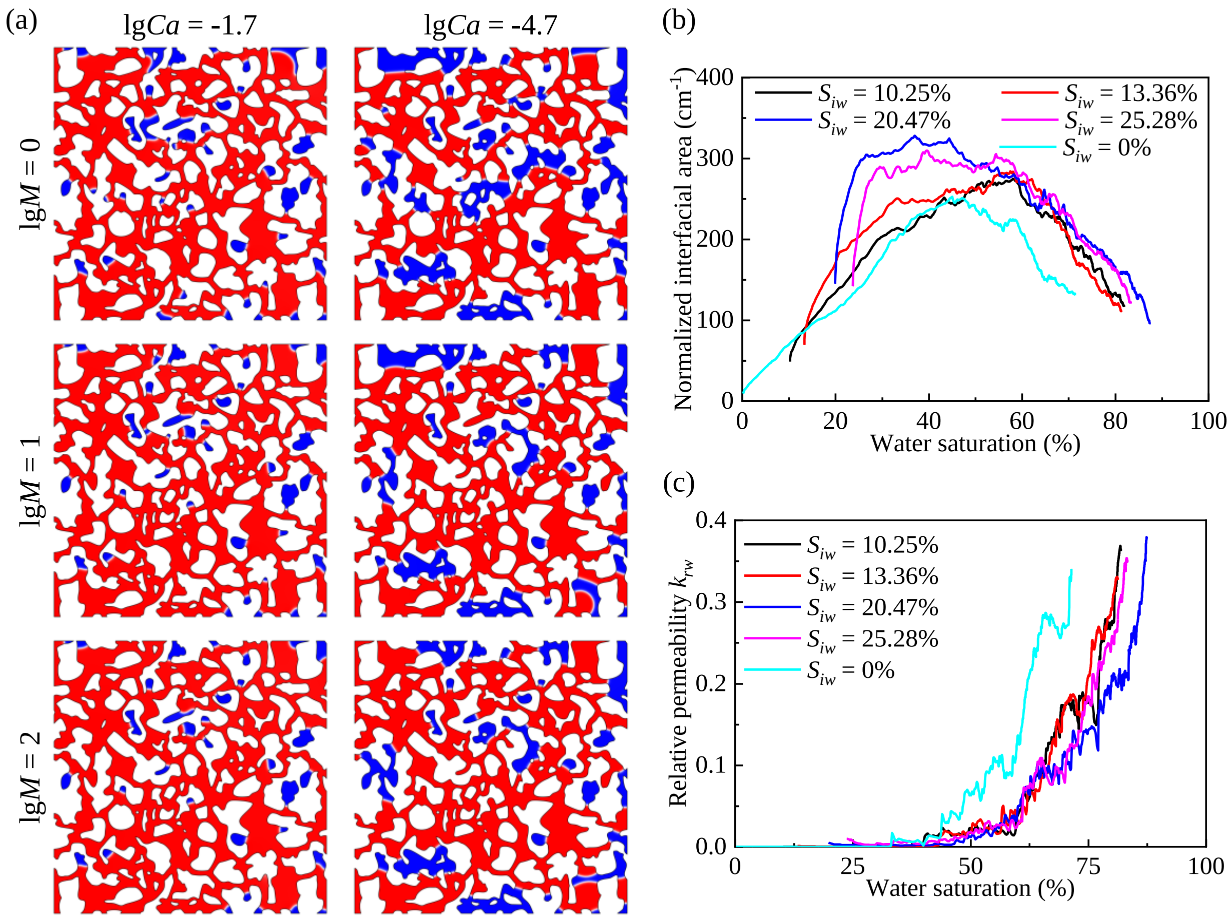

Fig. 3. (a) Effect of capillary number and viscosity ratio on spilled oil distribution. The spilled oil and water are marked in red and blue, respectively, (b) normalized interfacial area, and (c) relative permeability as a function of water saturation under different initial water saturation (He et al., 2024).

fully saturated the pore space after an oil spill accident, as shown in Fig. 3(a). Therefore, this assumption will inevitably affect the morphology of flow pathways within the porous media and the remediation efficiency. He et al. (2024) investigated the dynamic evolution of fluid interfaces under various initial water saturations. They indicated that the initial fluid distribution will significantly affect the interfacial area and relative permeability, as shown in Figs. 3(b) and 3(c). Moreover, the morphology of contaminant distribution within soil pores significantly influences its dissolution behavior in the injection solvents (Hu et al., 2021). In this sense, the effect of initial contaminant distribution on remediation efficiency should be highlighted in future studies of oil-contaminated soil remediation.

5. Conclusion and prospect

Numerous decontamination techniques, such as hot water flooding and surfactant flushing, have been proposed for oil-contaminated soils. Similar to P&T technology, these methods also require injection and extraction well systems. The decontamination process can be considered as an immiscible fluid displacement process due to the low solubility of spilled oil in groundwater. Macroscopic flow phenomena are governed by microscopic flow features. Therefore, conducting pore-scale studies on the remediation of oil-contaminated soil can strengthen the understanding of the remediation mechanisms under various pumping methods.

Although numerous pore-scale studies have been conducted to investigate the immiscible fluid displacement process, fewer studies have focused on the remediation of oil-contaminated soil. Previous researches on soil remediation typically assume that the pore space is fully saturated with spilled oil before the remediation, which is inconsistent with the actual spill scenario. The unsaturated initial state is the crucial difference between soil remediation and other immiscible fluid displacement scenarios. Additionally, soils are classified as saturated and unsaturated based on water saturation levels. Current studies mainly focus on the decontamination process in saturated soils. The presence of the gas phase in unsaturated soils further complicates the migration of contaminants within the soil pores.

Future studies should sufficiently consider the real initial distribution morphology of spilled oil, including the oil-water distribution within saturated soils and the oil-air-water distribution within unsaturated soils. Undoubtedly, it will provide more accurate fluid dynamics characteristics and remediation mechanisms for oil-contaminated soils. The effect of geometric confinement, such as pore depth and connectivity, on the immiscible fluid displacement process should be quantified in microfluidic experiments. More realistic three-dimensional soil pore models should be adopted for soil remediation simulations, given the advancements in computing power and digital core technology. Furthermore, more complex fluid interaction mechanisms, contaminant dissolution, and chemical reactions between the injected solvent and the contaminant should be considered. Future field-scale studies can be conducted based on parameters such as relative permeability and capillary pressure curves obtained from pore-scale studies. This upscaling process allows the pore-scale results to guide the development of decontamination strategies.

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Conflict of interest

The authors declare no competing interest.

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