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Review

Advantages and challenges of Tween 80 surfactant-enhanced technologies for the remediation of soils contaminated with hydrophobic organic compounds

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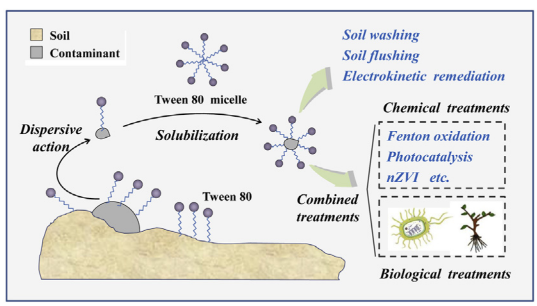
HIGHLIGHTS

- The application of Tween 80-enhanced technologies for soil remediation is reviewed.
- Tween 80 improved the performance of soil washing, flushing and other treatments.
- The performance of these technologies is largely affected by soil properties.
- Further work is required to investigate economic or operational problems.
- Possible improvements and outlooks of Tween 80-enhanced technologies are proposed.

GRAPHICAL ABSTRACT



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ABSTRACT

The occurrence of hydrophobic organic compounds (HOCs) in soil has become a highly significant environmental issue. This problem has been exacerbated by the strong sorption of HOCs to soil and the absence of remediation technologies that have been tested at full-scale. More and more studies show that surfactants are able to solubilize HOCs from the soil, thus to enhance the remediation efficiency. Among these surfactants, Tween 80 has gained particular interest due to its low cost, low polarity, low toxicity and high solubilization capacity. This review aims to highlight the development of Tween 80-enhanced technologies for HOCs removal. Specifically, it provides an overview of HOCs removal by Tween 80-enhanced soil washing, soil flushing and combined treatments (Tween 80 extraction coupling with Fenton processes, electrochemical oxidation, etc.). We present a general and rigorous review on this topic, from the basic understanding of Tween 80-enhanced solubilization of sorbed HOCs to the current new techniques developed until 2016. Besides, possible improvements and outlooks of Tween 80-enhanced technologies are proposed.

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Contents

1. Introduction

The occurrence of hydrophobic organic compounds (HOCs) in the soil is becoming a global problem because of their potential toxicity, carcinogenicity and mutagenicity [1]. HOCs such as petroleum hydrocarbons, polycyclic aromatic hydrocarbons (PAHs) and polychlorobiphenyls (PCBs) are characterized by a high octanol/water partition coefficient (Kow), and thus have a tendency to tightly bind onto the soil particles. The strong sorption of HOCs to the soil represents a significant limitation to the remediation of HOCs-contaminated soils. Therefore the slow desorption of HOCs from soil is a key problem to solve for improving the remediation efficiency [2].

A lot of studies have addressed that addition of surfactants can cause increased removal of soil-bound HOCs by washing or other remediation processes. Various recent reviews [3–8] show surfactants-enhanced technologies produce good results for HOCs removal and the current level of interest in this area. Surfactants are able to enhance the apparent solubility of HOCs, thereby enhancing the remediation efficiency. In general, surfactants are classified into cationic, anionic and nonionic by the chemical nature of the hydrophilic part [9]. A lot of studies have shown that nonionic surfactants are more desirable than cationic and anionic surfactants for soil remediation [10–13], since cationic surfactants would be easily sorbed onto soil particles [14], while anionic surfactants can be precipitated with the cations in soil [15]. Besides, nonionic surfactants generally have higher solubilization capacities and economic benefits [16].

Among the nonionic surfactants, polyoxyethylene-(20)-sorbitan monooleate (Tween 80, $C_{64}H_{124}O_{26}$), has gained particular interest. Tween 80 shows all the positive features of nonionic surfactants. Compared to other nonionic surfactants, it is of low cost and low toxicity on soil microorganisms [17,18]. Since its first use about 25 years ago [19], Tween 80 has been widely used in soil remediation to clean up HOCs. To our knowledge, several papers [1,3,4,20,21] and few book chapters [22,23] have summarized some data on the performance of Tween 80 in enhancing soil remediation. However, the information about Tween 80 are mixed with those of other surfactants, and to date, a sole evaluation of the studies of Tween 80 in soil remediation has not been reported.

The general focus of this review is to summarize the progresses made in developing Tween 80-enhanced technologies for the remediation of HOCs-contaminated soils. First, an overview of the desorption and solubilization of the sorbed HOCs by Tween 80, with special emphasis on the interaction of Tween 80 and soil particles, is described to present the fundamental of the Tween 80 related technologies. Then Tween 80-enhanced soil washing and flushing treatments are detailed to show the solubilization performance of Tween 80 in different systems. The joint application of Tween 80 solubilization and other techniques is also summarized and discussed. Besides, the outlooks of all these Tween 80-enhanced technologies for the upcoming future are envisaged.

2. Application of Tween 80 to soil–water systems

2.1. Desorption and solubilization of the sorbed HOCs

Solubilization and lowering of the surface and interface tension are the main mechanisms for a facilitated transport of hydrophobic pollutants adsorbed on the solid phase to the aqueous phase [24,25]. The proposed mechanisms for desorption and solubilization of HOCs in a soil–water system are shown in Fig. 1 [4]. Normally, the hydrophilic group of Tween 80 is apt to enter into the water and the lipophilic group tends to combine with soil particles or the pollutants. At a low concentration, Tween 80 monomers mainly accumulate at the soil–water interface [26,27], causing repulsion between the solid phase and the hydrophilic group of the Tween 80 molecule, thereby promoting the desorption of the pollutants [28]. As Tween 80 concentration increases, the interfacial solvent (water) would be gradually replaced by Tween 80 molecules, leading to a lower surface tension. When Tween 80 concentration further increases, and above a critical aqueous concentration, known as critical micelle concentration (CMC), spheroidal or ellipsoidal micelles form. In soil–water systems, the hydropho-

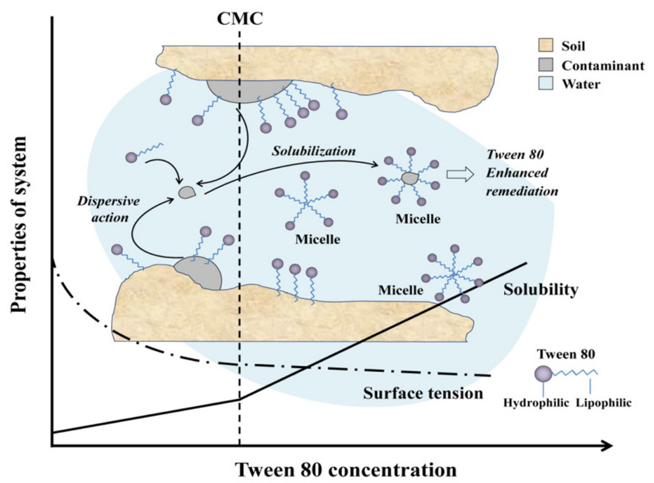


Fig. 1. Schematic of Tween 80-enhanced remediation of contaminated soils [4].

bic cores of Tween 80 micelles can compete strongly with the soil particles for the partitioning of hydrophobic pollutants. The solubilized HOCs in the water phase have better mobility, and therefore show better availability to the subsequent treatments (soil washing, soil flushing, etc.).

In the absence of surfactants, the distribution of HOCs in a soil–water system is mainly depend on soil organic matters (SOM), and according to the partition theory it can be determined by the soil–water distribution coefficient K_d [29]. In the presence of Tween 80 surfactant, some studies proposed that the distribution of HOCs in a soil–water system could be estimated by the following model [30,31]:

$$K_d^* = \frac{K_d(1 + f_{sf}K_{sf}/K_d)}{1 + X_{mn}K_{mn} + X_{mn}K_{mc}} \quad (1)$$

where K_d^* is the distribution coefficient for HOCs (L kg^{-1}) in a soil–water–Tween 80 system, K_d is the sorption coefficient of HOCs by soil in a Tween 80 free system (L kg^{-1}), f_{sf} is the amount of soil-sorbed Tween 80 (mg kg^{-1}), K_{sf} is the distribution coefficient of HOCs with the soil-sorbed Tween 80 (L kg^{-1}), X_{mn} is the concentration of Tween 80 monomers in water (g L^{-1}), X_{mc} is the concentration of Tween 80 micellars in water (g L^{-1}), K_{mn} is the partitioning coefficients of HOCs with Tween 80 monomers (L g^{-1}), and K_{mc} is the partitioning coefficients of HOCs with Tween 80 micellars (L g^{-1}).

Molar solubilization ratio (MSR) has been widely applied to assess the solubilizing efficiencies of HOCs by Tween 80. It can be calculated by the following equation [32]:

$$\mathrm{MSR} = \frac{S_{w-}S_{CMC}}{C_s - \mathrm{CMC}} \quad (2)$$

where Sw is the solubility of HOCs (mol L^{-1}) in micellar solution at Tween 80 concentration > CMC, S_{CMC} is the solubility of HOCs (mol L^{-1}) at the CMC, Cs is the Tween 80 concentration (g L^{-1}) at which Sw is evaluated in moles per liter. In other words, MSR can be obtained from the slope of the linear fitted line on the plot between the concentrations of HOCs against Tween 80 concentrations above CMC.

2.2. Soil absorption of Tween 80

An important consideration is that the sorption of Tween 80 onto soil may result in a proportion of Tween 80 being unavailable for the micellar solubilization of HOCs. Researchers have found that the solubilization of PAHs in the soil–water system occurs at Tween 80 concentration much higher than the clean water Tween 80 CMC [33,34]. Same phenomenon were observed in the systems using other nonionic surfactants, furthermore, the adsorption of nonionic surfactants onto soils was found highly associated with clay minerals and SOM [35–38]. Some researchers indicate that the sum of CMC and maximum sorption of Tween 80 onto the soil can be used for the estimation of the "effective CMC" in the soil–water system [34]. Further researches demonstrated that the sorption isotherms of Tween 80 onto soil can be well described by the linear Langmuir sorption model (Eq. (3)) [39,40]. And pollutants in the soil did not change the best fitted equations and shape of adsorption isotherm of Tween 80 to soil [40].

$$\frac{1}{q} = \frac{1}{q_m} + \frac{1}{Kq_m} + \frac{1}{c} \quad (3)$$

where q is the amount of soil-sorbed Tween 80 (mg g^{-1}), q_m is the maximum amount of soil-sorbed Tween 80 (mg g^{-1}), K is the adsorption constant of Tween 80, and c is the equilibrium concentration of Tween 80 in aqueous phase (mg L^{-1}).

Recently, Kang and Jeong [37] studied the sorption behaviors of Tween 80 by oxides (alumina, silica and hematite) and aluminosilicate minerals (plagioclase, vermiculite, kaolinite and montmorillonite). Results showed that the sorption by alumina and silica were well fitted into linear isotherms, and the sorption by hematite and aluminosilicate minerals can be described by Langmuir isotherm (Fig. 2). The authors concluded that, for alumina and silica,

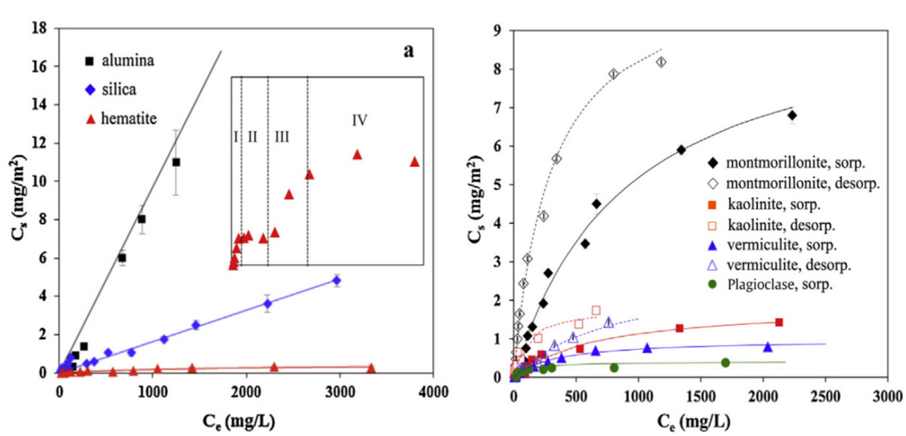


Fig. 2. Sorption isotherms of Tween 80 by oxides (a) and aluminosilicates (b). Batch compositions were 100 mM NaCl and 60 M carbonate buffer at pH 6.8. Error bars indicate one standard deviation. Lines are the data fits using linear or Langmuir isotherm models. The inset of part (a) is an enlarged plot of Tween 80 sorption by hematite. Adapted with permission from ref 37. Copyright 2015 Elsevier.

Tween 80 partitions into the interface between them and water; but for hematite and aluminosilicates, Tween 80 binds to the surface of them.

2.3. The toxicity of Tween 80

An additional concern is that Tween 80 may bring about some potential ecotoxicologic effects. Introduction of surfactants to the soil may alter the physicochemical properties of the soil [41]. More importantly, some surfactants can persist indefinitely in the soil and pose a potential threat to the environment [42]. As for Tween 80, it is considered non-toxic and it shows no carcinogenic activity [43]. The potential toxicological impacts of Tween 80 on soil environment have been studied by a lot of researchers. In one study, the tolerance of Tween 80 by 18 different fungal strains was investigated [44]. The results showed that the growth of all the strains was not affected by Tween 80. The findings are in agreement with the observations of many other researchers [18,45–49], who observed that Tween 80 has little or no negative effects on soil microorganisms.

Tween 80 treatments also showed no obvious inhibition to the plant growth. Zheng et al. [50] observed that Tween 80 have no apparent toxicity to the growth of ryegrass (*Lolium multiflorum*) and red clover (*Trifolium pretense*) in the range of tested concentrations (500–2500 mg kg^{-1}). The same authors also found Tween 80 (10 × CMC) has no phytotoxicity on the plants growth in an aqueous solution [51]. In some cases, the plant biomass yields with Tween 80 were even higher than that without Tween 80 [2,50]. This could be due to Tween 80 having potentially bioavailable carbon. Besides, Tween 80 may able to increase root permeability, leading to a more efficient uptake of nutrients from the soil.

The feasibility of Tween 80-enhanced treatments is also enhanced by the fact that Tween 80 will not bring long-term impact on soils. It was found that Tween 80 resulted to be extremely biodegradable and mineralisable under aerobic and anaerobic conditions also by not specialised soil bacteria [46,52].

3. Tween 80-enhanced soil washing treatments

Soil washing or solvent extraction technology is a mechanical process that uses liquids to remove the contaminants from the soil into the aqueous solution. Fig. 3 illustrates the general procedures of the ex-situ Tween 80-enhanced soil washing process [4]. Briefly, the excavated polluted soil is pretreated (e.g., remove the rocks and branches) and mixed with Tween 80 solution, and agitated. After the extraction process, the soil particles can be separated out and Tween 80 can be regenerated for next round use.

Studies have indicated the concentrations of Tween 80 plays a crucial role in the removal of HOCs. In soil washing processes, Tween 80 micelles are necessary for the solubilization of sorbed HOCs. As found by Grasso et al. [53], only negligible amount of PAHs were desorbed from a real polluted soil when the concentration of Tween 80 was below or close to the CMC. The CMC of Tween 80 in pure water is about 13.0 mg L^{-1}. But due to the absorption by soil, 1–10 g L^{-1} of Tween 80 was generally applied to achieve relatively high HOCs removal [16,54–58]. Some researchers investigated the further increase in Tween 80 concentration on the removal of HOCs. However, results showed the increase in Tween 80 concentration from 10 g L^{-1} to 30 g L^{-1} cannot cause a significant increase in the removal of PAHs [59].

The desorption efficiency of Tween 80-enhanced soil washing also depends on the SOM content [60]. It is generally accepted that the content of SOM is the most important parameter in the adsorption and mobility of HOCs [60,61]. On the one hand, the HOCs can be strongly sorbed to the SOM, leading to low desorption and mass transfer rates. On another hand, some researchers showed the adsorption of Tween 80 on soil is increased with increasing SOM content of the soils [62,63]. Compared to medium and coarse textured soils, the fine textured soils have higher sorption capacity and hence show less sensitivity to the contaminants leaching [64]. In particular, surfactant-enhanced washing can be ineffective for soils that have more than 20% SOM content [65].

3.1. Comparison between Tween 80 and other nonionic surfactants

According to the literature, apart from Tween 80, the other two nonionic surfactants (Brij 35 and Triton X-100) also have attracted much attention in surfactant-enhanced soil washing. There are many studies focus on the comparison of Tween 80 and the other two nonionic surfactants for washing HOCs from the soil [16,57–59,66–69]. In one study, Brij 35 gave slightly higher removal of PAHs compared to Tween 80, under the same conditions [16]. However, this result is not in agreement with that reported by Dhenain et al. [68]. The authors observed that Tween 80 gave

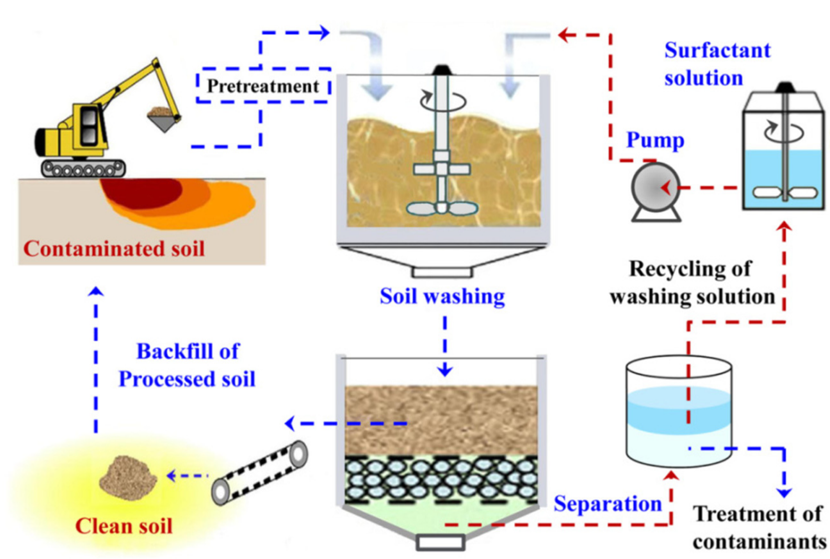
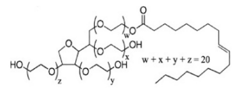
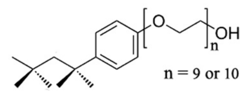
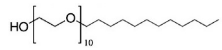


Fig. 3. General procedure of Tween 80-enhanced ex-situ washing for soil remediation [4].

Table 1
Physicochemical properties of the three widely studied surfactants.

Compound	Formula	Structure	Molecular weight	CMC[b] (mg L^{-1})	HLB[c]
Tween 80	$C_{18}S_6$[a] $(CH_2CH_2O)_{20}$		1310	13.0	15.0
Triton X-100	$C_8H_{17}C_6H_4O(CH_2C\ H_2O)_{9.5}H$		628	142.6	18.7
Brij 35	$C_{12}H_{25}O(CH_2CH_2O)_{23}H$		1200	51.4	22.2

[a] S_6 is a sorbitan ring ($C_6H_9O_5$).
[b] CMC, critical micelle concentrations of different surfactants measured by Cheng and Wong [57].
[c] Hydrophile lipophile balances were adopted from Davies and Rideal [70].

better results than Brij 35 with PAHs removal, at rates of over 35% for several PAHs. To compare the results from different studies is difficult because the environmental and operational conditions (e.g., soil type, pollutants type and pollutants type) are often based on different assumptions. Consequently, they could lead to fairly different results. Nevertheless, a lot of authors have pointed out the preeminent role of Tween 80 for the extraction of HOCs from soils among the tested nonionic surfactants in their experiments under the same conditions [16,55,57–59,66,69]. For example, several researchers have observed that the extraction of phenanthrene from the soils was in the order of Tween 80 > Triton X-100 > Brij 35 in different experiments [57,58,66]. The better results with Tween 80 is probably due to the lower CMC and higher hydrophile-lipophile balance (HLB) compared to Triton X-100 and Brij 35 (Table 1) [70]. As shown in Table 1, Tween 80 has a C24 in the hydrophobic tail which is much longer than of Brij 35 (C12) and Triton X-100 (14). As a result, Tween 80 is more likely present in the water phase. Tween 80 was also demonstrated better than some other nonionic surfactants such as Tween 40 [58] and Tyloxapol [16] in washing HOCs out from the soil solid phase into solution. All these results suggest that Tween 80 has great potential for practical application of surfactants-enhanced soil washing treatments.

3.2. Sdbs–Tween 80 mixed surfactants system

Apart from the solubilization capacities of surfactants, the amount of sorbed surfactants is also the major factor influencing desorption and solubilization of HOCs during surfactants-enhanced remediation processes. To reduce the loss of Tween 80 by soil adsorption, SDBS–Tween 80 mixed surfactants have been developed. The mixed surfactants have gained practically interest due to their synergistic advantages compared with single surfactant [71]. The anionic surfactant (SDBS) can greatly decrease the adsorption of the nonionic surfactants (Tween 80) on soils [72], and the addition of Tween 80 could reduce the precipitation between SDBS and cations in soil [73,74]. Guo and co-workers [39] evaluated the sorption of Tween 80 by the soil in the mixed surfactant system with different Tween 80/SDBS ratio. Results showed that the amount of sorbed Tween 80 was significantly decreased when increasing SDBS/Tween80 ratio from 0 to 1:1 (Fig. 4). Similar results were obtained by some other researchers when studying the SDBS–Tween 80 mixed soil washing system [75,76]. It is because the addition of the SDBS can increase the negative surface charge of SDBS–Tween 80 mixed micelles, thereby decrease the electrostatic attraction between Tween 80 and soil particles [77,78]. However, at the condition of excess SDBS (SDBS/Tween 80 ratio above 1:1), the precipitation between the anionic surfactant and multivalent electrolytes would be

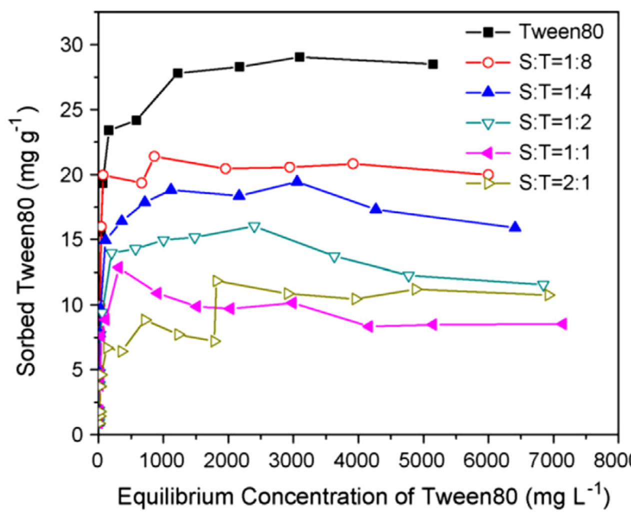


Fig. 4. Sorption isotherms of Tween80 by the soil particles. Adapted with permission from ref 39. Copyright 2009 Elsevier.

enhanced, and resulted in higher absorption amount of Tween 80 [15,79].

Compared with Tween 80, the anionic surfactant SDBS would lead to less desorption of HOCs, as the CMC of SDBS (418.2 mg L^{-1}) was much higher than that of Tween 80 (13.0 mg L^{-1}). For example, Deshpande et al. [28] found that 10 times more of SDBS than Tween 80 were required to obtain the same pollutants removal rate. Interestingly, the mixed SDBS–Tween 80 surfactants showed higher efficiency in solubilization of HOCs than that by single Tween 80. Guo and co-workers [39] found the MSR values were remarkably increased from 0.222 to 0.410 when SDBS/Tween 80 ratio increased from 0 to 1:1. In another work, the mixed SDBS–Tween 80 surfactants with mass ratios of 1:9, 3:7, and 5:5 all showed higher desorption efficiencies than that of single Tween 80 [76]. This is believed because SDBS molecules (the hydrophobic tails) could embed into Tween 80 micelles, leading to the formation of smaller mixed micelles and thus increased the micelles concentration [77,80–82]. More interestingly, studies have shown that the greatest desorption efficiency were achieved with the lowest SDBS/Tween80 ratio in the experiments [75,76], which indicated that the solubilization capacity of Tween 80 could significantly promoted by addition of a low amount of SDBS.

3.3. Other soil washing systems

Several studies showed that increasing the hydrophobicity of Tween 80 micelles cores is a feasible approach to promote the solubilization capacity of Tween 80 [83–87]. The results show some oil-in-water microemulsions, also called as oil-swollen micelles, are very useful for extracting HOCs in soil washing processes. In these systems, co-surfactants were applied to provide additional stabilizing area between oil molecules and Tween 80 tails and additional entropy of mixing, resulting in the formation of more hydrophobic cores [88]. Significant enhancements in the solubilization of dichlorodiphenyltrichloroethane (DDT) were achieved by oil-swollen micelles as compared to single Tween 80 [84,85]. It was speculated that both the oil solubilized in the micelle core and cosurfactant/surfactant interfacial layer could provide solubilizing spaces for the hydrophobic pollutants [89,90]. Results also showed that the solubilization capacity of oil-swollen micelles can be markedly increased with the increase of either cosurfactant/surfactant ratio or oil/surfactant ratio [84]. Further research revealed that the solubilizing efficiencies of oil-in-water microemulsion systems depend largely on soil types. It was found the saturation sorption amount of oil onto the clay soil is approximately 4 times of that onto loam soil, which has significant inhibitory effect on desorption of the pollutant [83]. As shown in Fig. 5, the desorption enhancement rates (DERs) for DDT are always positive in loam soil group, whereas the values are always negative in clay soil group. The results suggest oil-swollen micelles might exhibit their superiority over empty Tween 80 micelle to desorb HOCs only in soils with relatively low oil sorption capacity.

Chu and Kwan [86] reported that organic solvents (triethylamine or acetone)-incorporated Tween 80 micelles can improve the efficiency of Tween 80-enhanced soil washing. The enhancement is likely due to better desorption of HOCs from the soil assisted by the solvent, and the formation of solvent-incorporated Tween 80 micelles, which increase both the size and the affinity of micelles for more effective pollutant extraction. In another study, Cheng and Wong [91] reported that Tween 80 extraction of pyrene and phenanthrene can be facilitated by the co-existence of dissolved organic matter (DOM). Due to the formation of DOM–Tween 80 complex, the combination of Tween 80 and DOM showed greater extraction capacity than the additive effect of Tween 80 and DOM individually.

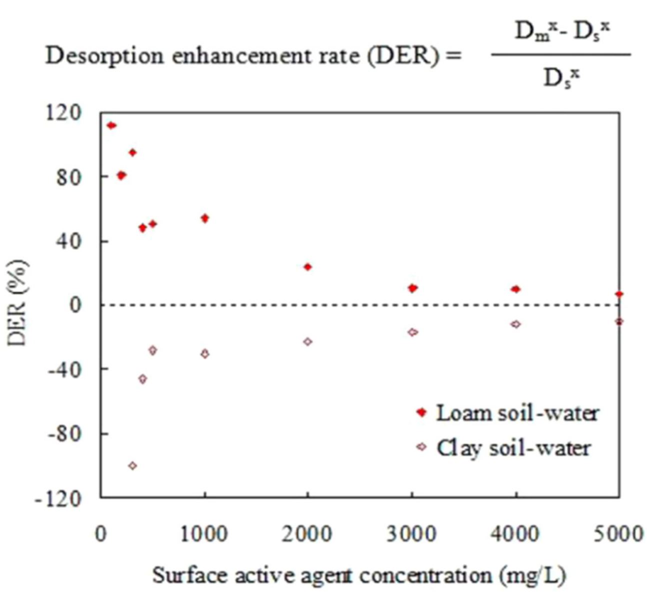


Fig. 5. Desorption enhancement rates (DER) during DDT desorption as a function of surface active agent concentrations in loam soil-water (◆) and clay soil–water (◇) systems. D_m^x represents desorption rate achieved by X mg L^{-1} of oil-swollen micelles, and D_s^x represents desorption rate achieved by X mg L^{-1} of empty micelles only. Adapted with permission from ref 83. Copyright 2012 American Chemical Society.

3.4. Remarks and future prospect

Table 2 summarizes the results of some of the most significant studies on Tween 80 related soil washing processes and includes the experimental conditions and maximum removals obtained. Tween 80-enhanced soil washing processes technologies are promising technologies for the removal of HOCs from soils, but research in this area is still far from the commercial stage and because of that, no rigorous economic assessments could be done nowadays.

In the soil–water systems, Tween 80 can desorb the HOCs from soil particles and solubilize them in the surfactant micelles. The dissolved HOCs in aqueous phase have better mobility, showing better availability for the washing treatment. Due to its lower CMC and higher HLB, in most studies, Tween 80 showed the best capacity for the extraction of HOCs from the soil among the tested nonionic surfactants. Attentions should be paid that sorption of Tween 80 onto soil (especially with high SOM content) may result in a proportion of Tween 80 being unavailable for the micellar solubilization of HOCs. Due to this reason, high concentrations (above 1 g L^{-1}) of Tween 80 were needed to achieve relatively high HOCs removals. The efficiency of Tween 80-enhanced soil washing can be increased by reducing the sorption loss of Tween 80 by the soil. One promising technology is the combination of Tween 80 with anionic surfactant SDBS, which can greatly decrease the adsorption of Tween 80 on soils. The mixed surfactants exhibits lower CMC value than either individual Tween 80 or SDBS, showing better solubilization efficiency. The efficacy of Tween 80-enhanced soil washing can also be enhanced by increasing the hydrophobicity of Tween 80 micelles cores (e.g., Tween 80 based oil-swollen micelles and solvents-incorporated Tween 80 micelles) to promote the solubilization capacity of Tween 80 micelles.

Despite the good results have been obtained in the laboratory, many improvements are required before the technology can be scaled up to pilot level. It would be injudicious to extrapolate these laboratory findings to real applications without first performing a scale-up study. In addition, many issues still need to be investigated at the lab-scale, especially in terms of obtaining a fundamental understanding of these processes, for example, the interaction between Tween 80 and soil components.

4. Tween 80-enhanced soil flushing processes

In-situ soil flushing with Tween 80 solutions is another practical strategy for the removal of HOCs in the soil. The in-situ soil flushing is appealing because it causes little disruption to soil environment and no extra costs are necessary for soil transportation and the stirring as compared with ex-situ soil washing. As shown in Fig. 6, the flushing eluents are injected into contaminated soil via injection wells, and passing through the contamination zone due to the pressure gradient. The contaminant-containing fluid can be pumped out for disposal, recirculation, or on-site treatment and reinjection [21]. A lot of works, including laboratory studies [46,92–98] and field studies [99–102] have indicated that adding Tween 80 to the soil flushing solution can remarkably enhance the treatment efficiency. One recently published column study shows the soil flushing treatment with 4 g L^{-1} of Tween 80 solution can obtain an 81.5% removal of phenanthrene in 12 h, compared to the removal of only 5.9% in the absence of Tween 80 [94].

A very promising pilot-scale study was demonstrated by Ramsburg and co-workers [100], in which Tween 80-enhanced soil flushing was used to remove tetrachloroethene (PCE) from the contaminated site located in northeastern Michigan, Oscoda, USA. In the field study, the concentration of PCE in the flushing effluent during Tween 80 flushing increased about 4-fold over the initial

Table 2
Combination of Tween 80 related soil washing processes.

Surfactant; concentration	Pollutant; concentration	Operation conditions	Maximum efficiency reported	Refs.
Tween 80; 10 g L^{-1} Brij 35; 10 g L^{-1} TritonX-100;10 g L^{-1}	p-cresol; 210 mg kg^{-1} (spiked)	Soil/water = 3.5 mL g^{-1}, 50 rpm, 20 °C, 96 h	58% removal 54% removal 43% removal	[55]
Tween 80;1.5 g L^{-1} Brij 35;1.5 g L^{-1} Triton X-100;1.5 g L^{-1}	Phenanthrene; 300 mg kg^{-1} (spiked)	0.5 g of soil, 10 mL of surfactant solution, 250 rpm, 25–28 °C, 24 h	Solubility: 37 mg L^{-1} Solubility: 17 mg L^{-1} Solubility: 26 mg L^{-1}	[57]
Tween 80; 10 g L^{-1} Tween 20; 10 g L^{-1} Brij 35; 10 g L^{-1} Tyloxapol; 10 g L^{-1}	Benzo[a]pyrene; 500 mg kg^{-1} (spiked)	2.5 g of kaolin, 2.5 g of Na_2SO_4, 50 mL of surfactant solution, 150 rpm, 30 °C, 3 d	80% removal 69% removal 84% removal 72% removal	[16]
Tween 80; 2 g L^{-1} Tween 40; 2 g L^{-1} Brij 35; 2 g L^{-1}	Phenanthrene; 200 mg kg^{-1} (spiked)	50 g of soil, 500 mL of surfactant solution, 160 rpm, 48 h	72.4% removal 55.5% removal 71.6% removal	[58]
Tween 80; 10 g L^{-1} Brij 35; 10 g L^{-1}	Fluoranthene; 500 mg kg^{-1} (spiked)	2.5 g of soil, 50 mL of surfactant solutions, 150 rpm, 30 °C, 48 h	86% removal 37% removal	[59]
Tween 80; 10 g L^{-1} TritonX-100; 10 g L^{-1}	Diazinon; 100 mg kg^{-1} (spiked)	2 g of polluted soil, 20 mL of surfactant solution, shaken end-over-end, 15 °C, 24 h	Relative desorption rates: 285% Relative desorption rates: 100%	[69]
Tween 80;4 g L^{-1} SDBS/Tween 80 = 1:2; total 6 g L^{-1} SDBS/Tween 80 = 1:1; total 8 g L^{-1} SDBS/Tween 80 = 2:1; total 12 g L^{-1}	p-Nitrochlorobenzene; 152 mg kg^{-1} (spiked)	2 g of soil, 20 mL of surfactants solution, shaken end-over-end, 22 ± 2 °C, 24 h	Solubility: 11.15 mg L^{-1} Solubility: 11.96 mg L^{-1} Solubility: 12.13 mg L^{-1} Solubility: 11.62 mg L^{-1}	[39]
Linseed oil/Tween80 = 0 Linseed oil/Tween80 = 1:10	DDT; not given (spiked)	0.5 g of loam soil, 1000 mg L^{-1} of Tween 80; 250 rpm, 25 °C, 96 h	43% desorption 68% desorption	[83]
Acetone/Tween80 = 0 Acetone/Tween80 = 15 Acetone/Tween80 = 70	4,4′-dichlorobiphenyl; 40 mg kg^{-1} (spiked)	0.5 g of soil, shaken, 20 °C, 24 h	61% desorption 73% desorption 78% desorption	[86]

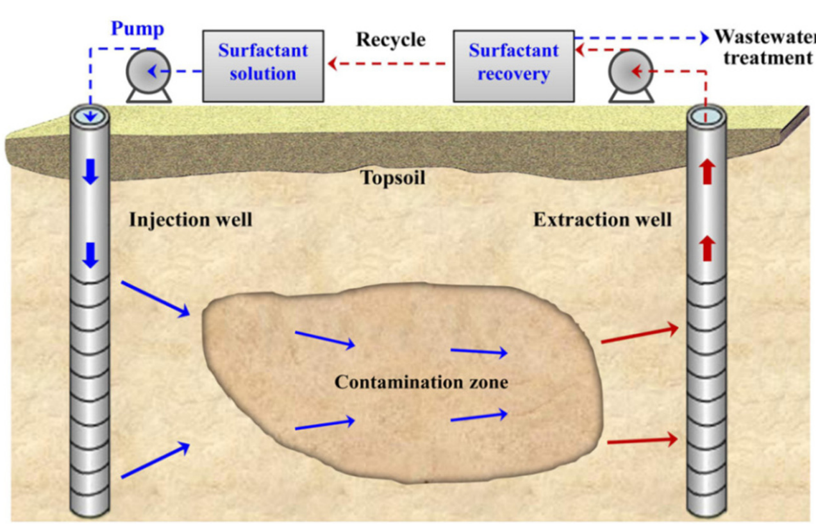


Fig. 6. General procedure of Tween 80-enhanced in-situ flushing for soil remediation.

level (water flushing, about 10 mg L^{-1}) (Fig. 7). After 47 days' Tween 80 flushing treatment, a total of 19 L of PCE were removed from the soil, indicating that Tween 80-enhanaced flushing is of great potential for the practical remediation. In this study, the longer-term impacts of the treatment on PCE concentrations in the contamination zone were also studied. The 450 days monitoring results demonstrated a two order-of-magnitude decrease in PCE concentration comparing with that measured before the flushing treatment. The cost for this Tween 80-enhanaced flushing treatment was calculated as \$2100 m^{-3}, which seems unacceptable for the full-scale operations. Note, however, that the costs of full-scale applications are generally much lower than those of similar treatments at pilot-scale. Besides, reusing the surfactant can further reduce the chemical cost of the treatment. It was reported that by hydraulic control throughout the treatment, about 95% of the injected Tween 80 can be recovered from the flushing effluent [100].

Some of the most significant studies on Tween 80-enhanaced flushing process are included in Table 3. In summary, Tween 80

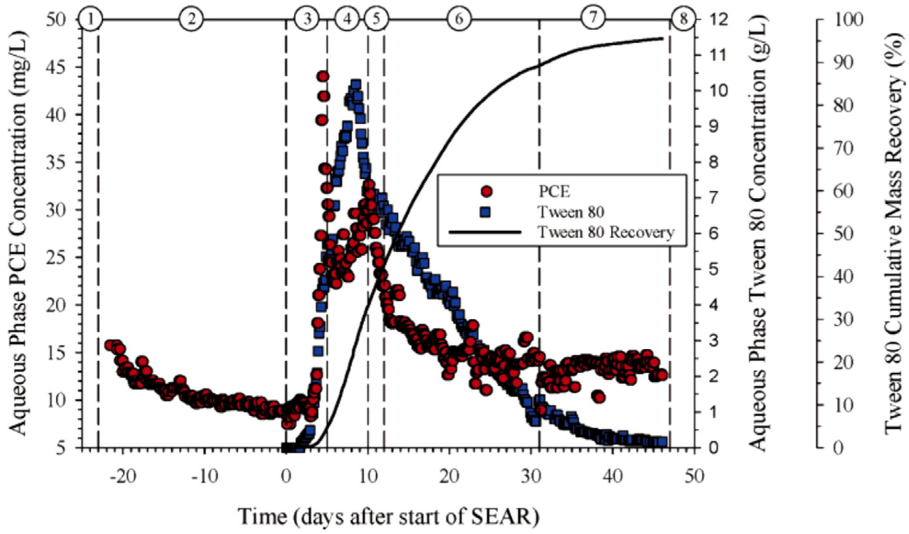


Fig. 7. Concentrations of PCE and Tween 80 in the extraction well. Vertical lines represent a change to the influent solution or total flow rate (1, extraction only; 2, water flood through S1, S2, S3, W1, W2, and W3; 3, fully screened injection of Tween 80 in S1, S2, and S3; 4, partially screened injection of Tween 80 through S2 and S3; 5, water flood through W1, W2, and W3; 6, extraction only; 7, increased rate of extraction; 8, pumping terminated). Adapted with permission from ref 100. Copyright 2005 American Chemical Society.

can enhance soil flushing processes mainly by dissolving of HOCs sorbed on soil particles and improving the permeability of the soil. The lab scale experiments in both spiked soils and real polluted soils were very successful. And several field studies have verified the feasibility of this technology in term of pollutants removal. Further works need be done to optimize the operation parameters and reduce the cost.

5. Tween 80-enhanced electrokinetic remediation

Over the last two decades, the applications of electrokinetic remediation technology have increased significantly. Instead of the pressure gradient, a voltage gradient is used as the driving force in this process. The anode and cathode electrodes are inserted into the soil to produce electric field as shown in Fig. 8 [103,104]. With the application of electric field to the contamination zone, contaminants are transported out of the soil mainly by electro-osmosis [22]. The flushing effluent at the electrodes can be pumped out for surfactant recovery and pollutants treatments (Fig. 8).

A number of laboratory studies have determined that the organic pollutants with a relatively high-aqueous solubility, such as trichloroethylene, benzene, toluene or m-xylene, could be easily removed by electro-osmosis [105,106]. However, due to the neutrality and hydrophobicity of HOCs, it is difficult to transport them out of the soil by electro-osmosis. In recent works, Tween 80 has frequently been used to increase the desorption and solubility of hexachlorobenzene [107], DDT [108], PAHs [109–112] and numbers of other HOCs [113–116] in electrokinetic flushing process. In general, high removal efficiencies of HOCs can be achieved if Tween 80 solubilizes the pollutants in the micelles and the electrokinetic flushing process maintains a high electro-osmotic flow (EOF) [117]. However, it should be noted that the use of Tween 80, particularly with the relatively high doses, tends to decrease EOF due to the changes in the interaction of Tween 80 solution with the soil particle surface, thus decrease the electric conductivity of the system [22].

Helmholz-Smoluchowski (H-S) equation [118] has been established to assess the EOF velocity (V):

$$V = -D\varepsilon_0\zeta Ex/\eta \quad (4)$$

where D represents the dielectric constant, ε_0 represents the permittivity of vacuum, ζ represents zeta potential, E_x represents the electrical gradient and η represents the viscosity. According to the H–S equation, the value of EOF is mainly depended on the electric-field strength and soil zeta potential. There is evidence that the addition of Tween 80 solution had little impact on the long-term current value of the sample [119]. On the other hand, the zeta potential of the soil was found to increase from -60 to −32 mV due to the soil absorption of Tween 80 [120]. Therefore, on the condition that the electric-field strength is basically constant, increase of zeta potential (caused by Tween 80) would lead to the decrease of EOF. Nevertheless, according to the literature, Tween 80 can enhance the electrokinetics remediation in synthetic and real soils with different success [109–112]. It was suggested that Tween 80 solubilization of HOCs plays a crucial role in the electrokinetic remediation processes. Besides, Tween 80 may acts as the dispersing agent to improve the permeability of the soil and thus make the pore fluid more smoothly [107].

5.1. pH control

In typical electrokinetic remediation processes, the oxidation and reduction of water at the anode and cathode would lead to the generation of H^+ and OH^- according to the following reactions:

$$\text{At the anode}: 2H_2O \rightarrow O_2(g) + 4H^+ + 4e^- \quad (5)$$

$$\text{At the cathode}:: 4H_2O + 4e^- \rightarrow 2H_2(g) + 4OH^- \quad (6)$$

The generated H^+ and OH^- would move towards to the oppositely charged electrode and consequently create an acidic (H^+)/alkaline (OH^-) front in the soil samples [121], resulting in changing the pH, with low/high pH values close to the anode/cathode chamber. Boulakradeche and co-workers [116] observed the pH value of the anolyte rapidly dropped to pH 3, and pH value of the catholyte increased up to 11 from initial pH 5.4. In this condition, EOF would be inhibited by the decrease of soil zeta potential by excess H^+ and heavy metal precipitation by excess OH^- [113,116].

Several studies have demonstrated that the EOF can be remarkably increased by adding a pH control solution to the anode chamber [110,122–124]. For instance, the addition of tris-acetate buffering solution maintained a basic pH (8.7–10) and increased

Table 3
Combination of Tween 80 related soil flushing processes.

Technology	Type of soil	Pollutant; concentration	Operation conditions	Maximum efficiency reported	Refs.
Tween 80-enhanced soil flushing	Sandy soil (89.7% sand, 10% silt, 0.3% clay)	Phenanthrene; 100 mg kg^{-1} (spiked)	Column: diameter = 5 cm, length = 37 cm; 4 g L^{-1} of Tween 80, 1 L min^{-1}, 12 h	81.5% removal	[94]
Tween 80-enhanced soil flushing	Glacial outwash sands from a contaminated site (field experiment)	Tetrachloroethene (PCE); 52 mg kg^{-1} (real polluted)	Scale: 17.6 m × 14.8 m × 7.6 m (58 × 50 × 15 grid nodes), 6% Tween 80, 1.9 L min^{-1}, 10 d	A total of 19 L of PCE and 95% of the injected surfactant were recovered	[100]
Tween 80- enhanced soil flushing	Sandy loam from a contaminated site (field experiment)	Xylene; 42.1 mg kg^{-1} (real polluted)	Scale: 5 m × 5 m × 3 m, 7800 L of 0.1–0.2% Tween 80,10 d	53.9% removal	[102]
Tween 80-enhanced electrokinetic soil flushing	Contaminated peaty clay collected from a former asphalt factory site	16 PAHs; total 3000 mg kg^{-1} (real polluted)	Reactor cell: diameter = 5 cm, length = 5 mm; Voltage gradient: 0.37 V cm^{-1}, 1% Tween 80, 9 d	Without Tween 80: 0.04% removal; with Tween 80: 30% removal	[109]
Tween 80-enhanced electrokinetic soil flushing	The soil at the site is located in river foreland (field experiment)	PAHs; 310–542 mg kg^{-1} (real polluted)	Reactor cell: 1 m × 1 m × 0.5 m, voltage gradient: 0.12 V cm^{-1}, 1% Tween 80, 159 d	Day 50, PAHs were found at the anode	[112]
Tween 80-enhanced electrokinetic soil flushing	Kaolin (4% sand, 18% silt,78% clay)	Phenanthrene; 500 mg kg^{-1} (spiked)	Reactor cell: diameter = 6.2 cm, length = 19.1 cm; voltage gradient: 1.0 V cm^{-1}, 3% Tween 80, 230 d	Total mass removal without pH control: 42 ug in 230 d; with pH control: 40 ug in 63 d	[122]
Tween 80- enhanced electrokinetic soil flushing	Kaolin (85% kaolin clay, 14% mica, and <1% quartz)	Three PAHs; each 500 mg kg^{-1} (spiked)	Reactor cell: diameter = 32 mm, length = 100 mm; voltage gradient: 3.0 V cm^{-1}, 1% Tween 80, 23 d	pH control: 46.27% removal; without pH control: 8.69%	[110]
Tween 80-enhanced electrokinetic soil flushing	Saudi Arabian silt soil (39% sand, 54% silt, 7% clay)	Phenanthrene; 1623 mg kg^{-1} (spiked)	Reactor cell: diameter = 5 cm, length = 6 mm; electrical current density: 0.127 A m^{-2}, 1% Tween 80, 10 d	86.93% removal	[111]
Tween 80-enhanced electrokinetic soil flushing	Olst silty loam (10% sand, 82% silt, 8% clay)	Phenanthrene; 1165 mg kg^{-1} (real polluted)	Reactor cell: diameter = 5 cm, length = 6 mm; electrical current density: 0.127 A m^{-2}, 1% Tween 80, 10 d	35.24% removal	[111]

the EOF from 3.7 mL d^{-1} to 11.6 mL d^{-1} [123]. The pH control at the anode has been shown to be effective to increase pyrene [110] and phenanthrene [122] solubilisation and migration from the soil. In the first study, with the pH control in the anode chamber (pH set as 7.0), the total recovery of phenanthrene was higher than 50%, compared to a removal of only 13.9% without pH control. In the second study, the residual phenanthrene in soil (6 cm from anode) with pH control in the anode chamber was only about one third of that in the system without pH control.

5.2. The effects of soil types

Another critical element of this technology is the influence of soil types. Kaolin clay was extensively studied as the model soil in the electrokinetic remediation experiments. Lab results showed that Tween 80-enhanced electrokinetic flushing process is effective to remove HOCs from Kaolin [107,110,122,125,126]. However, in some cases, huge differences have been obtained between the pollutants removal efficiencies in an actual polluted soil and a Kaolin soil. A good example can be found in a study by Reddy and Saichek [119], in which big difference in the removals of phenanthrene in glacial till and kaolin were observed due to the higher SOM content of glacial till compared to kaolin. The interaction of the SOM (2.8%) in the glacial till with Tween 80 largely restrained phenanthrene removals. The removal of HOCs from sandy soil is also found less efficient than from kaolin clay. Alcántara et al. [123] reported that about 65% and 85% of phenanthrene were removed from the sandy soil and kaolin clay, respectively.

The pilot-scale field study also demonstrated that significant improvement is required before applying this technology to actual remediation processes. A 159 days in situ Tween 80-enhanced electrokinetic remediation experiment was set up by Ana T. Lima and co-workers [112] in a PAHs contaminated site. The experiment was set up in a 1 m × 1 m × 0.5 m volume soil unit within a fine grained layer (loamy soil) located at a depth between 3.70 and 4.20 m below ground surface. In the experiment, PAHs did not found at the anode until day 50, and PAHs concentration in the electrolyte reservoir was only about 200 μg L^{-1}. Besides, PAH content of the soil was still very high after the 159 days treatment. This suggests there is still an important challenge in Tween 80-enhanced electrokinetic remediation of HOCs contaminated sites.

5.3. Remarks and future prospect

Some of the most significant studies on Tween 80-enhanaced electrokinetic remediation process are included in Table 3. According to the scientific literature, most studies have been conducted on spiked real or synthetic Kaolin clay at the lab-scale. Note, however, that kaolin clay's characteristics may largely differ from actual polluted soils. The complexity of real polluted soils may alter the performance of Tween 80 in these processes. Thus, significant attention should be paid over the next few years to the differences between current studies and treating actual polluted soils. Another important consideration is that reactor design has not been investigated, and most studies in this area use small electrochemical cells with relatively low voltage (1–3 V cm^{-1}). Larger reactors have been used in the field to demonstrate the removal efficiency of this system; however, these reactors have not been used to obtain information on system issues, such as economic or operational problems, that would arise for large scale systems. Besides, the pioneer research on the field application of Tween 80-enhanced electrokinetic remediation shows the performances were not quite satisfactory. Thus, the recommendations for future studies also include the system optimization.

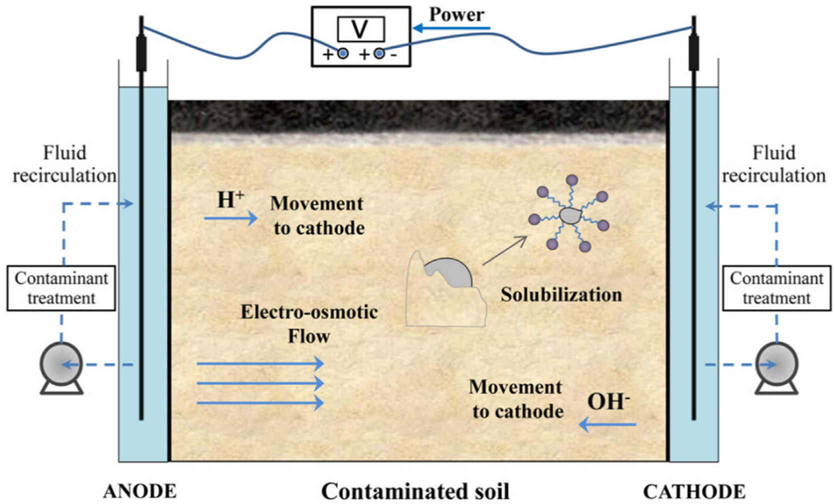


Fig. 8. General procedure of Tween 80-enhanced in-situ electrokinetic flushing for soil remediation.

6. Tween 80 solubilization integrated with other treatments

Tween 80-enhanced soil washing or flushing process is generally efficient to extract the pollutants; therefore a combined treatment is needed to destroy them. In the scientific literature, various studies have reported the use of soil washing/flushing in combination with other remediation techniques such as technologies based on Fenton reaction chemistry [127,128]. A good review in which the treatments of soil washing/flushing solutions are described exhaustively was recently published by Trellu and co-workers [3]; however, the review considers the treatment of various eluents (surfactants, biosurfactants, cyclodextrins, etc.) and does not focus specifically on Tween 80 solution. In the current review, we focus on the joint application of Tween 80 solubilization and chemical/biological process for the elimination of HOCs from the washing/flushing solutions and soil–water systems.

6.1. Coupling with oxidation process

Technologies based on Fenton reaction chemistry involve the generation of hydroxyl radicals ($\cdot OH$), the second strongest oxidizing agent with a redox potential of 2.8 V [129,130], have been developed in the last few years to treat washing/flushing solutions [131]. Among these technologies, electro-Fenton process, which consists of electro-catalytically assisted Fenton reaction (Eq. (7)), appears to be a promising way to destroy HOCs in the soil washing/flushing solutions [66,131,132]. Compared to the classical Fenton process, electro-Fenton process allows minimizing the cost of reagents since H_2O_2 is electro-generated on the cathode via oxygen reduction (Eq. (8)) [133]. Moreover, no iron would be needed in most cases because Fe^{2+} could be generated under the presence of the existent Fe in the soil washing/flushing solutions (directly extracted from soil and continuously electro-regenerated at the cathode following Eq. (9) [134].

$$Fe^{2+} + H_2O_2 \rightarrow Fe^{3+} + OH^- + \cdot OH \quad (7)$$

$$O_2 + 2H^+ + 2e^- \rightarrow H_2O_2 \quad (8)$$

$$Fe^{3+} + e^- \rightarrow Fe^{2+} \quad (9)$$

These continuously generated $\cdot OH$ are effective for the degradation of HOCs in the washing/flushing solutions until their overall mineralization. For instance, Huguenot and co-workers [132] reported the complete mineralization of the total petroleum hydrocarbons in the solution ($TOC = 5100\ mg\ L^{-1}$) after 32 h electro-Fenton treatment.

Photocatalysis is another $\cdot OH$ based oxidation technology, which makes use of the semiconductor metal oxide (TiO_2, ZnO, CdS, GaP, WO_3, etc.) as catalyst [135]. In particularly, titanium dioxide (TiO_2) in the anatase form has been proved to be the most appropriate one due to its cost-effectiveness, inert nature and photostability [136,137]. In recent years, some researchers investigated the photodegradation efficiencies of HOCs in soil with the elution of Tween 80. Xiong et al. [138] observed that endosulfan in the soil eluates can be completely degraded with visible-light irradiation in 4 h. In some previous investigations, it was found that the photodegradation of various aromatic pollutants is inhibited in the presence of surfactants [139]. Yang et al. [139] showed that Fenton reaction combination for photodegradation can overcome this problem. In the experiments, traditional photodegradation can only reach 44.63% removal of PAHs, but Fenton-photodegradation can remove 97.96% of PAHs.

Due to its strong oxidation characteristics, $\cdot OH$ is able to degrade most of the organic pollutants to the ultimate mineralization products of CO_2 and H_2O [140–142]. However, due to the non-selectivity of $\cdot OH$, photocatalysis and Fenton processes trend to degrade Tween 80 along with the target pollutants simultaneously [131]. This phenomenon can also be explained by the way of forming complexes between Tween 80 and the target pollutants. The pollutants molecule is trapped into Tween 80 micelle core, therefore $\cdot OH$ has to destroy the micelle before contacting with the target pollutants [131]. The relatively mild oxidation method, electrochemical oxidation, could be a good alternative. In this process, organic pollutants can be destructed by the redox reactions induced by electric current [59]. Using this technology, Tween 80 has been successfully recovered, while the target pollutants were degraded in the meantime [143,144]. For instance, Gómez et al. [143] showed that after a 95% removal of phenanthrene by electrochemical oxidation, the recovery rate of Tween 80 reached 82.4%. Electrochemical oxidation is recognized as an environmentally friendly technique because the electron is the main reagent. In addition, the economic cost of this system is very low, the experimental work found the electric power consumption of phenanthrene degradation is 0.45–1.96 $kW\ h\ g^{-1}$ [16,143].

6.2. Coupling with zero valent iron nanoparticles (nZVI)

nZVI has been found effective for the removal of chlorinated compounds in wastewater [145–148]. As for soil remediation, it is recognized the transport of nZVI is largely hindered by its aggregation and settling in the soil pores [149]. Researches have shown Tween 80 coupled with electrokinetics can facilitate the delivery of nZVI into low-permeability soils and overcome the mass transfer limitation [114,145,149–152]. Chen et al. [114] used nZVI-Tween 80-electrokinetics system for the degradation of perchloroethylene (PCE) in a glass sandbox (silica sand). They found that the nZVI activity was largely promoted by Tween 80 and electrokinetics, and resulted in the completely dechlorination of PCE. In another work, the same method was tested to enhance dechlorination of PCB in a two-compartment cell where the contaminated soil was stirred in slurry with Tween 80, nZVI, and a constant potential ($1\ V\ cm^{-1}$) [152]. Results showed that the use of nZVI with Tween 80 and the direct current gave the highest PCB removal among all tested groups. In this process, Tween 80 can enhance not only the mobility of nZVI but also the desorption of pollutants from soil particles. As illustrated by Zhang et al. [145], the pollutants degradation by nZVI was strongly limited by the desorption rates, especially for the soils with high SOM content. In their experiments, under otherwise identical conditions, in the absence of surfactant, about 82% of trichloroethylene in the farm soil (SOM content = 0.7%) was degraded in 30 h, compared to only 44% for potting soil (SOM content = 8.2%). And this work also shows that addition of very low concentration ($1 \times$ CMC) of Tween 80 to the potting soil can increase trichloroethylene degradation from 44% to 49%.

6.3. Coupling with microbial degradation

The addition of Tween 80 to enhance the bioavailability of HOCs has received considerable attention. For instance, the white rot fungus *Phanerochaete chrysosporium* was applied to degrade PAHs in the eluent after extraction from the contaminated soil with Tween 80 solution [153] and in a soil–slurry system [154] in the presence of the same surfactant. The results showed that Tween 80 was readily catabolized without any effect on the fungi growth and production of ligninolytic enzymes [153,154]. This result is in agreement with some other researchers, for example, Kotterman et al. [155] reported that *Phanerochaete chrysosporium* growth was not affected by Tween 80 with concentrations range from 250 to 10,000 mg/L in the medium. In most cases, Tween 80 can enhance the degradation efficiency by increasing dissolution of pollutants molecules in aqueous phase and transportation of pollutants molecules to the microbial cells. As for white rot fungi, Tween 80 can also promote the oxidation ability of manganese peroxidase and laccase, which are considered very important for the degradation of HOCs [156–159].

Apart from the Phanerochaete chrysosporium, many other strains (e.g., *Armillaria* [160], *Bacillus licheniformis* [161], *Burkholderia cepacia* [162], *Lasiodiplodia theobromae* [163], *Klebsiella oxytoca* [164], *Phanerochaete sordida* [165], *Pleurotus ostreatus* [166], *Pseudomonas aeruginosa* [167] and *Staphylococcus aureus* [168]) have been applied together with Tween 80 to remediate HOCs contaminated soil. Researches show Tween 80 not only exhibits the role to increase the apparent solubility of HOCs but also possess biological effects to modify the cell surface properties [164,169,170]. Several researchers speculated that the effect of Tween 80 on microbial biodegradation is related to cell surface hydrophobicity (CSH) [164,168]. CSH has been reported to be one of the most important parameters of microorganism, which affects the efficiency of various bioprocesses, such as adherence of cell to HOCs and transmembrane transport of HOCs [171]. Surfactants could change the CSH mainly by absorbing surfactant molecules on the cell surface or releasing of lipopolysaccharide (LPS) from bacterial cell surface. It is worth noting that higher concentration of Tween 80 was not always corresponding to a higher CSH. For example, Zhang and Zhu [164] found CSH of Klebsiella oxytoca PYR-1 increased initially and then slightly decreased with the increasing of Tween 80 concentration from $0.2 \times$ CMC to $8 \times$ CMC.

It should be noted that using Tween 80 on the biodegradation does not always yield positive results: in some cases, the addition of Tween 80 worsens the performance significantly compared to biodegradation without surfactant. For instance, Betancur-Corredor et al. [172] found that although desorption of DDT from the soil matrix was enhanced by Tween 80, but DDT degradation performance of the soil indigenous microorganisms was inhibited. Similar observations have been found by some other researchers when studying the biodegradation of PAHs in the contaminated soils [48,173,174]. The possible explanations for the inhabitation include (i) application of Tween 80 to the system may decrease the microbial populations due to the toxicity of Tween 80 on the microorganisms, (ii) the micellar-solubilized HOCs show lower availability to the microorganisms, and (iii) preferential microbial uptake of Tween 80 as substrate inhibits the direct cell-pollutants contact [161,171,172].

6.4. Coupling with phytoremediation

Phytoremediation is an emerging technology that utilizes plants to stabilize, transform or remove contaminants located in soils, sediments or water [175]. HOCs in the contaminated soil can be either adsorbed to the roots and taken up inside the plant tissues (phytoextraction) or degraded by the microorganisms in the root zone (rhizoremediation) [176].

Plants uptake of HOCs from the soil is largely hindered by the low availability of HOCs. Interestingly, the presence of Tween 80 even at very low concentrations resulted in significant positive effects on plants uptake of HOCs. A good example can be found in a study by Zheng et al. [50] in which $1 \times$ CMC of Tween 80 solution was used to facilitate the release of phenanthrene from the sorbed phase, leading to a significant promotion (2.16 folds) on the uptake of phenanthrene, compared to Tween 80-free control group. There are observations that co-addition of plant growth stimulating substances [177,178] or carboxylic acids [176] can further increase the efficiency of phytoremediation. In a promising study conducted by Sun et al. [177], the combination of Tween 80 and GA3 (plant hormone) increased Benzo[a]pyrene accumulation in the roots, stems and shoots by 4.14–5.86, 1.86–2.31 and 1.33–1.55 times, respectively, compared with the control plant.

In a previous study, Ramamurthy and Memarian [179] investigated the phytoremediation of engine oil-contaminated soil with the addition of Tween 80 at various concentrations (0.5–$2 \times$ CMC). The results showed Tween 80 could facilitate the utilization of HOCs by rhizosphere microorganisms. More importantly, they found that Tween 80 concentrations and the rhizodegradation rates of engine oil are positively correlated. To further investigate the effects of Tween 80 on rhizodegradation, the same authors [180] studied the variation of overall soil microbial activity by monitoring the basal soil respiration (BSR) rate. It was found the addition of Tween 80 could enhance the activity of microbe in terms of BSR for all concentrations of Tween 80 tested. In the experiments, Tween 80 showed no significant impact on plant growth; however, it seems to have the positive effect on the soil microbial population [179,180]. One possible explanation for this stimulatory effect is that Tween 80 could provide a suitable carbon source for the microorganisms. Another explanation might be that higher levels of dissolved organic matter released by Tween 80 in soil pore water could serve as a new carbon source for additional microbial growth [180].

6.5. Remarks and future prospect

In most of the cases, Tween 80-enhanced soil washing/flushing coupled with oxidation process could successfully eliminate HOCs from the soil. The main drawback of ·OH based oxidation technologies is Tween 80 would be degraded along with the target pollutants, which increases cost of treatment. Despite this drawback, fairly interesting results can be found in the literature because most of these technologies can completely mineralize the organic pollutants down to negligible concentrations. In particular, very promising results have been obtained for electro-Fenton processes. The versatility of electro-Fenton processes is enhanced by the fact that they can generate reagents in situ, which prevents risks associated with its transport, storage and handling. To reuse Tween 80 and reduce the cost of reagents, further research is needed to find a solution, either by separating Tween 80 and the pollutants before the oxidation processes or by selective degradation of the target pollutants. Based on several lab studies, electrochemical oxidation by using graphite electrodes could be a good alternative. But actually, to date, only very limit information in the scientific literature is available on this process. In the future studies, more effort should be conducted on this field, since this technology is both ecologically sound and cost-effective.

Recently, there was growing interest in using Tween 80 combined with electrokinetics to facilitate the delivery of nZVI into low-permeability soils. According to the good performance of this nZVI-Tween 80-electrokinetics system obtained in the lab, with the in-depth study of the related researchers, the synergistic system may provide cost-effective solution to remediate some of the most challenging environmental cleanup problems. However, there some environmental issues (such as the potential ecotoxicological effects of nZVI and anodic precipitation) that still require further studied before full-scale implementation. In addition, the extensions of this nZVI-Tween 80-electrokinetics system is worthy of studying. With some modifications, such as replace nZVI by Fenton reagents, this system may able to deal with more types of organic contaminations.

Bioremediation has been recognized a promising technology that is ecologically sound and cost-effective. However, bioremediation could be inefficient for treating of HOCs polluted soil since HOCs usually bind tightly onto soil particles, making them inaccessible to microorganisms and plants. A lot of studies have shown Tween 80 can enhance the biodegradation of HOCs by improving the availability of HOCs to the microorganisms. In some cases, Tween 80 was found able to improve the cell surface properties and promote the oxidation ability of the extracellular enzymes. Note, however, that the application of Tween 80 to biodegradation does not always yield positive results. The mechanism underlying the inhibitory effect of Tween 80 to biodegradation need further research, especially on Tween 80′s toxicity to some types of PAHs-degrading bacteria. Phytoremediation is a cost-effective and secure technology that can be applied for the in-situ cleanup of contaminated sites. Studies have shown that Tween 80 can enhance both the plant uptake and rhizodegradation of HOCs. However, research in this field is at a critical stage. Most studies to date have been conducted at the lab-scale using pot experiments. Thus, the feasibility of this technology needs to be tested by field studies, in which the soil properties, weather and other environmental issues should be taken into consideration.

7. Tween 80 recovery

In order to improve the cost-effectiveness of these processes, some authors investigated the possibility of Tween 80 recovery after the treatment. The technologies for recovering and reusing Tween 80 include selective adsorption [181–184], air stripping [100,185] and oxidation treatment [186,187].

Previous studies demonstrated that selective adsorption was potentially effective to regenerate Tween 80 from the soil washing/flushing effluent since the partitioning coefficients of HOCs are much higher than those of nonionic surfactants [181,182]. Rosas et al. [184] studied the recovery of Tween 80 from the washing effluent by selective adsorption of p-Cresol with activated carbon. The results showed that activated carbon can selectively remove over 90% of total p-Cresol from the solution. On the other hand, the level of p-Cresol removal from a new soil obtained with the recovered Tween 80 solution was similar to that obtained with a new Tween 80 solution at the same concentration. Note that the activated carbon needs to be recycled in the treatment due to its high cost. Nowadays, saturated activated carbon is often regenerated by the use of thermal processes [188]. However, it is an expensive process with high energy consumption. Further studies are needed in this area in order to develop an economically feasible technology.

Several researchers showed air stripping and related technologies such as vacuum stripping are very effective for removing contaminants from surfactant solutions. In a previous work, Kibbey et al. [185] have thoroughly discussed the issues related to air-stripper design for treating surfactant-containing wastewaters. They demonstrated that quantitative correction of Henry's law constants (K_H^0) for surfactant solubilization-induced volatility reduction can lead to accurate predictions of air-stripper performance. Base on this theory, Ramsburg et al. [100] designed a 2.4-meter-high stripper to regenerate Tween 80 from the washing effluent in a pilot-scale treatment. It was reported that 95% of the injected surfactant could be recovered by this stripper [100].

Since the pollutants are trapped into Tween 80 micelle core, the following treatments have to degrade the micelle before degrading the pollutants. For example, due to the non-selectivity of ·OH, Fenton processes trend to degrade Tween 80 along with the target pollutants simultaneously [131]. However, in some cases, due to their different reaction rates with ·OH, the pollutants can be selectivity degraded, while Tween 80 could be regenerated. It has been shown that it is possible to achieve complete removal of p-cresol ($20\ mg\ L^{-1}$), while less than 10% of Tween 80 ($0.86\ g\ L^{-1}$) was degraded by using Fenton process [186]. Similar results was found in a recent study, in which 79% of Tween 80 was recovered after electro-Fenton process, while the monitored pollutants (PAHs) were completely degraded (>99%) [187]. However, in another study, electro-Fenton treatment of Tween 80/phenanthrene solution allowed regenerating only 50% of Tween 80 [131]. This suggests the efficiency of the selective degradation could be largely influenced by operating conditions. Moreover, it is worth to note that the total mineralization of target pollutant has not been considered for these recycling strategies.

8. Discussion and conclusions

Due to its low cost, low toxicity, low polarity and high solubilization capacity, Tween 80 has been frequently used to enhance the removal of HOCs from soils. We provide an extensive view on the role of Tween 80 in different soil remediation systems, with the aim of improving the basic understanding of Tween 80-enhanced HOCs pollution remediation technologies including soil washing, soil flushing, electrokinetics remediation and combined treatments.

Tween 80-enhanced soil washing, soil flushing and electrokinetics treatments simplify the soil decontamination problem to a wastewater treatment problem. Tween 80-enhanced soil washing has been extensively studied due to the following reasons: (1) easy to implement, the actual process takes place in a closed system, which permits control of the ambient environmental conditions and (2) the time to complete the cleanup is relatively short: the washing process usually takes less than 3 d. However, the effectiveness of Tween 80-enhanced soil washing is largely affected by soil properties; in particular, this treatment could be ineffective for high fine grained clay soil. Besides, additional cost is required for transportation of soils from the contaminated site to treatment site. By comparison, Tween 80-enhanced soil flushing and electrokinetics remediation are conducted in situ. Therefore less cost for labour and transportation would be required, and less environmental disturbance would be caused. Tween 80-enhanaced flushing treatment has been successfully applied on some pilot sites. However, the efficiency of the flushing treatment is not comparable to that of washing treatment. Besides, the operation of Tween 80-enhanaced flushing is relatively complicated. The major problem of Tween 80-enhanaced electrokinetics remediation is Tween 80 tend to decrease the EOF. It is necessary to enhance the EOF using high voltage gradients, which make it difficult to be implemented in real applications.

The combination of Tween 80 solubilization and other technologies can transform the pollutants into harmless products. Compared with Tween 80-enhanced biological technologies, the oxidization processes are much more rapid and aggressive. For example, Fenton reaction and related processes can completely mineralize HOCs in hours. But the cost of these oxidization treatments is relatively high because Tween 80 together with SOM would also consume oxidant during the treatment. The electrochemical oxidation is very promising for treating the washing/flushing effluent; however, very little information on this technology is available in the literature. Tween 80-enhanced biological technologies are both ecologically sound and cost-effective. Also, the dosage of Tween 80 applied in the biodegradation and phytoremediation are much lower than that in soil washing and flushing processes. Note that the addition of Tween 80 would retard the degradation rate when Tween 80 is toxic to the microorganisms or preferential utilization of Tween 80 by the degraders as a nutrient. Tween 80-enhanced phytoremediation has high potential for the in situ remediation of HOCs-contaminated sites. Phytoremediation only requires modest nutrient input, but it offers many potential beneficial side-effects, such as carbon sequestration and feedstock for biofuel production.

Generally, the remediation efficiency of these Tween 80 enhanced technologies is largely hindered by the sorption of Tween 80 onto soil, specifically for the real polluted soils which usually have high SOM content. Promising results have been obtained using mixed surfactants, oil-swollen micelles and solvents-incorporated micelles, showing that these technologies may help to overcome the abovementioned problem in the near future. The Tween 80 micelle-solubilized HOCs in aqueous phase have better mobility, thus showed better availability to the subsequent chemical and biological treatments. In some cases, Tween 80 can also enhance the removal efficiency of the chemical and biological treatments by facilitating the delivery of oxidant/reluctant and promoting the degradation capacity of the microorganisms, respectively. Good results obtained in using these Tween 80-enhanced technologies means that a solution to the huge environmental issue of occurrence of HOCs in the soil is in progress, and more attention should be devoted in the future to scaling up these technologies to investigate their economic feasibility and problems arising from the scale-up.

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