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ABSTRACT

Perfluorooctanoic acid (PFOA) is a persistent, nonbiodegradable environmental pollutant progressively becoming more problematic. Due to its ubiquitous use, PFOA is one of the PFAS chemicals frequently detected in the environment. This study assessed electrokinetic (EK) technology as an in-situ remediation procedure to remove PFOA from kaolinite as a model soil. In this study, a 100 mg/kg of PFOA/kaolinite mixture was used, and the effectiveness of the unenhanced EK was compared with the surfactant-enhanced EK experiments with SDS, NaC, and Tween80. 5% (w/w) surfactants were added to the cathode in the EK experiments of one and two weeks long. The study evaluated the impacts of processing time and electric current on the EK performance. Electroosmosis and electromigration mechanisms affected PFOA transport in soil, causing PFOA to accumulate in the middle of the soil. However, the minimal accumulation of PFOA in the cathode region suggests that electromigration was the main transport mechanism. In a one-week EK experiment, the PFOA removal efficiency in the unenhanced EK process was 14.49% at 10 mA and 19.00% at 20 mA, while in surfactant enhanced EK, it was 17.67% at 10 mA and reached 32.66% at 20 mA. The PFOA removal in surfactant enhanced EK reached 75.58% in the two weeks EK experiments carried out at 20 mA. The NaC-EK experiment exhibited the highest PFOA removal, followed by the SDS-EK experiment and TW80-EK experiment. The outcomes show the feasibility of removing PFAS from contaminated soil using the surfactant-enhanced EK technique.

1. Introduction

Perfluorooctanoic acid (PFOA) belongs to a group of chemicals called per and polyfluorinated substances (PFAS). PFOA-contaminated environmental pollution is one of public health's most critical and emerging problems. Since the 1950s, PFAS

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have been widely employed in many applications due to their unique physicochemical properties (Teaf et al., 2019). These substances are used in industrial resources, waterproof surfaces, packaging materials, fire hydrant foams, surfactants, cosmetics and pesticide additives (van Asselt et al., 2011). The strong carbon–fluorine (C–F) bonds of PFOA make them chemically stable, very persistent in soil, and highly resistant to biodegradability. At the end of the chain, the hydrophilic functional group enables PFOA mobility in an aqueous medium (Deng et al., 2015). PFOA and PFOS are the most common PFAS compounds detected in the environment (Hassan et al., 2020). PFOA and PFOS manufacturing and using products containing them have been phased out. Their emissions have been reduced in many countries due to their toxic and persistent nature, which accumulates in the soil and plants posing a significant risk to public health through the food chain and drinking water (US EPA, NSW EPA, EU).

Conventional soil remediation methods such as mechanochemical treatment, ball stabilization and solidification, thermal treatment, electrochemical tests, and encapsulation have been applied to remove and mitigate PFOA and PFOS from polluted soils in situ and ex-situ (Bolan et al., 2021; Mahinroosta and Senevirathna, 2020; Senevirathna et al., 2021). Some of the remediation technologies mentioned above, such as thermal treatments, are not cost-efficient due to their high energy demands (Crownover et al., 2019; Jacobs, 2019). Stabilization and solidification treatments involve the adsorption of PFAS compounds onto adsorbent surfaces; however, it does not degrade the contaminants. Also, the presence of other organic contaminations in soil reduces the adsorption efficiency of PFAS. Encapsulation treatments mitigate the PFAS compounds transported across the soil; nevertheless, long-term leaching is still unknown. Excavation and landfilling raise exposure to risk while handling contaminated soils. Electrochemical studies demonstrated the feasibility of PFAS removal from contaminated soils. Their study evaluated the transport, mobilization, and desorption of PFAS under a low constant electric field. The EK experiments achieved a limited PFAS removal from the contaminated soil, and most PFAS accumulated in the middle section of the soil. To date, limited studies have been carried out on the surfactant-enhanced EK treatment of PFAS-contaminated soils. Hou et al. (2022) investigated electrochemical destruction and mobilization of PFOA and PFOS in saturated soil. They reported a high degradation rate of 92.1% and 92.2% in the cathode region and a lower rate of 55.1% and −37.1% in the anode region (Hou et al., 2022). The study also found that PFOA and PFOS were accumulated in the soil's middle section. Wang et al. (2020) studied the electrochemical degradation of PFOA by Ti/SnO2-Sb anode via peroxymonosulfate (PMS) activation. It was hypothesized that PFOA was converted into a shorter chain and that the k_{PFOA} value rose with the increase in current density (Wang et al., 2020). In another lab-scale study, electrodialytic remediation was applied to remove PFAS from contaminated soil (Ghobadi et al., 2020a). The study found that the predominant transportation mechanism for PFOA and PFOS was electromigration towards the anode. The study suggested the promising future of in situ remediation trials to remove PFOA and PFOS.

EK remediation is a well-known in-situ method for removing heavy metal ions from soil (Ghobadi et al., 2020a). The EK remediation uses a low-intensity direct electric current that can be used in low permeability soils and mixed contaminations. Electric current assists the transport of contaminants in the soil before precipitation near the cathode zone. A low-density direct current is applied through anode and cathode electrodes strategically placed in the contaminated soil. This approach enables the mobilization of contaminants in the soil matrix. The contaminants transported towards the cathode or anode electrode compartments are pumped out (Cameselle and Gouveia, 2018). Electromigration is the primary transport mechanism for negatively charged molecules in the EK process. However, several studies demonstrated the co-transport mechanism by electroosmosis for negatively charged molecules such as oxyfluorfen and phenol (Kuppusamy et al., 2017; Meng et al., 2019).

Conventionally, the removal of organic compounds by the EK process is more challenging than metal ions due to their low water miscibility, adsorption onto the soil, or neutral charge. Therefore, researchers used the surfactant-enhanced EK process to remove soil organic contaminants. Yuan et al. (2006) evaluated the removal of hexachlorobenzene from contaminated clayey soil using TW80 and β-cyclodextrin enhanced EK system. Results revealed a lower removal rate of hexachlorobenzene in the EK process with the TW80 enhancement agent. This study recommended sodium dodecyl sulphate (SDS) as a washing solution for soil remediation due to its biodegradability and low adsorption on the soil surface (Giannis et al., 2007). Researchers found that soil treatment by simultaneous use of SDS (catholyte) and TW80 (anolyte) resulted in improving the removal efficiency of polycyclic aromatic hydrocarbon (PAH) from the soil (Boulakradeche et al., 2015). SDS enhanced-EK remediation for removing kerosene from contaminated soils increased the removal of the contaminant from 40% in non-enhanced EK to 55% (Fardin et al., 2021). Using Tween80 and enhancement agent in the EK achieved 45% kerosene from the soil. Sodium cholate is a non-toxic biosurfactant with a large, rigid, and planar hydrophobic moiety of a steroid nucleus with two or three hydroxyl groups (Sugioka et al., 2003). Sodium cholate (NaC) was selected to represent biosurfactant due to its great solubilization capacities, high biodegradability and biocompatibility. A study reported that exposure to NaC did not affect cell morphology; therefore, it is an environmentally friendly surfactant (Dong et al., 2009). According to the research (Zeng et al., 2013), NaC demonstrated a high solubilization rate of 2,4,6-TCP and is an outstanding environmentally friendly surfactant for surfactant-enhanced soil remediation.

Earlier EK studies showed that PFAS compounds are transported in the soil by electroosmosis and electromigration mechanisms and accumulate in the midsections. Still, the main transport mechanism for PFOA was towards the anode by electromigration (Hou et al., 2022; Ghobadi et al., 2020a). One of the methods to enhance organic compound removal in the soil is surfactant-enhanced EK processes (Fardin et al., 2021). Three types of surfactants, nonionic (TW80), anionic (SDS), and biosurfactant (sodium cholate), were evaluated in this study for PFOA removal from kaolinite soils. The negatively charged SDS surfactant was introduced to the cathode zone to electromigrate towards the anode with the

Table 1
Kaolin soil properties.

Soil characteristics	Values
Particles size analysis	
Clay	46.81
Silt	51.17
Sand	2.02
Permeability (m/s)	4×10^{-10}
Density (g/cm^3)	1.45
Porosity (kg/m^3)	633
Organic matter	0.02
CEC (cmol/kg)	2.65
pH	4.68
Electrical conductivity (mS/cm)	0.26

PFOA molecules. Sodium cholate is another anionic biosurfactant introduced to the cathode to enhance PFOA removal towards the anode zone. TW80 was introduced to the anode and is expected to react with PFOA in the soil and transport towards the cathode electrode.

This study investigates the feasibility of the surfactant-enhanced EK process for PFOA removal. Kaolinite was used as a model soil to eliminate soil impurities (organic and inorganic matter) effect on the PFOA removal by the EK process. Surfactants are introduced to increase the mobility and solubility of PFOA, hence increasing the removal efficiency (Fardin et al., 2021). However, no study has investigated the surfactants' role in enhancing the EK removal of PFOA from soils. Three scenarios were evaluated for PFOA treatment in this study (i) conventional EK process, (ii) enhanced EK process using an anionic surfactant (iii) enhanced EK process using a nonionic surfactant. Anionic and nonionic surfactants were applied to the cathode or anode to study their impact on the EK performance. Anionic surfactants (SDS and sodium cholate) were added to the catholyte. Nonionic Tween80 was added to the anolyte and catholyte separately to evaluate the EK system's performance for removing PFOA from contaminated kaolin soils. It has been frequently used as a flushing solution and enhancing agent in electrokinetic treatments to remove HOC due to low toxicity, cost-effectiveness, low polarity, and high solubility capacity (Cheng et al., 2017). The study assessed the surfactant-EK process's performance when the test duration increased from one to two weeks.

2. Materials and methods

2.1. Materials and soil preparation

Kaolin clay obtained from Keane Ceramic Pty. (Australia) was selected as model soil for the EK experiments because of its low permeability, low carbon content and low cation exchange capacity. The supplier provides the characteristics of the kaolin soil used in the EK tests (Table 1). The absence of organic matter content and non-reactivity of the kaolin soil enables the study of PFOA mobility under direct electric fields in the EK system without any interference. The kaolin soil was artificially spiked with PFOA for all EK experiments with a 100 mg/kg target concentration. Previous studies reported 123.6 mg/L and 300 mg/L PFOA concentration in China and 33 mg/L to 373 mg/L PFOA concentration in the US in primary contaminated sites (Brusseau et al., 2020; Zhan et al., 2020). Also, the maximum PFOA concentration detected in primary source contaminated sites in Australia ranged from 10 mg/kg to 460 mg/kg (CARE C Assessment, 2017). Each experiment used 1000 g of kaolin. Initially, 1 g of PFOA was completely dissolved in 1000 mL of Milli-Q water, yielding a 1000 ppm stock solution. The stock solution was diluted to 100mg/L for each experiment, and prepared 1 L solution was mixed with 1000 g of kaolin. The spiked soil was then kept at room temperature for at least 72 h, with periodic stirring to maintain consistent PFOA homogeneity. The saturated soil was then loaded into the reactor in layers and uniformly compacted to ensure consistent distribution of PFOA. A multimeter was used to measure the pH and electric conductivity of soil specimens (model HACH HQ40D).

2.2. Electrokinetic cell setup

Fig. 1 depicts a schematic diagram of the EK test. The EK experiments were carried out in a plexiglass reactor measuring $23 \times 8 \times 11$ cm^3. The reactor had three compartments: two electrode chambers at each end of the sample compartment, separated by a plexiglass plate. The contaminated soil was 19 cm deep in the middle. A DC bench power supply (EA-PS 3016-10B, EA Electro-Automatik) was used to apply a constant current (10 and 20 mA), and the electric current was measured and recorded hourly with a multimeter (Keithley 175 Autoranging multimeter). Two graphite rod electrodes (15×1 cm) were inserted vertically into the reactor's electrode chambers on both sides. Graphite Australia Pty Ltd supplied the graphite electrodes. Two filter papers were placed between the soil and the electrode chambers and supported by a perforated plexiglass plate to keep the soil out of the electrolyte cells. The cationic surfactants SDS and NaC (purchased from Sigma Aldrich) were added to the catholyte solution in the cathode compartment, and the nonionic surfactant (TW80) was added to the anolyte solution in the anode compartment. To replenish water losses due to electroosmotic flow, distilled water was fed into the anolyte compartment regularly. Throughout the experiment, the current intensity and volume of the cathode effluent were measured regularly.
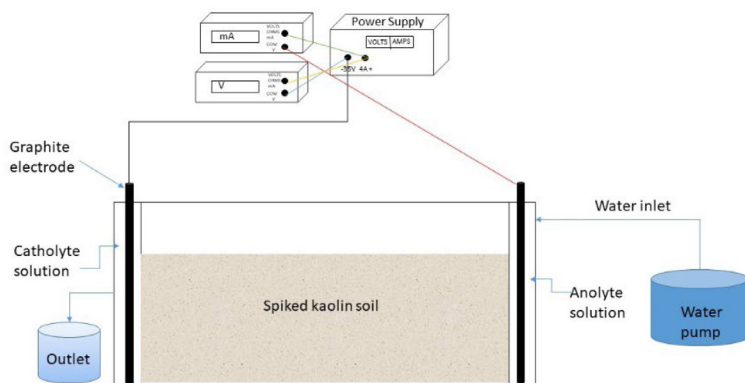
Fig. 1. Schematic diagram of the electrokinetic cell setup.

Table 2
Electrokinetic experiments with kaolinite soils spiked with PFOA.

Exp No.	Target Contamination	Concentration of target contamination (mg/kg)	Surfactant and dosing point	Current (mA)	Surfactant Concentration (% w/w)	Duration (says)
E1	PFOA	100	NA	10	–	7
E2	PFOA	100	NA	20	–	7
E3	PFOA	100	SDS/cathode	10	5	7
E4	PFOA	100	SDS/cathode	20	5	7
E5	PFOA	100	NaC/cathode	10	5	7
E6	PFOA	100	NaC/cathode	20	5	7
E7	PFOA	100	TW80/anode	10	5	7
E8	PFOA	100	TW80/anode	10	5	7
E9	PFOA	100	SDS/cathode	20	5	14
E10	PFOA	100	NaC/cathode	20	5	14

Table 3
Mass balance and removal efficiency of PFOA in EK experiments.

Experiment	Initial concentration in soil (mg/kg)	Duration (Days)	Residual PFOA mass (mg/kg)	Mass balance	Removal Rate (%)
E1	100	7	88.23	105.74	14.49
E2	100	7	81.28	95.29	19.00
E3	100	7	86.49	100.67	13.84
E4	100	7	84.21	106.70	15.73
E5	100	7	88.06	107.01	17.67
E6	100	7	67.03	97.50	32.66
E7	100	7	86.67	93.51	12.90
E8	100	7	92.91	96.59	7.65
E9	100	14	54.02	94.22	45.98
E10	100	14	24.30	92.16	75.68

2.3. Test design

Ten distinct EK tests with varied surfactants and experimental settings were undertaken at room temperature. Table 2 lists the results of the EK tests. The flushing solutions were SDS (5% m/m), Tween 80 (5% m/m), and sodium cholate (5% m/m), and the electrolyte solution was distilled water. All the tests employed an initial concentration of 100 mg/kg PFOA kaolin mixture. SDS and NaC, cationic surfactants, were placed into the cathode chambers, while Tween80, a nonionic surfactant, was loaded into the anode chamber; additionally, the fluid level in the inflow reservoir was kept constant to guarantee a constant steady hydraulic gradient throughout the soil. To evaluate the effect of EK remediation on PFOA removal from kaolin, E1 and E2 were performed without any enhancement, as indicated in Table 3. It was then utilized to examine the efficacy of surfactant-enhanced EK tests carried out under identical experimental settings. For one week, E4, E6, and E8 EK tests were carried out with a 20 mA constant current gradient and E3, E5 and E7 EK tests were carried out at a 10 mA constant current. The PFOA removal effectiveness was investigated over two weeks at a steady current of

20 mA in E9 and E10 EK tests. For one and two-week studies, the voltage and change in current of EK tests were recorded using a multimeter.

At the end of the experiments, aqueous solutions from anode and cathode chambers and electrode assemblies were collected. The soil sample was then divided into five equal portions, mixed and homogenized, and dried in an oven overnight at 105 °C, with 5 g of dried soil taken in duplicate from each section to extract and analyze PFOA concentrations. The PFOA content in the soil was determined using a triple methyl alcohol extraction method, in which 5mL of methyl alcohol was mixed with 5 g dry soil, shaken at 250 rpm and 22 °C for 60 min, sonicated at 30 °C for 30 min, and then centrifuged at 9000 rpm for 10 min. The supernatants after each extraction were combined, diluted, filtered (PTFE syringe filter), and put into vials for UHPLC-MS/MS analysis (LC/MS 8060, Shimadzu, shim pack column 1.6 m, 2.0 mm 50 mm). Recovery of the extraction was tested. Therefore, the data were calculated to determine the mass balance and removal efficiency. The removal efficiency was calculated from (Eq. (1)), where C_i (mg/kg) is the initial concentration of PFOA, and C_f (mg/kg) is the final PFOA concentration after EK experiments. Meanwhile, the pH and EC of soils prior to the experiment and each section post-experiment were measured by making a soil suspension with dry soil to DI water ratio of 1:5 (w/v).

$$\text{Removal Efficiency} = \frac{c_i - c_f}{c_i} \times 100 \tag{1}$$

3. Results and discussion

3.1. Electrical current

Fig. 2 presents changes in the electric current and voltage over time for experiments E1 to E10. EK experiments E1–E8 were carried out under a constant current of 10 mA and 20 mA for 168 h, and experiments E9–E10 were conducted under a constant current of 20 mA for 336 h. The electric potential across the EK cell varied significantly throughout all experiments. The voltage change with treatment time for the EK experiments performed at 10 mA for 168 h is shown in Fig. 2a. Because of the stable operating conditions, the current remained constant throughout the EK treatment, and the voltage change gradually increased over time. As shown in Fig. 2a, the E1 and E7 experiments reached a maximum voltage of 35 V after 144 h, the E5 experiment reached a maximum voltage of 35 V after 127 h, and the E3 experiment reached a maximum voltage of 19 V after 96 h and remained stable, with slight fluctuation, until the experiment ended. The gradual increase in voltage in experiments E1, E3, E5, and E7 could be attributed to increasing soil resistance over time caused by the precipitation of dissolved ions. The results showed that the voltage in the 10 mA experiments reached 35 V after about 140 h, while the voltage in the 20 mA experiments reached 35 V after about 48 h (Fig. 2c). The rapid increase in voltage in the 20 mA experiments is due to a faster electrolysis reaction, which leads to faster transport and the meeting of acid and alkaline fronts in the soil. The electric current remained constant at the start of the EK experiments, then nearly stabilized at 34 V from 48 h to the end of the experiments. As previously stated, an increase in voltage reflects soil resistivity to electric current caused by acid and alkaline fronts colliding in soil and ions precipitating (Eq. (4)). As demonstrated in previous studies, a decrease in electric current causes a gradual increase in voltage across the EK cell (Fardin et al., 2021; Ghobadi et al., 2020b).

The fluctuations in electric current were comparable in the EK experiments performed at 20 mA (Fig. 2b). The electric current initially remained constant at 20 mA for several hours due to the electrolysis reaction at the anode and increasing dissolved ions in the pore solution (Eqs. (2) and (3)) as the electric current through the soil cell is strongly correlated with the content of free ions; thus, the electric current is an important factor influencing the utilization of contamination transport through the soil. As shown in Fig. 2b, the maximum electric current in experiments E2 and E4 was 20 mA for 51 h. After 168 h, the current gradually decreased to 7.63 mA in E2, dropping sharply to 7 mA in E4 after 87 h before returning to 8.7 mA until the experiment ended. In E6, the current remained constant at 20 mA for 44 h, then fell to 14 mA after 94 h, leapt to 18 mA, and again gradually decreased to 10 mA at the end of the study. In E8 electric current fell from 20 mA after 34 h to 8.7 mA; in experiment E2, it stayed at 20 mA for 50 h. After that, it gradually dropped to 8 mA. Because of higher soil resistance, the decrease in current in experiments E2, E4, E6, and E8 correlates to an increase in electric potential (Fig. 2c).

Fig. 2b and c show the electric current and potential profile in experiments E9 and E10, conducted for 336 h. Although the duration of the experiments was doubled, the change in current and voltage was similar to 168 h experiments. In experiments E9 and E10, the electric current reached 19.22 mA and 19.46 mA, respectively, before dropping sharply after 45 and 39 h. Then, the electric current dropped steadily and reached 3 mA at the end of experiment E9 and 2 mA at the end of experiment E10. In general, the electric current at the end of experiments E9 and E10 was lower than that in experiments conducted for one week at 20 mA (Fig. 2b).

Oxidation at the Anode:

$$(2H_2O - 4e^- \rightarrow O_2 + 4H^+) \tag{2}$$

Reduction at the Cathode
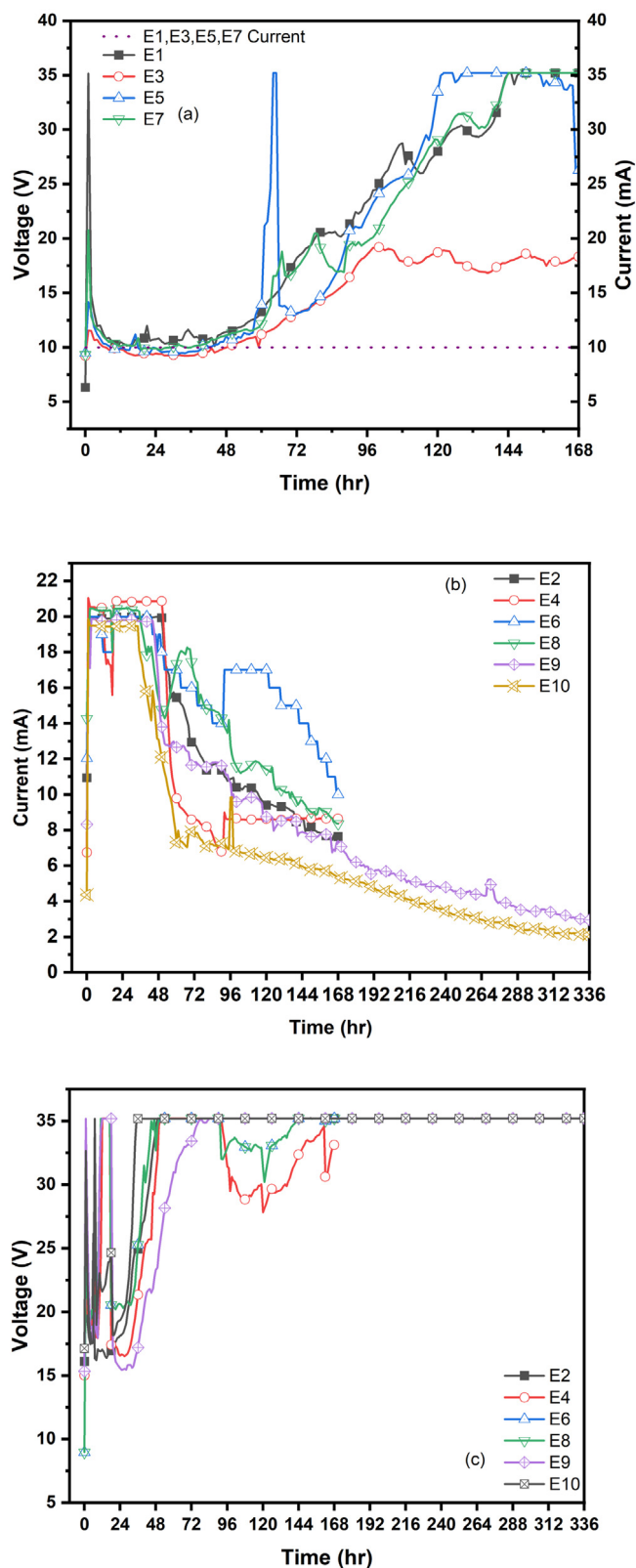
$$(2H_2O + 2e^- \rightarrow H_2 + 2OH^-) \tag{3}$$

Fig. 2. (a)The electric current and potential in the EK experiments at 10 mA for 168 h (E1,E3,E5,E7) (b) Change in current over time in the EK experiments at 20 mA for 168 and 336 h (c) change in voltage over time of EK tests at 20 mA for 168 and 336 h.
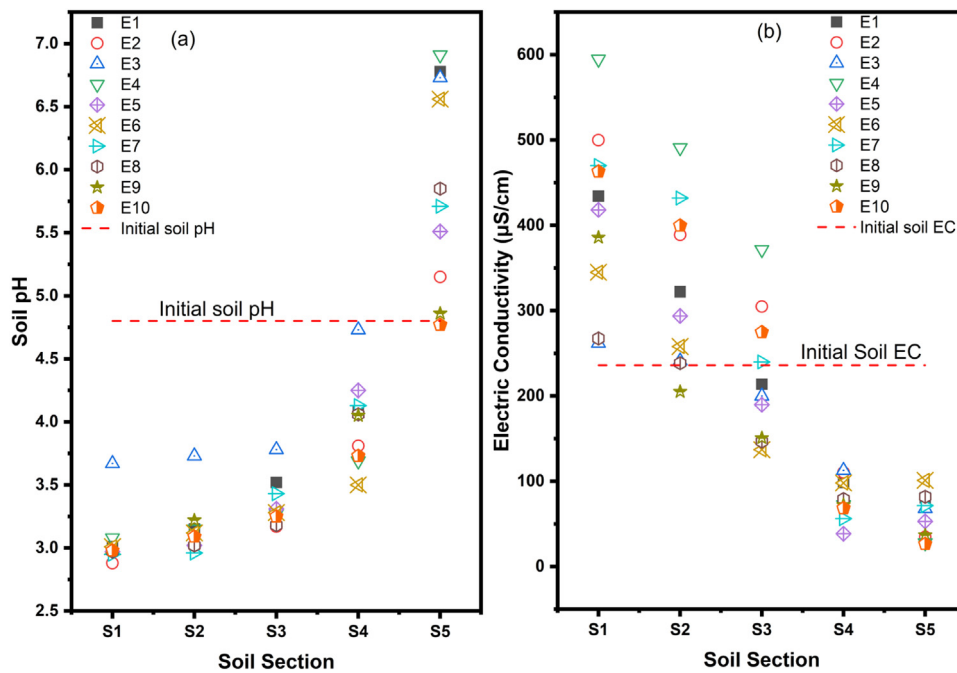
Fig. 3. (a) pH of the soil across soil sections for all experiments; (b) electric conductivity of the soil across soil sections for all experiments.

3.2. Soil pH and electric conductivity

The pH distribution across the soil from anode to cathode (sections S1 to S5) after the EK experiments is shown in Fig. 3a. All EK tests were conducted without pH control. After the EK treatment, soil pH in sections close to the anode compartment decreased to lower than the initial soil pH and gradually increased in the soil sections towards the cathode zone. However, the soil pH in section S4 was close to the initial soil pH (Fig. 3a). Soil pH distribution across sections S1–S4 was lower than the initial soil pH due to the electrolysis reaction at the anode and the transport of the H^+ from the anode to the cathode zone (Eq. (3)). An increase in the soil pH in the sections near the cathode is due to the production of hydroxide ions (OH^-) from the water electrolysis reaction at the cathode (Eq. (4)). The effective ionic mobility of the hydrogen ions is approximately 1.8 times higher than that of hydroxyl ions (Acar and Alshawabkeh, 1993). As observed in Fig. 3a, the low soil pH in sections S1 to S4 is due to the faster migration of hydrogen ions than hydroxide ions under the electric field. However, the soil pH approached the initial value in section S4 and raised significantly in section S5 due to the advancement of the alkaline front from the cathode. It is noteworthy that the sorption of PFOA in soils increases with decreasing pH (Oliver et al., 2019), as discussed in the following sections.

Fig. 3a displays the pH values of EK processes under a 10 mA constant current for 7 days. Experiment E3 exhibited slightly higher soil pH than other EK experiments. The change in pH is almost insignificant across the first three sections (S1–S3); however, there was a sudden spike in experiment E1 pH in sections S4 to S5. While experiments E1, E5 and E7 operated under the same experimental conditions, they exhibited a gradual increase in the soil pH from S1 to S5. The soil pH in the EK experiments carried out at 20 mA for 7 days is shown in Fig. 3a. The soil pH in sections S1 to S4 was lower than the initial soil pH (pH 4.4) in all experiments, although experiment E8 exhibited a slightly higher pH than other experiments. The higher soil pH is due to the advancement of the acid front in the soil from the anode to the cathode electrode. The soil pH increased over the initial soil pH in section S5 of all experiments due to the propagation of the alkaline front from the cathode zone. The highest pH was recorded in section S5 of experiment E4 (pH 6.8) with an SDS enhancement agent. Fig. 3a shows the soil pH of the EK experiments conducted for 2 weeks. The soil pH in experiment E9 was slightly higher than in experiment E10 in all soil sections. The soil pH increased gradually in sections S1 to S4 and surpassed the initial soil pH in section S5. Section S5 of Experiment E10 exhibited the lowest pH (pH 4.6) compared to other experiments due to the longer EK duration that allowed the acid front to sweep across the soil specimen.

After seven days of the EK treatment, the kaolinite soil's electric conductivity (EC) decreased in sections S1 to S4 and slightly increased in sections S4 and S5 (Fig. 3b). In the fourteen-day EK experiments, the EC of the soil sections decreased in sections S1 to S5 (Fig. 3b). The soil EC increased due to the higher concentration of soluble soil ions at the anode section (S1), as the acidic condition facilitated the dissolution of ionic species (Ghobadi et al., 2020b). The high value of EC is correlated to the presence of free protons. The EC decreased when the concentration of ionic species in soil pore fluid probably decreased due to meeting the acid and alkaline front in the soil section S5. Fig. 3b presents the EC of
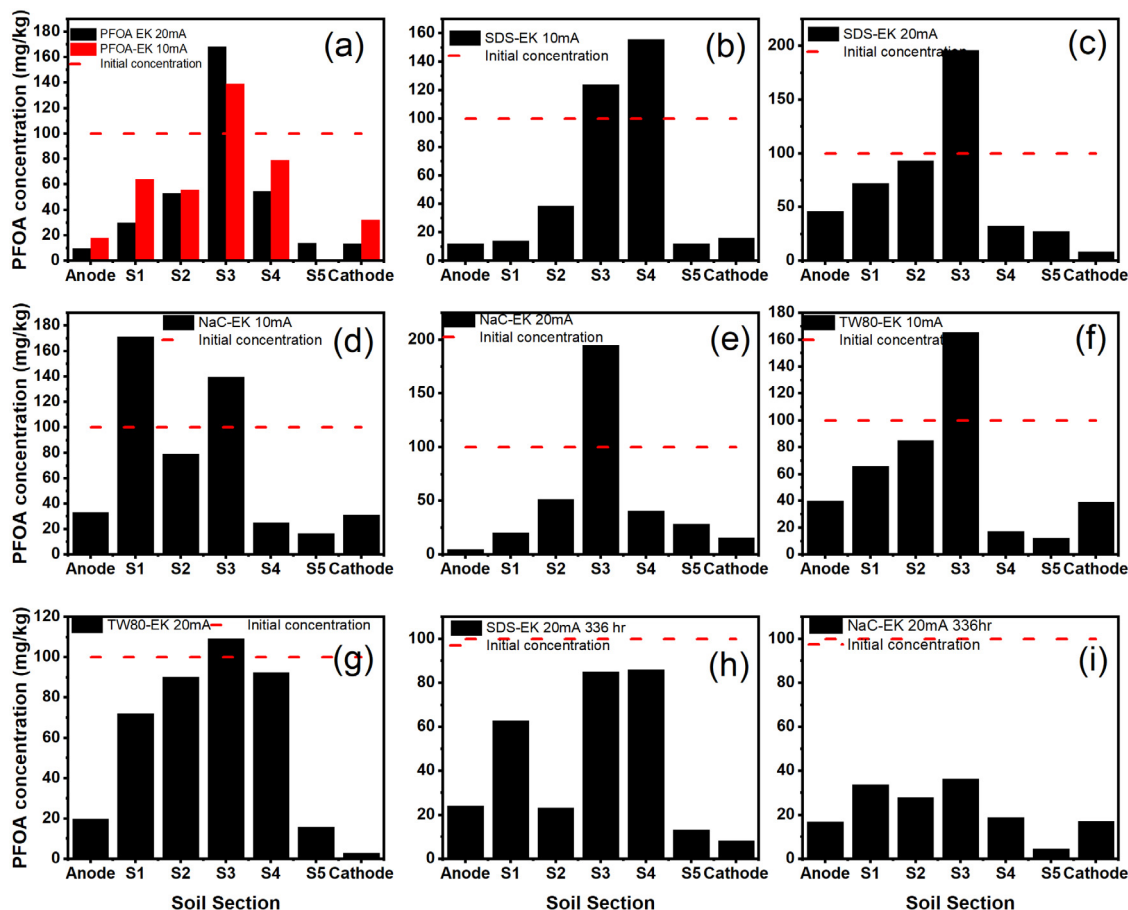
Fig. 4. PFOA concentration through the soil sections (a) E1–E2; (b) E3; (c) E4; (d) E5; (e) E6; (f) E7; (g) E8; (h) E9; (i) E10.

experiments E9 and E10 conducted for two weeks; both experiments showed a trend of a gradual decrease in soil EC from S1 to S5, with experiment E9 having the lowest EC values. Overall, the EC of experiments E9 and E10 was lower than that of experiments E1 to E8 due to the longer treatment time and the higher electric current than experiments conducted at 10 mA, i.e. E1 E3, E5, and E7. It should be mentioned that the enhancement agent could influence the soil EC due to the interaction between the OH⁻ and H⁺ ions generated at the cathode and anode with the surfactant agent, causing a fluctuation in the soil EC. Accordingly, the soil EC in section S1 was higher in Experiments E1 and E2 performed without enhancement agents.

3.3. Performance of EK with different surfactants

Fig. 4 displays the residual concentration of PFOA across the soil sections after the EK experiments. As shown in Fig. 4, after EK experiments, PFOA is transported from the anode and cathode zones and concentrated in the soil middle section (S3). These results agree with the earlier studies that showed PFAS compounds' transport and concentration in the soil's middle section (Niarchos et al., 2022). PFOA transportation was similar in soil sections S1, S2, S4 and S5. Studies on soil sorption of PFAS indicated that PFOA sorption increased with decreasing the soil pH (Oliver et al., 2019). Generally, PFOA is a negatively charged water-soluble contaminant with a carboxylic functional group (Fig. 5c). Therefore, PFOA would be readily available in soil pores for transportation by the electrokinetic process. Due to its negative charge, PFOA electromigration will be towards the anode, while electroosmosis transportation will be towards the cathode, leading to its distinctive distribution in the soil after the EK process. In addition, PFOA sorption will be affected by the soil pH due to the advancement of the alkaline and acid fronts in the EK process. The charge of soil will be positive at pH<pH $_{ZPC}$ and negatively charged at pH>pH $_{ZPC}$. The adsorption of negatively charged PFOA increases when the soil is positively charged near the anode since its pH<pH $_{ZPC}$ (Fig. 3a). In contrast, PFOA adsorption on the soil decreases near the cathode due to the high soil pH or when pH>pH $_{ZPC}$. Thus, PFOA removal was higher at the cathode zone due to (i) the electrostatic repulsion between PFOA and the soil surface and (ii) the electromigration of PFOA towards the anode. Another study also noted the
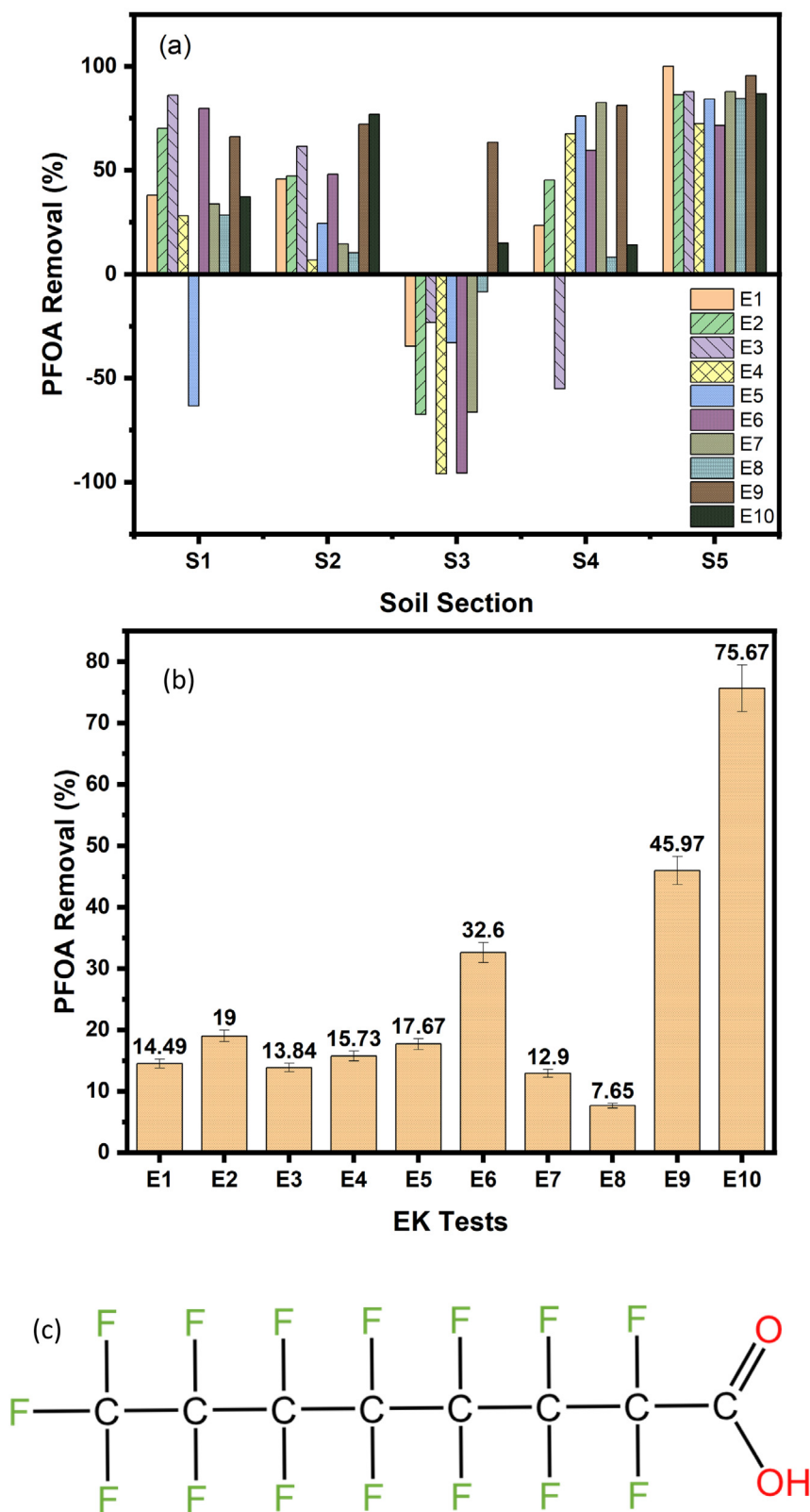
Fig. 5. (a) PFOA removal in each section for all EK experiments (b) PFOA removal efficiency (%) for EK experiments (c) PFOA chemical structure.

negative relationship of PFOA with pH, where an increase in pH resulted in decreasing sorption of PFOA for soil (Groffen et al., 2019). The accumulation of PFOA in the low pH region near the anode could be due to PFOA electromigration and electrostatic interaction with the positively charged soil surface. These findings would explain the PFOA concentration in sections S1 and S2 and a slight drop in sections S4 and S5. As soil pH increased in the latter sections, a tangible decline in the PFOA concentration was observed near the cathode zone. Considerable PFOA concentration in section S3 is attributed to PFOA transport from the cathode and anode by electromigration and electroosmosis mechanisms and accumulation in the middle section. Due to the charge difference, the negatively charged PFOA electromigrated from the cathode to the anode zone, whilst the electroosmosis flow carried water-soluble PFOA from the anode towards the cathode. This phenomenon was also observed in earlier studies (Hou et al., 2022; Sörengård et al., 2019). As shown in Fig. 4a–i, the accumulation of PFOA in the anode region indicates that electromigration is the main mechanism for PFOA transport in the soil sections near the cathode and is consistent with the pH distribution across soil sections. At the same time, electroosmosis was responsible for PFOA transport across the soil specimen and accumulations in the catholyte chamber. As displayed in Fig. 4a–b, the catholyte chamber contains more PFOA concentration than the anolyte chamber. Generally, the soil near the anode acquires a positive charge during the EK process, favoring PFOA adsorption. The effect of soil charge on the PFOA removal is due to the electrostatic interaction between negatively charged PFOA compounds and positively charged soil surface. It is noteworthy that significant PFOA removal from the soil Section 4 despite the soil pH is lower than pHzpc (Figs. 4 and 5a). The results agree with the literature (Niarchos et al., 2022; Sörengård et al., 2019), suggesting that electromigration is a preferable transport mechanism for PFOA near the cathode.

The profile of the PFOA distribution in the soil in the unenhanced EK experiments, E1 and E2 (Fig. 4a), was similar to the enhanced EK experiments, E3 to E8 (Fig. 4b–g). Increasing the EK duration to two weeks promoted PFOA transport from the cathode to the anode zone. As seen in Fig. 4h–i, the accumulated PFOA in soil sections is below the initial PFOA concentration. In contrast, in one-week-long experiments, the accumulated PFOA concentration in the anode region is higher than the initial value. However, the lower PFOA concentration close to the anode zone was probably due to soil compaction over time, reducing the electroosmosis flow and PFOA transport from the anode zone. Interestingly, increasing the electric current from 10 to 20 mA increased PFOA concentration in section S3 in experiments E4 and E6 (Fig. 4c and e). These results may be explained by the fact that higher electromigration and opposing electroosmotic flow met at a higher rate, leading to more PFOA accumulation in the middle section. However, the lowest concentration of PFOA across the soil sections was observed in experiment E10, followed by experiment E9, due to a longer treatment time under 20 mA electric current.

As shown in Fig. 5, PFOA removal in the unenhanced EK experiment E1 was 14.49%. An extra 31% PFOA removal was achieved when the electric current of the unenhanced EK process increased from 10 mA to 20 mA in experiment E2 (∼19% total PFOA removal). The PFOA contaminant was removed from soil sections S1, S2, S4 and S5 and accumulated in section S3 (Fig. 5a). For all experiments, the highest PFOA removal efficiency was between 85% and 95% in section 5, except for experiments E4 and E6, where the removal efficiency was 72%. Additionally, most PFOA concentrated in the soil section S3 of experiments E1 to E8. In contrast, experiments E9 and E10 exhibited 63.51% and 14.92% PFOA removal from the soil section S3, indicating a better PFOA removal efficiency in the EK experiments conducted for two weeks. In the enhanced EK processes conducted at 10 mA, PFOA removal was 13.84%, 17.67% and 7.65% in experiments E3, E5 and E7, respectively (Fig. 5b). The highest PFOA removal was achieved in experiment E5 with a sodium cholate enhancement agent. PFOA removal from the soil increased with increasing the electric current from 10 mA to 20 mA in experiments E4 and E6 conducted with anionic surfactants (SDS or NaC). Still, it decreased in experiments E8 conducted with nonionic surfactants (TW80). The increased electric current promoted the soil's electrolysis reaction and PFOA transport. However, the lower PFOA removal in experiment E8 could be due to the enhancement agent application in the anode that had an insignificant impact on the PFOA removal. The enhancement of PFOA removal by adding TW80 in the anode compartment was hindered due to kaolinite soil compaction over time, reducing the electroosmosis flow from the anode to the cathode zone. For all EK experiments, the electroosmotic flow was high at the beginning of the experiment, notably during the first 72 h, and reached the highest volume of 250–300mL in 24 h. However, after 72 h, the electroosmosis flow declined quickly when the pH across soil sections dropped due to acid front propagation across the EK unit. This phenomenon was observed in low pH buffering soils, resulting in a sharp decline in the electroosmosis flow and contaminants removal (Cameselle and Gouveia, 2018; Saichek and Reddy, 2003). Also, the low dielectric conductivity of TW80 causes low dissolution of ions, and the relative movement of ions could be responsible for the low electroosmotic flow in TW80 enhanced EK since the electroosmotic flow is directly proportional to dielectric conductivity (Cheng et al., 2017). TW80 nonionic surfactant has a low dielectric constant, high viscosity and low CMC; these factors contributed to the low electroosmotic flow and PFOA removal in the electrokinetic tests. Previous studies reported that TW 80 efficiency is highly hindered by sorption onto the soil, and its efficiency highly depends on the soil properties (Fardin et al., 2021).

The highest PFOA removal in one-week EK experiments was 32.6% in experiment E6 with sodium cholate enhancement agent, followed by 15.73% PFOA removal in experiment E4 with SDS enhancement agent. SDS was selected as a conventional enhancing agent to improve the transportation of PFOA to the anode. The movement of the negatively-charged SDS under an electric field is contrary to the direction of electrokinetic flow. However, the experimental results revealed less removal achieved in the SDS-EK, which can be caused by the reaction of SDS at the cathode producing salt and hindering the transportation towards the anode. Therefore, the SDS-enhanced EK system did not significantly improve the removal efficiencies. PFOA removal in experiment E6 was twice that in experiment E4. In experiments E9 and E10,

the duration of the EK process was extended to 2 weeks to investigate its impact on the EK performance. PFOA removal in experiment E9 was three times higher than in experiment E4, and in experiment E10, it was 2.3 times higher than in experiment E6. PFOA removal was 45.97% and 75.67% in experiments E9 and E10, respectively (Fig. 5b). In general, results revealed that sodium cholate experiments achieved higher PFOA removal from the kaolinite soil under 10 mA and 20 mA electric current for both one and two weeks experiments. TW80 enhanced EK experiments E7 and E8 achieved the lowest PFOA removal rate, 12.90% and 7.65%, respectively (Fig. 5b). The performance of sodium cholate enhanced EK experiments is better than the SDS, and TW 80 enhanced EK experiments and unenhanced EK experiments (Fig. 5b). The increased PFOA removal efficiency of sodium cholate enhanced EK tests could be due to the greater PFOA dissolution into surfactant micelles and the transport of PFOA-containing micelles under the induced electric potential. An earlier study observed that NaC had superior solubilization of 2,4,6-trichlorophenol compared to SDS and cyclodextrin, achieving a higher removal rate (Zeng et al., 2013). The study also concluded that sodium cholate is a bile salt with a lower micelle aggregation number than most typical aliphatic surfactants and a lower critical micelles concentration (CMC) than other surfactants such as the SDS. The strong interaction between PFOA and NaC biosurfactant assisted the transportation of PFOA to the anode.

The results underline the surfactant-enhanced EK process to improve PFOA removal from kaolinite soil, which was much higher than reported in previous studies using contaminated soils. In real soil, the expected PFOA removal could be lower than in the experiments with kaolinite soil due to the higher cation exchange capacity and presence of organic matter that influences PFOA transport in the soil. Unlike heavy metal contaminants, PFOA showed irregular transportation in the EK process due to its electromigration towards the anode and electroosmosis transport towards the cathode. PFOA's relative high solubility in water, about 9.5 g/L, promotes the electroosmosis transport mechanism towards the cathode. Besides, low soil pH near the anode would encourage PFOA adsorption, requiring a longer processing time for removal. It is evident that increasing the EK duration from 1 to 2 weeks resulted in 2.3 times higher PFOA removal in the NaC-enhanced EK process.

3.4. Impact of PFOA concentration

Surface-enhanced EK tests were performed to examine the influence of change in PFOA concentration on removal efficiency. Compared to Tween80 nonionic surfactant, NaC and SDS surfactant enhanced EK tests revealed a better removal rate. Furthermore, NaC and SDS surfactant-enhanced EK tests were carried out for 2 weeks at 10 mg/kg PFOA concentration to evaluate the impact of PFOA concentrations on its removal from kaolinite soil.

Fig. 6a. outlines the change in voltage and current over 336 h, demonstrating the voltage decreased initially and subsequently increased to 35.21 V before stabilizing after 48 h till the end of the experiment. Within the first 48 h of the NaC-enhanced experiment, there was a strong fluctuation in the EK voltage before it stabilized. Fig. 6b illustrates the change in current in both tests; throughout the first 48 h for the SDS enhanced EK test and 72 h for the NaC enhanced test, the current remained constant at 20 mA. The electric current gradually dropped to 2 mA and 3 mA after 336 h.

Change in pH in soil sections of EK tests enhanced with 5% NaC and SDS tests presented in Fig. 6c. pH in soil sections increased gradually towards S5. NaC enhanced EK experiment displayed slightly higher pH in S4 and S5 compared to SDS enhanced test. S4 in NaC enhanced EK test reached the initial pH, whereas SDS enhanced tests displayed a lower pH value. Both tests showed a gradual increase towards section S5. The change in pH trend is similar to other EK tests.

Fig. 6d illustrates PFOA concentration in each soil section in both EK tests with 10 mg/kg PFOA concentration. At the end of the experiments, the PFOA removal was 37.42% and 26.59% in NaC and SDS enhanced EK tests, respectively. Fig. 6d shows the PFOA removal percentage from each soil section at the end of the EK experiments. The highest PFOA removal efficiency in both experiments was between 85% and 55% in section S5. Section S1 in NaC enhanced EK test achieved 73% PFOA removal. Additionally, most PFOA concentrated in the soil section S3 in both tests. The overall PFOA removal rate in EK experiments with 10 mg/kg concentration was lower than 100 mg/kg PFOA concentration. In both EK tests removal rate has been decreased to half. The explanation for lower PFOA removal at 10 mg/kg could be due to the lower PFOA ions in soil pore fluid and readily available for migration and transport compared to the EK tests with 100 mg/kg PFOA concentration.

3.5. Energy consumption and mass balance

The energy consumed by EK studies is a significant element in determining the treatment's total cost. (Ghobadi et al. 2020b). The specific energy consumption (SEC) for all experiments was calculated using Eq. (4), where V_s is the total soil volume (kg), V is the average applied voltage (V), I is electric current (A), and t is the treatment time (h).

$$E_u = \frac{1}{V_s} \int VI \, dt \tag{4}$$

The EK tests' results indicate that raising the electric current resulted in a considerable rise in power usage (Fig. 7). Generally, the specific power consumption increased with the electric current increase of the EK process. In the unenhanced EK processes, the power consumption increased from 0.0255 kWh/kg in experiment E1 to 0.050 kWh/g in experiment E2 due to increasing the electric current from 10 mA in experiment E1 to 20 mA in experiment E2. A similar
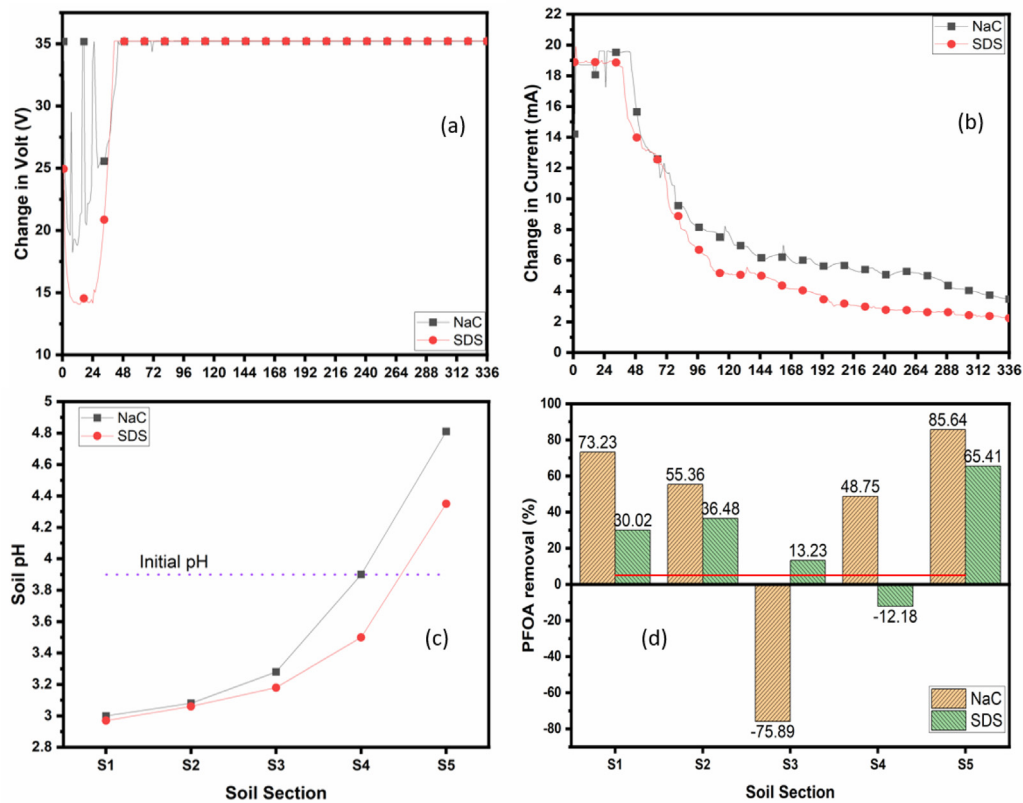
Fig. 6. (a) PFOA concentration in soil sections. (b) The PFOA removal rate in each soil section.
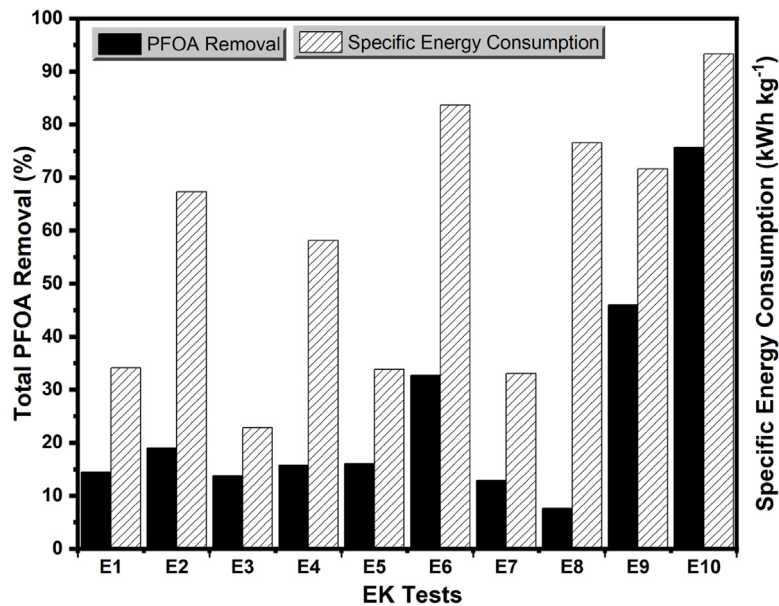

Fig. 7. Total removal of PFOA and Specific Energy Consumption.

observation was recorded in surfactant-enhanced EK processes; SEC was 0.017 kWh/kg and 0.044 kWh/kg in experiments E3 and E4, respectively. The SEC is proportional to the electric potential and current of the EK experiment; the higher the electric potential and current, the higher the SEC is (Eq. (4)). Accordingly, the SEC in experiment E6 was higher than in

experiments E4 and E8 due to the higher electric potential in experiment E6 (Figs. 2b and 2c). SEC in experiment E6 was 0.061 kWh/kg, and it was 0.04 kWh/kg and 0.057 kWh/kg in experiments E4 and E8, respectively. Eq. (4) also shows that the SEC increases with increasing the duration of the EK process in experiments E9 and E10. At the end of the EK process, the SEC was 0.0699 kWh/kg in experiment E10 and 0.052 kWh/kg in experiment E9. For EK processes conducted at 20 mA, Experiment E10 recorded the highest SEC, followed by experiment E6> experiments E8> experiment E9> experiment E2> experiment E4. It is worth noting that although experiment E6's duration was longer than experiments E6 and E8, the SEC in experiment E9 was lower than in experiments E6 and E8. The higher average electric potential and current in experiments E6 and E8 was probably the reason for the higher SEC in these experiments compared to experiment E9.

The mass balance and total PFOA removal rate in the unenhanced and enhanced EK experiments are depicted in Table 3. Each experiment's mass balance was estimated as the total PFOA residual in the soil and PFOA residues in the electrolyte solutions/pore water, divided by the initial quantity of PFOA in the soil (Ghobadi et al., 2021). As indicated in Table 3, the mass balance of unenhanced EK studies ranged between 84.72 and 87.93%. Enhanced EK treatments resulted in a mass balance of 92.16–106.7%. Changes in the processing time substantially influenced the efficiency of PFOA removal. EK tests conducted under the same experimental settings but with a different processing period demonstrated a twofold improvement in PFOA removal efficiency (Table 3). Interestingly, TW80-enhanced EK had the lowest PFOA removal rate, whereas NaC-enhanced EK experiments showed the highest PFOA removal rate of 32.66% among all EK experiments performed within one week. After two weeks of the EK treatment at 20 mA, NaC-enhanced EK experiments achieved 75.68% PFOA removal, followed by 45.98% PFOA removal in the SDS-enhanced EK experiment.

4. Conclusion

This study investigated the remediation of PFOA-contaminated kaolin as a model soil on a laboratory scale. Initially, PFOA removal was studied with conventional EK treatment without enhancement agents to compare with the surfactant-enhanced EK experiments. It was hypothesized that the EK process alone would not provide new data for removing PFOA from contaminated soil due to its persistent nature in the environment. Therefore, introducing different surfactants assisted PFOA removal from the kaolinite soil. Anionic and nonionic surfactants were considered in the EK process, i.e. sodium cholate, SDS and TW80 agents. The lowest removal rate was obtained with TW80 EK at 10 mA, and 20 mA treatment settings for enhanced EK treatments, followed by SDS enhanced EK experiments. At a constant current of 20 mA, the NaC enhanced EK experiment, experiment E6, demonstrated 35% removal efficiency in one week, and the highest removal efficiency among all one-week experiments. The removal efficiency of PFOA was then measured for two weeks to see whether there was any improvement in the EK system over time. The PFOA removal efficiency rose to 75.68% and 45% in the EK tests with sodium cholate SDS enhancement agents. PFOA removal rose nearly twice as much in the NaC and SDS enhanced EK testing conducted in one-week experiments. Increasing the experimental duration from one to two weeks resulted in a substantial increase in the removal efficiency. The study concluded that surfactant-enhanced EK experiments have great potential in improving PFOA removal from contaminated soil. Nevertheless, the improvement of PFOA removal was significantly dependent on the surfactant type, with NaC biosurfactant exhibiting the highest removal efficiency. Future work should include (i) the removal of different PFAS compounds from kaolinite by surfactant enhanced-EK process, (ii) PFAS removal by surfactant-enhanced EK from real soil, and (iii) pilot study on the surfactant-enhanced EK process for PFAS treatment and surfactant biodegradability in soils.

CRediT authorship contribution statement

Namuun Ganbat: Conceptualization, Methodology, Formal analysis, Investigation, Data curation, Writing – original draft, Writing – review & editing. **Ali Altaee:** Conceptualization, Methodology, Formal analysis, Investigation, Writing – original draft, Writing – review & editing, Supervision. **John L. Zhou:** Formal analysis, Investigation. **Thomas Lockwood:** Resources, Investigation. **Raed A. Al-Juboori:** Formal analysis, Writing – original draft, Writing – review & editing. **Faris M. Hamdi:** Writing – review & editing. **Elika Karbassiyazdi:** Formal analysis, Investigation. **Akshaya K. Samal:** Formal analysis, Investigation. **Alaa Hawari:** Resources, Formal analysis. **Hadi Khabbaz:** Formal analysis, Investigation.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

All data obtained have been shared in the form of figures and tables.

Acknowledgment

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