

Effect of processing fluid and initial concentration on electrokinetic removal of environmental hormone—nonylphenol (NP) from soil matrix

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Abstract This study was aimed to investigate the electrokinetic removal of environmental hormone—nonylphenol (NP)—from soil matrix under potential gradient of 1 V cm^{-1} for 5-day treatment. The EK experiments were conducted with four processing fluids of de-ionized water, citric acid, NaOH, and methanol in a Pyrex glass cylindrical cell. Results showed that the electrokinetic removal efficiency of the above-mentioned processing fluid was 29, 36–38, 43–53, and 53–69%, respectively. It was found that the removal of NP in EK system was highly related to the solubility of NP in processing fluid. Approximate 88.8–94.2% of NP removal was collected in the cathode reservoir after EK treatment, which was 7.9–16.2 times greater than that collected in the anode reservoir. It was concluded that NP was mainly removed from anode to cathode by electroosmosis flow. The electrokinetic phenomenon of current density, electroosmotic permeability, and power consumption were also investigated.

Keywords Electrokinetic process · Environmental hormone · Nonylphenol · Processing fluid · Soil remediation

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1 Introduction

Currently, 80% of the most of the non-ionized detergent contains 4-nonylphenol polyethoxylates (NPnEO). They are common nonionic surfactants applied in the industrial production of cleaning products, textiles, petroleum, pulp and paper, and pesticides formulation [1]. When NPnEO mode surfactants were dumped into effluent waters, it would yield less biodegradable products such as NP1EO, NP2EO, or nonylphenol (NP) [2]. Since the structure of NP is very similar to animal estrogen (Fig. 1), NP possesses the ability to mimic natural hormones by interaction with estrogen receptors. It should be noted that these water pollutants exhibit estrogenic activity at a tissue concentration of $1 \mu\text{M}$ [3]. The use of NPnEO surfactants has been banned in domestic formulations in the United Kingdom, Germany, Switzerland and USA and they have also been listed as toxic chemical materials in Taiwan since 2007 [4]. Their use is restricted to industrial application in which the specific nature of the properties required makes it more difficult to substitute them. Such household and industry waste water dumped into rivers will create vast and serious impact on the environment.

The distribution of NP/NPnEO in environmental matrix was shown in Fig. 2. As NPnEO surfactant released to environment, it will degrade through natural biodegradation to daughter compounds with shorter EO chain, such as NP1EO, NP2EO, or NP. These daughter compounds have relative lower water solubility than NPnEO and will adsorb to suspended solids and sediments. Among which, NP is ~ 10 times more toxic than its ethoxylate precursor [5]. NP in wastewater extracted from digested sewage can pass into rivers. Consequently, it will raise a significant concern about the long-term impact on metabolism of all the biota within the food-chain cycle.

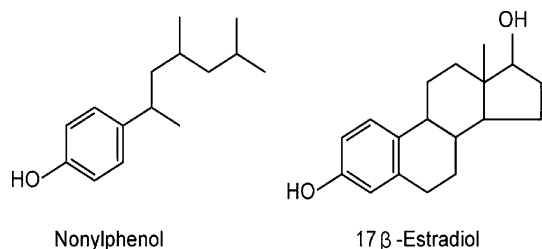


Fig. 1 Structure of nonylphenol and estrogen

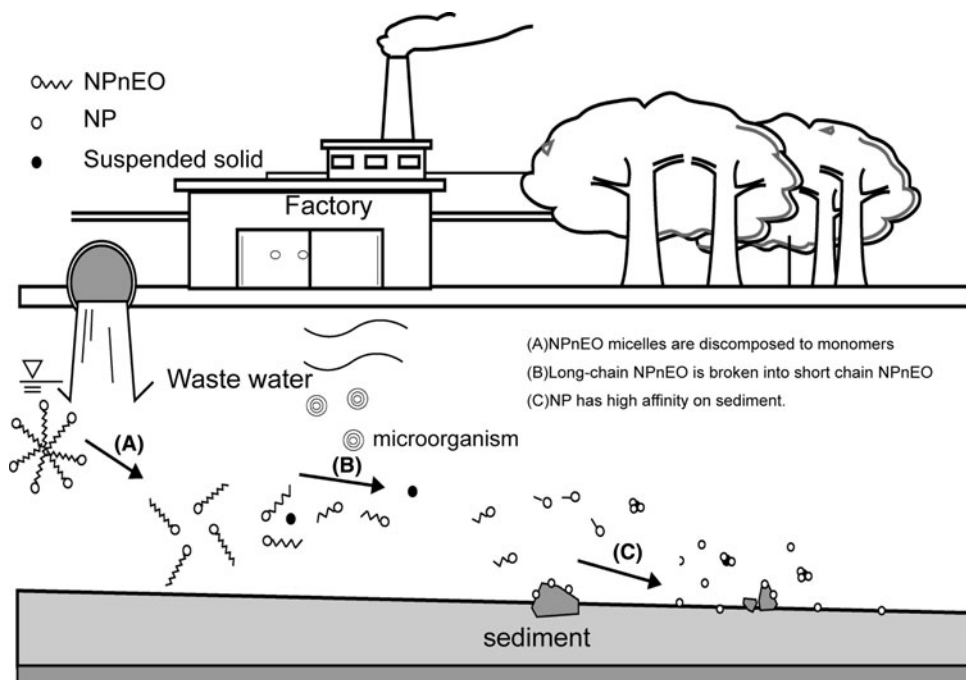
Trials on treatment of NP by adsorption and photodegradation in aqueous have been reported. Experiments to investigate the adsorption of NPs in aqueous phase onto four activated carbons have indicated that coconut shell and activated carbon had high adsorption power because they had the smallest mean pore diameter [6, 7]. It was reported that NP could be completely degraded by TiO_2 photodegradation within 80 h in aqueous [8]. Less research was focused on remediation of NP-contaminated sediment and soils. Chang et al. [9] reported that aerobic degradation rate constant and half-lives ($t_{1/2}$) for NP (50 mg kg^{-1}) in sand clay loam and silty clay soil was $0.098\text{--}0.126 \text{ day}^{-1}$ and 5.5–8.4 day. In river sediment, aerobic degradation performance of NP was more worse, which rate constant and half-lives ($t_{1/2}$) for 2 mg kg^{-1} of NP contaminated sediment was $0.007\text{--}0.058 \text{ day}^{-1}$ and 13.6–99.0 day [10]. The bioremediation of NP in environmental matrix is highly related to soil/sediment characteristics, microorganisms, pH, temperature, and ionic species. It was noted that bioremediation would be a time consuming technology. A more efficient

remediation technology for NP removal from soil matrix is highly desired.

Electrokinetic (EK) remediation process is a promising innovative technology for decontamination in low hydraulic permeability media. Basically, EK technique is upon the action of an electric field generated between inserted electrodes in the medium by applying a direct current or a constant voltage. The applied potential causes the migration of electrolyte solution and soluble contaminants toward the electrodes via electromigration, electroosmosis, and electrophoresis [11, 12]. Various enhancement techniques, i.e. careful management of the pH within the electrode [13, 14], chelating agent [15, 16], and enhancing reagent [17, 18] in the cathode reservoir, have been proposed to improve effectiveness in EK system. It was concluded that the key feature for improving EK performance of removing contaminants was highly dependent on the interaction between contaminants and processing fluid. Additional parameters affected EK performance were included potential gradient and processing time, which was dependant on the characteristics of soils and contaminants. Many researches had shown excellent electrokinetic removal efficiencies for metals from low permeable subsurface environments [19, 20].

NP, a kind of environmental hormone, is a chemical compound that is widely proliferated in the streams and creeks of country. It has been confirmed that once NP runs into human body it would decrease male sperm count and interfered normal metabolism [21–23]. It would also reduce immunity, interfere physiology, and cause breast and

Fig. 2 Distribution of NP/ NPnEO in environmental matrix



testicular cancer when exposures at NP-contaminated environment in a long time [24]. Nonylphenol would also reduce and inhibit estrogen efficacy in animals. It were reported that NP would stimulate synthesis of vitellogenin and inhibit growth of testicle of male rainbow trout [22, 25, 26]. Sublethal toxic effect of NP on zooplankton in natural water interferes with their sex determination and development [27].

This study was aimed to investigate the electrokinetic removal of environmental hormone—nonylphenol (NP)—from soil matrix under potential gradient of 1 V cm^{-1} for 5-day treatment. Four type of processing fluid, de-ionized water, citric acid, sodium chloride, and methanol, were applied in EK process, respectively. The effectiveness of NP removal and electrokinetic behavior in EK process were investigated.

2 Experimental materials and methods

2.1 Characteristics of Nonylphenol (NP)

The target compound, NP, was purchased from Aldrich and used for GR grade for all the experiments. The characteristics of NP were summarized in Table 1. It is lighter than water with specific gravity of 0.95. It is difficult to diffuse to air with vapor pressure of 4.55 mPa and low Henry's law constant of $11.2 \text{ Pa m}^3 \text{ mol}^{-1}$. NP could dissolve in most organic solvents but less soluble in aqueous solution with aqueous solubility of $5.4\text{--}8.0 \text{ mg L}^{-1}$. NP has high affinity in soil phase with high $\text{Log } K_{ow}$ value of 4.20–4.48 so it resulted in low mobility in subsurface environment.

2.2 Soil and processing fluid

Soil samples were collected from a depth of $\sim 0.7\text{--}1.5 \text{ m}$ below the surface within a farmland located in southern Taiwan, which characteristics as listed in Table 2. The texture of soil sample was classified as clay by particle size analysis. A soil organic matter content of 3.73% was

Table 1 Characteristics of NP [8]

Property	Specification
Synonyms	4-nonylphenol, <i>p</i> -nonylphenol
Molecular formula	$\text{C}_{15}\text{H}_{24}\text{O}$
Melting point ($^{\circ}\text{C}$)	-8 [28]
Boiling point ($^{\circ}\text{C}$)	$295\text{--}320$ [28]
Specific gravity	0.95
$\text{p}K_a$	10.7 [28]
Vapour pressure (mPa)	4.55 [28]
Henry's law constant ($\text{Pa m}^3 \text{ mol}^{-1}$)	11.2 [28]
$\text{Log } K_{ow}$	$4.2\text{--}4.48$ [29, 30]
Solubility (mg L^{-1})	$5.4\text{--}8$ [31]

Table 2 Characteristics of investigated soil

Items	Values	Items	Values
Clay (%)	70	pH	8.5
Sand (%)	20	pH_{ZPC}	2.4
Silt (%)	10	BET area ($\text{m}^2 \text{ g}^{-1}$)	16.4
Organic matter content (%)	3.73	CEC (meq g^{-1})	6.6×10^{-3}
Solid density (g cm^{-3})	2.64	Hydraulic conductivity (cm s^{-1})	$<10^{-8}$

observed by the combustion method [32]. Soil pH of 8.5 was measured in water suspensions at 1:1 ratio of soil to solution by volume and soil pH_{ZPC} of 2.4 was determined by a zeta potential instrument (Pen Kem-Laser Zee 3.0, USA). A soil solid density of 2.64 g cm^{-3} was determined according to the method of ASTM D854-92. The soil specific surface areas of $16.4 \text{ m}^2 \text{ g}^{-1}$ was measured by a BET surface areas analyzer (ASAP 2010, Micromeritics, USA). The hydraulic conductivity of the soil sample was less than $10^{-8} \text{ cm s}^{-1}$ measured by a falling-head parameter.

After removing debris and air-drying, the soil sample was sieved to less than 2 mm in diameter and then treated by pressure sterilization of 118 kPa for 5 min. The NP-contaminated soil was prepared by adding specific amount of 0.09 mL NP, 100 mL of methanol, and 900 g of sieved soil into a 2 L glass beaker. The mixture was then stirred to let methanol vaporize completely and achieve uniform NP spiked soil. To measure the NP concentration the soil sample was then extracted 5 g of spiked soil with 15 mL of methanol at a speed of 150 rpm for 20 min and the concentration of NP in extract was determined by means of HPLC–UV (Hitach, L-2000 series, Japan) coupled with column of Hypersil APS-2NH₂ ($250 \times 4.6 \text{ mm}$, $5 \mu\text{m}$) and detected at a wavelength of 277 nm. Triplicate soil samples were analyzed for quantification. This NP spiked soil was then ready for following EK experiments.

The solubility of NP was considered as key feature to select processing fluid. NP was more soluble in high pH solution and in polar solvent. Hence, sodium hydroxide (0.11–0.33 M) and methanol (10–30%) were selected as tested processing fluids. Another two processing fluids of citric acid (0.038–0.060 M) and De-ionized water were tested for compare.

2.3 Set up of EK experiments

Twelve EK experiments were conducted in an acrylic cell of $4.2 \text{ cm} (\varphi) \times 22 \text{ cm} (\text{L})$, consisting of three compartments: cathode reservoir with 5 cm in length, anode reservoir with 5 cm in length, and soil specimen chamber with 12 cm in length, which was shown in the Fig. 3. In this lab-scale EK system, the heterogeneous characteristic of soil

was assumed to be less and neglected. The processing fluid was initially added into both anode and cathode reservoirs and replenished in the anode reservoir every half day. All experiments were conducted under potential gradient of 1.0 V cm^{-1} for 5 day. The electric current, reservoir pH, concentration of NP in reservoir, and electroosmotic flow were monitored during the test periods. The current density was calculated as dividing electric current by crossing area of EK cell.

The pH of soil specimen was determined after treatment for each test according to ROC-EPA method. Among them, two were sampled at the end of anode and cathode side and the other six were sampled every 2 cm of soil specimen. Add 5 g of sectioned soil specimen into 15 mL of methanol solution and rotate for 20 min at a speed of 2000 rpm. The concentration of NP in methanol was analyzed by means of HPLC–UV (Hitach, L-2000 series, Japan) coupled with column of Hypersil APS-2NH₂ (250 × 4.6 mm, 5 μm) and detected at a wavelength of 277 nm.

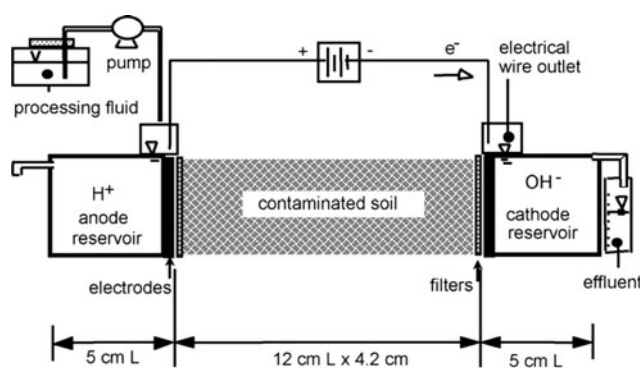


Fig. 3 Schematic setup of electrokinetic apparatus

Table 3 Summary results in EK system

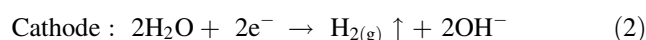
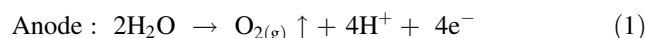
Test no.	NP (mg/kg)	Processing fluid	Processing gradient (V cm^{-1})	Processing time (day)	Q_e ($\text{cm}^3 \text{ day}^{-1}$)	k_e ($\text{cm}^2 \text{ V}^{-1} \text{ s}^{-1}$)	Power consumption (kWh m^{-3})	Removal efficiency (%)
1	50	DI water	1.0	5	8.2	6.9×10^{-6}	380	29
2	50	0.036 M citric acid	1.0	5	8.0	6.7×10^{-6}	378	36
3	50	0.060 M citric acid	1.0	5	7.9	6.7×10^{-6}	374	38
4	50	0.066 M NaOH	1.0	5	7.7	6.4×10^{-6}	387	43
5	50	0.110 M NaOH	1.0	5	7.4	6.2×10^{-6}	385	47
6	50	0.220 M NaOH	1.0	5	7.8	6.5×10^{-6}	407	52
7	50	0.330 M NaOH	1.0	5	7.8	6.6×10^{-6}	411	53
8	50	10% CH ₃ OH	1.0	5	7.5	6.7×10^{-6}	352	53
9	50	20% CH ₃ OH	1.0	5	7.0	5.9×10^{-6}	344	65
10	50	30% CH ₃ OH	1.0	5	6.7	5.6×10^{-6}	325	69
11	25	0.110 M NaOH	1.0	5	8.0	6.7×10^{-6}	381	62
12	100	0.110 M NaOH	1.0	5	8.5	7.1×10^{-6}	388	22

3 Results and discussion

Experimental results are summarized in Table 3 and further discussed in the following sections.

3.1 Electrokinetic behavior in EK systems

The generation of H⁺ and OH⁻ under an applied electric field (Eqs. 1, 2) will result in movement of acid and basic fronts in EK system and will change the soil pH drastically during EK process [33, 34]. Moreover, consumption of hydroxyl ions in precipitation, surface complexation and adsorption on the soil, and formation of complexes in the bulk solution are additional factors affected the soil pH in EK system [35].



The soil pH profiles along EK cell in investigated systems are shown in Fig. 4. A general trend of low pH near the anode and high pH near the cathode was found in all EK systems (Fig. 4a–d). As using citric acid (CA) as processing fluid (Fig. 4a), the acid front generated at anode reservoir flushed across the soil specimen, consequently lowered the soil pH from 8.5 to 2.0–2.8 near the anode side for all cases. At the cathode side, the migration of OH⁻ advancing toward the anode and the concentration differences of OH⁻ between reservoirs and soils would increase soil pH to 11.0–12.1. Similar trend of soil pH was also found in EK/NaOH systems (Fig. 4b) and EK/Methanol systems (Fig. 4c). Owing to NaOH is a basic solution, the soil pHs of both anode and cathode side were 3.2–3.3 and 12.2–12.9, respectively, which were higher than those in EK/CA systems. With methanol solution as

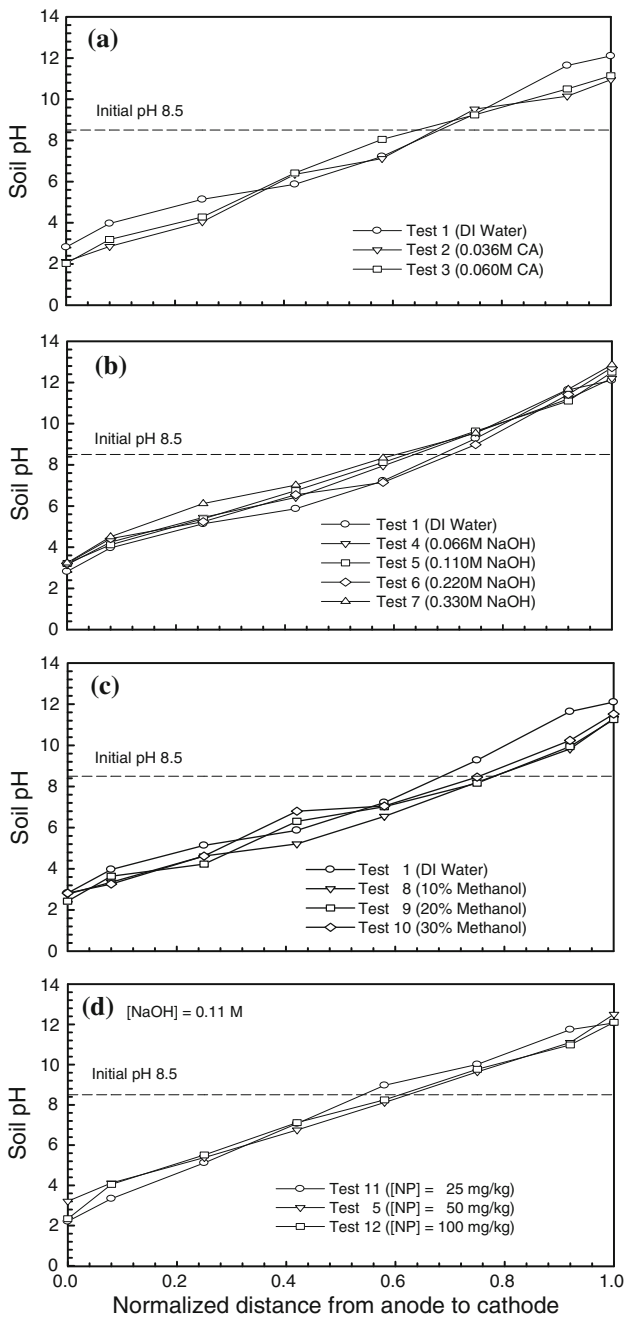


Fig. 4 Soil pH profiles in EK systems for the case of various processing fluid (a–c); and various concentration of NP (d) along EK cell

processing fluid (Fig. 4c), the soil pH of anode and cathode side were similar as those in EK/Di-ion water systems. Results also showed that no evident correlations between soil pH and concentration of NP in EK/NaOH systems (Fig. 5d).

In EK process, the movement of electrolyte solution driven by an electrical field, which carries the soluble pollutants toward the electrodes, is considered to be one of the mechanisms leading to the pollutants removal from

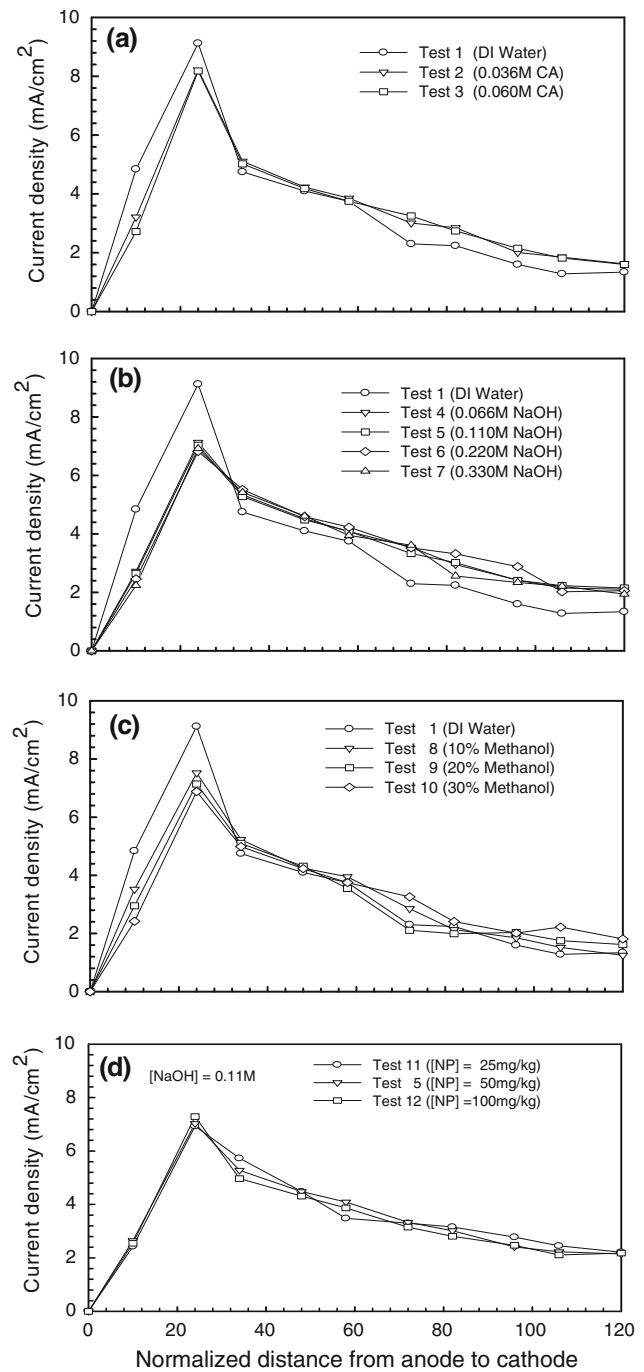


Fig. 5 The effect of processing fluid (a–c) and concentration of NP (d) on current density in EK systems

soils. The movement of electrolyte solution, i.e., electroosmotic flow, Q_e (mL day⁻¹), for a cylindrical soil core is direct proportional to the applied electric potential gradient, i_e (V cm⁻¹):

$$Q_e = k_e \times i_e \times A \tag{3}$$

where A (cm²) is cross-section area of soil core and k_e (cm² V⁻¹ s⁻¹) is electroosmosis permeability. As shown in

Table 3, the electroosmosis flow rate was in the range of $6.7\text{--}8.5\text{ cm}^3\text{ day}^{-1}$ and it was increased with value of k_e for all experiments. The values of k_e in EK/NaOH systems ($6.2 \times 10^{-6}\text{--}6.6 \times 10^{-6}\text{ cm}^2\text{ V}^{-1}\text{ s}^{-1}$, Tests 4–7 in Table 3) were lower than those in EK/De-ionized water systems ($6.7 \times 10^{-6}\text{--}6.9 \times 10^{-6}\text{ cm}^2\text{ V}^{-1}\text{ s}^{-1}$, Tests 1–3 in Table 3). This might largely because of clogging of precipitates in soil pores under basic environment. The lowest values of k_e were found in EK/Methanol systems ($5.6 \times 10^{-6}\text{--}6.7 \times 10^{-6}\text{ cm}^2\text{ V}^{-1}\text{ s}^{-1}$, Tests 8–10 in Table 3), which was attributed to the low conductivity of methanol solution.

The variations of current density as function of time in EK systems are shown in Fig. 5. Current density was gradually increased to 9.1 mA cm^{-2} within 24 h for Test 1. Similar phenomenon was shown for all other EK systems, however, lower current density was found.

The current density of investigated systems was all decreased to less than 2.5 mA cm^{-2} within 96 h. It was largely because the clogging of precipitates in the soil pore resulted in a low current density. This might be resulted from: (a) metal hydroxide precipitates formed at basic environment transported to cathode by electromigration; and (b) precipitates caused by processing fluid reacted with ions in soil matrix. As such, it was also explained why the peak current density of EK/CA system in Fig. 5a ($8.2\text{--}8.3\text{ mA cm}^{-2}$) was greater than that in EK/NaOH systems ($6.8\text{--}7.1\text{ mA cm}^{-2}$ in Fig. 5b) and EK/Methanol systems ($6.9\text{--}7.5\text{ mA cm}^{-2}$ in Fig. 5c). It was shown that the variance of current density was more significantly related to type of processing fluid (Fig. 5a–c) rather than concentration of NP (Fig. 5d). Results of current density were quite higher than those in soil matrix observed by Zhou et al. [14], Yuan et al. [18], and Yuan et al. [36].

3.2 Removal of NP

As shown on Table 3, results indicated that electrokinetic remediation efficiency of NP was highly related with processing fluid. At the case of using citric acid as processing fluid (Tests 2–3), only 36–38% of NP was removed, however, which was higher than that in EK/DI water system by a factor of 1.2–1.3. The removal efficiency of NP was increased to 43–53% in EK/NaOH systems (Tests 4–7) and it increased with concentration of NaOH increasing. This might largely attribute to solubility enhancement of NP in basic solution [37]. Results were also showed that the removal efficiency of NP in EK/methanol system (Tests 8–10) was up to 53–69%, which was 1.8–2.4 times greater than that in EK/DI water system. It might be resulted from solubility enhancement of NP in methanol solution. As shown in Table 4, the solubility of NP was 15.2 mg L^{-1} , 28.4 mg L^{-1} , and 52.6 mg L^{-1} in 10, 20, and 30% methanol solution, respectively. A better removal performance of NP was found

Table 4 Solubility of NP in solutions

Solutions	Solubility of NP (mg L^{-1})
Citric acid	
0.036 M	9.7
0.060 M	15.1
NaOH	
0.066 M	46.9
0.33 M	48.3
Methanol	
10%	15.2
20%	28.4
30%	52.6

in EK/Methanol system, hence, it was concluded that solubility of NP in processing fluid would be a critical parameter for removal performance in EK process. Higher concentration of NP in EK system would diminish removal performance due to mass transfer inhibition of NP in processing fluid. It was shown that as concentration of NP increased to 100 mg kg^{-1} (Tests 11–12), NP removal in EK/NaOH system was decreased to 22%. As a consequence, to achieve a successful EK process, a good selection of processing fluid (i.e. type and concentration) should be highly considered.

Energy expenditure in EK systems is calculated as follows:

$$E_u = \frac{P}{V_s} = \frac{1}{V_s} \int VI dt \quad (4)$$

where E_u = energy expenditure per unit volume of soil (kWh m^{-3}); P = energy expenditure (kWh); V_s = volume of soil (m^3); V = voltage (V); I = current (A); t = time (h). In the tests of constant-voltage condition, the energy expenditure is directly related to the time integral of the current across the cell. The calculated energy consumption was 380, 374–378, 387–411, 325–352 kWh m^{-3} in EK/DI water, EK/CA, EK/NaOH, and EK/Methanol systems, respectively. It was apparent that processing fluid was the controlling factors with respect to energy consumption in EK process [36]. Considering NP removal performance and power consumption in this study, the best result was found in EK/Methanol system. It was clear that energy applied was not enough for completion of remediation. However, this can be improved by either selecting other processing fluid to enhance NP solubility or prolonging the treatment periods [38].

3.3 Removal mechanisms of NP

Four mechanisms of electrolysis, electroosmosis flow, electromigration and electrophoresis were dominant for

Table 5 Fractional distribution of NP in EK systems

Test No.	(1) [NP] (mg/kg)	(2) Soil (g)	(3) NP (mg)	Fraction of NP			(7) Mass balance (%)
				(4) Soil phase (mg)	Reservoir		
					(5) Anode (mg)	(6) Cathode (mg)	
1	50	224	11.2	7.92	0.24 (11.2%) ^a	1.82 (88.8%) ^b	89.0
2	50	223	11.2	7.22	0.23 (8.1%) ^a	2.60 (91.9%) ^b	90.1
3	50	224	11.2	6.90	0.19 (5.8%) ^a	3.10 (94.2%) ^b	90.9
4	50	226	11.3	6.44	0.22 (6.2%) ^a	3.35 (93.8%) ^b	88.4
5	50	223	11.2	5.95	0.24 (6.9%) ^a	3.23 (93.1%) ^b	84.3
6	50	225	11.2	5.36	0.31 (8.0%) ^a	3.58 (92.0%) ^b	82.3
7	50	222	11.1	5.21	0.26 (6.5%) ^a	3.77 (93.5%) ^b	83.1
8	50	226	11.3	5.31	0.43 (9.4%) ^a	4.13 (90.6%) ^b	87.4
9	50	223	11.2	3.87	0.41 (7.0%) ^a	5.46 (93.0%) ^b	87.2
10	50	226	11.3	3.51	0.52 (7.7%) ^a	6.24 (92.3%) ^b	91.0
11	25	225	5.6	2.15	0.21 (7.4%) ^a	2.64 (92.6%) ^b	88.9
12	100	223	22.3	17.37	0.22 (8.1%) ^a	2.48 (91.9%) ^b	90.0

$$(3) = (1) \times (2)$$

$$(7) = [(4) + (5) + (6)] / (3) \times 100\%$$

$$^a (5) \div [(5) + (6)] \times 100\%$$

$$^b (6) \div [(5) + (6)] \times 100\%$$

contaminant removal in EK process [18]. Owing to NP were not huge agglomerates, electrophoresis would not play a key role in removal performance. The removed NP would either migrate toward anode reservoir by electromigration or toward cathode reservoir by electroosmosis flow. The fractional distribution of NP and mass balance in investigated EK systems were shown in Table 5. By summarizing mass of NP collected in soil and reservoir, the mass balance of NP in investigated systems were in the range of 82.3–91.0%. In EK/DI water system (Test 1 in Table 5), 88.8 and 11.1% of NP removal was collected in cathode and anode reservoir, respectively. For other EK systems, above 90% of NP removal was collected in cathode reservoir and much less was found in anode reservoir. This might largely because NP was remained as nonionic form when pH value was less than 10.7 (as shown pK_a value in Table 1). As a consequence, NP was migrated with EO flow from anode toward cathode. With increasing solubility of NP in processing fluid, more NP would be dissolved into reservoirs. As such, it was found that the largest collected NP in anode and cathode reservoir was found in EK/methanol systems, which was 0.41–0.52 mg and 4.13–6.24 mg, respectively (Tests 8–10 in Table 5).

It was concluded that both electroosmosis flow and electromigration were contributed to the NP removal in EK systems. Among which, electroosmosis flow were dominant for NP removal rather than electromigration.

4 Conclusions

This study shows that the electrokinetic remediation of NP spiked soil was highly related with processing fluid and NP concentration. The important conclusions in this study have been summarized as follows:

- (1) NP removal in EK systems was highly related to processing fluid. The removal efficiency of NP in EK/CA system, EK/NaOH system, and EK/Methanol system with potential gradient of 1.0 V cm^{-1} for 5 days was 36–38, 43–53, and 53–69%, respectively. Highest solubility of NP in methanol solution was resulted in highest removal efficiency. The solubility of contaminant in processing fluid might become an important selection factor.
- (2) Above 90% of NP removal was collected in cathode reservoir and much less was found in anode reservoir in investigated EK systems. Based on removal mechanism analysis, the electroosmosis mechanism was dominant in NP removal rather than electromigration mechanism. This might largely because NP was remained as nonionic form when pH value was less than 10.7.
- (3) To study performance more thoroughly, fining processing fluid with higher solubility of NP and prolonging the EK/CNT experiments are recommended in the future investigations. For full-scale

EK system, accurate dosage of processing fluid was important to achieve better remediation efficiency.

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