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Electrokinetic-enhanced Phytoremediation of Uranium-contaminated Soil using Sunflower and Indian Mustard

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Abstract

Electrokinetic-enhanced phytoremediation is an effective technology to decontaminate heavy metal contaminated soil. In this study, we examined the effects of electrokinetic treatments on plant uptake and bioaccumulation of U from soils with various U sources. Redistribution of uranium in soils as affected by planting and electrokinetic treatments was investigated. The soil was spiked with 100mg kg^{-1} UO_2 , UO_3 , and $\text{UO}_2(\text{NO}_3)_2$. After sunflower and Indian mustard grew for 60 days, 1 voltage of direct-current was applied across the soils for 9 days. The results indicated that U uptake in both plants were significantly enhanced by electrokinetic treatments from soil with UO_3 and $\text{UO}_2(\text{NO}_3)_2$. U was more accumulated in roots than in shoots. Electrokinetic treatments were effective on lowering soil pH near the anode region. Overall, uranium (U) removal efficiency reached 3.4–4.3% from soils with UO_3 and uranyl with both plants while that from soil with UO_2 was 0.7–0.8%. Electrokinetic remediation treatment significantly enhanced the U removal efficiency (5–6%) from soils with UO_3 and uranyl but it was 0.8–1.3% from soil with UO_2 , indicating significant effects of U species and electrokinetic enhancement on U bioaccumulation. This study implies the potential feasibility of electrokinetic-enhanced phytoremediation of U soils with sunflower and Indian mustard.

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Preface

This study was conducted for the U.S. Army Corps of Engineers under Project 458170, titled, "Depleted Uranium (DU) Clearance from DoD Ranges." The Grant Officer's Technical Representative was Mr. John H. Ballard, Office of the Technical Director for Installations and Operational Environments, ERDC-EL-EZT and the Technical Point of Contact was Dr. Steven L. Larson, Environmental Engineering Branch, ERDC-EL-EPE.

The work was performed by the Environmental Engineering Branch of the Environmental Processes Division, U.S. Army Engineer Research and Development Center, Environmental Laboratory (ERDC-EL). At the time of publication of this Miscellaneous Paper, Ms. Brooke Petery was Acting Branch Chief; Dr. Brandon Lafferty was Acting Division Chief; and Dr. Elizabeth Ferguson was the Technical Director for Installations and Operational Environments. The Acting Deputy Director of ERDC-EL was Dr. Justin Berman and the Acting Director was Dr. Jack Davis.

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The Commander of ERDC was COL Teresa A. Schlosser and the Director was Dr. David W. Pittman.

Electrokinetic-enhanced phytoremediation of uranium-contaminated soil using sunflower and Indian mustard

Introduction

Depleted uranium (DU) is the by-product of the extraction of ^{235}U in natural uranium. In recent years, DU weapons such as DU containing penetrators have been used in war frequently, leading to DU contaminated soil and water in combat areas (Bleise *et al.* 2003; Mirjana *et al.* 2008). Meanwhile, DU mine tailings and ore wastes around abandoned mines are continuous sources of DU in pollution of soils and groundwater.

Contamination of soil and water with DU has increased public health concerns due to its chemical toxicity. Long-term exposure to uranium (U) causes potential dangers to human health with the greatest health risk of kidney failure (Bleise *et al.* 2003). Moreover, its radioactivity poses increased risks of lung and bone cancers (Malaviya and Singh 2012). The remediation technology for the uranium polluted soils includes physical, chemical, and biological remediation (Meng *et al.* 2017, 2018). Bioremediation methods consist of phytoremediation and microbial remediation which are appropriate for large areas of soil and water cleaning up. However, phytoremediation has several limitations at actual contaminated sites. The use of plants for cleaning the environment often takes longer time than other remediation techniques and is best suited for places where the elements are present within the range of plant roots (Pilon-Smits and Freeman 2006; Doran 2009).

Sunflower is recognized as a potential phytoremediation plant due to its huge biomass and high tolerance to environmental stresses. Previous studies showed that sunflower was better than other plants in accumulating uranium from contaminated soil (Shahandeh and Hossner 2002; Laurette *et al.* 2012). Indian mustard also was used for phytoremediation of U, Hg, and other heavy metal contaminated soils (Duquène *et al.* 2009; Han *et al.* 2012; Jagetiya and Sharma 2013; Meng *et al.* 2018).

Increasing the efficiency of phytoremediation is important for field application. Electrokinetic remediation (EKR) is an environmental restoration technique especially designed for the *in situ* treatment of contaminated soils (Reddy and Cameselle 2009; Mao *et al.* 2016a,b,c, 2017, 2018). Using electrokinetic equipment after acidic washing for uranium contaminated soil requires a long decontamination time and a significant amount of electric power. To overcome these limitations, coupling phytoremediation with EKR technology,

which involves the application of a low DC electric potential in the soil, has been proposed. Studies showed that electrokinetic treatment enhanced phytoremediation efficiency of metal contaminated soils (Cameselle *et al.* 2013; Mao *et al.* 2016a). When a low intensity electric field is applied, H^+ is generated around the anode electrode through water electrolysis. As a result, more metal(loid)s are mobilized under the acid condition around the anode electrode (Thangavel and Subbhuraam 2004; Mao *et al.* 2016a,c, 2018).

The accumulation and distribution of uranium in plants was greatly influenced by U speciation in soil (Duquène *et al.* 2009; Véra-Tome *et al.* 2009; Laurette *et al.* 2012; Jagetiya and Sharma 2013; Meng *et al.* 2018). Soils from two US Army sites were characterized including a sand soil ($3210 \text{ mg U kg}^{-1}$) and a sandy silt soil ($1560 \text{ mg DU kg}^{-1}$) (Christine *et al.* 2006). About 83% of DU in the fine fraction was associated with carbonate, iron, and manganese oxide and organic matter fractions of the material. Sandy silt soil fine fraction contained 64% of DU as soluble U (VI) minerals or insoluble U (IV) (Christine *et al.* 2006). In less contaminated area, sorption and co-precipitation of DU are in aged, iron-rich soils, where uranium binding to carbonates and iron/manganese oxides became significant (Mirjana *et al.* 2008). In order to better assess the bioavailability of metals, chemical association of metals with soil components needs to be determined. However, the effects of U forms in soils on plant uptake and bioaccumulation under electrokinetic conditions have not been understood.

Nevertheless, there is little understanding on coupling efficiency of electrokinetic and phytoremediation of uranium soils with various U forms (Dushenkov *et al.* 1997; Ebbs *et al.* 1998). At the same time, U in the earth crust has three major isotopes U238, U235, and trace amount of U 234. U238 (DU) has major alpha decay mode with a long-term $t_{1/2}$ (4.468×10^9 years). It is a weak radionuclide. Therefore, for seasonal experiments on plant uptake and bioaccumulation, DU radioactivity toxicity should be much smaller than its chemical toxicology.

The objectives of this study were: (1) to investigate the coupled electrokinetic phytoremediation to remove DU from polluted soils with sunflower and Indian mustard; (2) to study bioavailability of U in Mississippi River Delta paddy soil with various U sources under electrokinetic field (EKF) after a growing season, (3) to determine DU speciation and its contribution to DU bioavailability in paddy soils under EKF treatment, and (4) to examine DU uptake and distribution in sunflower and Indian mustard in contaminated soils.

Materials and methods

Soil and experimental design

The fresh surface paddy soil (0–30 cm) was collected from Mississippi River Delta. The soil was air-dried at room temperature and then passed through 2-mm mesh and mixed well before potting test.

Plant culture and DU exposure

Analytically pure grade uranium nitrate $UO_2(NO_3)_2$, uranium trioxide UO_3 , uranium dioxide UO_2 were added at 100 mg kg^{-1} level to the soil. About 500 g of air-dried soil was weighed and placed into each plastic pot with 5.5 cm diameter and covered with aluminum foil. In order to save the disposal cost, each treatment was replicated. Every replicate pot took three plant samples for analysis. The sunflower and Indian mustard seeds were purchased from Ferry-Morse (USA).

EKR setup and determination of metal solubility and bioaccumulation

Figure 1 shows the experimental setup. A DC power supplier (0–60 V, 0–3 A) was used as electrical power source. Graphite electrode rod (0.95 cm diameter, 30.5 cm length) was used as both anode and cathode due to its low cost and inertness. A DC electrical field with constant intensity of 1 V cm^{-1} was applied to soil in plastic pots for a period of 9 days. A pair of graphite electrode rods was vertically inserted into both sides of each pot with 13 cm below the soil surface (Figure 1). The copper wire was used to connect electrode rods with the power source. The electrical equipment was set up after 5 weeks of growing sunflower and Indian mustard.

U fractionation in soils

Soil U was fractionated into the following solid phase components according to Tessier *et al.* (1979) and Han *et al.* (2007).

S1: Soluble plus exchangeable fraction

The exchangeable (EXC) fraction includes soluble plus EXC fraction on the surface of soil particles. Twenty-five mL of

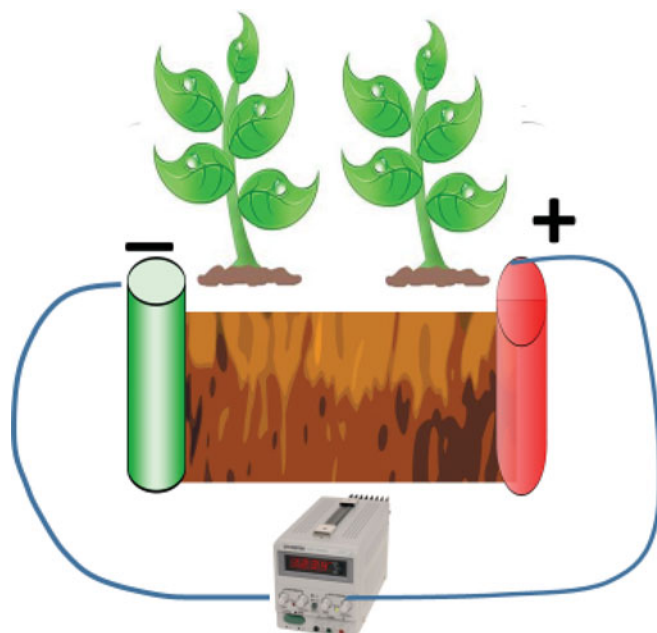


Figure 1. The diagram showing the experiment setup of electrokinetic enhanced phytoremediation of U soil.

1 M NH_4NO_3 with pH adjusted to 7.0 with $\text{NH}_3\bullet\text{H}_2\text{O}$ were added into 50 mL polytetrafluoroethylene centrifuge tube containing 1 g air-dried soil. The mixture was shaken for 30 min in a temperature controlled shaker at 25 °C. The suspension was centrifuged at 3000g for 15 min. The supernatant was poured out and filtered through 0.45- μm filter for U analysis with inductively coupled plasma mass spectrometry (ICP-MS). The soil residue was retained for the next step. The following extraction steps used the same centrifugation-decantation-filtration processes.

S2: the fraction bound to carbonate

This U fraction bound to carbonate (CARB). Twenty-five mL of 1 M CH_3COOH with pH adjusted to 5.0 with CH_3COOH were added into the soil residue from step one. The tubes were shaken for 6 h at 25 °C.

S3: the fraction bound to easily reducible oxides

This U fraction mostly bound to easily reducible oxides (ERO), such as Mn-oxides (Han *et al.* 2007). Briefly, 25 mL of 0.1 M $\text{NH}_2\text{OH}\cdot\text{HCl}$ + 0.01 M HCl solution (pH 2.0) was added into the residue soil from the last step. The mixture was shaken for 30 min at 25 °C.

S4: the fraction bound to organic matter fraction

This fraction mainly bound to soil organic matter (OM) and microorganisms, such as HA. Three mL H_2O_2 + 5 mL 0.01 HNO_3 were added into the residue soil from the last step. The mixture was put into a water bath at 80 °C. After 2 h, additional 2 mL H_2O_2 were added into mixtures for one more hour in water bath at 80 °C. Then, 15 mL 1 M NH_4NO_3 were added and the tubes shaken for 10 min.

S5: the fraction bound to amorphous iron oxides (AmoFe)

Twenty-five mL of 0.2 M $(\text{NH}_4)_2\text{C}_2\text{O}_4$ + 0.2 M $\text{H}_2\text{C}_2\text{O}_4$ buffer solution (pH 2.89) were added into the residue soil from the last step and shaken in a temperature controlled shaker for 4 h at 25 °C under dark.

S6: the fraction bound to crystalline iron oxides (CryFe)

Twenty-five mL of 0.04 M $\text{NH}_2\text{OH}\bullet\text{HCl}$ + 25% CH_3COOH solution were added into the residue soil from the last step in water bath for 3 h at 95 °C.

S7: Residue fraction (RES)

Twenty-five mL 4 M HNO_3 was added into the residue soil from the last step, in a water bath for 16 h at 80 °C (Han *et al.* 2012).

Pseudo-total U concentration

The pseudo-total U concentration was determined in soil after digestion with HCl-HNO_3 mixture. Briefly, weighed 0.5 g soil sample in a 50 mL polytetrafluoroethylene vessel

with 4 mL of 3 HCl :1 HNO_3 (3 vol:1 vol) mixture and heated on a hot oven (100 °C, Environmental Express Ltd.). At the end, extracts were filtered through 0.45- μm membrane into 50-mL glass flasks, filled to the mark with distilled water and the U was analyzed with ICP-MS.

All extraction experiments were performed in duplicates. A series of standards were run in ICP-MS for every 30 samples to calibrate any drift to the instruments.

Determination of U concentration in plants

Dried plant samples were digested on hot block with wet acid digestion. Briefly, weighed about 0.5 g plant samples into a 50 mL polytetrafluoroethylene vessel with 6 mL concentrated HNO_3 and 2 mL H_2O_2 (30%) mixture. The digestion solution was diluted to 40 mL with distilled water. The solution was filtered through 0.45- μm filter and U was analyzed with ICP-MS, where U concentration should be below 100 $\mu\text{g L}^{-1}$. Blanks were prepared in the same way as the sample and each sample was duplicated.

Determination of soil pH

About 10 g of each soil sample was mixed with deionized water at soil/water ratio of 1:2.5. Stirred the mixture vigorously and allowed the slurry to set for about 15 min. A pH meter was used to measure the pH of the slurry after calibration using pH 4, 7, and 10 buffers. The electrode of the pH meter was directly placed into soil slurry and then pH was read and recorded. Room temperature (20–25 °C) was ensured throughout the pH measurement.

Statistical analysis

The data were processed with Micro software Excel and Origin 8.0 (OriginLab, USA).

U removal efficiency was determined with plant uptake amount and amount of U in soil. The equation is as follows:

$$w(\%) = m_r \cdot w_r / C \cdot M \text{ or } w(\%) = m_s \cdot w_s / C \cdot M,$$

where $w(\%)$ is the U removal efficiency by plants in %; m_r and m_s are the U concentration in plant roots and shoots in mg kg^{-1} , respectively; w_r and w_s are the plant root and shoot biomass weight in kg; C and M are the U concentration in soils in mg kg^{-1} and total soil mass in kg, respectively.

Results and discussion

Effect of the EKF on soil pH

The changes of soil pH under EKF showed the similar trend with sunflower and Indian mustard (Figure 2). Soil pH near the anode decreased, while pH increased near the cathode. For example, for sunflower experiment, pH of the control soil without metal (CK) was 7.65 but decreased to 5.24 near the anode and increased to 9.38 near the cathode. Similarly, pHs changed in both anode and cathode regions in U-treated soils. The soil pHs with UO_2 , UO_3 , and $\text{UO}_2(\text{NO}_3)_2$

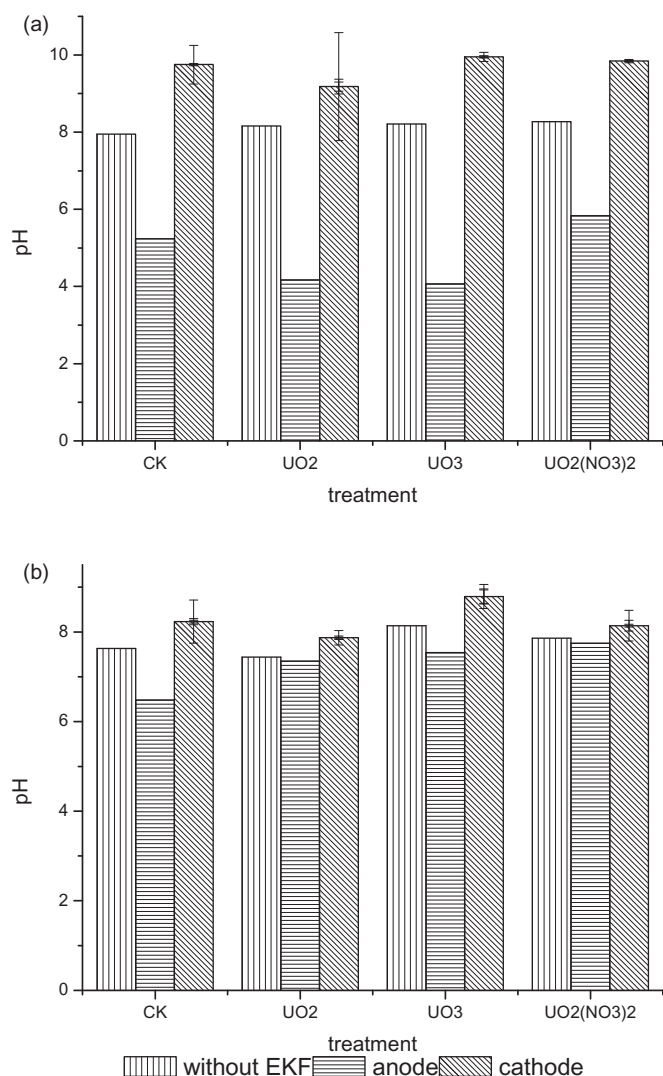


Figure 2. Effect of EKF on pH changes of depleted U (DU)-contaminated soil after growing sunflower (a) and Indian mustard (b).

treatments were around 8.02–8.20, but reduced to 4.07–5.86 near the anodes while increased to 8.19–9.48 near the cathodes. Among all U treatments, UO₃ had the strongest acidification in the anode (4.07) but UO₂(NO₃)₂ had the strongest alkalization (9.48) in the cathodes. It was noticed that sunflower growing had lower pH in soils than Indian mustard (Figure 2). This might be related with the amount and type of root exudates (such as organic acid and polysaccharide) (Mench *et al.* 1988; Kim *et al.* 2010).

Results indicated a significant acidification and alkalization of soils under operational electrical conditions at DC electrical field intensity of 1 V cm⁻¹ for a period of 9 days (24 h per day). The pH changes were ascribed to the generation of H⁺ and OH⁻ ions from water electrolysis under DC treatments. EKF seemed an effective technique to change soil pH. The application of the electric potential induces a variety of reactions and transport processes in the contaminated soil, which result in the mobilization and transport of the contaminants toward the anode or cathode electrodes for subsequent removal out of the contaminated soil (Cameselle *et al.* 2013). Thus, these changes shifted ion concentrations in the pore water of soil, contaminant

speciation, and contaminant dissolution or precipitation (Reddy and Cameselle 2009). Lowering soil pH often increases bioavailability of most metals in soil.

U concentration and uptake in sunflower and Indian mustard

Uranium forms significantly affected plant uptake of U as well its bioaccumulation in shoots and roots (Figure 3). Both sunflower and Indian mustard accumulated more U in both shoots and roots from soils with U(VI) such as UO₃ and UO₂(NO₃)₂ than from soil with U(IV) such as UO₂ ($p < 0.05$) (Figure 3). This was related to the solubility of U compounds. UO₂ was highly insoluble in soil, while UO₃ and UO₂(NO₃)₂ had high solubility (Meng *et al.* 2018). Moreover, more U was accumulated in roots than in shoots.

Meanwhile it was found that EKF significantly increased U uptake in both shoots and roots of sunflower and Indian mustard (Figure 3), especially for soils with UO₃. In other words, EKF increased bioavailability of U in soils compared to the no-EKF treatment. U concentrations in sunflower shoots with EKF increased by >50–100% from soils with UO₂ and UO₃ (both >100%) and uranyl (>50%). U concentrations in the Indian mustard roots with EKF also increased by >50% compared to those without EKF for soils with UO₂, UO₃, and UO₂(NO₃)₂. A similar increase in U concentrations with EKF was found in the roots of sunflower compared to those without EKF. The lower applied voltages significantly increased U in both shoots and roots compared to no-EKF treatment for both UO₂ and UO₃ treatments ($p < 0.05$). There were possible mechanisms responsible for electrokinetic enhancement of DU on plant uptake: (1) Acidification in the anode section; (2) potential enhanced mobilization processes from electro migration of uranium ions across the soil profile and electro-osmosis of soil solution/water moving toward the cathode section (Mao *et al.* 2016c).

U fractionation in soil and EKF effects

Uranium underwent different redistribution processes among various solid phase components (Figure 4). The solid phase component distribution of U highly depended on the U sources. EKF often enhanced such a redistribution process.

In the control soil after a growing season of sunflower without EKF, 38.7% of the total U in the soil was bound in the CARB fraction, followed by the ERO fraction (22.9%) with 0.26% total U in EXC fraction (Figure 4a). Uranium in soils with UO₂ was mainly presented in the AmOFe (70.8%), followed by CryFe (22.3%) (Figure 4a). The CARB fraction (35.8%) and AmOFe (22.5%) fraction, and ERO fraction (21.9%) were the dominant solid-phase fractions in UO₃ contaminated soils, while the CARB (36.26%) and OM fractions (36.7%) were the major solid-phase fractions for soils with UO₂(NO₃)₂ (Figure 4a). Results indicated the similar portion of the CARB fraction in soils with UO₃ and UO₂(NO₃)₂, but more OM fraction was found in soils with UO₃(NO₃)₂. Numerous studies showed that carbonates had a strong complexing capacity with U (Waite *et al.* 1994;

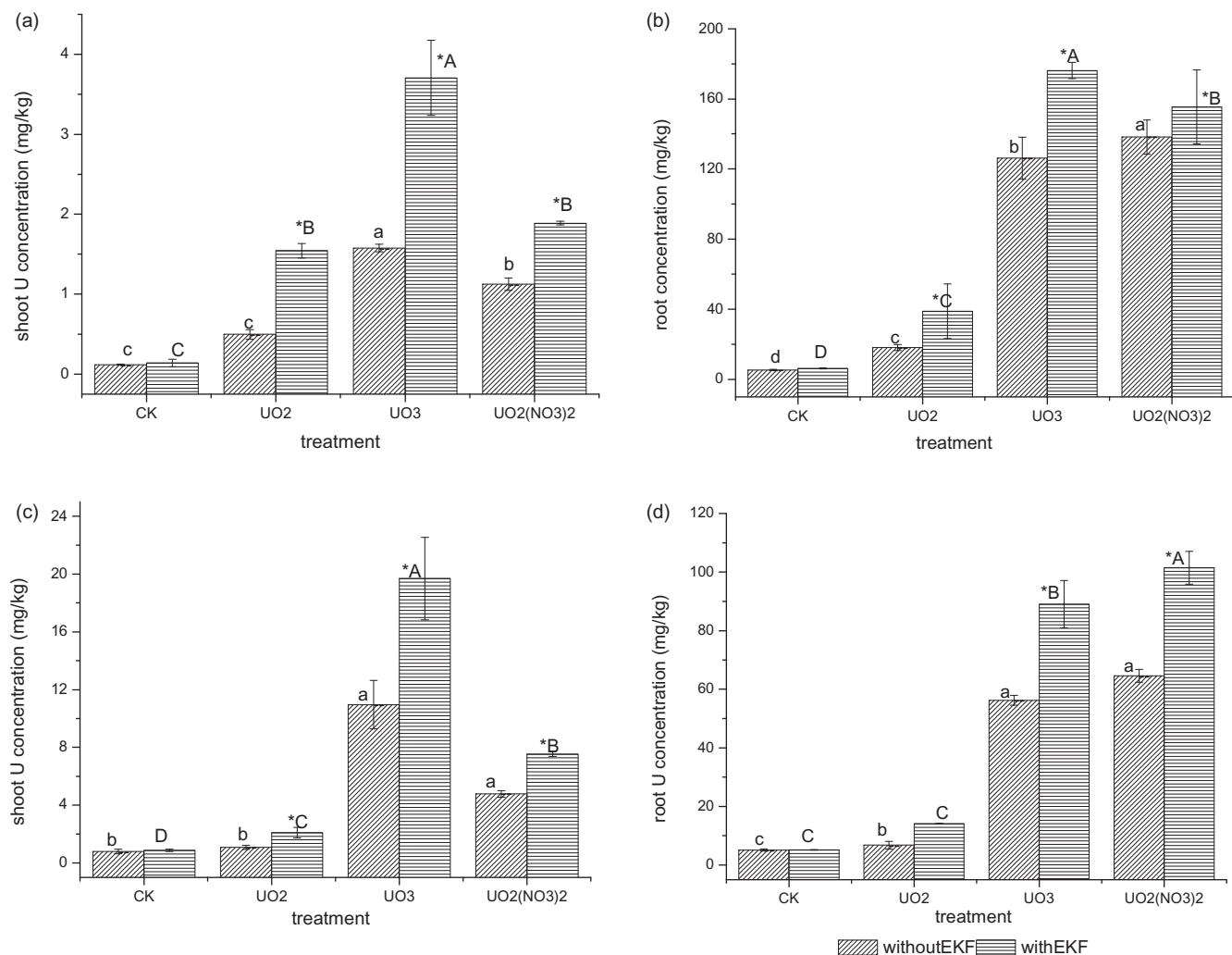


Figure 3. U UPTAKE and bioaccumulation in shoots and roots of sunflower (a,b) and Indian mustard (c,d) from soils with U sources. The same label among various U species indicated no significant difference at $p = 0.05$ probability level and the same alphabet whether capital or low case within the same U specie indicated no significant difference at $p = 0.05$ probability level between EKF and no EKF treatments.

Pabalan and Turne 1997; Pabalan *et al.* 1998). Pollutants associated with oxidizable phase of soil such as organic matter bound are assumed to remain immobilized for longer periods and become available through decomposition processes of organic matter (Filgueiras *et al.* 2002; Han, 2007).

EKF treatment significantly shifted redistribution of U among solid-phase components in soil. In the anode of the control soil after a growing season of sunflower, the main U fractions were the CARB and ERO fractions (47.5% and 26.6%, respectively) (Figure 4a). In the anode with UO₂, U was mainly presented in the AmoFe (59.4%), followed by the CryFe (13.8%) (Figure 4). The other fractions decreased in the order: ERO > CARB > OM > RES > EXC. In the anode of soils with UO₃, U was mainly present in the CARB (29.8%), ERO fraction (25.8%), OM (18.9%), and AmoFe (18.4%) fractions. While the CARB (41.5%) and OM (20.8%) fractions were the major solid-phase fractions in soils with UO₂(NO₃)₂ (Figure 4a).

In the cathode of the control soil, the main fractions were the CARB and AmoFe fractions (55.4% and 12%, respectively) (Figure 4a). In the cathode of soil with UO₂,

U was mainly present in the AmoFe (68.7%), followed by the CryFe fraction (21.4%). In the cathode with UO₃ contaminated soils, the carbonate (30.4%) and reducible (29.5%) fraction were the major ones with AmoFe (17.4%) and OM (15.3%). In the cathode of soils with UO₃(NO₃)₂, the CARB (42.4%) and OM (22.8%) were the main solid-phase fractions (Figure 4). Moreover, U in soils with various U sources after a season of Indian mustard had the similar U redistribution process among solid-phase components, but U was more in the AmoFe fraction and less in the CARB fraction (Figure 4b)

U sources significantly affected U solubility and potential bioavailability in soil. EXC and to some extent CARB fractions are considered to be rapidly bioavailable to plants (Han 1997). In the control soil without EKF, more than 1.6% of the total U in the soil was bound in the EXC and CARB fractions. In the UO₂ soil, 2.1% of the total U was bound in the EXC and CARB fraction. However, both EXC and CARB fractions increased to 42.8% and 53.9%, respectively, in soils with UO₃ and UO₃(NO₃)₂. EKF did not show significant effects on U redistribution among solid-phase components.

Compared to those in the anode, U in EXC and CARB fractions significantly reduced in the cathode in soils with all three U forms.

The removal efficiency of uranium with sunflower and Indian mustard

Phytoremediation technology is feasible to remediate a large area of fields with low concentration of pollutants. Advantages

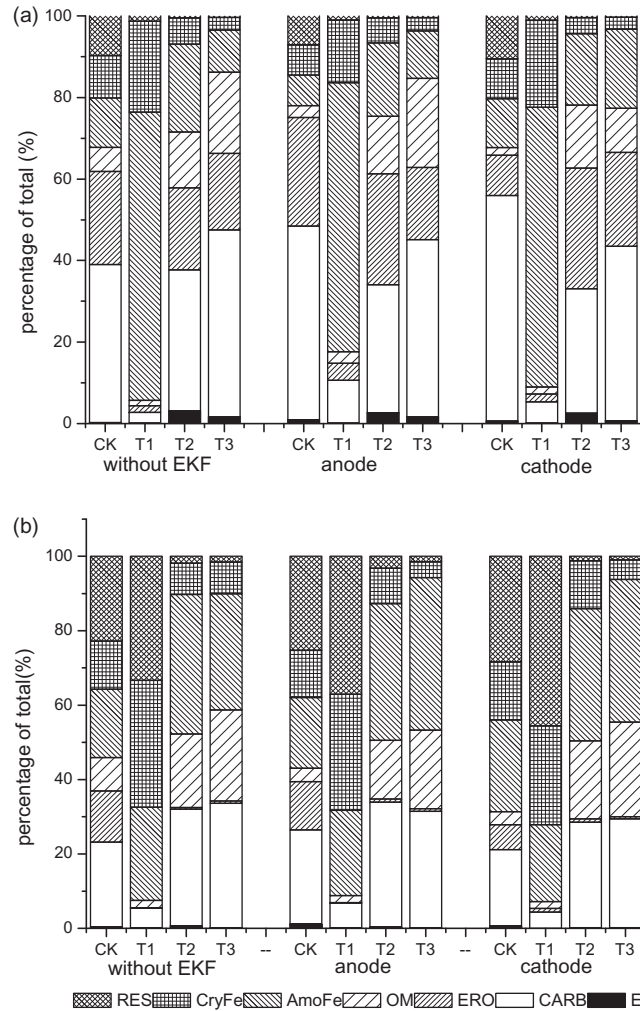


Figure 4. The fractionation of U in soils after 3 months of growth of sunflower (a) and Indian mustard (b) with and without EKF treatments. (T1, T2, T3 were soils with UO_2 , UO_3 , and uranyl, respectively).

included economic viability, effectiveness, no secondary pollution, and relatively little disturbance to the environment. The current study showed the efficiency of removing U depended upon original U source forms in soils. Both sunflower and Indian mustard showed higher U removal efficiency from soils with UO_3 and $\text{UO}_3(\text{NO}_3)_2$ than from soils with UO_2 . Without EKF the U removal efficiency in sunflower reached 3.7%, 3.4%, and 0.69% from UO_3 , uranyl, and UO_2 , respectively. Similarly, Indian mustard had the U removal at 4.3%, 3.9%, and 0.8% from soils with UO_3 , uranyl and UO_2 , respectively. Indian mustard showed a little higher U removal efficiency than sunflowers from soils with different U sources (Table 1). Meng *et al.* (2018) reported the similar range of removal efficiency of U from soils with various U sources. Since U was bio-accumulated in roots, roots of both sunflower and Indian mustard had a majority of contribution to the overall U removal. Low translocation factors of uranium in sunflowers and Indian mustard contributed the low removal of U in shoots (Rodríguez *et al.* 2006). It was noticed that nine days of consecutive treatment with DC electrical field intensity of 1 V cm^{-1} resulted in slight reduction of biomass production for both plants (data not shown).

EKF treatment significantly increased U removal efficiency of both plants compared to no EKF treatments (Table 1). With EKF the U removal efficiency in sunflower and Indian mustard reached 5.7–5.8%, 5.0–5.3%, 0.9–1.3% from soils with UO_3 , uranyl, and UO_2 , respectively. EKF treatments increased U removal efficiency by 35–50%, 35–47%, and 26–62% from soils with UO_3 , uranyl, and UO_2 , respectively. Similarly, the increase in U removal efficiency was the contribution of shoots and roots upon EKF treatments. Thus, the current study showed the potential of Indian mustard and sunflower for phytoremediation of U(VI) polluted soils coupled with electrokinetic treatments.

U is primarily in the (+VI) oxidation state in surface soils, which is the most mobile form (Christine *et al.* 2006, Meng *et al.* 2017, 2018). At low pH and in the absence of potential ligands, uranyl cation (UO_2^{2+}) is predominant. During oxidation of DU metal, DU was converted into UO_2 , then to UO_3 and other hydrate and complexes. Therefore, UO_2 , UO_3 , and uranyl species are the major U species in soils. Of course spiking soils with various U species may vary from the actual status of contaminated soils with its bioavailability and toxicity, but as close proxies these DU

Table 1. U removal efficiency (%) by Indian mustard and sunflower with EKF after a growing season.

Plant	Treatment	Without EKF			With EKF		
		Shoot	Root	Total	Shoot	Root	Total
Sunflower	CK	–	–	–	–	–	–
	UO_2	$0.09 \pm 0.01\text{b}$	$0.6 \pm 0.03\text{b}$	$0.69 \pm 0.02\text{b}$	$0.17 \pm 0.02\text{b}$	$0.7 \pm 0.03\text{b}$	$0.87 \pm 0.3\text{b}$
	UO_3	$0.29 \pm 0.03\text{a}$	$3.4 \pm 0.3\text{a}$	$3.69 \pm 0.3\text{a}$	$0.46 \pm 0.05\text{a}$	$5.1 \pm 0.6\text{a}$	$5.56 \pm 0.12\text{a}$
	Uranyl	$0.33 \pm 0.02\text{a}$	$3.1 \pm 0.6\text{a}$	$3.43 \pm 0.5\text{a}$	$0.44 \pm 0.03\text{a}$	$4.6 \pm 0.7\text{a}$	$5.04 \pm 0.1\text{a}$
Indian mustard	CK	–	–	–	–	–	–
	UO_2	$0.3 \pm 0.04\text{b}$	$0.5 \pm 0.02\text{a}$	$0.8 \pm 0.05\text{b}$	$0.6 \pm 0.45\text{b}$	$0.7 \pm 1.2\text{b}$	$1.3 \pm 0.12\text{b}$
	UO_3	$1.3 \pm 0.3\text{a}$	$2.5 \pm 0.3\text{b}$	$4.3 \pm 0.7\text{a}$	$1.9 \pm 0.3\text{a}$	$3.9 \pm 1.3\text{a}$	$5.8 \pm 0.1\text{a}$
	Uranyl	$1.2 \pm 0.41\text{a}$	$2.4 \pm 0.5\text{b}$	$3.9 \pm 0.4\text{a}$	$1.5 \pm 0.53\text{a}$	$3.7 \pm 0.62\text{a}$	$5.3 \pm 0.12\text{a}$

U removal efficiency was determined with plant uptake amount (the product of U concentrations in plants multiplied with biomass) and the amount of U in soil (the product of U concentrations in soils multiplied by soil mass). The same labels in the same column within the same plant indicated no significant difference at $p = 0.05$ probability level.

species to mimic actual DU contaminated soils was acceptable (Meng *et al.* 2018).

Conclusions

This study clearly showed the electrokinetic-enhanced phytoremediation of DU contaminated soil. EKF treatments significantly decreased the soil pH in the anode region and increased the soil pH in the cathode region. Therefore, U redistribution among various solid-phase components differed in both anode and cathode regions. EFK treatments increased U removal efficiency by 35–50%, 35–47%, and 26–62% from soils with UO_3 , uranyl, and UO_2 , respectively. This study implies the potential feasibility of coupled electrokinetic and phytoremediation of U polluted soils with various U forms by using sunflower and Indian mustard.

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14. ABSTRACT Electrokinetic-enhanced phytoremediation is an effective technology to decontaminate heavy metal contaminated soil. In this study, we examined the effects of electrokinetic treatments on plant uptake and bioaccumulation of U from soils with various U sources. Redistribution of uranium in soils as affected by planting and electrokinetic treatments was investigated. The soil was spiked with 100mg kg ⁻¹ UO ₂ , UO ₃ , and UO ₂ (NO ₃) ₂ . After sunflower and Indian mustard grew for 60 days, 1 voltage of direct-current was applied across the soils for 9 days. The results indicated that U uptake in both plants were significantly enhanced by electrokinetic treatments from soil with UO ₃ and UO ₂ (NO ₃) ₂ . U was more accumulated in roots than in shoots. Electrokinetic treatments were effective on lowering soil pH near the anode region. Overall, uranium (U) removal efficiency reached 3.4–4.3% from soils with UO ₃ and uranyl with both plants while that from soil with UO ₂ was 0.7–0.8%. Electrokinetic remediation treatment significantly enhanced the U removal efficiency (5–6%) from soils with UO ₃ and uranyl but it was 0.8–1.3% from soil with UO ₂ , indicating significant effects of U species and electrokinetic enhancement on U bioaccumulation. This study implies the potential feasibility of electrokinetic-enhanced phytoremediation of U soils with sunflower and Indian mustard.					
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