Multi-electrode System for Electrokinetic Remediation of Paddy Soil to Remove Toxic Metals

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Electrokinetic remediation technology is a promising method to recover metal-contaminated soil. In the present study, the experiment was carried out in a reactor cell which was made of bricks (length × width × height = 560 mm × 560 mm × 300 mm), containing 20 kg soil over 60 days. Six electrodes were vertically inserted as anodes on both the sides of soil column, while two electrodes were vertically inserted in the middle of soil area as cathodes. And a voltage gradient of 1.4 V/cm was constantly implemented in the experimental system. The solution of 0.1 M citric acid + 0.1 M sodium chloride (pH=3.24) was added into the experiment as electrolyte. After 60-day treatment, high concentration of residual lead and cadmium were obtained in the center of soil column. Compared with 57.02 % and 69.95 % of lead and cadmium removal efficiencies in whole experiment, 63.93% and 76.87% of lead and cadmium removal efficiencies were obtained in soil column excluding the central zone. X-ray Fluorescence Spectrometry (XRF), X-ray Powder Diffraction (XRD) and Scanning Electron Microscopy (SEM) studies were used to detect changes of soil particle in soil samples.

1. INTRODUCTION

Now the extensive deposition of hazardous heavy metals like Hg, Cd, Pb, Cr and Cu in the soil can lead to the biological toxicity and inhibits the plant growth [1]. With the significant development

of industry, human activities are the key factors for metals pollution in soil due to industrial waste, mining activities, mine tails and metal refineries [1-4].

The remediation technologies of metals contaminated soil are categorized into physical, chemical and biological remediation [5]. Electrokinetic remediation is one of the physical technologies to recover metal-contaminated soil which forces pollutants to move out from soil by electromigration, electroosmotic flow, electrophoresis and diffusion [6-7]. In addition, electrokinetic remediation also protects the natural environment at low cost as it can be easily installed and operated. There are many studies on electrokinetic remediation of heavy metals from soils which have shown high metals removal efficiencies [8-12].

Lead and cadmium could pose a threat to human beings by food and water [13]. There are many reports about recovering lead and cadmium contaminated soil by electrokinetic remediation [8,11]. And, the main mechanisms of lead and cadmium migrating out of soil by electrokinetic remediation test are desorption, dissolution, diffusion and electromigration [14]. And, the mobility and availability of lead and cadmium in soil are determined by pH, redox potential, minerals, and organic matters [15]. When the citric acid was dissolved in the electrolyte, low pH environment was created due to numerous deal of H^+ ions, which accelerates the lead and cadmium moving out from the soil particle. In addition, the organic matters of two functional groups (carboxyl and carbonyl) from citric acid will chelate with metals and transform into soluble citric salt [16]. Furthermore, sodium chloride is one available salt which has good current conductivity when it dissolves and generates quantity of Na^+, Cl^-, OH^-, H^+ ions in solution. And, the cations of Pb and Cd complex with Cl^- to form Pb/CdCl and then migrate out from soil matrix [17]. The potential redox in the experiment change the mobility and toxicity of trace metals. Since sodium chloride was mixed in the electrolyte, the electrical conductivity of solution was increased sharply. The rising thermal loss surged the temperature and redox potential in solution. The oxidation and reduction of lead and cadmium can be extracted from soil by citric acid and sodium chloride.

During electrokinetic remediation process, the type and configuration of electrode system are key to pollution removal efficiency. Most studies were tested in uniform electric field distribution of two plate electrodes. Generally, the surface area of the plate electrode is large than other ones which intensify the polar reaction and cause large power consumption. It is necessary to operate the experiments to evaluate the removal of treatment with different electrode configuration. As a result, there have been some reports about electrokinetic remediation by multi-electrode system [18-20]. Those works were carried out at large scale and implemented with cylinder electrodes in non-uniform electric fields. The hexagonal electrikinetic systems was used to remediate the saline agricultural lands [21]. Approximately 30 % of K and Ca were cleaned from the EK reactor (1m × 1 m × 0.25 m) during the 14-day test. Turea assessed effect of octagonal electrode configuration on the efficiency of electrokinetic remediation and compared with two-plate electrode arrangement [22]. Kim estimated the metals removal efficiencies of electrode configuration on in situ electrokinetic remediation of metal-contaminated soil [19]. In the experiment, a great number of electrode were involved in configurations which could cause high current and raise soil temperature.

Therefore, in present study we investigated the feasibility of lead and cadmium from paddy soil by multi-electrode system for 60 days. The changes of soil properties before and after the treatment were tested in detail.

2. MATERIALS AND METHODS

2.1 Paddy soil preparation

The paddy soil used in this experiment was sampled from a rice farmland of Yunnan Province, China, which was extremely polluted by metals. For the experiment, the soil was air-dried and sieved through a 10-mesh sieve (≤2 mm), then mixed uniformly by spade.

Table 1. The properties of lead and cadmium accumulated paddy soil

Parameters	Value
pH	8.97
Soil texture (%)	
Sand (2.00-0.05 mm)	18.36
Silt (0.05-0.002 mm)	58.08
Clay (<0.002 mm)	23.56
Electrical conductivity(μS/cm)	217
moisture content(%)	35.18
Initial concentrations of metals (mg/kg)	
Pb	944.91
Cd	7.53

Table 1 shows the main physical and chemical properties of paddy soil. The soil was classified into sand, silt and clay, which took up 18.36%, 58.08% and 23.56% in the soil. The pH of initial sample was 8.97 and the average moisture was 35.18%. The concentrations of lead and cadmium in initial sample were 944.91 and 7.53 mg/kg respectively.
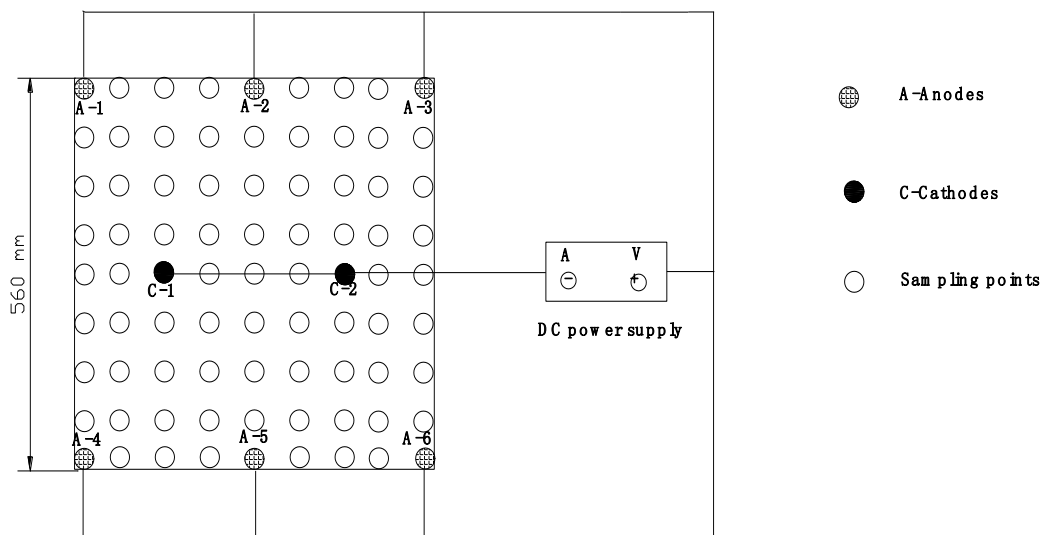
2.2 Experimental design

Figure 1. Schematic diagram of electrode configuration and sampling points

In this experiment, paddy soil was padded into a reactor cell which made of bricks (length × width × height= 560 mm × 560 mm × 300 mm). Fig 1 shows the schematics representation of test system. In the system, six electrodes were vertically inserted as anodes in both sides of soil column, while two electrodes were vertically inserted in the middle of soil column as cathodes. All the anodes and the cathodes used in this experiment were high purity graphite electrodes with a large quantity of holes which allow the electrolyte to pass through freely. The permeable geotextile was placed between electrolyte well and soil column to block the soil particle enter to the electrolyte wells. In order to improve the water tightness of brick, some more impermeable coating was added into the surface of soil reactor. In addition, a waterproof geotextile was placed under the soil to prevent the possible leak. After 2 months electrokinetic remediation, 73 soil samples were collected at the indicated points by soil sampler (Eijkelkamp, Agrisearch Equipment, Netherlands).

20 kg paddy soil (dry weight) was used in the experiment, and the thickness of soil column was 80 mm. In the experimental system, a voltage gradient of 1.4 V/cm was constantly implemented. The solution of 0.1 M citric acid + 0.1M sodium chloride (pH=3.24) was added into the experiment as an electrolyte. Before to start the experiment, the electrolyte was added into the soil column to soak the soil particle for two days. In the process of electrokinetic remediation, most of metals migrate to the electrolyte wells. As the experiment extended, the metals concentration in electrolyte wells was increased. In order to remove metals from soil, we refresh the electrolyte regularly in the two-month remediation. The dewatering electrolyte information shows in table 2.

Table 2. Information of dewatering electrolyte (the volume of extract solution was 5 liters each time)

Treating period	Dates of dewatering	Pb value (mg / L)	Amount of removal Pb per day (mg)	Cd value (mg / L)	Amount of removal Cd per day (mg)
13/10-14/10	14/10	123.45	617.25	0.91	4.55
14/10-16/10	16/10	131.45	328.63	0.86	2.15

16/10-21/10	21/10	182.33	182.33	1.89	1.89
21/10-25/10	25/10	100.28	125.35	1.46	1.83
25/10-1/11	1/11	152.17	126.81	1.56	1.3
1/11-4/11	4/11	98.46	123.08	0.22	0.28
4/11-7/11	7/11	89.49	149.15	0.26	0.43
7/11-10/11	10/11	88.16	110.2	0.28	0.35
10/11-13/11	13/11	78.55	130.92	0.26	0.43
13/11-20/11	20/11	170.15	106.34	0.41	0.26
20/11-25/11	25/11	89.12	89.12	0.43	0.43
25/11-30/11	30/11	85.55	85.55	0.45	0.45
30/11-5/12	5/12	86.15	86.15	0.41	0.41
5/12-10/12	10/12	72.18	72.18	0.36	0.36

The currents of anodes and cathodes were measured for every two-hours from 8 AM to 8 PM and counted the average values for every day during the 60-day operation.

2.3 Analytical method

The electrical conductivity (EC) and pH of soil before and after electrokinetic remediation were measured by an electrical conductivity meter (DDS-307 Shanghai Leici, China) and a digital acidity meter (PHS-29A Shanghai Dapu, China) respectively by mixing 10 g soil with 50 ml deionized water. After the experiment, the soil samples were collected by soil sampler (Eijkelkamp, Agrisearch Equipment, Netherlands). And then, those samples were air-dried and sieved through a 10-mesh sieve.

The total lead and cadmium in the soil samples were extracted by $HCl + HF + HClO_4 + HNO_3$ digestion. The digestion process of soil as follow: 1 g soil was added into 150 ml polytetrafluoroethylene tube mixing with 10 ml HCl at temperature of 130 ℃, then 20 ml HF, 20 ml $HClO_4$ and 10ml HNO_3 were injected into the test tube. And then the sample was heated at 160 ℃ until all the soil particle was digested. The final digestion solution was infused into 50 ml colorimetric tube with 5 % HNO_3. The values of lead and cadmium in solution were detected by thermo atomic absorption spectrometer (iCE 3300, Thermo Scientific, UK). As to insure the correctness and repeatability of the experiment, the new tubes and high purity graphite electrodes were used in the experiment. All chemicals used in the experiment were of analytical grade. The statistical analysis of experiment data was determined three times each sample and analyze by Origin 8.5.

2.4 Power consumption

The equation was used to calculate accumulated power consumption is listed as below [23].
$$W=\int_0^t UIdt$$
Where W is the power consumed, U is the voltage (V), I is the electric current (A), t is time implemented in the experiment (h).

3. RESULTS AND DISCUSSION

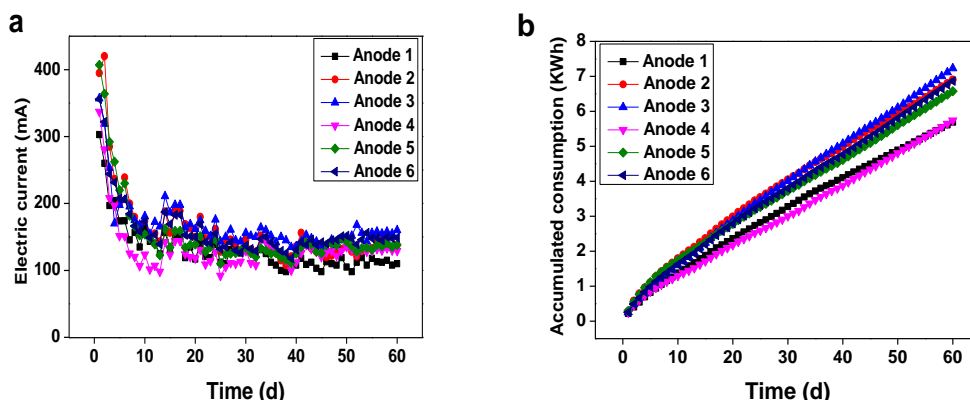*3.1 Electrical current changes and power consumptions.*

Figure 2. a Current changes and **b** accumulated consumptions of anode 1 – anode 6

Fig 2a is indicated the currents changes from anode 1 to anode 6 were slight different during the treatment. The maximum currents achieved at first day of electrokinetic remediation were 303, 395, 357, 337, 407 and 356 mA, respectively from anode 1 to anode 6. That because of many ions were solubilized from soil particle in the presence of sodium chloride which lead to electrical currents rose sharply at start time of the treatment [24]. As the operating time extended, anions and cations were moved toward to the anodes and cathodes respectively which made the resistance of soil growing drastically. At 13th day of treatment, the currents of all six anodes were dropped sharply (125, 143, 166, 98, 123 and 139 mA) which is nearly equal to one third of the values at test beginning. And then, the values of currents were waved slightly up to 43th day. Eventually, the currents were maintained at steady level due to the augment of soil resistance [25]. A similar decreasing tendency of total current was obtained and is shown in Fig 3a.
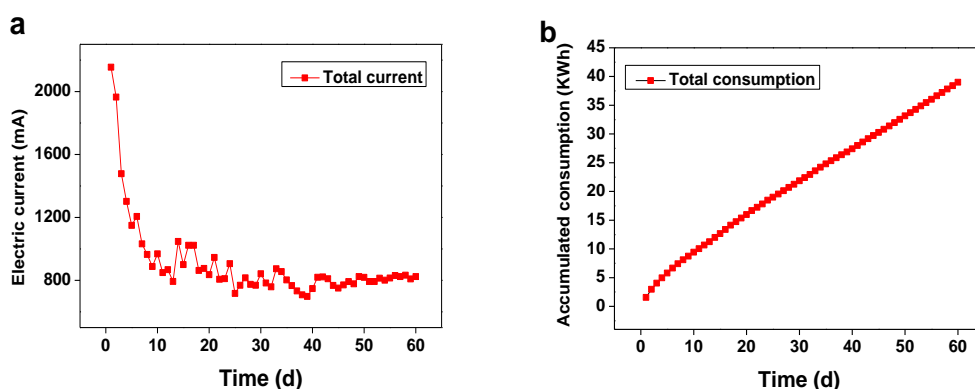

Figure 3. a Total current change and **b** total accumulated consumption

During the whole remediation process (2 months), a voltage gradient of 1.4 V/cm was constantly applied in the experiment system and the slight different currents implemented in anode 1 to anode 6 cause different power consumptions. Fig. 2b shows the increase power consumption order of

W (anode 1) = 5.69 kWh < W (anode 4) = 5.74 kWh < W (anode 5) = 6.57 kWh < W (anode 6) = 6.87 kWh < W (anode 2) = 6.90 kWh < W (anode 3) = 7.23 kWh. Six anodes and two cathodes were vertically installed in the soil column that cut the spacing of electrodes and greatly improved the power consumption in the whole experiment. Furthermore, the use of conductive solution intensifies the current consumption [26]. The total power consumption in the whole experiment was 39.00 kWh as the sodium chloride was presented in the electrolyte.

Fig. 3 shows the total current change and whole energy outflow in the experiment. Since the sodium chloride was injected into the soil column, the conductivity of electrolyte was significantly enhanced. The maximum total current obtained in the whole experiment at first day was 2155 mA. And then, the total current was plummeted into 794 mA at 13th day of treatment. Ultimately, the current kept at nearly 800 mA until the end of recover experiment. In the whole process of remediation, the total current consumption was 39.00 kWh.

3.2 Soil pH and electrical conductivity distribution

The water close the electrode was electrolyzed when electricity applied in the experiment. A great deal of H^+ and OH^- were generated at anodes and cathodes respectively and migrated to the oppositely electrodes which caused the whole soil column pH fluctuated. The initial soil pH was 8.97. In order to improve the desorption rate and enhance the activities of metals [27]. Citric acid (pH=3.24) was infused into the soil column to control the solution pH. Fig 4 shows the soil pH values increased from the both two sides of nearly 4 to the center of nearly 12 in soil column after the soil treatment. Contribute to the H^+ ions were generated from anodes, the soil pH values close those anodes were maintained low level. Ascribe to the slight different energy consumption for each anode that cause the different intensity of solution electrolysis close each electrode. After the treatment, soil pH values areas close anode 3 and anode 6 were higher than the ones close anode 1 and anode 4 due to more power consumptions of anode 3 and anode 6.
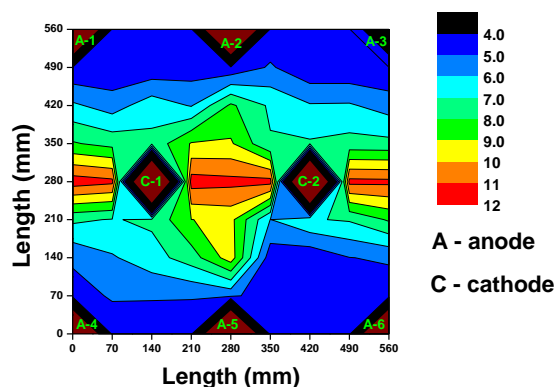

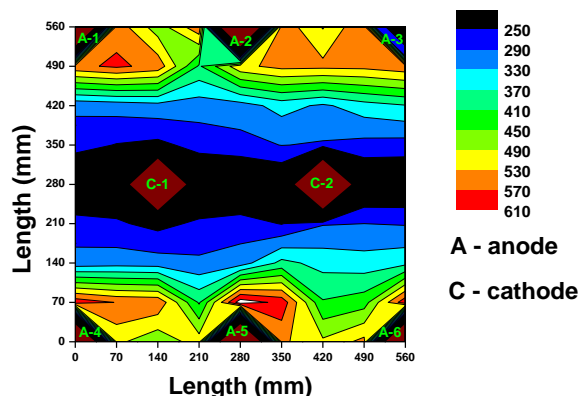**Figure 4.** Distribution of soil pH after electrokinetic remediation (initial=8.97)

Figure 5. Distribution of electrical conductivity after electrokinetic remediation (initial=217μS/cm)

Soluble salts, clay content, mineralogy, soil water content, bulk density, organic matter, and soil temperature in soil are the key parameters of electrical conductivity [28]. Compare with cations, the dissolved anions are more responsible for the electrical conductivity growth [21]. Since sodium chloride was presented in the electrolyte, numerous of Cl^- ions were found which significantly improve the electrical conductivity. The average of soil electrical conductivity after electrokinetic remediation was increased by 76.46%, from 217 μS/cm to 383 μS/cm. Fig. 5 shows the distribution of electoral conductivity after electrokinetic remediation. In contrast to the distribution of soil pH, the high electrical conductivity areas were located in low level of pH zones. The electrical conductivity values decrease from the middle zones of soil column to the cathode on both sides approximate of 200 μS/cm. This distribution of electrical conductivity is due to the high activity and the concentration of chloride ions which was accumulated in middle zones of soil column.

3.3 The dewatering electrolyte data and the removal of Pb and Cd

The chelating agent of citric acid was injected into soil reactor, which could not only control the electrolyte pH but also integrate with metals to form soluble complexes [29]. In the electrokinetic remediation, the main mechanisms of trace lead and cadmium migrated out from soil are electromigration and electroosmotic flow [30,31]. With time elapsed, the metals concentrations in electrode wells were increased. Then, the solution was periodically replaced by equal volume of fresh one. The information of dewatering electrolyte shows in table 2. The maximum values per day were obtained in electrolyte solution (first day, 617.25 mg Pb and 4.55 mg Cd) due to a large amount of metals were desorbed, dissolved and moved to the electrode wells. The amount of Pb and Cd removal per day was decreased drastically from the beginning day to 19th (123.08 mg Pb and 0.28 mg Cd) which only equal to 19.94% and 3.70 % of first day values. For this migration of metals out the soil column, current was the main source. When this experiment extended, current was maintained constant at low level in the experiment, the amount of transfer of metals per day from electrode wells was small.

From the literature we know that, citric acid in which it has chelating effect was widely used in electrokinetic remediation as electrolyte to separate the trace metals from soil particle by forming

metal-chelant [32]. Those metallic compounds have strong soluble in water. When electrical field was implemented in the soil column, the metal-chelant moved toward the cathodes and extracted with electrolyte.
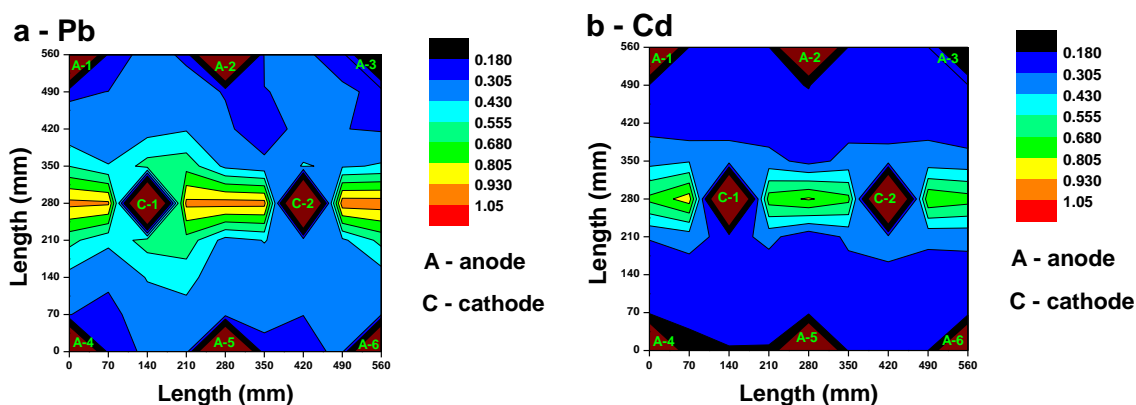

Figure 6. Distribution of residual Pb and Cd after electrokinetic remediation (C/C_0)

Thermodynamics and kinetics were the key parameters of metals removal efficiencies in the remediation system. Fig 6 shows the residual distribution of Pb and Cd after treatment. C and C_0 were the residual and initial values of metals respectively. After 60-day treatment, most of the metals had been moved out from the soil. But, some of anions (CO_3^{-2}, SO_4^{2-}, HPO_4^{2-}, OH^-, Cl^-) were accumulated in the central area of soil column. And, those anions combined with metals to form insoluble hydroxides and salts which caused a serious problem for the decontamination in the area. 97.88 % and 76.86 % of initial lead and cadmium were found in the center of soil column. Oppositely, few metals were achieved in the rest of soil column as the soil pH maintained at low level that due to the H^+ ions were attended from electrolysis of solution and citric acid. Generally, the low level pH condition determines the desorption efficiencies of lead and cadmium from soil particle and the stability constants of lead/cadmium-chelant complexes. In addition, the configuration of electrodes cut down the spacing between electrodes which strengthened the current in the whole experiment. Compared with 57.02 % and 69.95% of lead and cadmium removal efficiencies were obtained in the whole experiment. 63.93% and 76.87 % of lead and cadmium removal efficiencies were achieved excluded the central area of soil column. The high accumulated metals soil in central zone can be remove out the soil reactor and treat by electrokinetic remediation again. As a consequence, the configuration of electrodes can improve the metals removal efficiencies and short the remediation period.

3.4 Trace elements changes after experiment.

Trace elements changes in the soil after the experiment provides useful information which is important for agriculture [33,34]. X-ray Fluorescence Spectrometry (XRF), X-ray Powder Diffraction (XRD) and Scanning Electron Microscopy (SEM) were used to detect soil samples and obtain the exact information.

Table 3. The main elements volume changes before and after treatment.

Elements	Initial soil (%)	After soil (%)	Deposition in cathode wells (%)
Si	24.88	26.16	1.22
Al	11.19	11.81	0.70
Fe	3.54	2.96	0.29
Ca	2.96	0.21	31.87
K	2.67	2.91	0.15
Others	54.76	55.95	65.77

Table 3 shows the main elements volume changes in whole soils after the experiment. The main constituents of initial soil were Si, Al, Fe, Ca, K which took up 24.88 %, 11.19 %, 3.54 %, 2.96 %, 2.67 % respectively. After the remediation, those trace elements were occupied 26.16 %, 11.81 %, 2.96 %, 0.21 %, 2.91 % respectively. In the cathode wells, there had some white powder sediment in which Ca was occupied 31.87% far higher than one in initial soil of 2.96 % and one in after experiment soil of 0.21%. During the process of electrokinetic remediation, calcium completed with metals for complexing of citric acid and sodium chloride. The excess chelant integrated with mobile cations and moved to cathode wells by electromigration and electroosmotic flow. The trace elements of Si, Al, Fe and K were occupied small percentage of 1.22 %, 0.70 %, 0.29 % and 0.15% in the whole powder sediment of cathode wells.

The mobility of trace elements in soil depends on the process of precipitation-dissolution, sorption-desorption and reduction-oxidation [35]. Soil pH and redox potential are the key parameters of oxidation and reduction in soil. Usually, there are large percentage of trace element exists in nature soil as oxide-bound [36]. XRD results had indicated that several oxide-bound minerals were found in the agriculture soil. Such as silicon oxides, aluminum oxides.

Figure 7. a Surface change of soil particles before, **b** after in **electrokinetic remediation** and **c** deposition in the cathode wells

Some chemical and physical reaction (particularly, the surface reaction) had been found in soil particle since the presence of electrolyte and current. Those reactions accelerate molecular diffusion, dissolved constituents which were depleted soil particle surface. After soil treatment, the surface of soil particle (Fig 7 b) was more smooth than the initial one (Fig 7 a). In the process of electrokinetic remediation, most of calcium ions complexed with citric acid and sodium chloride. Eventually

complexes migrated to cathode wells by electromigration and electroosmotic flow. Fig 7 c shows the image of powder sediment in cathode which deposited by needle shaped crystals.

4. CONCLUSION

The experiment was carried out to evaluate the lead and cadmium removal efficiency in paddy soil with multi-electrode system under electrokinetic remediation. Six anodes and two cathodes were installed in the soil column which cut the spacing of electrodes. The configuration of electrodes can improve the metals efficiencies and shorten the remediation period. 57.02 % and 69.95% of lead and cadmium removal efficiencies were obtained in the experiment. It is increased to 63.93% and 76.87 % for lead and cadmium if we excluded the central area of soil column. The high accumulated metals soil in central zone can be removed out the soil reactor and treat by electrokinetic remediation again. As a consequence, the configuration of electrodes can improve the metals removal efficiencies and short the remediation period.

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