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# **Development of electrokinetic remediation for caesium: A** feasibility study of 2D electrode configuration system

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Abstract. Agar matrix was artificially contaminated with caesium and subjected to rapid assessment of electrokinetic treatment on the basis of the 2D electrode configuration. The effect of caesium concentration on the process was investigated using different electrode configuration (i.e. rectangular, hexagonal and triangular). During treatment the in situ pH distribution, the current flow, and the potential distribution were monitored. At the end of the treatment, the caesium concentration distribution was measured. The results of these experiments showed that for caesium contamination, pH control is essential in order to create a suitable environment throughout the agar matrix to enable contaminant removal. It was found that the type of electrode configuration used to control the pH affected the rate of caesium accumulation. All of the electrode configurations tested was effective, but the highest caesium extraction was achieved when the hexagonal pattern was used to control the pH. After 72 h of treatment at 50 mA, the concentration of caesium decreased gradually from the second and first layer of agar matrix throughout the cell, suggesting that most of the caesium was concentrated on the cathode part.

#### 1. Introduction

Now day Japan has faced a crisis on the radionuclide spreading after the accident in Fukushima Nuclear Power plant after hits by a tsunami following the M8.7 earthquake on 11 March 2011. Based on data released by MEXT and other resources, the radionuclide contaminants (Cs-137) are spreading in the soil and also water around 30 km from the accident location[1]. The deposition of <sup>137</sup>Cs occurs primarily in association with precipitation [2], and its initial depth distribution in the soil layer is affected by infiltration of precipitation into soil and adsorption of radionuclides by the soil particles of the surface horizon [3]. On one hand, shallow penetration and slow downward migration rate of <sup>137</sup>Cs result in its long residence time in plant root zone. On the other hand, deeper penetration and fast migration in soil may increase the risk of water contamination by the radionuclide. Even though today the radioactive concentration is still far below from the indexes of drinking water and food safety limit reported by the Nuclear Safety Commission of Japan, but it was still necessary to restore these contaminants location including around the nuclear reactor.

Recently there has been considerable interest in the international community in a range of in-situ and ex-situ remediation technologies for radioactively contaminated soil and groundwater, including a low expense and large scale application of phytoremediation [4, 6]. While these technologies have been applied in a range of environments, their application in clay and silt-rich soils may have raised the problem, due largely to the low hydraulic conductivities of these soils. Also, a major factor which

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influences the mobility of contaminants in soil is the soil pH. One emerging technology that has, however, received much attention as a practical in situ remediation technology for low permeability clay-rich soil is electrokinetic remediation, which applying a dc current to the soil results in the generation of acid at the anode and alkali at the cathode [5, 7, 8]. This leads to an acid front at the anode and a basic front at the cathode. In the presence of the electric field, the acid front tends to migrate towards the cathode and the basic front advances towards the anode by electromigration. This method has been successfully employed at laboratory scale on a range of radioactively contaminated soil and groundwater [9]. A common problem with electrokinetic removal of heavy metals is precipitation of the metal ions which has been found to occur close to the cathode, hindering the extraction process [10, 11]. The generation of alkali conditions at the cathode tends to immobilise heavy metal contaminants by hydroxide precipitation. A low pH environment can promote the removal of the metallic contaminant from fine grain soils as a low pH promotes desorption (by ion exchange) of most heavy metals and cations from the soil surface. The latest results has confirmed that the combined process of electrokinetic with other methods, such as permeable reactive barrier (PRB) material using drinking water sludge as an entrapping zone (i.e. entrapping zone electrokinetic remediation-EZ-EKR) and phytoremediation using Kentucky bluegrass as an accumulator species (i.e. electro-assisted phytoremediation-EAPR), has a promising result to apply for the removing heavy metal from contaminated soil [12, 13] and water [14].

An open electrode configuration allowing the exchange of solution between the electrode and the subsurface environment is essential for proper functioning of the electrokinetic process. The electrodes can be installed horizontally or vertically. Most bench-scale and large-scale laboratory and pilot-scale field studies on electrokinetic remediation and electro-assisted phytoremediation (EAPR) system performed to date have been horizontally one-dimensional (1D) [15, 16, 17]. Limited research has been conducted to study the effect of the vertically 1D electrode configuration on the efficiency of electrokinetic remediation and EAPR system [18, 19, 20]. Limited numerical sensitivity analyses have been performed on two-dimensional (2D) or axisymmetrical configurations (Renaud and Probstein 1987; Jin and Sharma 1991) and also on EAPR system [22]. In that configuration, the cathode is placed at the center, and the anodes are placed on the perimeter to maximize the spread of the acidic environment generated by the anodes and to minimize the extent of the basic environment generated by the cathode. These configurations of electrodes generate 2D non-linear electric fields. In a 1D configuration, the electric current density, i.e. current per unit area, is independent of location. In 2D configurations, however, the electric current density increases linearly with distance towards the cathode [23]. Therefore, the electric field strength also increases linearly with distance towards the cathode.

The purpose of this study was to evaluate a designed 2D electrode configuration on the electrokinetic system for removal of Cs from soil for further application in the field at Fukushima area, Japan. The study was conducted using an agar matrix for rapid assessment on the basis of pH changes as an indicator the effectiveness of the electrokinetic process.

## 2. Methodology

## 2.1. Designed electrokinetic cell

The study was focused on the development of modified electrokinetic system using 2D electrode configuration as shown in Figure 1. This reactor was provided to evaluate the application of vertical upward current, which is characterized by the removal of radionuclide from a lower subsurface soil (i.e. depth  $5 \sim 10$  cm). Furthermore, a low cost/free waste recycling and re-utilization material, such as dewatering drinking water sludge (DWS), will be applied as an adsorbent material for the combined electrokinetic and entrapping zone system as shown in Figure 2. This material in Japan is now a day produced in large quantity and dumped for the landfill as aluminum drinking water treatment residuals (Al-WTRs). Through the introduction of a ready cheap and available material, the method can be applied in low-cost operation and environmentally friendly technology.



**Figure 1.** The electrokinetic apparatus used in this study. (a) the cathode electrode made from stainless still bar (10 mm (W) x 200 mm (L), 3 mm thickness) and net wire (150 mm (W) x 200 mm (L), 10 mesh); (b) anode electrode made from graphite (100 mm (L),  $\emptyset$  58 mm); (c) dimensional electrokinetic cell (200 mm (W) x 300 mm (L) x 100 mm (H)); (d) lateral images of full scale 2D electrode configuration, respectively.



Figure 2. Schematic diagram of modified 2D reactor for EK/EZ system.

#### 2.2 Evaluation of 2D electrode configuration of electrokinetic cell

The aim of the study was to evaluate whether the construction of 2D electrode configuration could be used efficiently to enhance the electromigration of caesium ion from contaminated plume during the

electrokinetic process. The test was preceded in the agar matrixes representing as a liquid media, where rapid assessment of the caesium ions movement in a short time (i.e. 72 h) could be easily evaluated. The contaminated part was set-up in the bottom tray; meanwhile another part in the tray was filled with clean agar.Each layer of agar matrix was prepared by pouring of 30 g agar in 4 L of 0.01 M KCl solution. The contaminated agar matrix was prepared by addition of 100 mg/L of CsCl. The results from this step were further used to design the effectiveness of electrode configuration in the soil matrix for the electrokinetic process.The studies in agar matrix were carried out in three different electrode configurations such as rectangular, hexagonal and triangular patterns as shown in Figure 3. All studies were run in 50 mA of constant current.



**Figure 3.** 2D electrode configuration for the electrokinetic system. (a) rectangular configuration needs for 20 electrodes, (b) hexagonal configuration needs for 36 electrodes, and (c) triangular configuration needs for 18 electrodes, respectively. The black dot representing the electrode position and the connection of electric wire between points were represented by dashed yellow line.

## 3. Discussions And Implications

A series of experiments were carried out by using different 2D electrode configuration (i.e. rectangular, hexagonal and triangular) for 72 h electrokinetic process. Figure 4 shows the pH profiles from contaminated agar for rectangular electrode configuration. The decreasing agar pH which was indicated by purple color would increase the solubility of Cs ions. At first, of 2 h electrokinetic process acid pH gradually changed into mild acid as indicating with dark red color (i.e. pH range  $4.2 \sim 6.2$ ). After 72 h of the electrokinetic process, totally the agar media changed the acidic environment as indicating with red color. However, the pink color in the upper layer of agar matrix representing the alkaline environment (i.e. pH range  $7.8 \sim 10.0$ ). Also, a similar result has been obtained when hexagonal electrode configuration was applied in the system as shown in Figure 6. In this study, the anode position changed after 24 h electrokinetic process to spread the current for another half part of the electrokinetic cell.

Figure 5. shows the average caesium concentration in the upper layer of the electrokinetic cell was much increase after the test. The concentration of caesium much decreased from the contaminated layer (i.e., second layer) toward the first layer and an upper layer of agar matrix, suggesting that most of the caesium was concentrated on the cathode part. Furthermore, the distribution of caesium at upper layer much concentrated in the left part (S8 ~ S12) compared to the right part of the cell (S1 ~ S7) indicating the electromigration of Cs ion in the cell was not normally spreading due to unbalance a current distribution. However, for the hexagonal electrode configuration (figure 7), the electromigration of caesium at upper layer was grouped into two part of the hexagonal cluster. For example, the Cs ion has a high concentration at the right hexagonal group (i.e. S1 ~ S8) and left the hexagonal group (i.e. S9 ~ S15) indicating the current distribution in the cell tends to concentrate on the hexagonal structure. These results suggested that the electrokinetic process could be enhanced by 2D electrode configuration system.

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**Figure 4.** pH profiles from contaminated agar for rectangular electrode configuration in the electrokinetic process. The Color degradation is representing of the pH profiles in the electrokinetic cell, i.e. purple for bromophenol blue at pH range  $3.0 \sim 4.6$ ; dark red for methyl red at pH range  $4.2 \sim 6.2$ ; and pink for phenolphthalein at pH range  $7.8 \sim 10.0$ .



**Figure 5.** Caesium migration profiles from contaminated agar matrix toward cathode rectangular electrode configuration. (a) Sampling points of agar in the electrokinetic cell; (b) Concentration of Cs in each sectional of agar at sampling points.

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**Figure 6.** pH profiles from contaminated agar for hexagonal electrode configuration in the electrokinetic process. The Color degradation is representing of the pH profiles in the electrokinetic cell, i.e. purple for bromophenol blue at pH range  $3.0 \sim 4.6$ ; dark red for methyl red at pH range  $4.2 \sim 6.2$ ; and pink for phenolphthalein at pH range  $7.8 \sim 10.0$ .



**Figure 7.** Caesium migration profiles from contaminated agar matrix toward cathode hexagonal electrode configuration. (a) Sampling points of agar in the electrokinetic cell; (b) Concentration of Cs in each sectional of agar at sampling points.

## 4. Conclusions

The shape of the caesium concentration profile would depend in particular the pH of agar matrix. Regions with low pH would be expected to show higher rates of extraction, as the caesium is more readily dissolved in the pore fluid as indicating with red color (i.e. pH range  $4.2 \sim 6.2$ ). However, the pink color in the upper layer of agar matrix representing the alkaline environment (i.e. pH range  $7.8 \sim$ 

10.0) which indicating of heavy metal precipitation. The electrokinetic process could be enhanced using both of 2D electrode configurations (e.g. rectangular and hexagonal).

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#### References

- [1] Endo S, Kimura S, Takatsuji T, Nagasawa K, Imanaka T and Shizuma K 2012 Measurement of soil contamination by radionuclides due to the Fukushima Dai-ichi Nuclear Power Plant accident and associated estimated cumulative external dose estimation *J. EnviRadioact* 111 18-27
- [2] Tamura T 1964 Selective sorption reaction of caesium with mineral soils *Nuclear Safety* **5** 262-268
- [3] Ritchie J C and McHenry J R 1990 Application of radioactive fallout Caesium-137 for measuring soil erosion and sediment accumulation rates and patterns a review. J. EnviQual 19 215-233
- [4] Campbel L S I and Davies B E 1997 Experimental investigation of plant uptake of caesium from soils amended with clinoptilolite and calcium carbonate *Plant and Soil* **18** 65–74
- [5] J Trombly 1994 Electrochemical remediation takes to the field *Environ. Sci. Technol.* 28 289–291
- [6] Uchida S, Tagami K, Shang Z R and Choi Y H 2009 Uptake of radionuclides and stable elements from paddy soil to rice: a review *J. Enviro. Radio.* **100** 739–745
- [7] Acar Y B and Alshawabkeh A N 1993 Principles of electrokinetic remediation *Environ. Sci.* Technol. 27 2638–2647
- [8] Acar Y B, Gale R J, Alshawabkeh A N, Marks R E, Puppala S, Bricka M and Parker R 1995 Electrokinetic remediation: basics and technology status *J. Hazard. Mater.* **40** 117–137.
- [9] Al-Shahrani S S and Robersts E P L 2005 Electrokinetic removal of caesium from kaolin J. Hazard.Mater. B122 91 – 101
- [10] Lageman R, Pool W and Seffinga G 1989 Electro-reclamation: theory and practice *Chem. Ind.* 585–590
- [11] Eykholt G R and Daniel D E 1994 Impact of system chemistry on electro osmosis in contaminated soil J. Geotech. Eng. 120 797–815
- [12] Putra R S and Tanaka S 2011 Aluminum drinking water treatment residuals (Al-WTRs) as an entrapping zone for the lead in soil by electrokinetic remediation Sep.Pur Techno 79 208-215
- [13] Putra R S, Ohkawa Y and Tanaka S 2013 Application of EAPR system on the removal lead from sandy soil and uptake by Kentucky bluegrass (*Poapratensis L.*) Sep.Pur Techno. 102 34–42
- [14] Putra R S, Cahyana F and Novarita D 2015 Removal of lead and copper from contaminated water using EAPR system and uptake by water lettuce (*PistiaStratiotes L.*) Procedia Chemistry 14 381-386
- [15] O'Connor C S, Lepp N W, Edwards R and Sunderland G 2003 The combined use of electrokinetic remediation and phytoremediation to decontaminate metal-polluted soils: a laboratory-scale feasibility study *Enviro*. *Monit*. Assess. 84 141-158
- [16] Aboughalma H, Bi R and Schlaak M 2008 Electrokinetic enhancement on phytoremediation in Zn, Pb, Cu and Cd-contaminated soil using potato plants J. Enviro. Sci. Health Part A 43 926-933
- [17] Lim J M, Salido A L and Butcher D J 2004 Phytoremediation of lead using Indian mustard (Brassica juncea) with EDTA and electronics, *Microchem.* J., **76**, 3-9.

- [18] Wang J Y, Huang X J and Kao J C M 2006 Stabnikova, Removal of heavy metals from kaolin using an upward electrokinetic soil remediation (UESR) technology, J. Hazard. Mater., B136 532-541.
- [19] Zhou D M, Chen H F, Cang L and Wang Y C 2007 Ryegrass uptake of soil Cu/Zn-induced by EDTA/EDDS together with a vertical direct-current electrical field, *Chemosphere*, 67, 1671-1676.
- [20] Cang L, Wang Q Y, Zhou D M and Xu H 2010 Effect of electrokinetic-assisted phytoremediation of multiple metal contaminated soil on soil metal bioavailability and uptake by Indian mustard, *Separ., Pur., Techno.*, **79**, 246-253.
- [21] Renaud P C and Probstein R F 1987 Electroosmotic control of hazardous wastes, PCH *PhysicoChemical Hydrodynamics*, **9(1/2)** 345-360.
- [22] Hodko D, Hyfte J V, Denvir A and Magnuson J W 2000 Methods for enhancing phytoextraction of contaminants from porous media using electrokinetic phenomena, U.S. Patent No. 6,145,244, Nov. 14, 2000.
- [23] Alshawabkeh A N, Yeung A T and Bricka M R 1999 Practical aspects of in-situ electrokinetic extraction, *J. Environ. Eng.*, ASCE, **125 (1)** 27-35.