Improved electrokinetic process for capturing heavy metals in contaminated soil

by Romina Ghobadi

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Doctor of Philosophy in Environmental Engineering

under the supervision of Professor John L. Zhou and Dr. Ali Altaee

University of Technology Sydney
Faculty of Engineering and Information Technology

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CERTIFICATE OF ORIGINAL AUTHORSHIP

I, ROMINA GHOBADI declare that this thesis, is submitted in fulfilment of the requirements for the award of *PhD*, in the *School of Civil and Environmental Engineering/Faculty of Engineering and Information Technology* at the University of Technology Sydney.

This thesis is wholly my own work unless otherwise referenced or acknowledged. In addition, I certify that all information sources and literature used are indicated in the thesis.

This document has not been submitted for qualifications at any other academic institution.

This research is supported by the Australian Government Research Training Program.

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LIST OF PUBLICATIONS

As the 1st author, I have published three peer-reviewed articles derived from this thesis:

Romina Ghobadi, Ali Altaee, John L. Zhou, Peter McLean, Sudesh Yadav *"Copper removal from contaminated soil through electrokinetic process with reactive filter media"* **Chemosphere**, 252 (2020): 126607.

Romina Ghobadi, Ali Altaee, John L. Zhou, Peter McLean, Namuun Ganbat, Donghao Li "*Enhanced copper removal from contaminated kaolinite soil by electrokinetic process using compost reactive filter media*", **Journal of Hazardous Materterials,** 402 (2021) 123891.

Romina Ghobadi, Ali Altaee, John L. Zhou, Elika Karbassiyazdi, Namuun Ganbat "*Effective remediation of heavy metals in contaminated soil by electrokinetic technology incorporating reactive filter media*", **Science of the Total Environment**, 794 (2021) 148668.

In addition, I co-authored 2 peer-reviewed publications during PhD Study:

Sudesh Yadav, Ibrar Ibrar, Ali Altaee, Akshaya Kumar Samal, **Romina Ghobadi**, John Zhou "*Feasibility of brackish water and landfill leachate treatment by GO/ MoS2-PVA composite membranes*", **Science of the Total Environment** 745 (2020) 141088.

Namuun Ganbat, Ali Altaee, Uttam Kumar, Andrew Hursthouse, Sudesh Yadav, John Zhou, **Romina Ghobadi** "*Soil remediation techniques from organic and inorganic pollutants: review*", **Journal of Environmental Management.** Under review.

ABSTRACT

Soil contamination is a worldwide problem affecting human health. Electrokinetic (EK) remediation is a promising electrochemical remediation technology for soil decontamination using low-intensity direct current and resulting in the mobilization of chemical contaminants through electromigration and electroosmosis. EK application in the heavy metals contaminated soil faces with a few obstacles such as high pH distribution and contaminants extraction from the zone near the cathode region. Therefore, this study focused on integrating the EK technique with different reactive filter media (RFM) of activated carbon (AC) and biochar (BC), for the first time, without adding chemicals to facilitate the removal of copper ions from the contaminated kaolinite soil. This study initially selected copper as the heavy metal contaminant due to many reports on soil contamination by copper ions. RFM serves as an adsorbent during the electroosmosis and electromigration processes to facilitate contaminants removal. The performance of these RFMs in the EK system has been examined to select the best in terms of EK performance, regeneration, and contaminant removal. Tests based on EK (control test), EK coupled with AC (EK-AC), and EK combined with BC (EK-BC) were performed under an electric potential of 10 V, and the overall removal efficiency of copper ions decreased as EK-BC > EK-AC > EK. The results show that 27% of copper in the soil was captured by BC, compared with only 10% by AC. Additional EK-BC test performed under a constant current (20.00 mA) revealed that the acid front swept across the soil, resulting in 70.6%-95% copper removal from soil section 4 to section 1 close to the anode region, with more copper accumulation in section 5. Similar to the EK-BC test under a fixed voltage, BC captured 26% of copper in the soil during EK-BC treatment under a constant current, although with higher energy consumption. Moreover, RFM was regenerated by flushing with an acid solution,

achieving 99.3% of the copper recovery in BC and 78.4% in AC. Although the permeability of AC-RFM was higher than that of BC-RFM, the copper contaminant was more easily leached out from the BC-RFM.

Another recyclable RFM called compost (C) was proposed for the first time in this study to be applied in the EK system for copper removal from contaminated kaolinite soil. The feasibility of regeneration of compost was also studied for its reuse. Similar to other RFM, compost placed near the cathode served as an environmentally friendly adsorbent to bind copper ions while buffering the advancement of the alkaline front in the soil. The total copper removal rate increased from 1.03% in EK to 45.65% in EK-C under an electric potential of 10 V. Further experiments conducted by using compost/biochar (C+BC) mixture RFM at different ratios showed total Cu removal efficiency decreasing as EK-100%C > EK-(10%BC+90%C) > EK-(20%BC+80%C) > EK-(30%BC+70%C) > EK. The application of a constant electric current of 20.00 mA further enhanced copper removal to 84.09% in EK-100%C, although it did not show significant enhancement in EK-(BC+C). The compost RFM was regenerated by acid extraction and then reused twice, achieving a total removal of 74.11%. The findings demonstrated compost as a promising and reusable RFM for the efficient removal of copper in contaminated soil.

In addition, compost RFM was incorporated in the EK process for simultaneously treating multi-heavy metals contaminated real soil. Six EK operations were performed to investigate the performance of EK-RFM under different operating conditions such as the electric current, processing time, and the amount of the RFM. The electric current and treatment time demonstrated a significant impact on removing Zn, Cd, and Mn ions, while changing the amount of the RFM had an insignificant impact on heavy metals removal efficiency. The results showed that 51.6% to 72.1% removal of Zn, Cd, and Mn was achieved at 30.00 mA electric current and 14 days of operation. The energy consumption of the EK process was 0.17 kWh

kg^{-1}. The organic matter in the natural soil had a detrimental effect on the mobilization and migration of heavy metals such as Cu and Pb in soil. The results reported in this study should be useful in optimizing the design of the EK-RFM system and extending it to the field-scale applications.