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Research Article
Removal of Cu, Mo and As from Contaminated Industrial Soil by Electrokinetic Remediation

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Abstract
Background and Objective: Electrokinetic (EK) remediation used to remove organic and inorganic compounds by using an electric potential. The purpose of this study was to reduce Cu, Mo and As contents from contaminated industrial soil using the EK remediation. **Materials and Methods:** The soil samples were collected at 0-100 cm depth in industrial area, Erdenet mine, Mongolia. The Cu (7312 mg kg^{-1}), Mo (176 mg kg^{-1}) and As (114 mg kg^{-1}) content in topsoil sample was exceeded the toxic level of Mongolian National Soil Quality Standards (500, 20, 30 mg kg^{-1}, respectively). The heavy metals removal from contaminated soil by EK remediation was studied via comparing electrolyte solution (de-ionized water and 0.1 M NaCl), voltage (10-20 V) and time (108-200 h). **Results:** Based on results, the maximum removal of Cu was 90% of initial concentration occurred near the anode, in the condition of 0.1 M NaCl electrolyte solution, 15 V constant voltage and 156 h. Besides, the contents of Mo and As in anode and cathode were removed almost 89 and 79%, respectively. **Conclusion:** This study showed that EK remediation is a viable, environmentally friendly method for treatment of the copper contaminated soil.

Key words: Heavy metals, industrial area, contaminated soil, electric field, removal efficiency

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Data Availability: All relevant data are within the paper and its supporting information files.

INTRODUCTION

The soil is the basic materials of constituting the ecosystem and human life[1]. The contamination of soil usually arises from anthropogenic such as pesticide, wastewater, urban, industrial, traffic, mining, petroleum, motor oil and military areas[2-4]. Then, soil contaminated by organic and inorganic compounds has become hazard to the environment, human and animal health, food safety and agriculture[5]. Therefore, proper attention should be devoted to reduce or eliminate contaminants from the soil as much rapidly as possible. Depending on the origin and characteristics of soil pollution, the remediation has been already successfully introduced in practice on earth[6]. Nevertheless, our country as compared to the developed countries, the investment and research in the remediation of contaminated soil was not far enough[7]. The aim of soil remediation is not only to eliminate contamination from the soil but also to regenerate the quality of the soil. Environmental remediation technologies apply physical, chemical or biological processes to remove, reduce, stabilize or separated contaminants[6,8]. Physical methods of soil reclamation are the simple engineering methods such as soil spading, new soil importing and soil replacement. These techniques are not responsible to change the chemical properties of the pollutants accumulated in the soil to be cleaned[9]. The chemical reclamation method uses techniques such as washing, leaching, fixation, EK and vitrifies technology using chemical reagents and chemical processes[10]. Biological remediation methods are based on animals, plants (Phytoremediation) and microorganisms (Bioremediation), which are commonly used for the reclamation of soils polluted by heavy metals[11]. The nature and concentration of the contaminant are one of the most important factors to be taken into account in remediation technologies. Another factor is the economic cost[12]. From those reclamation methods, the advantage of electrokinetic remediation technique is technologically easy, low cost, the high removal efficiency of pollutants, less time consuming and a low electric potential is applied across the anodes and the cathodes[13]. Electrokinetic remediation is to remove organic and inorganic compounds by using an electric current[14]. Contaminants are transported by 3 contributive processes: electromigration (migration of ions species to the oppositely charged electrode), electroosmosis (movement of liquid containing ions) and electrophoresis (the movement of charged colloids and particles to the oppositely charged electrode)[15]. The purpose of this study was to reduce Cu, Mo and As content from contaminated industrial soil using the EK remediation.

MATERIALS AND METHODS

Study object is Erdenet copper mine, it's an open-pit mine that has been operated[16] since 1978. Every year, 22.23 million t of ore are extracted from this mine, with 126700 t Cu and 1954 t Mo produced. The research team was assessed the topsoil pollution around the Erdenet city and Erdenet mine, the study found that content of Cu, Mo and As in the topsoil of the industrial zone was high in the 4 seasons[17].

This study was conducted in the Laboratory of Environmental Analysis, the National University of Mongolia from January-October, 2019.

Soil properties: The soil samples (0-100 cm depth) were taken from Erdenet mine industrial area (north latitude 49°02'35.9", east longitude 104°08'38.9") that are shown[17] in Fig. 1 and 2. Soil color was classified via the Munsell methods[18]. The topsoil samples were dried at room temperature (25°C) for 2-3 days and soil pH and electrical conductivity (EC) were measured in a 1:2.5 soil/water suspension using a Mettler Toledo S47 pH meter and a Horiba B-173 electrical conductivity meter, respectively. The content of organic carbon (OC) was measured using the loss of ignition (LOI) method[19]. The concentration of elements in the soil samples was measured using an "Axios-Panalytical" X-ray fluorescence (XRF) spectrometry.

Electrokinetic remediation: The EK remediation experiments were carried out in a rectangular translucent plastic test reactor with the following dimensions:

Length = 12.0 cm
Width = 10.0 cm
Height = 10.0 cm

The graphite electrodes were used for the test, the surface area of the electrode was 22.58 cm^2. For each EK remediation test, approximately 400 g of dry soil sample was loaded into a reactor. At the end of each test, in order to study the soil was extruded from the reactor and divided into 2 equal sections between the anode and the cathode.

Statistical analysis: The electrokinetic remediation experiment has performed a frequency of 3 times and was calculated the amount of variation of heavy metals in the soil by the standard deviation. Also, the average soil sample of anode and cathode zones was prepared by systematic grid-square methods[20].

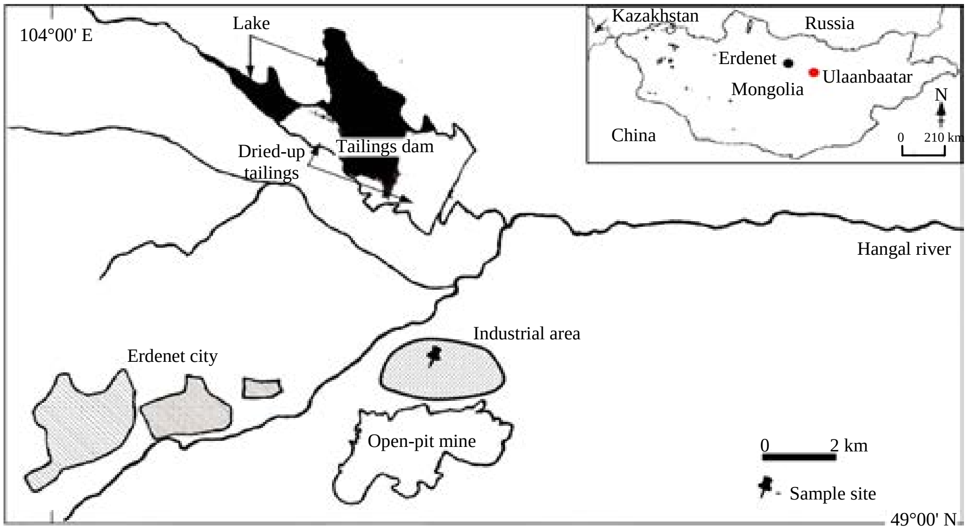

Fig. 1: Study area

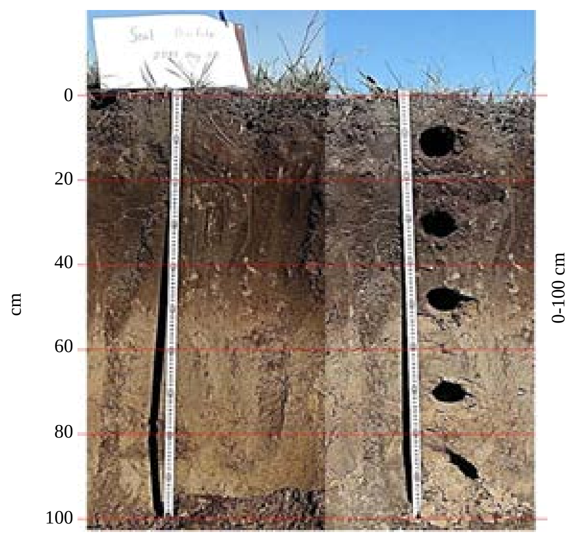

Fig. 2: Soil profile

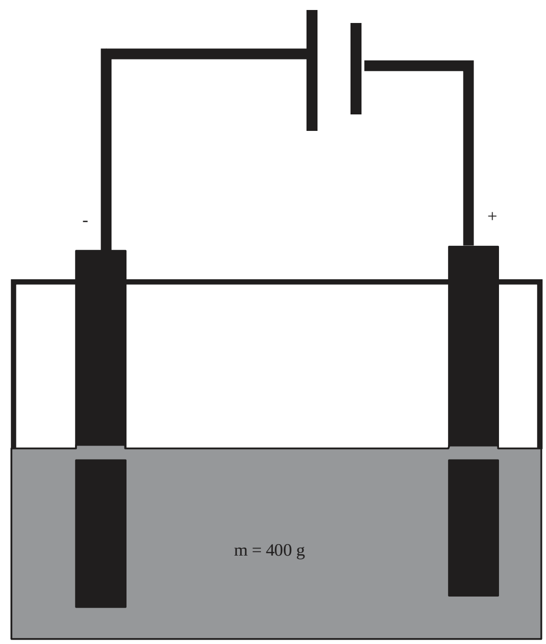

Fig. 3: Electrodes were placed directly on the soil

To compare of electrolyte solution and position of electrodes: The electrokinetic remediation must shows positive impact on the environment. Therefore, the electrolyte solution is directly linked to reducing pollution and distilled water and 0.1 M sodium chloride (NaCl) was used as the electrolysis solution. Depending on the position of the electrodes, the following comparative experiments were performed (Table 1). Figure 3 shows, the electrodes of the first reactor scheme was placed directly on the soil. Whereas, a graphite electrode assembly of the 2nd reactor scheme is placed inside both the anode and cathode chambers as shown in Fig. 4. Also, a glass fiber membrane was used between the electrode chambers and the soil cell to avoid any possible leakage. A control test was carried at 10 V and 108 h.

To compare of voltage and duration changes: In order to determine the optimum conditions for copper content, was studied the relationship between voltage and time after the position of the electrolyte solution and the electrodes. Ten reactor schemes with peak outputs of 10, 15 and 20 V were connected to identical and investigate the effectiveness of electrokinetics to remove the copper after 108, 132, 156 and 200 h of remediation. 0.1 M sodium chloride was used as an electrolysis solution and the electrodes were placed in the electrode chamber, on both sides of the soil (Fig. 4) in these tests.

Table 1: Electrolyte solution and position of electrodes

Test number	Electrolyte solution	Position of electrodes
1	H_2O	Electrodes were placed directly on the soil
2	NaCl	
3	H_2O	Electrodes were placed in electrode chamber, on both sides of the soil
4	NaCl	

Table 2: Content of some trace elements in the soil

	Elements (ppm)							
Profiles	As	Cu	Cr	Mo	Ni	Pb	V	Zn
0-20 cm	114	7312	58.47	176	16.91	22.72	42.30	183.7
21-40 cm	27.83	130.8	57.12	3.477	27.20	17.92	48.14	82.48
41-60 cm	12.14	22.93	22.03	2.016	5.098	9.955	6.277	72.52
61-80 cm	12.14	36.01	43.79	3.520	4.333	9.602	38.12	71.49
81-100 cm	2.62	40.69	29.30	4.371	12.93	12.14	22.89	75.49
0-100 cm	40.1	3263	43.60	7.90	29.60	20.9	33.57	94.00
MNS 5850:2008*	30	500	400	20	1000	500	600.00	600

*Mongolian soil quality standard (MNS 5850:2008)

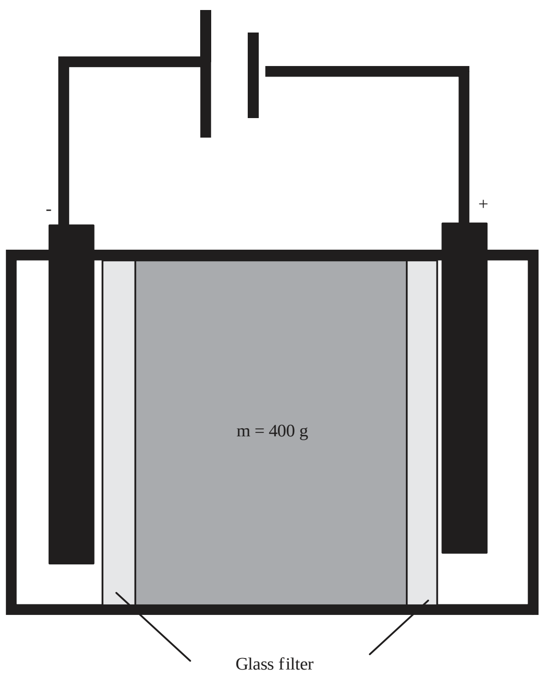

Fig. 4: Electrodes were placed in electrode chamber, on both sides of the soil

RESULTS

Soil properties: The Cu and As concentrations in the soils around the Erdenet mining area, measured by XRF and compared with the toxicological level of The Mongolian Soil Quality Standard (MNS 5850:2008)*, these are shown in Table 2.

The topsoil (0-20 cm) of the industrial area was contaminated with inorganic contaminants such as copper, molybdenum and arsenic. The concentration of Cu, Mo and As in topsoil samples were 7312, 176 and 114 mg kg^{-1}, respectively. Whereas, 21-100 cm depths soil not contaminated by those elements. Hence, due to the pollution of topsoil, the content of the Cu (3263 mg kg^{-1}) and As (40.1 mg kg^{-1}) in 0-100 cm soil was high predict.

The mineralogical analysis indicates the presence of quartz (SiO_2), albite ($Na(AlSi_3O_8)$), anorthite ($CaAl_2Si_2O_8$), magnetite (Fe_3O_4), hematite (Fe_2O_3) and chalcopyrite ($CuFeS_2$) in the study area. Therefore, we predict that the high content of Cu in the soil of the industrial area is correlated to minerals from the mining or smelting activities of this area.

Electrokinetic remediation

To compare of electrolyte solution and position of electrodes: The heavy metal removal from contaminated soil by EK remediation is due to combining the effects of electrodes position along with the change in the electrolyte solution during the process. The changes of total Cu concentration in the soil sample during EK remediation are displayed in Fig. 5.

The contents of Cu and As in the soil near anode were decreased by about 67 and 63% (maximum), respectively. Also, sodium chloride was removed copper from the soil than H_2O.

To compare the voltage and duration time: Ten reactor schemes with peak outputs of 10, 15 and 20 V were connected to identical and investigate the effectiveness of electrokinetics to remove the copper after 108, 132, 156 and 200 h of remediation.

As shown in Fig. 6, before the treatment the concentration of Cu was 3263 mg kg^{-1} and Cu was removed by 67%, near the anode after the remediation (15 V

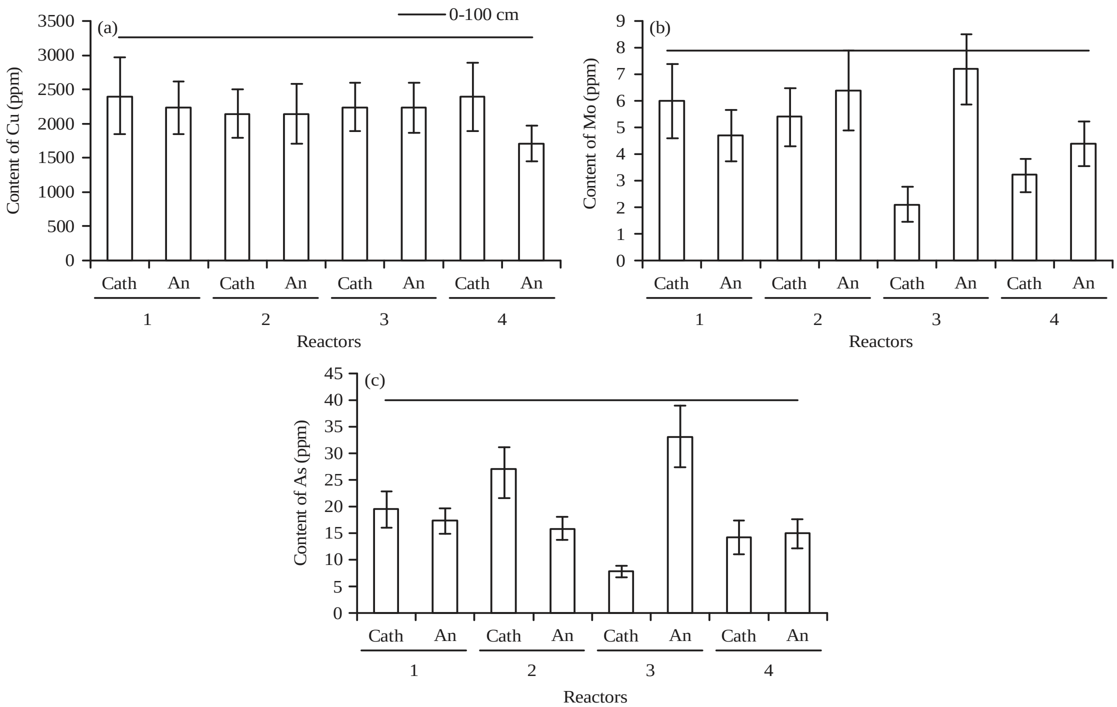

Fig. 5(a-c): Comparison of electrolyte solution and position of electrodes (mg kg^{-1}) (a) Cu, (b) Mo and (c) As
Cath: Cathode, An: Anode

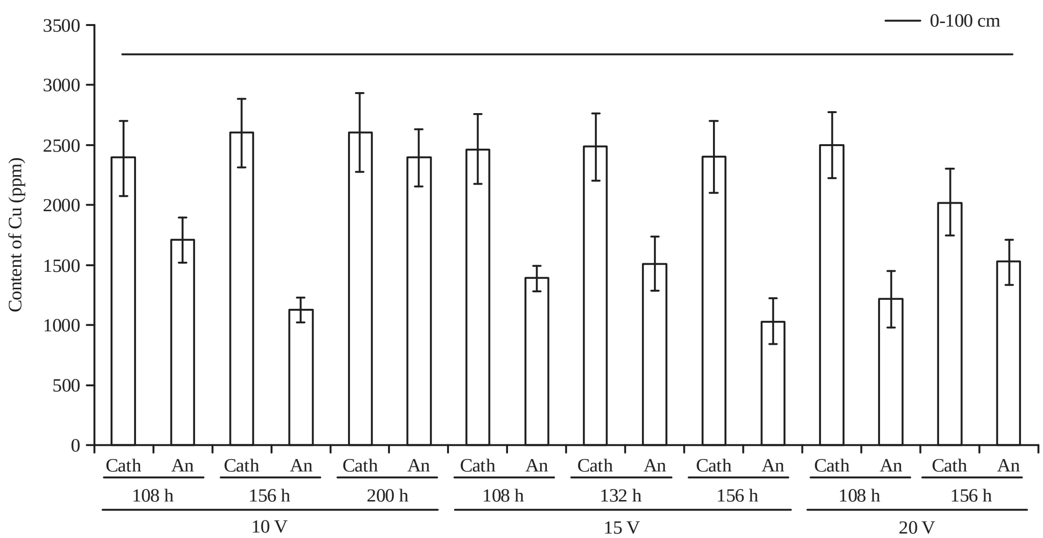

Fig. 6: Comparison of electrolyte solution and position of electrodes (mg kg^{-1})
Cath: Cathode, An: Anode

and 156 h). Thus, higher copper removal from the anode section can be concluded that the transport of copper in the soil pore fluid depends principally on the electro-migration and electro-osmosis. Results from these studies have shown that the optimal conditions of remediation were 15 V, 156 h used by 0.1 M NaCl.

EK remediation of topsoil (0-20 cm) sample: The most optimal condition of the previous study was used in this experiment. During this experiment, the grey coating had arisen on the surface of the soil after 96 h and is shown in Fig. 7. The grey coating was 1.5-2 cm distance from the anode, 1.5-3 cm in distance from the cathode and the coating

Fig. 7: Grey coating on the soil

thickness was approximately 0.1 mm. In order to determine the elemental analysis, the grey coating was cut from anode and cathode soils. Figure 8 also shows the comparison (%) of copper concentration in the soil after the EK remediation test to the initial concentration in the topsoil.

Results from studies have shown that the copper content by EK remediation was removed by 90% in the anode region and up to 42% in the cathode region. Besides, the contents of Mo and As in anode, cathode and grey coating was removed by about 89 and 79%, respectively.

DISCUSSIONS

This study identified Cu that is extremely accumulated in the topsoil (0-20 cm) of the industrial area. Besides, the Cu, Mo and As concentration of the topsoil was reduced by about 90, 89 and 79%, respectively by electrokinetic reclamation.

Charzynski *et al.*[21] identified technogenic soil that has been affected by the effects of human activity such as urban, industry, transportation, mining and military. Our study results showed that technogenic soil in the industrial area has been creating by the mining activity such as ore extraction, concentration and transportation for 40 years. Dasgupta *et al.*[22] and Zhou *et al.*[23] removed 74 and 81%, respectively of the copper pollution in the soil by the support

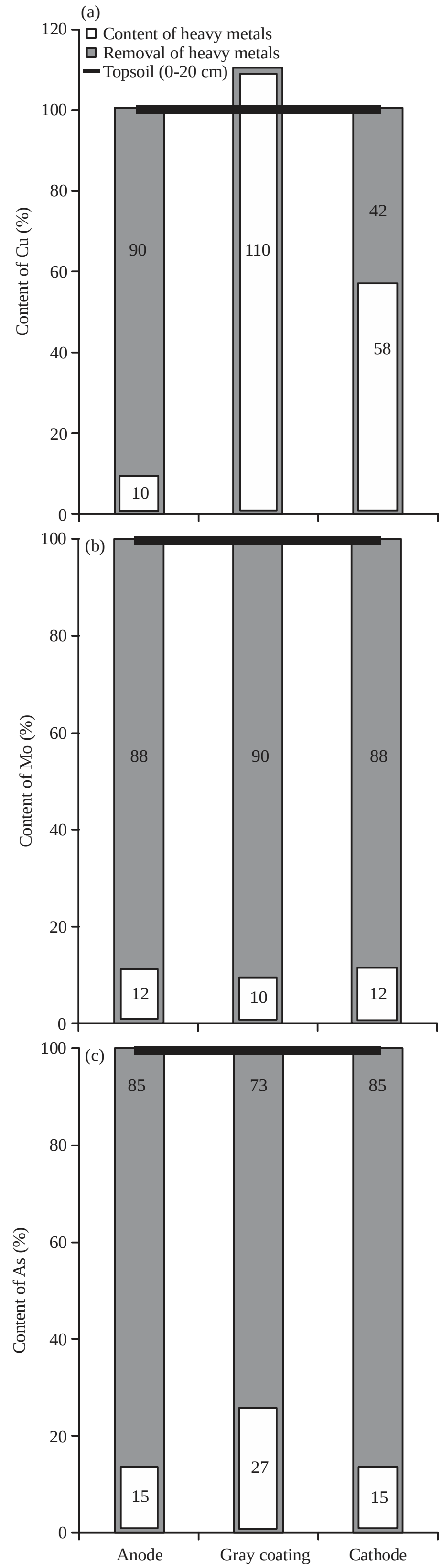

Fig. 8(a-c): (a) Cu, (b) Mo and (c) As concentration in topsoil sample after electrokinetic remediation (mg kg^{-1})

of the organic solution. Because organic solvents have a negative effect on the environment, distilled water and sodium chloride was selected as an electrolyte solution in the experiment. Figure 5 shows the 0.1 M NaCl solution has removed the heavy metal in the soil than distilled water during the electrokinetic remediation test. Because the NaCl solution has a higher electrical conductivity than distilled water as explained by Golnabi *et al.*[24].

Electrokinetic remediation method was combining effects of electroosmosis, electromigration and electrophoresis along with the change in the soil pH. In our study, substantially decreased of the copper content in the soil near the anode and this result was in agreement with previous studies by Hassan and Mohamedelhassan[7]. Whereas, the copper concentration in the grey coating was 10% increased than the initial topsoil by effects of electro-osmosis process was high accumulated.

This study was successfully reduced heavy metals in the contaminated soil using low voltage for short-term.

CONCLUSION

The highest concentrations of Cu (7263 mg kg^{-1}), As (114 mg kg^{-1}) and Mo (176 mg kg^{-1}) were found in topsoil samples (0-20 cm depth) from the Erdenet mining industrial area. Also, the content of Cu and As in the 0-100 cm depth soil samples were 3263 and 40.1 mg kg^{-1}, respectively. Those contents exceed their toxicological level stated in the Soil Quality Standards of Mongolia (MNS 5850:2008), whereas other microelement concentrations were much lower than their limits listed in these Standards.

The optimal conditions of EK remediation experimental are constant 15 V, 156 h, 0.1 M NaCl electrolyte solution and reactor with an anolyte and a catholyte chamber.

The copper content of topsoil was decreased about 90% (maximum) near anode using by the optimal condition. Besides, the contents of Mo and As was removed by about 89 and 79%, respectively.

SIGNIFICANCE STATEMENT

This study discovered the remediation method that can be beneficial for contaminated soil by Cu, Mo and As in industrial area moreover, this study will help the researchers to uncover the critical areas of environmental chemistry and soil remediation that many researchers were not able to explore. Thus a new theory on electrochemical of contaminated soil may be arrived at.

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REFERENCES

1. Cameselle, C., S. Gouveia, D.E. Akretche and B. Belhadj, 2013. Advances in Electrokinetic Remediation for the Removal of Organic Contaminants in Soils. In: Organic Pollutants-Monitoring, Risk and Treatment, Rashed, M.N. (Ed.)., IntechOpen Limited, London, ISBN: 978-953-51-5347-4.
2. Morel, J.L., C. Chenu and K. Lorenz, 2015. Ecosystem services provided by soils of urban, industrial, traffic, mining and military areas (SUITMAs). J. Soils Sediments, 15: 1659-1666.
3. Ferreira, C.S.S., Z. Kalantari, L. Salvati, L. Canfora, I. Zambon and R.P.D. Walsh, 2019. Chapter six-urban areas. Adv. Chem. Pollut. Environ. Manage. Prot., 4: 207-249.
4. Stoker, H.S. and S.L. Seager, 1977. Organic Chemical Pollution: Petroleum, Pesticides and Detergents. In: Environmental Chemistry, Bockris, J.O. (Ed.)., Springer, Boston, MA., ISBN: 978-1-4615-6923-7, pp: 401-427.
5. Ashraf, M.A., M.J. Maah and I. Yusoff, 2014. Soil Contamination, Risk Assessment and Remediation. In: Environmental Risk Assessment of Soil Contamination, Soriano, M.C.H. (Ed.)., IntechOpen Limited, London, ISBN: 978-953-51-4235-5, pp: 3-56.
6. Yao, Z., J. Li, H. Xie and C. Yu, 2012. Review on remediation technologies of soil contaminated by heavy metals. Procedia Environ. Sci., 16: 722-729.
7. Hassan, I. and E. Mohamedelhassan, 2012. Electrokinetic remediation with solar power for a homogeneous soft clay contaminated with copper. Int. J. Environ. Pollut. Rem., 1: 67-74.
8. Gill, R.T., M.J. Harbottle, J.W.N. Smith and S.F. Thornton, 2014. Electrokinetic-enhanced bioremediation of organic contaminants: A review of processes and environmental applications. Chemosphere, 107: 31-42.
9. Maini, G., A.K. Sharman, C.J. Knowles, G. Sunderland and S.A. Jackman, 2000. Electrokinetic remediation of metals and organics from historically contaminated soil. J. Chem. Technol. Biot., 75: 657-664.
10. Chang, J.H., Z. Qiang and C.P. Huang, 2006. Remediation and stimulation of selected chlorinated organic solvents in unsaturated soil by a specific enhanced electrokinetics. Colloids Surf. A: Physicochem. Eng. Aspects, 287: 86-93.

11. Ojuederie, O.B. and O.O. Babalola, 2017. Microbial and plant-assisted bioremediation of heavy metal polluted environments: A review. Int. J. Environ. Res. Public Health, Vol. 14, No. 12. 10.3390/ijerph14121504.
12. Gavrilescu, M., L.V. Pavel and I. Cretescu, 2009. Characterization and remediation of soils contaminated with uranium. J. Hazard. Mater., 163: 475-510.
13. Cai, Z.P., D.R. Chen, Z.Q. Fang, M.Q. Xu and W.S. Li, 2015. Enhanced electrokinetic remediation of copper-contaminated soils near a mine tailing using the approaching-anode technique. J. Environ. Eng., Vol. 142, No. 2. 10.1061/(ASCE)EE.1943-7870.0001017.
14. Zhao, S., L. Fan, M. Zhou, X. Zhu and X. Li, 2016. Remediation of copper contaminated kaolin by electrokinetics coupled with permeable reactive barrier. Procedia Environ. Sci., 31: 274-279.
15. López-Vizcaíno, R., A. Yustres, M.J. León, C. Saez, P. Cañizares, M.A. Rodrigo and V. Navarro, 2017. Multiphysics implementation of electrokinetic remediation models for natural soils and porewaters. Electrochim. Acta, 225: 93-104.
16. Dejidmaa, G. and M. Naito, 1998. Previous studies on the erdenetiin ovoo porphyry copper-molybdenum deposit, Mongolia. Bull. Geol. Surv. Jpn., 49: 299-308.
17. Yondonjamts, J., B. Oyuntsetseg, O. Bayanjargal, M. Watanabe, L. Prathumratana and K.W. Kim, 2019. Geochemical source and dispersion of copper, arsenic, lead and zinc in the topsoil from the vicinity of Erdenet mining area, Mongolia. Geochem.: Explorat. Environ. Anal., 19: 110-120.
18. Kichiro, I. and K. Kazuyuki, 2004. Standard soil color charts. Japan.
19. Turin, I.V., 1965. The soil organic/sulphide matter and its role in fertility. Italy.
20. Wollenhaupt, N.C. and R.P. Wolkowski, 1994. Grid soil sampling. Better Crops, 78: 6-9.
21. Charzyński, P., P. Hulisz and R.M. Bednarek, 2013. Polskie Towarzystwo Gleboznawcze. [Technogenic Soils of Poland]. Polish Society of Soil Science, Toruń.
22. Dasgupta, R., K. Venkatesh and R.P. Tiwari, 2016. Electrokinetic remediation of copper contaminated soils using EDTA. Int. J. Innov. Stud. Sci. Eng. Technol., 2: 18-23.
23. Zhou, D.M., C.F. Deng and L. Cang, 2004. Electrokinetic remediation of a Cu contaminated red soil by conditioning catholyte pH with different enhancing chemical reagents. Chemosphere, 56: 265-273.
24. Golnabi, H., M.R. Matloob, M. Bahar and M. Sharifian, 2009. Investigation of electrical conductivity of different water liquids and electrolyte solutions. Iran. Phys. J., 3: 24-28.