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Efficiency of EDTA and TWEEN 80 for the Removal of Chromium using Soil Washing Technologies

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Abstract: The study on the efficiency of chelate and surfactant for the removal of chromium using soil washing technology of which the chelate is EDTA and surfactant is TWEEN 80 at concentrations of 8, 16 and 32 millimoles was separately conducted. The experiment soil was synthetic soil with chromium contamination for 2 years. In this study, 20 g of contaminated soil would be washed with 200 milliliters of EDTA and TWEEN 80 with pH values of 2, 4, 6, 8 and 10 before being shaken at speeds of 90, 120 and 150 rpm for 15, 30, 60, 90, 120 and 180 min and at every 3 h until it reached its equilibrium point at room temperature. After that it was filtered with filter paper before digestion with acid whereupon the obtained solution would be further analyzed. The study result revealed that EDTA could remove chromium best at a concentration of 32 millimoles with pH of 10 and shaking speed of 120 rpm and it could eliminate chromium at the level of 38.54%. In the meantime, TWEEN 80 could remove chromium best at a concentration of 32 millimoles with pH of 10 and shaking speed of 150 rpm and it could eliminate chromium at the level of 9.86%. Therefore, when comparing efficiency between EDTA and TWEEN 80, the researcher concluded that EDTA had higher efficiency in removing chromium from soil than TWEEN 80.

Keywords: Soil Washing, Chromium, EDTA, TWEEN 80, Wastewater

Introduction

Contamination with heavy metals is a major problem that happens all over the world. Heavy metal contamination in soil can potentially lead to high environmental risks to human health and ecological security and it is necessary to remediate in such contaminated areas (Udovic and Lestan, 2008; Xuhui *et al*., 2015). Chromium (Cr) is one of the heavy metals most commonly found in the environment and normally highly absorbed in soil. Generally, it has been known that heavy metals are absorbed in soil with ion exchange and surface precipitation. In other words, it is sphere surface complexation and this is the term that has been widely used (Zhang *et al*., 2008).Chromium has two forms based on the principle of oxidation states including Hexavalent (Cr^{6+}) and Trivalent (Cr^{3+}); the quality of chromium depends on the molecular structure of the chromium composition, especially the oxidation state or oxidation number (Hawley *et al*., 2004).

Recently, technology of soil washing with proper extraction shows potential and is another alternative in treating and restoring contaminated soil (Maturi and Reddy, 2008; Dermont *et al*., 2008). Soil washing is a remediation process which is primarily used to treat soils and sludge which are contaminated with only one or two groups of contaminants (e.g., metals and/or volatile organic compounds) (Bilgin and Tulun, 2015). This process, however, has not been extensively employed on soils that are contaminated with pesticides in addition to metals and volatile organic compounds (Semer and Reddy, 1996). Soil washing is normally employed with different extracts such as acids, bases, chelating agents, electrolytes, oxidizing agents and surfactants (Zou *et al*., 2009). Chelating agents such as Ethylene Diaminetetraacetic Acid (EDTA) have the ability to capture heavy metals in stabilized form. More than that, they can be reformed to be soluble and movable in water. Therefore, EDTA is another substance that contributes to the removal of heavy metals from soil (Zhang and Lo, 2008; Sampanpanish and Pojanaporn, 2014). The washing method is utilized in restoring contaminated soil and it is acceptable when soils contain delicate soil content of less than 30%. In fact, delicate soil content has size smaller than 0.075 millimeters, such as sands and silt or clayey sands mixed with organic and inorganic matter. However, for washing contaminated soil, it should have the composition of delicate content or muddy content. If the composition of delicate soil is more than 30%, this method can be exercised but it also has limitations (Reddy and Supraja, 2000). For surfactants, it has been reported that this type of agent has good potential for removing metals in soil. However, popular surfactants are the cationic, anionic and nonionic types that can be used for washing (Mulligan *et al*., 1999) such as TWEEN 80, Triton X-100, Brij-35 (nonionic surfactant), SDS, AOT, SDBS (anionic surfactant) and hydrochloric acids (Kos and Lestan, 2004; Cheng *et al*., 2011; Xuhui *et al*., 2015; Min *et al*., 2017). According to the findings in many studies, filling surfactants in contaminated soil could enhance the emission and dissolution of contaminated organic matter because the solvent as cooled water needs to have a low Critical Micelle Concentration (CMC) related to a low value of toxic microbes (Cheng and Wong, 2006). Thus, this study was designed to show the efficiency of the chelating agent (EDTA) and surfactant (TWEEN 80) in removing chromium from contaminated soil using soil washing technology.

Materials and Methods

Chemicals Preparation

The chelating agent in this study is EDTA, purchased from Ajax (A.R. grade) and the nonionic surfactant is TWEEN 80 purchased from Rankem (Lab grade).

Soil Preparation

The soil in this study was chromium-contaminated soil through the study of plant toxicity and synthetic soil filled with potassium dichromate ($K_2Cr_2O_7$) with 2-year contamination. Basically, the soil property analysis included soil texture, pH, Cation Exchange Capacity (CEC), Electrical Conductivity (EC), Organic Matter (OM) and Total Chromium (TCr).

Experimental Design of Soil Washing

Each set of experiments of soil washing would use 20g of soil mixed with EDTA and/or TWEEN 80 as the solution for soil washing with concentrations of 8, 16 and 32 millimoles and pH of 2, 4, 6, 8 and 10 adjusted with HCL or NaOH. After that it would be mixed by shaking at speeds of 90, 120 and150 rpm for 15, 30, 60, 90, 120 and 180 min at every 3 h at ambient temperature until reaching the substance equilibrium. Finally, the clear part would be poured and filtered with filter paper for further analysis.

Samples Analysis

The concentration of chromium in the solution was analyzed with microwave digestion according to the principles of USEPA method 3051A (USEPA, 1998). After that, it was analyzed by Atomic Absorption Spectrophotometer (AAS).

Statistical Analysis

The statistical analysis was conducted through Analysis of Variance (ANOVA). In the case of different values, the comparison would be carried out with Duncan's New Multiple Range Test (DMRT) using the Statistical Package for the Social Sciences (SPSS) program.

Results and Discussion

Physical and Chemical Properties of Contaminated Soil

In this experiment the researcher used soil contaminated with synthetic chromium for 2 years which had been used for experiments with plants as a toxicity indicator. The results of the physical and chemical analysis of soils (Table 1) revealed that the soil was silty clay with a ratio of sand: Silt: Clay at 65.20: 9.20: 28.60. Soil with large amounts of sand could be well washed (USEPA, 1993). The soil pH was 3.57 which was deemed to be extremely acidic (FES, 2005). For ion exchange, the cation was 3.3 centimole/kilogram ($cmol_{(c)}kg^{-1}$) and conductivity was 0.37 decisemen/meter (dS/m) while organic matter in the soil accounted for 0.6% which was regarded as soil with organic matter as normal, since good soil should contain more than 1.5% organic matter (Yongyut, 2000). Concentration of all chromium in the soil was 466.4 milligram/kilogram which considerably exceeded soil quality standards for residential and agricultural purposes.

Table1: Physical and chemical properties of soil

Soil properties	Values measured
Sand (%)	65.20
Silt (%)	9.20
Clay (%)	28.60
Soil texture	Sandy clay loam
Soil pH	3.57
Nitrogen (%)	0.03
Phosphorus (mg/kg)	8.00
Potassium (mg/kg)	564.00
Cation exchange capacity ($cmol_{(c)}kg^{-1}$)	3.30
Electrical conductivity (dS/m)	0.37
Organic matter (%)	0.60
Total chromium (mg/kg)	466.40

Removal of Chromium with EDTA

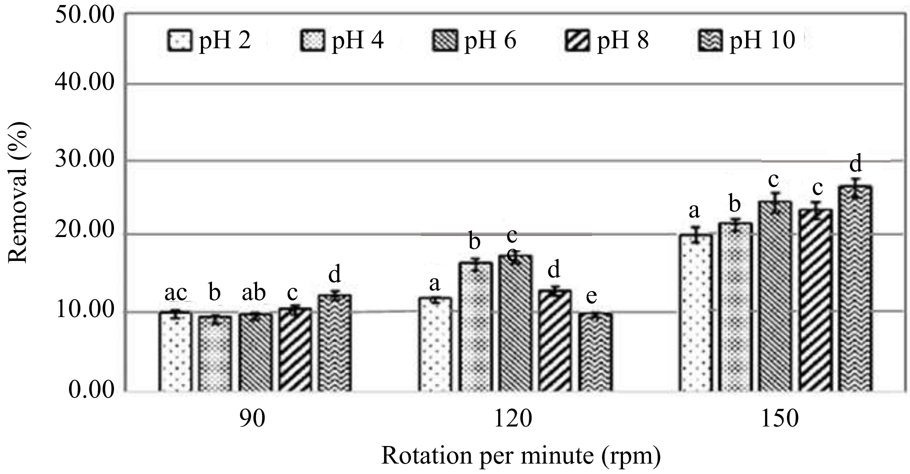
The experimental result on the efficiency of chromium-contaminated soil washing showed that the proper time for soil washing or the time of equilibrium was 30 min. Therefore, reporting the findings would take into account only these times. Nevertheless, the results on studying the chelating agent EDTA indicated that a concentration of 8 millimoles and pH of 10 provided the best condition for soil washing which was statistically significantly different from pH at 2, 4, 6 and 8 ($p \le 0.05$) at shaking speeds of 90 and150 rpm with the percentages of chromium removal at 12.77 and 26.84%, or 59.56±0.45 and 125.18±0.66 milligram/kilogram, respectively. This result also confirmed the findings of Zou *et al*. (2009) who reported that consecutive extractions using low concentrations were more effective than a single extraction with concentrated EDTA if the same dose of EDTA was used. Meanwhile, at a shaking speed of 120 rpm and pH of 6, the percentage of chromium removal was 17.91% as shown in Fig. 1a while at a concentration of EDTA at 16 millimoles, the findings revealed that at every shaking speed with pH 10, soil could be best washed, which was significantly distinct from other pH values ($p \le 0.05$) and the percentages of chromium removal were 16.93, 26.76 and 35.57% or 78.96±0.54, 124.81±0.98 and 165.9±0.85 milligram/kilogram, respectively, from high to low shaking speeds as shown in Fig. 1b. In addition, at an EDTA concentration of 32 millimoles, the findings indicated that at every shaking speed with pH 10, the soil could be washed best and it was statistically significantly different from other pH values ($p \le 0.05$). At shaking speeds of 90,120 and150 rpm, the percentages of chromium removal were 17.71, 38.5% and 35.8% or 82.6±0.43, 179.75±0.37 and 166.97±0.59 milligram/kilogram as shown in Fig. 1c (Dave *et al*., 2011). Normally, solutions in acid condition can leach heavy metals better and more effectively than those in alkalinity. Chromium-contaminated soil washing with EDTA solution will result in reaction of chromium complex compounds ($C_{10}H_{13}CrN_2O_8$) affecting chromium solubility. Nevertheless, when adjusted to be in acidity, the chromium complex compounds ($C_{10}H_{13}CrN_2O_8$) will turn to be $Cr(OH)_3$ with decreased solubility. In the meantime, when pH is increased or in alkalinity, the chromium complex compounds ($C_{10}H_{13}CrN_2O_8$) will turn to be CrO_4^{2-} with increasing solubility (Bibhabasu and Amit, 2011; Kawalpreet and Michelle, 2014). Furthermore, according to the outcome of the soil washing experiment with EDTA, the efficiency in removing chromium was at an improper or ineffective level because chromium has the anion structure as shown in Equation 1:

$$2CrO_4^{2-} + 2H^+ \leftrightarrow 2HCrO_4^- \leftrightarrow Cr_2O_7^{2-} + H_2O \qquad (1)$$

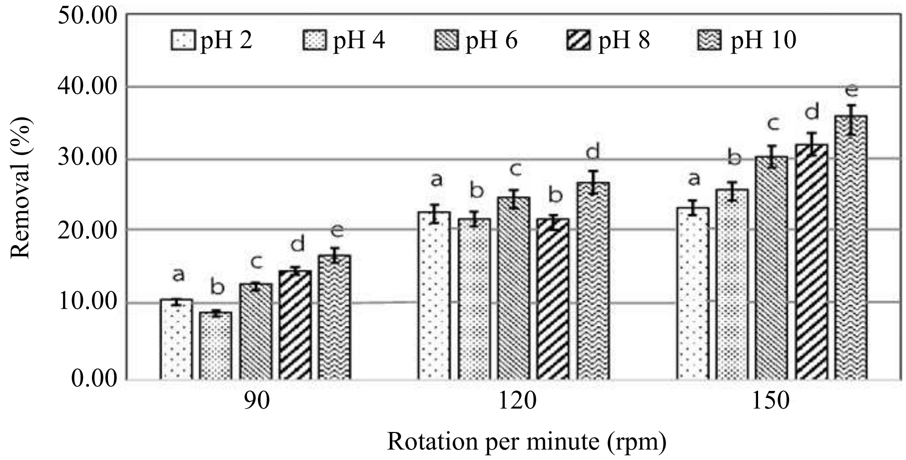
However, while chromium solution could be in EDTA in anion form, it could not be captured with the structure of EDTA; thus, this experiment was consistent with Pichtel and Pichtel (2009) who measured chelating agents of EDTA and NTA and the anion surfactant of SDS. They found that EDTA was more efficient than NTA in eliminating lead and chromium. In addition, efficiency would be enhanced when the concentration of the chelating agents increased; at 0.1 mol concentration of EDTA, it could remove lead and chromium accounting for 96.2 and 84% respectively at pH 12. In the meantime, SDS could remove lead and chromium at 30-40.5 and 29.35% respectively (Salehian *et al*., 2012). Additionally, the findings correspond to those of Lim *et al*. (2004) who examined the use of nitric acid and the chelating agent EDTA in washing heavy metals contaminating soil; EDTA was a composition that had efficiency in removing lead and cadmium in soil with a high pH exceeding that of nitric acid. The findings also proved that only 15 min of shaking time could remove heavy metals contaminating soil. Hence, removing chromium with a chelating agent may have low efficiency and the concentration of the chelating agent may be increased for more effective chromium removal. This result also corresponds to that of Sun *et al*. (2001) who suggest that the lability of metals in soil, the kinetics of metal desorption/dissolution and the mode of EDTA addition were the main factors controlling the behavior of metal leaching with EDTA.

Removal of Chromium with TWEEN 80

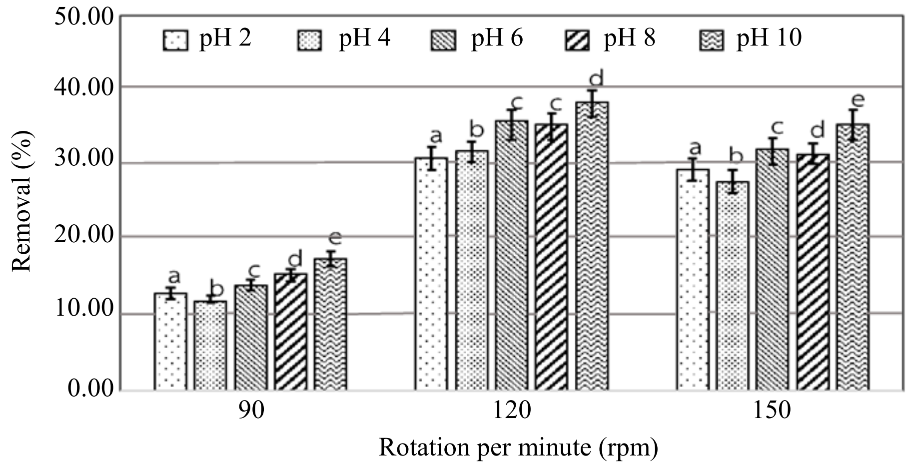
According to the experiment on the efficiency of chromium contaminated soil washing at 30 min, which was the substance equilibrium point with the surfactant TWEEN 80 at 8 millimoles concentration at its lowest, a shaking speed at 120 and 150 rpm and pH of 10 was the condition which removed chromium with the highest efficiency and this was different from other pH values with statistical significance ($p \le 0.05$) as shown in Fig. 2a. The result was similar to that of Oudghiri *et al*. (2015) who used the chelating agent to remediate the sediment. The result promoted the notion that the addition of EDTA with pH 8 causes more positive outcomes than pH 3.8. The percentages of chromium removal were 3.48 and 9.67% or 16.23±0.61 and 45.1±0.34 milligram/kilogram, respectively. However, chromium was mostly eliminated at a shaking speed of 90 rpm, pH 2, accounting for 5.91% or 27.56±0.83 milligram/kilogram. Meanwhile, at 16 millimoles concentration, 90, 120 and 150 rpm shaking speed and pH 10 of TWEEN 80, soil could be best washed which was statistically significantly different from other pH values ($p \le 0.05$) with chromium removal percentages of 5.98, 4.31 and 9.79% or 27.89±0.96, 20.10±0.94 and 45.66±0.43 milligram/kilogram, respectively (Fig. 2b).



(a)



(b)



(c)

Fig. 1: Removal of chromium using EDTA at a concentration level, (a) 8 mM (b) 16 mM and (c) 32 mM; **Note:** The different English characters represent the statistical difference 95 percent confidence interval between different sets of the experiments using One Way ANOVA and to compare the difference in data using Duncan's new Multiple Range Test (DMRT)

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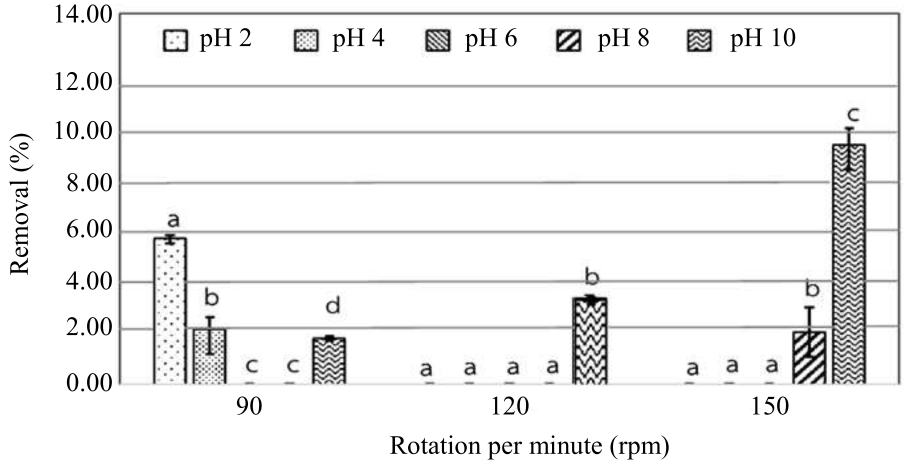
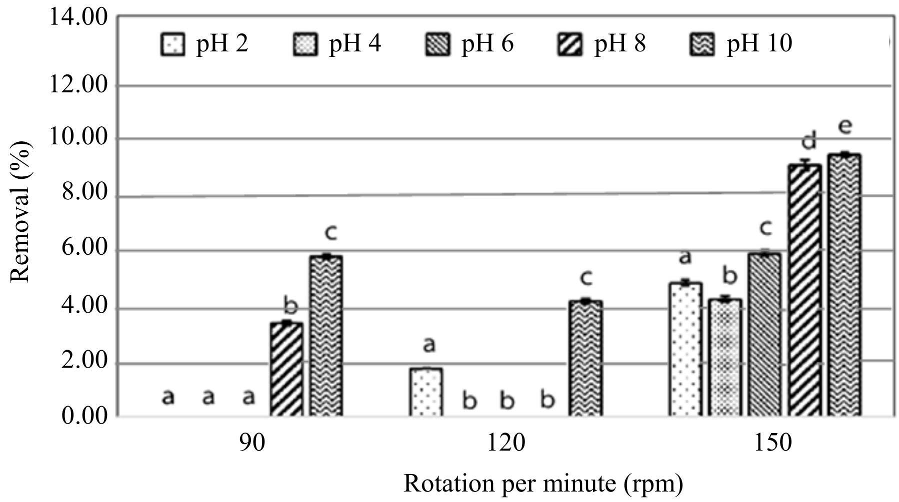
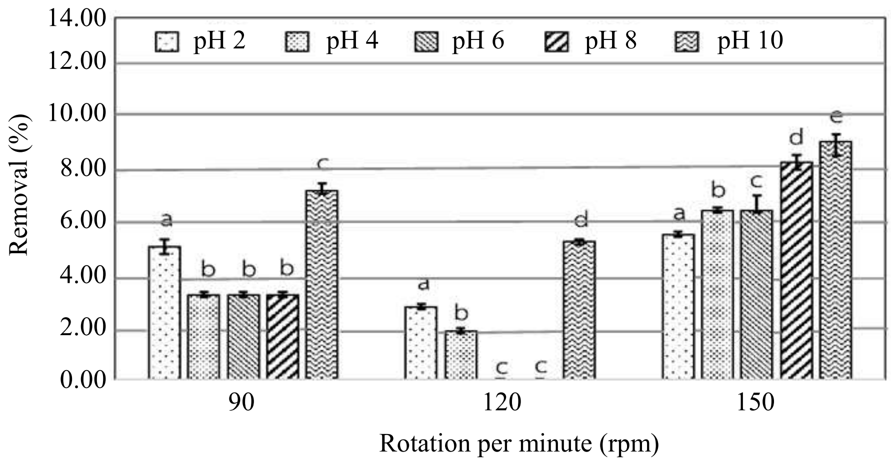


(a)



(b)



(c)

Fig. 2: Removal of chromium using TWEEN 80 at a concentration level, (a) 8 mM (b) 16 mM and (c) 32 mM; Note: The different English characters represent the statistical difference 95 percent confidence interval between different sets of the experiments using One Way ANOVA and to compare the difference in data using Duncan's new

Moreover, at 32 millimoles concentration of TWEEN 80, which was the highest concentration, the findings indicated that at shaking speeds of 90, 120 and 150 rpm and pH 10, soil could be best washed which was statistically significantly different from other pH values ($p \leq 0.05$) with chromium removal percentages of 7.79, 5.69 and 9.86% or 36.33±0.53, 26.54±0.98 and 45.98±0.19 milligram/kilogram (Fig. 2c). Furthermore, this study also discovered that the efficiency of chromium removal with TWEEN 80 was substantially lower due to the surfactant which reduced the surface tension in the form of an electric charge (Xuhui *et al*., 2015). When dissolved in water, cations would be decomposed resulting in the chromium being impossible to remove because of this reason. This aligned with Villa *et al*. (2010) who reported the use of TX-100 surfactant which is a nonionic agent which, when changing concentration of TX-100 from 3 to 12 cmc, could remove chromium from 0.12 to 0.14 milligram/liter and was not statistically significantly different. Tween 80 is a nonionic surfactant. According to the study of Bloor *et al*. (2006) examining effect of acidity-alkalinity on quality of Nonionic Surfactant Micellar, the findings revealed that the increase of pH in alkalinity possibly affects size of Nonionic Surfactant Micellar and it is likely to become smaller when being in high alkalinity. In addition, Neelam and Mamta (2008) performed an experiment and discovered that size of Micellar influenced aqueous solubility provided that minute Micellar will be more efficient in solubility than larger ones. Nevertheless, it is because small Micella contains more surfaces than bigger Micellar. The findings conformed to this study that high pH or alkalinity enhances efficiency of soil washing.

Comparison of Efficiency for the Removal of Chromium between EDTA and TWEEN 80

Comparing EDTA and TWEEN 80 at every concentration, at shaking speeds of 90, 120 and 150 rpm, the researcher found that EDTA could remove chromium at every pH value because EDTA could be combusted with heavy metals in a wide range of pH values. It could eliminate chromium best at pH 10 accounting for 38.54% at 32 millimoles concentration of EDTA and shaking speed of 120 rpm. In the meantime, TWEEN 80 could remove chromium best at pH 10. For other pH Values, it could eliminate little or no chromium; moreover, TWEEN 80 could remove chromium at most at only 9.86% at 32 millimoles concentration and shaking speed of150 rpm. According to the findings, chromium removal with EDTA and TWEEN 80 had low efficiency; however, EDTA could eliminate chromium 33.91 times or 20% more than TWEEN 80. Thus, comparing efficiency between EDTA and TWEEN 80, the researcher would say that EDTA could remove chromium better than TWEEN 80; in other words, TWEEN 80 was not powerful enough in eliminating chromium with the washing method. This finding concurred with Khalil *et al*. (2015) who found the capability of EDTA in the removal of copper to be better than SDS. Moreover, Luis *et al*. (2012) found that soil washing to remove arsenic and zinc using TWEEN 80 could remove 42.6 and 85.4%, respectively. These results were similar to the reports of Metka and Domen (2009) and Abumaizar and Smith (1999), who stated that EDTA addition can cause positive effects in terms of heavy metal removal with the soil washing process. The results were similar to those of Gitipour *et al*. (2011) who studied the effects of soil washing on the removal of chromium and cadmium contaminated sludge from oil refinery ponds. The sediment samples were collected from various sources in the pond and the sediment was washed with EDTA and hydrochloric acid. The results showed that the addition of 0.3 MHCl and EDTA 0.1 M have a positive effect on chromium and cadmium removal from soil, reaching levels of more than 70%.

Conclusion

This study concluded that the chelating agent EDTA could be used for soil washing with more efficiency in chromium removal than the surfactant TWEEN 80, which barely eliminated chromium. The best concentration for EDTA and TWEEN 80 was 32 millimoles and the best pH value was 10. Shaking speed slightly affected soil washing compared with concentration and pH at the same level. Nevertheless, since Chromium has different valences, such as Chromium trivalent (Cr^{3+}) and Chromium hexavalent (Cr^{6+}) which could have oxidized or reduced in natural conditions, EDTA had low efficiency in removing chromium and TWEEN 8 had no efficiency in eliminating chromium. However, to enhance the efficiency of chromium removal with the washing method there should be further studies on other types of chelating agents and surfactants with cation, anion, or no charge and studies on washing soil contaminated with different heavy metals or the use of both substances in simultaneous or alternating ways which may provide the enhancement of heavy metal removal using soil washing methods. Furthermore, future studies should investigate functional groups of chromium by employing Fourier-Transform Infrared Spectroscopy (FTIR) in order to explain mechanism of reaction or bonding between chromium with complex compounds and soil surfactants.

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Author's Contributions

This article is original research paper. Authors participated in all experiments, coordinated the data-analysis and contributed to the written and read of this manuscript. Authors give final approval of the version to be submitted this journal.

Ethics

The authors declare no conflicts of interest and confirm that the manuscript has been submitted solely to this journal and is not published, in press, or submitted elsewhere.

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