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Effects of operating parameters on the performance of washing–electrokinetic two stage process as soil remediation method for lead removal

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A B S T R A C T

The application of washing–electrokinetic two stage process on remediation of lead (Pb) contaminated soil was investigated. The process consisted of an initial soil washing, followed by an electrokinetic process. The use of electrokinetic process in soil washing could provide additional driving force for transporting the desorbed Pb away from the soil even in the absence of pore flow. Thus, high usage of wash solution may be mitigated. In this study, the effect of operating conditions such as electric potential difference, wash solution concentration and initial Pb concentration on the feasibility of washing–electrokinetic two stage process on Pb removal was investigated using response surface methodology based on Box–Behnken design. The wash solution used was citric acid and three main aspects were examined, namely: (i) removal efficiency, (ii) effluent generated, and (iii) power consumption. The results revealed that the increase in electric potential difference and wash solution concentration generally enhanced Pb removal efficiency and the interactions of these parameters were significantly positive. However, undesirable high effluent generation and power consumption were also caused by these increments. Optimisation study revealed that 84.14% removal efficiency with zero effluent generation and a power consumption of 2.27 kW h/kg Pb removed could be achieved at 7.58 V and 0.057 M citric acid concentration. In comparison with normal soil washing, washing–electrokinetic two stage process showed an enhancement in removal efficiency by ≈16% via electromigration under optimum conditions using similar solution: soil ratio of <0.8 mL:1 g soil. The study reveals that incorporation of electrokinetic process in soil washing is feasible as it not only enhances Pb removal efficiency at minimum wash solution usage with respect to normal soil washing, but also provides *in situ* Pb recovery in cathode chamber via electrodeposition.

1. Introduction

Lead (Pb) is a highly toxic substance to living organisms. Its ability to bio-accumulate often causes acute and chronic illnesses which damage human body systems when it is inhaled and ingested [1,2]. Pb has been extensively used as a raw material in manufacturing processes such as ammunition, batteries, bearings, plumbing, ceramic glazes, weights, caulks, dyes, pigments and pesticides [2,3]. In order to support huge demand for Pb in the world, global Pb production as high as 10,654,000 tonnes was reported in 2012. As a result of its huge production and usage, humans can be easily exposed to Pb via variety pathways. Soil contamination is one of the pathways for Pb exposure especially from industrial lands with the activities like battery manufacturing, gould casing, scrap Pb handling [4] as well as Pb smelting and mining [1,2]. High Pb concentration in the range of 751.98–138,000 mg/kg was reported in the soil from these industries [3,5–8]. Thus, a proper treatment to these soils is necessary.

Soil washing has been reported as one of the effective soil remediation methods for removing Pb and heavy metals [9]. However, it is worth noting that soil washing requires high amount of wash solution for effective treatment. A high solution: soil ratio of 3.33–20 mL:1 g [10–12] shows the disadvantage of applying soil washing as the amount of spent wash solution that requires post treatment is large. In contrast, electrokinetic process could reduce the amount of wash solution needed in soil remediation.

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Electrokinetic process is one of the soil remediation methods that show great potential to remove organic compounds and heavy metals [13–15]. This process is conducted by inducing low magnitude direct current through the soil as the driving force for contaminants removal [13,15,16]. For metal ions removal, electro migration is the main mechanism for the transport in the soil under electric field influence towards their respective electrodes. It is worth noting that electromigration is independent of the pore fluid movement [13]. This suggests that electrokinetic process has potential to be incorporated into soil washing to reduce the wash solution usage in the remediation of high permeability soil.

Electrokinetic process has been applied in the treatment of soils with high permeability [15,17–21]. The work of Kim et al. [19] in treating Co and Cs contaminated sandy soil showed that electrokinetic process not only provided favourable removal efficiency but also lower effluent generation which was only 5% of the effluent generated by soil washing. Furthermore, Kim et al. [19,20] reported that further enhancement in the removal efficiencies of Co and Cs were achieved when electrokinetic process was incorporated into soil flushing. However, this was found to increase the effluent generation slightly in comparison with normal electrokinetic process. Recently, a study on the incorporation of electrokinetic process into soil washing as a washing–electrokinetic two stage process (also known as Two-Stage Electrokinetic Washing) for Pb removal from sandy soil has been investigated [22]. This process of soil remediation method which consisted of: (i) initial soil washing, and (ii) electrokinetic process was conducted at different stages in a single equipment, as shown in Fig. 1 [22]. The process was initiated by filling up the anode chamber with wash solution such as $NaNO_3$, HNO_3, citric acid and EDTA while the cathode chamber was left empty. Due to hydraulic gradient difference between the chambers, soil washing occurred via the diffusion and advection of wash solution from anode chamber to the cathode chamber through the soil column. This provides a "flushing" effect for Pb desorption and transport to the cathode chamber during wash solution filling process. When the cathode chamber was completely filled up, soil washing stopped, as shown in Fig. 1. Then, a constant voltage was applied through the soil as second stage of the treatment by providing electrical driving forces to further transport Pb away from the soil while preventing local concentration polarisation for Pb desorption. Previous work showed that washing–electrokinetic two stage process enhances soil remediation efficiency in comparison to soil washing [22]. Among the solutions investigated, citric acid was identified as the best wash solution as it had provided high enhancement in Pb removal efficiency mainly via electromigration even at low wash solution consumption of <1 mL:1 g. This eliminated the disadvantage of soil washing having high wash solution consumption. However, it is worth noting that significant volume of effluent was still generated via electroosmosis, which is not desirable. Hence, in order to limit the electroosmosis while maintaining high removal efficiency, the effects of operating parameters such as electric potential difference and wash solution concentration must be evaluated.

The main goal of the present study is to investigate the effect of operating parameters such as electric potential difference, wash solution concentration and initial Pb concentration on the performance of washing–electrokinetic two stage process as soil remediation method for Pb removal. Unlike most of the studies for electrokinetic process, this study was conducted using statistical response surface methodology based on Box–Behnken Design such that the effects of the parameters and their mutual interactions can be adequately analysed. The performance of the system was evaluated for: (i) removal efficiency, (ii) effluent generated, and (iii) power consumption. Finally, an optimisation test was conducted to investigate the possibility of washing–electrokinetic two stage process to obtain high removal efficiency at negligible effluent generation and low power consumption.

2. Materials and methods

2.1. Chemicals and soil

All the chemicals used in this study were of analytical grade and were supplied by R&M Chemicals, Malaysia. The soil used was taken from Hulu Langat, Malaysia and was sieved to a particle size of <0.85 mm. The soil was classified as sandy soil according to USDA Soil Classification as it had 92% sand content, with 8% silt and clay. General properties of uncontaminated soil are as shown in Table 1. The soil had high iron (Fe) and mineral content and posed potential risk for Pb contamination via adsorption with a maximum contamination level of 1000 mg/kg [23], which is higher than the regulatory limit in Malaysia [24]. In this study, artificially contaminated soil was prepared by spiking the soil with $Pb(NO_3)_2$ solution to create Pb contaminated soil with desired contamination level. The slurry was then homogenised using spatula and dried for one week. The contaminated soil was then stored in a dark place.

2.2. Response surface methodology

Response surface methodology (RSM) was used in this study to evaluate the effect of operating parameters on Pb removal in a washing–electrokinetic two stage process. RSM is a collection of mathematical and statistical techniques that are useful for modelling and analysis of problems for which a response on outcome is influenced by several variables and the objective of RSM is to optimise this response [25]. The use of RSM and Analysis of Variance (ANOVA) could give a suitable approximation for the true functional relationship between the response and the set of independent variables. In this study, a polynomial equation as shown in Eq. (1) was generated, where y is the response, β is the regression coefficient, and x is the independent parameter [25].

$$y = \beta_0 + \sum_{i=1}^{3}\beta_i x_i + \sum_{i=1}^{3}\beta_{ii} x_i^2 + \sum_{i=1}^{2}\sum_{j=i+1}^{3}\beta_{ij} x_i x_j \tag{1}$$

Box–Behnken Design was used in this study in combination with the Design Expert software. This design is a spherical three level design by combining 2^k factorial with incomplete block design and it is most efficient in terms of number of required runs in comparison to Central Composite Design [25]. Three parameters were investigated, namely electric potential difference (A), wash solution concentration (B) and initial Pb concentration (C). The levels of each parameter were coded as -1, 0 and 1 and the ranges of the parameters are shown in Table 2. The responses investigated in this study were (i) removal efficiency, (ii) effluent generated, and (iii) power consumption. Based on Box–Behnken design, 17 experiments were conducted with five replicates of centre point experiments to estimate pure error for the models generated. The experiments were conducted in a random sequence to improve the precision of the experiments. The results obtained were then analysed using ANOVA and F-test with 95% confidence level. The numerical models generated from this method were used to analyse the importance of the parameters and their interaction effects. Finally, optimisation of these parameters was conducted based on the mathematical models generated.

2.3. Experimental

Fig. 2 illustrates the schematic diagram for washing–electrokinetic two stage process. A PTFE soil column with a dimension of

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