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Removal of heavy metal from sludge by the combined application of a biodegradable biosurfactant and complexing agent in enhanced electrokinetic treatment

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HIGHLIGHTS

- The combined biosurfactant and complexing agent were used to enhance heavy metals removal in the EK treatment.
- GLDA and rhamnolipid could produce higher electric current and conductivity in the EK treatment.
- The GLDA obtained higher pH at the anode, the rhamnolipid obtained lower pH at the cathode.
- The combined chelating agent and biosurfactant to result in better heavy metals removal efficiencies.
- Exchangeable and reducible fractions of heavy metals had an apparent reduction.

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ABSTRACT

In this work, the heavy metal removal potentiality of an electrokinetic (EK) decontamination treatment enhanced by a biodegradable complexing agent Tetrasodium of N, *N*-bis (carboxymethyl) glutamic acid (GLDA) also in combination with a biodegradable biosurfactant (rhamnolipid) was investigated to decontaminate heavy metals from the sludge. The main results explored that the nature of sludge and their interactions with different improving agents significantly influenced the electrokinetic removal processes. A general increase of pH values from anode to cathode in the sludge-cell was observed due to the strong buffering capacity of carbonates. Compared with the deionized water, the use of GLDA as an electrolyte, Cu, Zn, Cr, Pb, Ni and Mn removal efficiencies were 53.2 ± 3.12%, 67.4 ± 3.45%, 59.2 ± 4.78%, 45.4 ± 4.15%, 72.8 ± 3.68% and 45.0 ± 4.85%, respectively, whereas a further improvement heavy metals removal efficiencies (Cu, Zn, Cr, Pb, Ni and Mn removal efficiencies were 64.8 ± 2.34%, 56.8 ± 4.12%, 49.4 ± 4.45%, 46.6 ± 2.35%, 60.4 ± 3.45% and 69.6 ± 3.54%, respectively) were achieved by repalcing rhamnolipid as the electrolyte. Significantly higher removal efficiencies (Cu, Zn, Cr, Pb, Ni and Mn removal efficiencies were 70.6 ± 3.41%, 82.2 ± 5.21%, 89.0 ± 3.34%, 60.0 ± 4.67%, 88.4 ± 4.43% and 70.0 ± 3.51%, respectively) were obtained by the simultaneous use of GLDA and rhamnolipid due to their synergic action in electrokinetic process.

1. Introduction

The application of municipal sludge to agricultural land is implemented worldwide, and it is an effective sludge disposal technique that has benefit of adding organic matter and plant nutrients (nitrogen, phosphorus and potassium) to the soil (Zhu et al., 2015). However, this treatment method may give rise to a potential threat to the environment (Wu et al., 2015a). Because the sludge contains the potentially high heavy metal content, an environmental problem that may be aggravated if toxic metals (Gao et al., 2013a,b). Heavy metals are transferred in the soil and then taken up by plants or migrated in the drainage waters, leading to their accumulation in the environments due to their high environmental persistence and non-biodegradability (Suanon et al., 2015). Heavy metals accumulation may happen in the natural environment and human tissues, such as liver, kidney and spleen (Gao et al., 2013b; Ebbers et al., 2015). The bio-magnification through the food chain

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damaged human health and produced environmental problem (Gao et al., 2013a,b). A significant way for solving the problem is to extract heavy metals from sludge. Hence, many methods such as bioleaching, chemical extraction, electrokinetic, ultrasound-assisted citric acid and super critical fluid extraction have been used by extracted heavy metals from sludge (Shi et al., 2013; Wang et al., 2015; Zhu et al., 2015).

Compared with other techniques, EK technology contains the application of a low direct current (0–1 A) or a low direct electric-voltage gradient (0–2 V cm^{-1}) through electrodes inserted into the sludge to move contaminants towards the electrodes (Fu et al., 2017). The mobilization of heavy metal ions through the media towards the electrodes. Because the four main mobilization mechanisms: (1) electroosmosis, (2) electromigration, and (3) electrophoresis, (4) coupled with electrolysis and geochemical reactions (Zhu et al., 2015; Fu et al., 2017). Heavy metal ions are dominantly mobilized by electromigration, which is the migration of heavy metal ions an oppositely charged electrode (Chowdhury et al., 2017; Fu et al., 2017). In the EK process, water at the anode is primarily oxidized ($2H_2O \rightarrow 4H^+ + 4e^- + O_2$), undergoing a reduction reaction ($2H_2O + 2e^- \rightarrow H_2 + 2OH^-$) at the cathode (Mena et al., 2015). Among these reactions, the electrolysis of water is the main reaction, then H^+ and OH^- are mobilized through the sludge via electrolysis, and that changes the pH values through a sludge level during electrokinetic process. In the EK process, sludge acidification may happen at low pH zone, which can lead to heavy metal ions desorption from the sludge. Conversely, sludge alkalization may happen at the high pH zone, which can result in heavy metal ions precipitation surface or inside the sludge (Peng et al., 2011; Chowdhury et al., 2017; Fu et al., 2017). In order to enhance the effectiveness of the process in terms of removing heavy metals from sludge, previous studies have conducted different applications such as conditioning the catholyte pH, adding multilayer of anion exchange resin, using catholyte circulation to control anolyte pH, using ion selective membranes (Peng et al., 2011; Gao et al., 2013a; Cherifi et al., 2016). Therefore, the suitable enhancing chemical agents can promote the heavy metal removal effectiveness, which then reduces the remediation costs and time.

For the synthetic chelates, commonly complex agents have been applied to extracted heavy metals from contaminated soils, sediments and sludges in washing treatment or as enhancing agents in EK decontamination (Kulikowska et al., 2015; Suanon et al., 2015; Villen-Guzman et al., 2015). These complex agents have a high effective chelating agent for heavy metal decontamination. However, the traditional chelators and complexants are costly and have a high environmental persistence, toxicity and solubility in the soils, sediments and sludges. Therefore, searching for the environmentally friendly, economically viable alternatives is imperative (Wu et al., 2015a; Falciglia et al., 2016). The tetrasodium of N, *N*-bis (carboxymethyl) glutamic acid (GLDA) has been introduced as a biodegradable chelant, which shows high chelating capacity towards a plethora of heavy metal ions, it also the extraction possesses outstanding biodegradability, with more than 60% of GLDA degraded within 28 days (Wu et al., 2015b).

Biosurfactants are bioavailable surface-active compounds mainly produced by bacteria, fungi and yeast. In comparison with synthetic surfactants, the biosurfactants often have more functional groups and larger molecular structure, environmental compatibility characteristic, a low toxicity and favorable biodegradability, which facilitate the biodegradation of contaminants and promote the self-degradation of biosurfactants (Chen et al., 2015; Ye et al., 2016). Consequently, biosurfactants can effectively solubilize, disperse and desorb both organics and heavy metals (Cao et al., 2013). Especially the rhamnolipid is a representative biosurfactant, which has low toxicity and critical micelles concentration (CMC) as well as relatively high solubility and compatibility, is considered as a promising and cost-effective solution (Mao et al., 2015).

Although many previous studies have been conducted on EK remediation of heavy metals, to the best of our knowledge, both GLDA and rhamnolipid were application as an electrolyte to enhance removal heavy metals from sludge in the EK process has not been explored. Moreover, the heavy metals removal efficiencies and fractions variation during the EK process were rarely investigated, and lacking of studies on the evolution of pH over time and the factors affecting pH during the EK process. The objective of this study is to supplement the research in these areas. GLDA and rhamnolipid were selected as electrolytes, which shared the similar biodegrability and compatibility, and hold the potential to be used as excellent electrolytes in EK remediation.

This study aims to investigate the influences of these electrolytes (GLDA, rhamnolipid, the combined GLDA and rhamnolipid) on EK sludge remediation. The specific tasks are (1) the factors affecting electrolyte and soil pH, which would be conducive to control the proper pH values during the EK process and enhance the heavy metals removal in the further studies. (2) heavy metals removal from sludge by electrokinetic with different electrolytes were investigated and the effects of size of the electrokinetic cell on EK remediation was compared, and the combined biosurfactant and complexing agent is conducive to improve heavy metals from dewatered sludge. (3) the heavy metals fractions variations in the different sections also investigated during the EK process, which can help us better elucidate heavy metals removal from the sludge under the different electrolytes.

2. Materials and methods

2.1. Sludge sample

Sewage sludge sample was collected from the Qun Li Urban Wastewater Treatment Plant (Harbin, China), which treats both industrial and domestic wastewater with domestic wastewater accounting for 60% of total wastewater. The collected sludge sample was divided into two parts. One part of sample was dried in an oven at 105 °C for 24 h, followed by grinding and sieving to a #100 (0.149 mm), then was analysed the sludge properties, and main characteristics of the sludge samples are summarized in Table 1. The other parts of sludge samples were air-dried, and then followed by grinding and sieving to a #100 (0.149 mm) before EK experiments, then the treated sludge sample was collected and stored in desiccators at room temperature.

Table 1 indicated heavy metals concentrations of the sludge sample collected after detailed characterization. The Zn and Cu concentrations were 731.58 ± 8.98 and 1352.11 ± 11.21 mg kg^{-1}, respectively, both of which exceed the environmental quality standard for soils and the control standards for pollutants in sludge from agricultural use. Consider the heavy metal accumulation in the environment, therefore, the six heavy metals (Cu, Zn, Cr, Pb, Ni, Mn) were investigated in this work.

2.2. GLDA and rhamnolipid characteristics

Tetrasodium of N, *N*-bis (carboxymethyl) glutamic acid (GLDA) was purchased from Akzo Nobel Chemicals Co., Ltd., Shanghai, China. It has a relative molecular mass of 351.1, solid content is more than 47.00%, the chemical agent exhibits good solubility in aqueous solutions over a wide range of pH value. The structure of GLDA can be depicted in Fig. S1a (Kołodyńska, 2013).

The rhamnolipid (purity over 85%) was purchased from Daqing Victex chemical Co., Ltd and used in this experiments, which was