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Combination of bioleaching by gross bacterial biosurfactants and flocculation: A potential remediation for the heavy metal contaminated soils

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HIGHLIGHTS

- Combining bioleaching-flocculation effectively treated heavily metal-polluted soil.
- Removal metals by the gross biosurfactants were 2 times of that by 0.1% of rhamnolipid.
- The gross biosurfactants transformed metals to more easily migrating speciation.
- Contents of metals in the used bio-leachate treated by PAC met limits of GB8978-1996.

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ABSTRACT

Combining bioleaching by the gross biosurfactants of *Burkholderia* sp. Z-90 and flocculation by poly aluminium chloride (PAC) was proposed to develop a potential environment-friendly and cost-effective technique to remediate the severely contaminated soils by heavy metals. The factors affecting soil bioleaching by the gross biosurfactants of *Burkholderia* sp. Z-90 were optimized. The results showed the optimal removing efficiencies of Zn, Pb, Mn, Cd, Cu, and As by the *Burkholderia* sp. Z-90 leachate were 44.0, 32.5, 52.2, 37.7, 24.1 and 31.6%, respectively at soil liquid ratio of 1:20 (*w/v*) for 5 d, which were more efficient than that by 0.1% of rhamnolipid. The amounts of the bioleached heavy metals by the *Burkholderia* sp. Z-90 leachate were higher than that by other biosurfactants in the previous studies, although the removal efficiencies of the metals by the leachate were relatively lower. It was suggested that more heavy metals caused more competitive to chelate with function groups of the gross biosurfactants and the metal removal efficiencies by biosurfactants in natural soils were lower than in the artificially contaminated soils. Moreover, the *Burkholderia* sp. Z-90 leachate facilitated the metals to be transformed to the easily migrating speciation fractions. Additional, the results showed that PAC was efficient in the following flocculation to remove heavy metals in the waste bio-leachates. Our study will provide support for developing a bioleaching technique model to remediate the soils extremely contaminated by heavy metals.

1. Introduction

Contamination of heavy metals in soils has been a serious environmental issue in the last decades (Wu et al., 2010; Zhang et al., 2013; Liu et al., 2017; Yang et al., 2017). It is primarily caused by anthropogenic activities such as mining, smelting, electroplating, irrigation with wastewater and fertilizer abuse (Tai et al., 2013; Bolan et al., 2014; Chai et al., 2017a; Yang et al., 2018). The elevated heavy metals are detrimental due to their biological virulence, chemical or biological non-degradation and difficult separation in soils (Wu et al., 2010). Therefore, it is a huge challenge to effectively remove hazardous heavy metals from soils.

As one of remediation technologies to remove heavy metal

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pollutants from soils, leaching has been intensively studied in recent years (Mulligan et al., 2001b). Compared with other methods, soil leaching is an alternative method due to its simplicity in operation, low cost and high efficiency (Jang et al., 2007; Wang et al., 2014). Many chemicals including inorganic compounds, chelators and surfactants have been used to extract heavy metals from soils (Mulligan, 2005; Kuo et al., 2006; Makino et al., 2006). Among these agents, EDTA is widely utilized to remove heavy metals from soils (Zou et al., 2009; Voglar and Lestan, 2012; Zhang et al., 2014). However, its high cost and poor biodegradability easily to cause secondary pollution in the treated soil limit its application (Lan et al., 2013; Wang et al., 2014). Therefore, searching for environment-friendly, biodegradable and cost-effective agents becomes one of critical steps in remediation on the metal-contaminated soil by leaching.

The valence of +2 of heavy metals (such as Zn, Pb, Mn, Cd and Cu), and As(III) and As(V) of As are dominant in soils of which the elevated are toxic to indigenous organisms, although they have many oxidation states (Tchounwou et al., 2012; Yang et al., 2015; Chai et al., 2018). One of the main detoxification strategies to the toxic metals of the indigenous organisms is to remove the metals from their adjacent soil micro-environment (Nies, 2003; Olaniran et al., 2013). Biosurfactants exhibit pronounced surface activity, major of which are polysaccharides, lipids, lipopeptides, glycolipids and neutral lipid derivatives produced by microorganisms, plants or animals (Soubes and Bonilla, 2001). Bioleaching by biosurfactants mainly aims to remove metals from soils by chelating metals with their function groups and transforming metals' speciation fractions to increase their mobility (Gadd Geoffrey et al., 2011; Mao et al., 2015).

Biosurfactant, an alternative biodegradable leaching agent, has received increasing attention. Many studies have demonstrated the potential of rhamnolipid on heavy metal removal in the contaminated soils (Mao et al., 2015). For example, Juwarkar et al. (2007) investigated that 92% of Cd and 88% of Pb in the contaminated soil were removed by the purified rhamnolipid produced by *P. aeruginosa* BS2 in the column leaching experiments for 36 h. For another example, the batch washing experiments by Venkatesh and Vedaraman (2012) revealed that the removing efficiencies of Cu reached 71% and 74% by rhamnolipid in the contaminated soils with the initial concentrations 474 and 4484 ppm, respectively. But their high cost of preparing materials, difficulty of separation and purification restrict the large-scale application of biosurfactants (Pansiripat et al., 2010; Sarachat et al., 2010). Moreover, the separation and purification account for 60% of the total production cost (Desai and Desai, 1993; Sen and Swaminathan, 2005). Thus, it potentially facilitates application of biosurfactants to avoid the downstream processes of separation and purification.

Leaching by a biosurfactant is mainly transferring pollutants from the contaminated soil to the leachate. As a result, purification of leachate is another vital step in soil remediation by leaching. Flocculation is widely used to purify leachate because of its simple operation, high efficiency and low process cost (Lee et al., 2014; Suarez Meraz et al., 2016; Yang et al., 2016a). The main flocculants are inorganic (aluminium and iron salts), organic polymer (polyacrylamide (PAM)) and biological flocculants (Li and Yang, 2007; El-Gohary and Tawfik, 2009). El Samrani et al. (2008) showed that low dosages of $FeCl_3$ and poly aluminium chloride (PAC) can efficiently remove turbidity and heavy metals (Cu, Zn and Pb). Chen et al. (2016) revealed the removal efficiency of Cu increased significantly and then decreased with the increasing dosage of PAM.

A novel biosurfactant producing bacterial strain *Burkholderia* sp. Z-90 isolated from a sewer of cafeteria has been previously revealed that it has the efficient ability to remove heavy metals from the contaminated soil. The main mechanism of metal removal is possibly complexation with carboxyl groups and metals (Yang et al., 2016b). Here, to widely apply such bacterial strain, an environment-friendly and cost-effective strategy to combine bioleaching by its gross bacterial biosurfactants and flocculation was proposed. The feasibility of bioleaching metals from the severely contaminated soils was systematically investigated using the mixture of biosurfactants of the bacterial strain and the flocculation process by PAC which was adopted for the following treatment for the waste bio-leachate. Our study will benefit to develop the technique model combining bioleaching and flocculation process for remediation on the soils contaminated by heavy metals and provide support for full-scale application of the technique.

2. Materials and methods

2.1. Sampling and soil physiochemical properties analyses

The soil sample used in this study was collected from vicinity of a Pb-Zn smelting factory in Hengyang, Hunan, China (112°35′12.25″E-112°35′8.74″E, 26°35′38.71″N-26°35′42.17″N). The soils have been highly polluted with heavy metals and metalloids of mining and smelting activities for decades. The soils of 10–30 cm depth were collected and then mixed, air-dried, sieved through a 4.75 mm nylon mesh for the bioleaching experiment. The air-dried soils were also passed through a 0.15 mm nylon mesh for determining total contents of the heavy metals (Deng et al., 2012, 2013). 0.2000 g of the sampled soils was digested with acid mixture (5 mL of HCl, 5 mL of HNO_3, 2 mL of $HClO_4$ and 4 mL of HF), and the concentrations of Zn, Pb, Mn, Cd and Cu were determined by an inductively coupled plasma-optical emission spectrometer (ICP-OES, Perkin-Elmer Optima 5300 DV, MA, USA) (Yang et al., 2016b). The total content of As was determined by an atomic fluorescence spectrometry (AFS-810, Beijing Titan Instrument, Beijing, China) after digesting samples with HNO_3-HCl (1:3 *v/v*) in a boiling water bath (Chai et al., 2016). The concentrations of Zn, Pb, Mn, Cd, Cu and As were 22374 ± 500, 2472 ± 270, 1840 ± 110, 447 ± 14, 709 ± 20 and 562 ± 12 mg kg^{-1}, respectively, all of which exceed the limits for vegetation normally growing (level 3) of the environmental quality standard for soils (Ministry of Environmental Protection of the People's Republic of China, 1995). Especially, the total contents of Cd, Zn and As exceed 447, 44 and 14 times of that of the levels, respectively.

2.2. Preparation of Burkholderia sp. Z-90 leachate

The biosurfactant producing bacterial strain *Burkholderia* sp. Z-90 has previously been isolated and identified (Yang et al., 2016b). The strain was maintained in the LB medium (pH = 7, 10 g L^{-1} of tryptone, 5 g L^{-1} of yeast extract, 10 g L^{-1} of sodium chloride) with 0.4% volume of vegetable oil. The liquid medium was sterilized at 115 °C under 0.15 MPa for 30 min. And then 2 mL of strain suspension was inoculated in 198 mL of the modified LB liquid medium in a 500-mL shake flask. Flask was incubated on a gas bath shaker at 35 °C with a speed of 160 rpm for 5 d to obtain the leachate. The surface tension of the leachate was 27.5 mN/m using an automatic surface tensiometer (BZY-2, Shanghai Hengping Instrumen, Shanghai, China).

2.3. Soil bioleaching batch experiments

The influences of soil sterilization, soil solution ratio, solution pH and leaching time on the removal efficiencies of the six metals by *Burkholderia* sp. Z-90 leachate were individually investigated. The initial bioleaching experiment was carried out in a 500-mL shake flask with 10.0 g of soil and 200 mL of the leachate