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Feasibility of bioleaching combined with Fenton-like reaction to remove heavy metals from sewage sludge

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HIGHLIGHTS

- Bioleaching combined with Fenton-like reaction was feasible for heavy metal removal.
- Zn and Cd were efficiently solubilized within 5-day bioleaching.
- Cu and Pb solubilization was effectively enhanced Fenton-like reaction.
- Heavy metal solubilization depended on their forms in sludge.

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ABSTRACT

Feasibility of bioleaching combining with Fenton-like reaction to remove heavy metals from sewage sludge was investigated. After 5-day bioleaching, the sludge pH decreased from 6.95 to 2.50, which satisfied the acidic conditions for Fenton-like reaction. Meanwhile, more than 50% of sludge-borne heavy metals were dissolved except for Pb. The bioleached sludge was further oxidized with Fenton-like reaction, with an optimal H_2O_2 dosage of 5 g/L, the Cu, Zn, Pb and Cd removal reached up to 75.3%, 72.6%, 34.5% and 65.4%, respectively, and the residual content of heavy metals in treated sludge meets the requirement of Disposal of Sludge from Municipal Wastewater Treatment Plant – Control Standards for Agricultural Use (CJ/T 309-2009) of China for A grade sludge. Bioleaching combined with Fenton-like reaction was the most effective method for heavy metal removal, compared with 15-day bioleaching and inorganic acid leaching with 10% H_2SO_4, 10% HCl and 10% HNO_3.

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1. Introduction

Activated sludge process is widely used for municipal sewage treatment (Wang et al., 2012), which produces a large amount of sludge with complex components. With popularization of sewage treatment, the production of excess sewage sludge greatly increases, which makes the proper sludge treatment and disposal technologies become more and more important (Marcus and Torsten, 2011).

Land application is considered as one of the most economical and effective ways for sludge final disposal as it combines recycling of plant nutrients and sludge disposal (Babel and Del Mundo Dacera, 2006). Unfortunately, heavy metals are accumulated in the sewage sludge through solubilization, cation exchange, precipitation, adsorption, complexation and other reactions during wastewater treatment, especially when the industrial wastewater flows into the wastewater treatment plants. The sludge containing heavy metals has serious potential risk to human health, and the heavy metals, which exceed the standards, become the main restricting factor for sludge land application. So, removing the heavy metals efficiently from the sludge is the key of sludge land application.

Several chemical and microbial technologies for heavy metal removal from sewage sludge have been reported (Wang et al., 2010; Zhang et al., 2012). Bioleaching is one of the microbial technologies for sludge treatment, which has gained increasing attention since it is environmentally friendly and economical (Peng et al., 2011). By bioleaching, the heavy metals can be dissolved due to the acidic condition resulted from *Thiobacillus* and then be removed through separation of solid and liquid. However, the bioleaching time is much longer than that of chemical method, the long reaction time has become the main barrier for application.

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The reaction of hydrogen peroxide with ferric ions, which is referred to as Fenton-like reaction, produces hydroxyl radicals (·OH) under acidic condition by iron-catalyzed decomposition of hydrogen peroxide (Pham et al., 2010). The (·OH) is the main reactant decomposing a number of organic substances (Liu et al., 2012). The Fenton-like reaction as one of the advanced oxidation processes (AOPs) has been used as an alternative sludge conditioning technology (Dewil et al., 2005). On the contrary, there is little information on the effectiveness of Fenton-like reaction for heavy metal removal from sewage sludge.

In the present study, the combination of bioleaching and Fenton-like reaction was applied to heavy metal removal from sewage sludge. The bioleaching provides the acidic condition and catalyst for the Fenton-like reaction, while the Fenton-like oxidation shortens the bioleaching period, which has been scarcely reported. The main objective of this study was to evaluate the feasibility and treatment efficiency of the combined process by comparing with the traditional bioleaching and chemical leaching.

2. Methods

2.1. Materials

Sewage sludge, which was thickened and dewatered, was obtained from a local full-scale wastewater treatment plant in Changsha, China. To avoid the change in physicochemical properties, the sewage sludge was transported to laboratory in an airtight polythene cask and stored at 4 °C for further study. The main characteristics of raw sludge were measured as below: solid content, 14.82%; Cu content, 545.6 mg/kg dry sludge (DS); Zn content, 1516.7 mg/kg DS; Pb content, 133.2 mg/kg DS; Cd content, 12.8 mg/kg DS. Compared with the Disposal of Sludge from Municipal Wastewater Treatment Plant – Control Standard for Agricultural Use (CJ/T 309-2009) of China for A grade sludge (Zn, <1500 mg/kg; Cu, <500 mg/kg, Pb, <300 mg/kg, Cd, <3 mg/kg), the Pb content in raw sludge was lower than the corresponding standard, the Cu and Zn content in raw sludge slightly exceeded the corresponding standards, while the Cd content was 3.2 times higher than the corresponding standard. After dilution with deionized water to a solid content of 2%, the sludge pH and oxidation–reduction potential (ORP) were 6.95 and −76 mV, respectively.

All chemicals used were of analytic grade and were used as received. Deionized water was used for all analysis.

2.2. Preparation of inocula for bioleaching

Fresh sewage sludge was collected from the sludge thickener in the same wastewater treatment plant as the seed sludge to enrich and culture the indigenous iron-oxidizing bacteria. About 150 ml seed sludge at a 2% solid content was added into a 250 ml Erlenmeyer flask, and 20 g/L $FeSO_4 \cdot 7H_2O$ was fed as the energy source. The Erlenmeyer flask was agitated on a gyratory shaker (ZHWY-1102, China) at 150 r/min and 30 °C until the pH of seed sludge dropped to 2.0. The acidified sludge of 7.5 ml was then transferred into 150 ml fresh sewage sludge, under the same conditions the iron-oxidizing bacteria were enriched and cultured twice again. The obtained sludge was used as the inocula for sludge bioleaching.

2.3. Experiments for heavy metal removal

The experiments for heavy metal removal included three methods, namely inorganic acid leaching, bioleaching, and bioleaching combining with Fenton-like reaction.

For inorganic acid leaching, 150 ml sewage sludge was acidified to a pH of 2.5 with 10% H_2SO_4, 10% HNO_3 and 10% HCl in flasks and agitated at 150 r/min and 30 °C on a gyratory shaker (ZHWY-1102, China) for 24 h. After inorganic acid leaching, the sludge taken from the flasks was centrifuged at 12,000 r/min for 15 min. The heavy metal concentration in supernatant was measured.

Bioleaching experiment was carried out in flasks with 300 ml sludge sample, which was mixed with 5% (v/v) inocula and 10 g/L (w/v) $FeSO_4 \cdot 7H_2O$ as the substrate. The sludge bioleaching lasted 15 d. During the bioleaching, the soluble heavy metal concentration, pH and ORP were measured once a day from the first day to the fifth day, then every other day until 15 days.

The combination of bioleaching and Fenton-like reaction consisted of two stages. The first stage was completed by 5-day bioleaching to a sludge pH lower than 2.5. Then, the Fenton-like reaction was performed by adding H_2O_2 into the bioleached sludge for 1 h. A control without H_2O_2 addition was carried out for comparison. After the Fenton-like oxidation of sludge, the heavy metal concentration in supernatant was determined.

During the bioleaching experiments, the water loss via evaporation was daily compensated by adding distilled water based on weight loss. All experiments were done in duplicate.

2.4. Analysis

The sludge solid content was measured by gravimetric analysis. Cu, Zn, Pb and Cd in the raw sludge were extracted by the five-stage sequential extraction procedure (Tessier et al., 1979) and the heavy metal concentration in liquid was measured with an atomic adsorption spectrometry (Perkin Elmer AA700, USA). The pH and ORP were monitored with a pH and ORP analyzer (Dr. Kornder K100, Germany).

3. Results and discussion

3.1. Change of pH and ORP during bioleaching

The bioleaching was performed at first. The sludge pH and ORP are widely known to be the important parameters influencing heavy metals solubilization during bioleaching, which can demonstrate the activity of iron-oxidizing bacteria (Zhang et al., 2009). Fig. 1 shows the change of pH and ORP during sewage sludge bioleaching using $FeSO_4 \cdot 7H_2O$ as energy substrate. The ORP and pH change showed opposite trend. When the pH decreased, the ORP

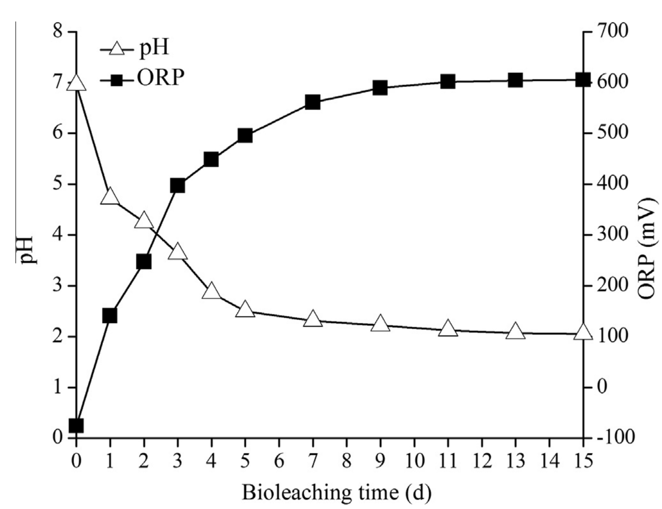

Fig. 1. Change in pH and ORP during bioleaching.

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