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Effective bioleaching of chromium in tannery sludge with an enriched sulfur-oxidizing bacterial community

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HIGHLIGHTS

- Bioleaching of chromium in tannery sludge using an enriched microbial community.
- Maximum bioleaching efficiency of Cr from tannery sludge was up to 96.8%.
- Forms of chromium in sludge affected the bioleaching performance.
- Some indigenous bacteria in sludge played a supporting role in bioleaching.

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ABSTRACT

In this study, a sulfur-oxidizing community was enriched from activated sludge generated in tannery wastewater treatment plants. Bioleaching of tannery sludge containing 0.9–1.2% chromium was investigated to evaluate the effectiveness of the enriched community, the effect of chromium binding forms on bioleaching efficiency, and the dominant microbes contributing to chromium bioleaching. Sludge samples inoculated with the enriched community presented 79.9–96.8% of chromium leaching efficiencies, much higher than those without the enriched community. High bioleaching efficiencies of over 95% were achieved for chromium in reducible fraction, while 60.9–97.9% were observed for chromium in oxidizable and residual fractions. *Acidithiobacillus thiooxidans*, the predominant bacteria in the enriched community, played an important role in bioleaching, whereas some indigenous heterotrophic species in sludge might have had a supporting role. The results indicated that *A. thiooxidans*-dominant enriched microbial community had high chromium bioleaching efficiency, and chromium binding forms affected the bioleaching performance.

1. Introduction

There are more than 1800 tanneries in China, which generate large quantities of tannery sludge during the wastewater treatment process in tannery. Tannery sludge contains 1–4% chromium, and it is classified as a dangerous waste in many countries (Chuan and Liu, 1996; Skrypski-Mäntele and Bridle, 1995). To reduce chromium content in tannery sludge to an acceptable level is a challenge for sustainable development of leather industry.

Removal of chromium from tannery sludge can be achieved by either physico-chemical or biological technologies. Although conventional physico-chemical methods are widely used for metal-contaminated sludge, in practice, they have inevitable limitations, such as secondary pollution, high installation and labor cost, and operational difficulties (Dewil et al., 2007; Pathak et al., 2009). Among the biological technologies, bioleaching has prominent advantages, because of its low cost, simplicity, low energy demand and environmental friendliness (Akinci and Guven, 2011). Sreekrishnan and Tyagi (1996) reported that compared to acidification or iron oxidation, bioleaching via microbiological sulfur oxidation is the cheapest method at low plant capacities and high solid operation. Bioleaching has been actively studied for years to deal with metal-contaminated sludge and environments, such as sediment and soil (Chen and Lin, 2004; Chen and Pan, 2010; Deng et al., 2012; Tyagi et al., 1996). Removal efficiency of most heavy metals is usually higher than 85%, but that of chromium is generally lower than 40% (Blais et al., 1993; Sreekrishnan et al., 1993). Reports on bioleaching of tannery sludge, in which the main heavy metal contaminant is chromium, are limited to date (Fang and Zhou, 2007).

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During the bioleaching process, addition of exogenous microorganisms, to strengthen the bioleaching efficiency, is generally critical. Single strains or artificial microbial consortiums capable of sulfur or ferrous oxidation are common choices for most researchers (Bharadwaj and Ting, 2013; Hocheng et al., 2014; Naresh Kumar and Nagendran, 2009; Yang et al., 2014), even though the cultivation of pure culture strains is laborious, and the structure stability of artificial consortiums is difficult to maintain (Zhu et al., 2014). However, enriched natural microbial communities can effectively overcome these shortcomings, especially those enriched directly from the pollution source will show better efficiency of metal solubilization (Chen and Pan, 2010; Mousavi et al., 2007). Indigenous microbes from sludge or the environment will interact with exogenous microbes, and are considered to affect their function, which will influence the bioleaching efficiency. In addition, some indigenous microbes themselves may have bioleaching capacity. However, most bioleaching studies reported use of sterilized soil or sludge (Akinci and Guven, 2011; Deng et al., 2012), and the function and effect of the indigenous bacteria were not considered.

Metals are present in different binding forms in soil or sludge, which will show different performances during bioleaching. There have been studies on the distribution of heavy metals in sediment (Ma et al., 2016), sewage sludge (Liu and Sun, 2013) and combustion waste (Karwowska et al., 2015). Heavy metal fractionation in sediment (Akinci and Guven, 2011) and soil (Naresh Kumar and Nagendran, 2009), before and after bioleaching, has also been reported. However, variation in chromium chemical forms in tannery sludge during bioleaching, and the influence of chromium binding forms on bioleaching efficiency have not been reported to date.

In the present study, a natural microbial community with bioleaching capability was enriched, using tannery sludge as microbial seed, and used as inoculums for bioleaching of unsterilized tannery sludge from three different tanneries. Using bioleaching process without inoculum of enriched natural microbial community as a control, the contribution of enriched natural microbial community on chromium solubilization, and the major functional microbes during bioleaching process were investigated. In addition, the binding forms of chromium in tannery sludge, before and after bioleaching, were determined to reveal the effect of binding forms on bioleaching efficiency.

2. Materials and methods

2.1. Tannery sludge

The three tannery sludge samples used in the study were collected from three different tanneries in China. They were named as H (from Haining), G (from Guangzhou), and Z (from Guangzhou). pH, water content, total organic carbon (TOC), total nitrogen and sulfur contents, organic content, ash content, total heavy metal content, and the forms of chromium were analyzed. The results have been presented in Table 1.

2.2. Enrichment of natural microbial community for bioleaching

Activated sludge, taken from an aeration tank in a tannery wastewater plant, was used as the seed for enrichment. A modified Starkey medium was used to enrich sulfur-oxidizing bacteria. The medium consisted of the following basal salts (g/L): $(NH_4)_2SO_4$ 2.0, $MgSO_4{\cdot}7H_2O$ 0.5, K_2HPO_4 0.5, KCl 0.1, and $CaCl_2{\cdot}2H_2O$ 0.25, and trace elements (mg/L): $FeCl_3{\cdot}6H_2O$ 11.0, HBO_3 2.0, $CuSO_4{\cdot}5H_2O$ 0.5, $MnSO_4{\cdot}H_2O$ 2.0, $Na_2MoO_4{\cdot}2H_2O$ 0.8, $CoCl_2{\cdot}6H_2O$ 0.6, and $ZnSO_4{\cdot}7H_2O$ 0.9. The pH of the medium was adjusted to 4.0 using H_2SO_4. In the enrichment process, 12.5 mL activated sludge was inoculated into 250 mL medium supplemented with 1.0% (w/v) sulfur powder, which was used as an energy source, in a 500 mL Erlenmeyer flask. The cultivation was carried out at 30 °C with an agitation speed of 180 rpm in a thermostat water bath. The pH of the culture, used as an indicator of the activity of the microbes, was determined every two days, during the enrichment process. When pH was below 2, the culture was transferred into fresh 250 mL medium. The enrichment was repeated until the sulfur oxidation capacity of the community stabilized. The culture was then used as an inoculum for the bioleaching experiment. The bacterial community of the enriched culture was analyzed using 16S rRNA gene clone library.

2.3. Bioleaching experiment

Bioleaching experiments on three different sludge samples were performed, with and without the inoculum of enriched culture. Experiments with the inoculum of enriched culture were named HB, GB, and ZB, whereas those without the inoculum were

Table 1
Physico-chemical characteristics of raw tannery sludge samples.

Parameter	H	G	Z
Organic content (g/g-dry sludge, %)	38.26 ± 0.49	40.75 ± 0.31	36.87 ± 0.48
Ash (g/g-dry sludge, %)	61.75 ± 0.49	59.25 ± 0.31	63.13 ± 0.38
Moisture (%)	60.01 ± 0.14	81.56 ± 0.11	75.82 ± 0.19
pH	7.32 ± 0.02	8.04 ± 0.04	7.21 ± 0.02
TOC (mg/kg-dry sludge)	882.10 ± 10.26	651.15 ± 32.17	453.05 ± 4.14
Cr (mg/kg-dry sludge)	12,204.49 ± 270.65	9,237.96 ± 502.18	9,085.75 ± 170.90
Cr (exchangeable and acid soluble fraction, %)	0.36 ± 0.09	12.13 ± 3.18	1.96 ± 0.17
Cr (reducible fraction, %)	2.31 ± 0.59	18.82 ± 3.76	3.32 ± 0.04
Cr (oxidizable fraction, %)	68.83 ± 13.08	49.84 ± 2.14	62.11 ± 1.29
Cr (residual fraction, %)	28.50 ± 3.68	19.21 ± 5.99	32.60 ± 1.17
Cu (mg/kg-dry sludge)	366.27 ± 17.13	349.84 ± 31.05	408.14 ± 81.90
Zn (mg/kg-dry sludge)	1,016.70 ± 298.62	945.92 ± 77.63	811.23 ± 16.69
Pb (mg/kg-dry sludge)	508.90 ± 55.49	995.39 ± 33.53	865.05 ± 39.30
Cd (mg/kg-dry sludge)	17.07 ± 4.04	9.61 ± 1.77	8.71 ± 0.32
Nitrogen (%)	2.21 ± 0.03	1.78 ± 0.01	1.99 ± 0.01
Sulfur (%)	5.63 ± 0.09	4.55 ± 0.04	5.24 ± 0.16
C/N ratio	6.44 ± 0.07	5.52 ± 0.01	6.57 ± 0.02

Data represents average ± standard deviation.