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Simultaneous metal leaching and sludge digestion by thermophilic microorganisms: Effect of solids content

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ABSTRACT

High concentrations of heavy metal in livestock manures limit land application of their sludges. A practical and economical method of sludge treatment is important for converting the livestock sludge into soil conditioners or fertilizers. In this study, the effect of solid contents on the simultaneous aerobic digestion and metal leaching at thermophilic condition were investigated in a batch reactor. Different solid contents in the range of 0.5–4% (dry-w/v) were studied. The results showed that an increase of solid content decreased the pH reducing rate. It was the result of increase in buffering capacity and possible microbial inhibition at a higher solid content. Similar results were also found in the variations of ORP and sulfate concentrations during this process. In most cases, this biological process is able to solubilize 82–99% of heavy metals from the livestock sludge. It was found that the efficiency and rate of metal solubilization decreased with increasing solid contents. In addition, 54–80% of organic matter in the sludge was degraded after 28 days of reaction. A low sludge digestion efficiency was found at a high solid content. Moreover, the dewaterability of sludge was improved and the fertility (N, P and K) of sludge did not change significantly after this bioprocess.

1. Introduction

Due to limited land in Taiwan, the management of swine farming tends toward a high intensity confinement system instead of a pasture-based system. In general, the wastewater produced from swine farms was one of the main sources polluting rivers in Taiwan. To protect the waterways and the sustainable growth of the swine production industry in Taiwan, the government imposed effluent standards for the industry. Therefore, the management of wasted sludge produced from the wastewater treatment facilities becomes a pressing issue for the swine farms. Rather than the landfills and incineration for sludge treatment, the swine sludge containing abundant nutrients and hydrocarbons is favourably applied to agricultural lands as soil conditioners or fertilizers. This way of sludge disposal achieve dual benefits of nutrient recycling and waste minimization of swine farms. Although such application may be beneficial to soil, the negative impacts such as groundwater pollution, pollutant accumulation in soil, etc., should be also considered [1]. Elevated heavy metals concentration in sludge limits the utilization of swine sludge in agriculture and land application [2,3]. There is a need to find an economically feasible and environmentally friendly process to remove heavy metals from swine sludge.

Although some physical or chemical techniques have been implemented for treating the sludge containing heavy metals such as acid/solvent extraction, acid/alkaline thermal hydrolysis and Fenton's peroxidation, the shortcomings of the requirement of large amount of chemicals, high operating cost, the operational difficulties and the secondary pollution problems restrict their applications [4,5]. Because of simplicity, low operating and capital costs, and environmentally friendly [6,7], the microbial leaching is considered as an efficient and economical technology to remove heavy metals from sludge, soil or sediment [8–11]. Tyagi et al. [12] showed that the microbial leaching is 80% cheaper in terms of chemical consumption compared to the traditional chemical methods employed for metal leaching from the sludge and recovery of metals from the leachate. Moreover, Sreekrishnan and Tyagi [13] further reported that the microbial leaching process is less costly as compared to the conventional aerobic digestion and metal leaching by acid addition or iron-oxidizing process, especially at small-scale operation and at high solids content. In the microbial leaching process, the removal of heavy metals from sludge is mainly achieved by the oxidation and acidification reactions caused by sulfur-oxidizing bacteria. The microorganisms mainly involved in the microbial leaching process belong to the genus *Acidithiobacilus* or *Thiobacillus*, specially including *At. thiooxidans*, *At. ferrooxidans* and *T. thioparus* [8]. Generally, acclimated indigenous microorganisms are used as

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the inoculums in the microbial leaching process to attain the high efficiency of metal solubilization [14]. However, when the acclimated indigenous microorganisms are inoculated in the microbial leaching process, the degradation of organic matter by indigenous heterotrophic microorganisms in the sludge also occurs, simultaneously coupled with the metal leaching reaction. Therefore, a process called simultaneous sludge digestion and metal leaching has been developed for sludge treatment [15].

It has been known that the sludge solid contents in the microbial leaching process play a significant role in determining the reactor capacity. In general, a low solid content increases the rate of microbial leaching; however, a large volume of reactor is required. Contrarily, a high solid content increases the treatment capacity, which results in a slow rate of microbial leaching and a long residence time. Kim et al. [16] showed that the rate and efficiency of metal solubilization decreased as an increase in the solid content in a microbial leaching system. Liu et al. [17] reported that the oxidation activity of *At. ferrooxidans* was inversely affected by the solid contents in the microbial leaching process because of collision and friction among solid particles. In addition, the microbial leaching process mostly performed under mesophilic conditions has shown slow metal leaching rates. However, due to higher metal tolerance capacity and metabolic characteristics at high temperature, the rates of metal leaching under thermophilic conditions are considerably enhanced [18–20]. Meanwhile, literature in simultaneous metal leaching and sludge digestion process at the thermophilic conditions is very limited. The purpose of this study was to investigate the effects of the solid contents on the performance of thermophilic simultaneous metal leaching and aerobic digestion of livestock waste sludge. The dewaterability and fertility (N, P and K) of treated sludge were also evaluated in this study.

2. Materials and methods

2.1. Sludge sampling

The sludge was obtained from an anaerobic treatment unit of the wastewater treatment plant in a local swine farm of northern Taiwan. The sludge was then screened through a 20-mesh (0.84 mm) sieve, well mixed and stored at 4 °C before the study. The swine sludge was analyzed for pH [21], total solids (TS) volatile solids (VS), suspended solids (SS), volatile suspended solids (VSS) [22], nutrient (N, P) contents and heavy metal contents. For metal content analysis, the sludge sample was digested with HNO_3–HF–HCl (5:4:1, v/v) mixture in the microwave digestion system (Model MARS Xpress, CEM), according to USEPA3052 method [23]. The total nitrogen content of the sludge was measured using a Herneus CHNOS Rapid elemental analyzer and the total phosphorus content was determined according to Method 424 C&D in Standard Methods [22]. The basic characteristics of the swine sludge are shown in Table 1.

2.2. Acclimation of sulfur-oxidizing bacteria

For acclimation, 15 g of sterilized elemental sulfur was mixed with 3 l of the swine sludge (1%, dry-w/v) in a batch reactor with an agitation speed of 200 rpm and the temperature of 55 °C. The acclimation activity of sulfur-oxidizing bacteria was determined based on sludge pH. The acclimation procedure was matured when sludge pH dropped to about 2.0. Then, a 300 ml of the acidified sludge sample was transferred to another reactor with 3 l of fresh sludge and 15 g of elemental sulfur, and the acclimation procedure was repeated at least three times. The acclimated sludge was then used as an inoculum for the simultaneous metal leaching and sludge digestion experiment.

Table 1
The characteristics of sludge used in this study.

Property	Value[a]
pH	7.3 ± 0.1
TS (%, w/w)	6.36 ± 0.12
VS (%, w/w)	3.08 ± 0.12
SS (mg l^{-1})	55420 ± 960
VSS (mg l^{-1})	26410 ± 530
Cu (mg kg^{-1})	1105 ± 74
Zn (mg kg^{-1})	3310 ± 90
Mn (mg kg^{-1})	1824 ± 56
Ni (mg kg^{-1})	84 ± 7
K (mg kg^{-1})	7271 ± 345
Total-N (mg kg^{-1})	33190 ± 940
Total-P (mg kg^{-1})	2140 ± 60

[a] Mean ± standard deviation (*n* = 10).

2.3. Simultaneous metal leaching and aerobic digestion experiments

The experiment of simultaneous metal leaching and aerobic digestion was performed in a completely mixed batch reactor containing, 1.5 l of inoculants, 50 g of elemental sulfur, and 10 l of sludge with different solid contents (0.5–4%, dry-w/v). The batch bioreactor was maintained at 55 °C with an agitation speed of 200 rpm. The sludge in the reactor was aerated with an airflow rate of 0.3 vvm (volume of air (volume of sludge)$^{-1}$ min^{-1}). A control test (2% solid content) without inoculation and elemental sulfur addition was also carried out under the same conditions. The pH and oxidation–reduction potential (ORP) of sludge were measured continuously by an on-line pH/ORP meter (Suntex, model PC-310). During the experiments, sludge samples were periodically withdrawn for SS and VSS analyses [22]. Meanwhile, the sludge sample was filtered through a 0.45 μm filter, and the filtrate was used for analyzing sulfate concentration [22] and heavy metals. The concentrations of heavy metals were determined by a flame/graphite atomic absorption spectrophotometer (Shimadzu AA-6200). At the end of the experiments, the sludge was directly taken out for the analysis of dewaterability index (specific resistance to filtration, SRF) [24] without any neutralization and conditioning. The sludge retaining on the filter papers in SRF analysis was air dried at room temperature. The sludge was scraped and analyzed for total nitrogen, total phosphorus and potassium.

In order to confirm the degradation of solids caused by acidification, 3 l of sludge (2% solid content) was abiotically acidified by a daily stepwise addition of sulfuric acid followed a similar biotic reactor pH profile in a stirred tank reactor. This experiment of sludge acidification was also carried out at 55 °C. The pH after each step of acid addition was recorded and the sludge samples were withdrawn to determine the SS and VSS.

3. Results and discussion

3.1. Variations of pH and ORP

The variations of pH in the simultaneous metal leaching and aerobic digestion experiments at the thermophilic conditions are shown in Fig. 1. The growth of thermophilic sulfur-oxidizing bacteria converting elemental sulfur into sulfuric acid caused the decrease of sludge pH in this study. As indicated in Fig. 1a, that decrease in pH was found during the simultaneous metal leaching and aerobic digestion. The pH after 28 days dropped from 7.3–7.6 to 1.7–3.0 with different solid contents. It was found that the higher pH decreasing rates was observed at lower solid contents. Generally, the biological and chemical oxygen demands are significantly