Research on applicability of anaerobic passive bioreactor to acid mine drainage treatment in Japan

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Abstract In past experiments, the applicability of the treatment system for mine drainage with the anaerobic bioreactor containing rice husk and composted bark with cow manure was investigated. This time, the relationship between a metal removal process and ORP was figured out and moreover, it is cleared that anaerobic passive treatment for mine drainage can operate at lower cost using cheaper contents in bioreactors.

Keywords passive treatment, sulfate-reducing bacteria, anaerobic bioreactor, rice husk

Introduction

Passive treatment system which uses natural purification processes has attracted a lot of attention as a low cost and maintenance mine drainage treatment technology, and many research results of verification tests on the passive system have been reported mainly in the United States and Europe (*e.g.* Gusek *et al.* 1998, Younger *et al.* 2003). In Japan, however, no passive treatment system is actually introduced. All mine drainage treatment plants use active systems where neutralizer, electricity and daily maintenance are required. However, recently, from the viewpoint of cost reduction for the mine drainage treatment, the research on the applicability of passive treatment system in some domestic abandoned metal mine sites has been started by Japan Oil, Gas and Metals National Corporation.

In this research, some fundamental tests were carried out in order to design the treatment system with anaerobic bioreactors. In the bioreactor containing sulfate reducing bacteria (SRB), dissolved metal ions are capably removed as metal sulfides because sulfide is more insoluble compared with hydroxide.

We have succeeded to continuously treat the neutral pH mine drainage for over 800 days under the appropriate condition, water retention time for 50 hours and water temperature of over 15 °C, using the bioreactors filled with rice husk and composted bark with cow manure. Similar application has been experimentally and successfully utilized for the continuous treatment of acid mine drainage for over 500 days(Furuya *et al.* 2012).

As described above, It has been found that the passive treatment system using anaerobic bioreactor can be applied to mine drainage at various pH levels, although it is essential to further understand the detailed reaction mechanism for the design of the actual equipment.

This paper shows the relationship between oxidation-reduction potential (ORP) and metal ions removal phenomena in the reactor. Moreover, in order to reduce the material cost of anaerobic treatment system, the applicability of cheaper organic material for SRB has been investigated.

Methods

Mine drainage

The mine drainage for the continuous test was sampled at the abandoned mine in Akita prefecture located in the northern part of Japan. The concentrations of zinc, copper and iron in the drainage are shown in Table 1. The values exceed the national effluent standard, and so, the drainage has been selected as the sample in this investigation.

Apparatus

A schematic diagarm layout of the continuous experimental apparatus with the anaerobic column is shown in Fig. 1. The diameter and height of column is 10 cm and 45 cm, respectively, and 5 intake holes (numbered as 1st, 2nd, 3rd, 4th, and 5th from upper side to lower side in order) are equipped in the wall of the column. The column was filled with rice husk and composted bark with cow manure as organic carbon resources for SRB and other microorganisms. These materials were found to be effective as organic carbon resources for the related microbes in the previous research (Furuya *et al.* 2011). Granular silicates and lime stones (diameter : 5 ≈ 10 mm) were utilized to maintain the inner structure of the reactor and to control pH respectively.The volume and weight fractions of each component in the bioreactor are shown in Table 2.

Process

The column was filled with the mixture of carbon resources and the mine drainage to the height of 40 cm. The contained SRB was incubated for 3 weeks in the mine drainage. SRB was supplied from the rice husk and composed bark with cow manure and no other bacteria source was added to the system. After then, the mine drainage was pumped up and introduced to pass the column from the top to the bottom with gravity flow. The hydraulic re-

	pH	Zn (mg/L)	Cu (mg/L)	Fe (mg/L)	SO_4^{2-} (mg/L)
Mine drainage (min—max)	3.3—3.8	15—18	3—10	33—38	350—400
National effluent standard	5.8—8.6	2.0	3.0	10	

Table 1 *Water quality of the mine drainage and the national effluent standard values.*

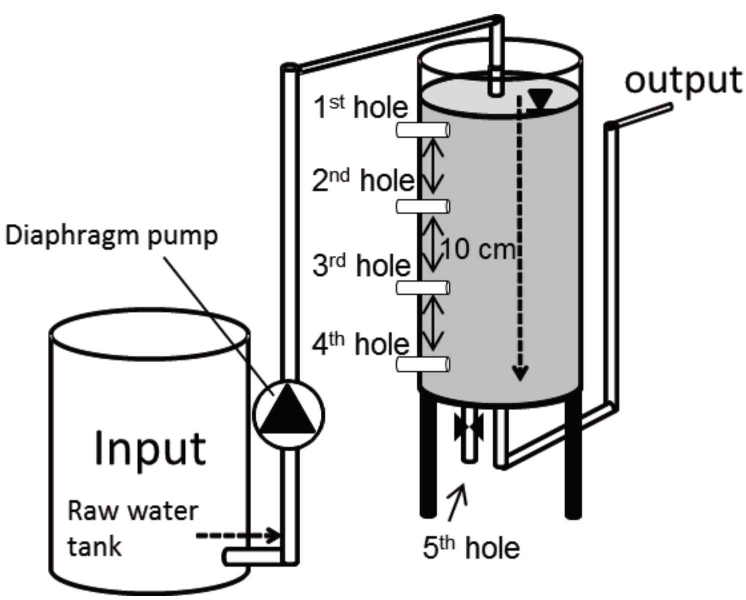

Fig. 1 *A schematic diagram of the setup for continuous treatment test with anaerobic bioreactor.*

Components	Rice husk	Composted bark with cow manure
Vol.%	99.7	0.3
Wt.%	95.0	5.0

Components	Crushed silica stone	lime stone
Weight,g	1050	300

Table 2 *Composition of mixture in the bioreactor.*

tention time was 50 hours, which was calculated from the void volume, and the flow rate was 60 mL/h. The treated water was discharged from the silicone tube which extended from the bottom to a static water surface level.

Results and discussion

Relationship of ORP and metal removal

Before starting the treatment test, while SRB was incubated, the inside of the column was reductive. The upper part of the column subsequently changed into an oxidative condition in a week after starting to introduce the raw water, the sampled mine drainage (Fig. 2(a)). This could be mainly caused by the introduction of the raw water of which ORP was 400 ≈ 500 mV and the fast oxidation of ferrous ions by iron-oxidizing bacteria at the upper part of the column. Although the upper part rapidly became oxidative as mentioned above, the lower part maintained the reductive condition with ORP of -100 ≈ -200 mV for over 100 days. The pH of the lower part was kept at 6 ≈ 7. Therefore, the activity of SRB could be allowed long-term.

It was observed throughout the monitoring period that sulfate ions drastically decreased at the upper part of the column, 350 mg/L to 300 mg/L, and at the lower part, 280 mg/L to 220 mg/L (Fig. 2(b)). The first decrease at the upper part could not be mainly caused by sulfate reduction by SRB because the upper part of the column was in the oxidative condition. It could be possibly caused by sorption to lime stones and organic materials. However, the drastic decrease of sulfate ions at the lower part of the column could be probably considered to be caused by the sulfate reduction by SRB.

Dissolved metal ions, especially zinc ions, drastically decreased from the middle part toward the lower part (Fig.2(c)). This result is well correlated with changes in the oxidation-reduction condition in the column. Zinc ions drastically decreased at reductive area where the activity of SRB was promoted, suggesting that most zinc ions passed through oxidative area without any chemical reaction and then zinc ions reacted with sulfide ions which were generated through sulfate ions reduction by SRB in the reduction condition, and finally removed as Zn sulfides. This was also correlated with the decrease of sulfate ions.

Sulfide ions were continuously detected from the outflow water. They were assumed to be generated by SRB, and surplus for the formation of metal sulfide. Removal of dissolved zinc ions continuously occurred even when almost all area of the column except for the lowermost part became oxidative. However, the concentration of total-zinc in the outflow water started to increase from around 100 days after the start of itroduction to the column, and exceeded the national effulent standards (2 mg/L) after 130 days.

Ferrous ions were removed through two-step process. Fig.2(d) shows that most of ferrous ions were removed at the upper part and the remaining portion were removed at the lower part. The first step removal, from the input to the 1st intake hole, was assumed to be oxidated by iron-oxidizing bacteria and removed as precipitates of ferric hydroxide. As the second step, from 4th to 5th, the remaining ferrous ions were supposed to be removed as ferrous sulfide.

Influence of leaking out of precipitates from the column

As described above, the concentration of total-zinc in the outflow water exceeded the national effulent standard 130 days after starting the introduction to the column(Fig.2(e)). However, the concentration of zinc ion became under the effulent standards after the filtration of the outflow water with 0.45 μm filter. This result was explained by suggesting that fine zinc sulfide precipitates were flowed out as a suspention form.

The investigation of cost reduction

So far, in the continuous treatment tests, the composted bark with cow manure and rice husk were utilized as the source of SRB and orgnic carbon resource for SRB. Rice husk is a common agricultural waste in Japan and it is easily available without cost. However, composted bark is a little costly. Since the real operation of mine drainage treatment needs a large amount of composted bark, high initial cost would be problematic.

Therefore, in order to reduce the material cost, the applicability of the soil which is available from mine sites as an alternative for composted bark was investigated.

Two polyethylene bottles were filled with 15 g rice husk, 15 g Lime stones, and 150 mL of

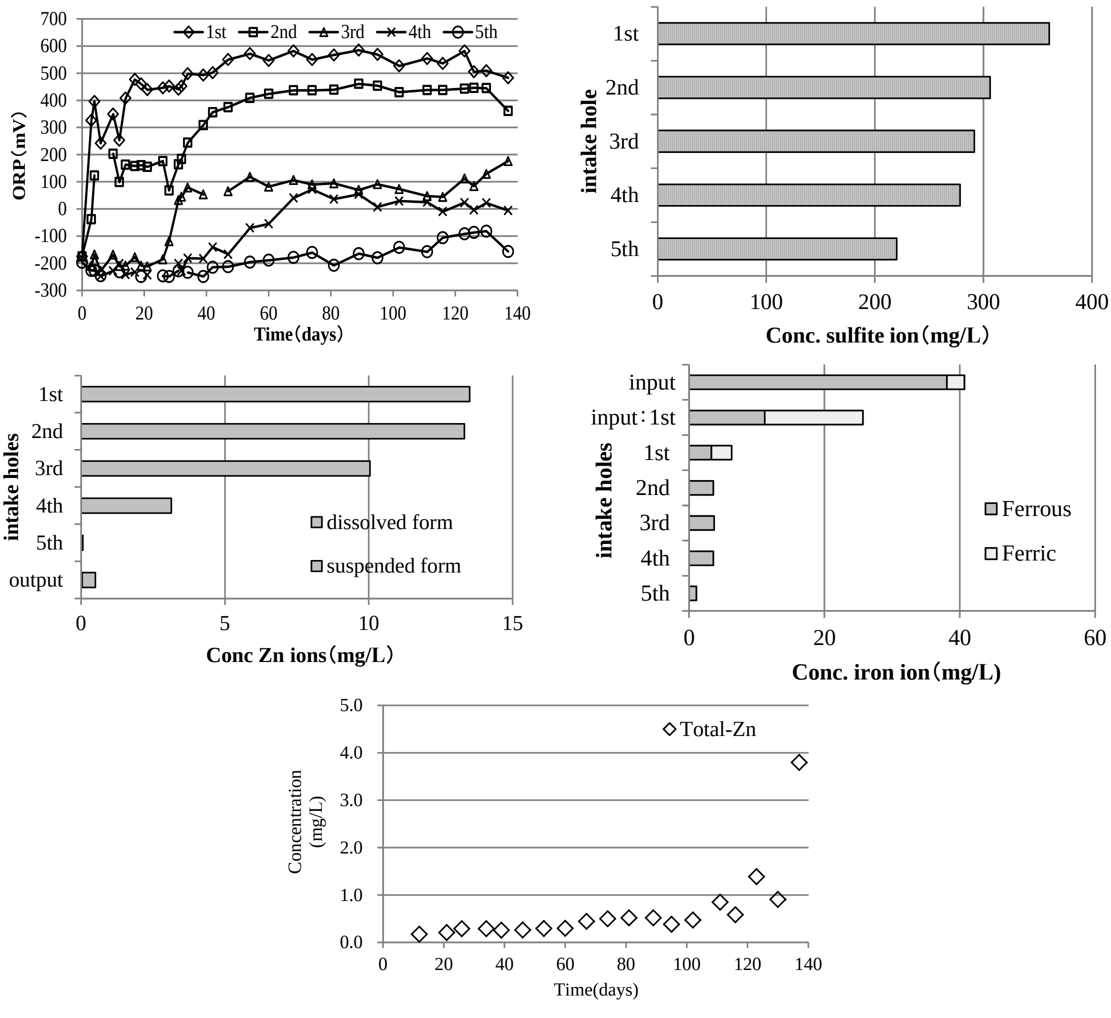

Fig. 2 *Water quality changes in the column*

the mine drainage containing sulfate ion. And then 1 g composted bark, or soil sampled from the mine site was added respectively. The ORP and the sulfate ions concentration of the solution in both bottles were regularly measured during the incbation.

As a result, both solutions almost became to be reductive conditions in around a same time (Fig.3(a)), and after 12days of incbation, the concentration of sulfate ion was also similar between both solutions (Fig.3(b)). This shows that the soil sampled from the mine site has the same effect as composted bark. Therefore, it would be probably expected that anaerobic passive treatment for mine drainage could operate with lower cost by using cheaper contents such as the soil at the mine site for bioreactors.

Conclusions

The correlation between the metal removal efficiency and the oxidation-reduction potential in the column was successfully evaluated. In fact, dissolved metal ions, especially zinc ions, reacted with sulfide ions which were generated through reduction of sulfate ions by SRB in the reduction condition. Moreover, the treatment of the acid mine drainage could be continued for 130 days. These results suggest the applicability of this system on acid mine drainage. Although leak-out of zinc sulfide precipitate was observed, the phenomena could be prevented by improving the structure of the bioreactor.

In addition, the experimental results showed that the appropriate soil at mine sites had almost same capability as composted bark. Therefore, it is expected that anaerobic passive treatment for mine drainage can operate with lower cost by using cheaper contents for the bioreactors.

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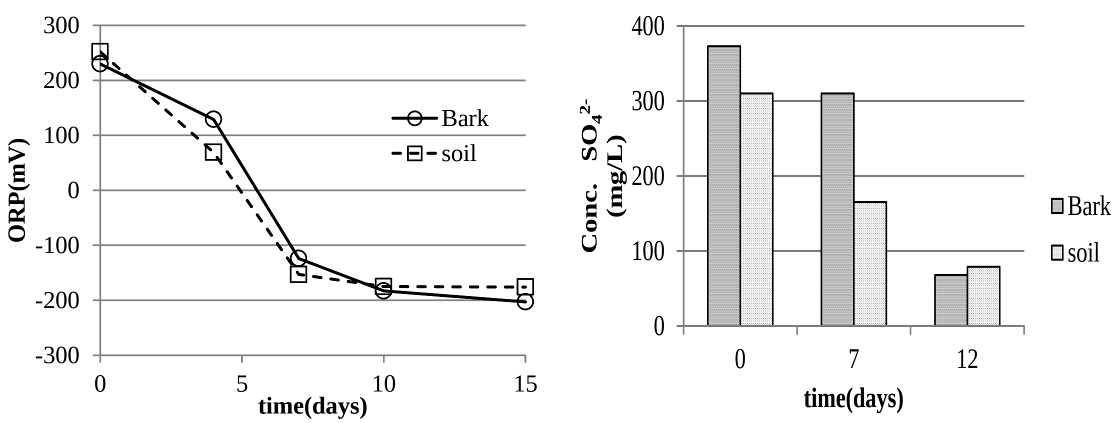

Fig. 3 *Comparing of capability between composted bark and soil*

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