

## BIOLOGICAL METAL REMOVAL FROM MINE DRAINAGE

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**Abstract.**--Mine drainage contaminated with heavy metals requires proper treatment to avoid pollution of surface waters. The aim of this work was to improve on known biological methods and to develop an economic, cost-effective pilot plant-scale process for removal of metal in mine drainage. Good results were obtained with a continuous-flow fluidized bed-type anaerobic reactor, with a working volume of 180L, in a continuous flow system using Desulfovibrio vulgaris as the principal sulfate-reducing bacteria.

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### INTRODUCTION

There are a number of inactive and abandoned underground mines in Japan from which acid drainage with heavy metals flows out continuously. To alleviate the acidic condition and metal content in the drainage, the following chemical treatment has been carried out: neutralization by slaked lime and powdered limestone. This chemical treatment, however, is not economically attractive because of its high cost and the occurrence of a large quantity of waste sludge. Therefore, alternative methods are needed.

Since dissolved metals can be removed as sulfide precipitates by anaerobic sulfate-reducing bacteria, various attempts have been made to treat acid mine drainage by this biological methods (Wakao, et al. 1979, Maree and Strydom 1985, 1987).

In this work basic experiments were carried out using Desulfovibrio vulgaris in both laboratory and pilot-scale tests in a continuous flow system. In developing the process, the following aspects were investigated:

- (1) Optimum conditions of a process to maintain the biological and chemical reactions: amount of organic carbon, pH level, inflow of raw water, and

temperature for reactions

- (2) Type of reactor required for effective separation of sulfide precipitate
- (3) Continuous process to remove metal from raw water.

### LABORATORY-SCALE EXPERIMENT

A strain of Desulfovibrio vulgaris was placed in culture medium shown in table 1 and incubated at 30°C. After incubation for six days, the bacteria grew and the medium turned black in color. The degree of reduction of sulfate to sulfide is shown in figure 1. This enriched culture was used as an inoculum of the culture of sulfate-reducing bacteria used in this experiment.

The experiment was performed in a continuous-flow fluidized bed-type anaerobic reactor with a working volume of 10L. The water used as feed was synthetic, composed of the same elements as shown in table 1. The water was fed continuously to the reactor after sufficient growth of the bacteria. Hydrogen sulfide was bubbled into the solution for mixing and coagulant, polyacrylamide, was added in the reactor to step up precipitation during the run.

Both the  $\text{Fe}^{2+}$  and  $\text{Zn}^{2+}$  ion contents in the feed water were set at 50mg/L respectively. Quantity of these metal ions and the sulfate concentration value in the effluent was used as the indicator of the reactions.

The experiment was performed changing

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Table 1.--Chemical characteristics of the culture medium used in the incubation of bacteria and laboratory-scale experiment.

Component	Value
K <sub>2</sub> HPO <sub>4</sub>	0.5g
NH <sub>4</sub> Cl	1 g
Na <sub>2</sub> SO <sub>4</sub>	1 g
MgSO <sub>4</sub> ·7H <sub>2</sub> O	2 g
Na-lactate	3.5g
Yeast extract	1 g
FeSO <sub>4</sub> ·(NH <sub>4</sub> ) <sub>2</sub> SO <sub>4</sub> ·6H <sub>2</sub> O	0.5g
Cu <sup>2+</sup> [CuSO <sub>4</sub> ·5H <sub>2</sub> O]	3 g
Pb <sup>2+</sup> [Pb(NO <sub>3</sub> ) <sub>2</sub> ]	10 mg
Zn <sup>2+</sup> [ZnSO <sub>4</sub> ·7H <sub>2</sub> O]	50 mg
Cd <sup>2+</sup> [CdSO <sub>4</sub> ·nH <sub>2</sub> O]	1 mg
As <sup>3+</sup> [NaAsO <sub>2</sub> ]	1 mg
Tap water	1,000 mL
pH	6.0-6.5

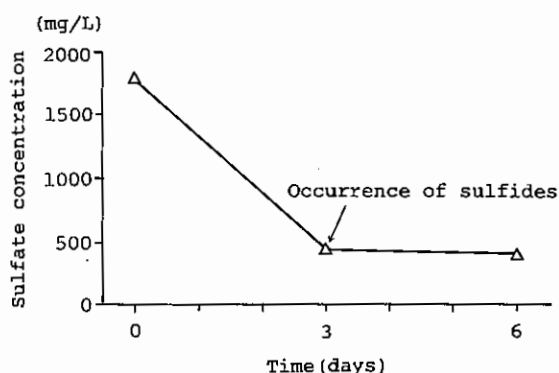


Figure 1.--Degree of decrease of sulfate by sulfate-reducing bacteria, *Desulfovibrio vulgaris*.

inflow stepwise at 10, 20, 40, and 80L/day, with the pH level between 5.0 and 6.5 and temperature in the reactor at 15°C and 30°C.

Sodium lactate used as the organic carbon source was added first at 3.5g/L and reduced to 1g/L. Yeast extract was started at 1g/L, then cut to zero finally.

Most of the metals in the feed water were separated as sulfides consistently during the periods of continuous operation: metal content in the effluent was kept below the emission standard in Japan (Fe < 10 mg/L, Zn < 5mg/L, Cu < 3mg/L, Pb < 1mg/L, Cd < 0.1mg/L, As < 0.5mg/L).

Concentration of the bacteria in the reactor was measured by the Thoma-Zeiss counting chamber once a week.

The results of the experiment was shown in table 2 and was summarized as follows.

- (1) Sodium lactate can be cut down to 1g/L at 20°C and pH5, and to 2g/L at 10°C and pH6
- (2) Retention time was 3 hours(80L/day inflow) at 15°C, pH6, sodium lactate 3.5g/L and yeast extract 1g/L, and 6 hours(40L/day inflow) at pH5

Table 2.--Result of the laboratory-scale experiment.

Inflow (L/day)	Retention time (hours)	Temp. (°C)	Effluent	
			pH	Zn <sup>2+</sup> (mg/L)
10	24	30	6.21-8.01	0.09-0.82
20	12	30	6.19-6.53	0.14-0.30
40	6	30	6.14-6.78	0.08-1.47
80	3	30	6.23-7.00	0.10-0.68
40	6	20	6.20-7.00	0.03-0.31
40	6	15	6.50-6.71	0.13-0.18
80	3	15	6.40-6.60	0.16-0.26

- (3) Concentration of the bacteria typically ranged from about 4-5X10<sup>9</sup> cells/mL throughout the experiment.

## PILOT PLANT-SCALE TEST

### Materials and Methods

A pilot plant-scale test was performed at Yanahara Mine in the Okayama Prefecture, based on the results of the laboratory experiment.

The test was in a continuous flow process using a fluidized bed-type anaerobic reactor with a working volume 180L, which was scaled-up version of the laboratory experiment system.

A schematic diagram of the plant and flow diagram of the process used are illustrated in figure 2 and figure 3, respectively.

The culture of the bacteria used for the test was the same as the previous laboratory experiment.

Mine drainage of Yanahara Mine, the quality of which is shown in table 3, was supplemented with sodium lactate, yeast extract, and inorganic materials. This mixed water was fed to the anaerobic reactor.

As the raw drainage was too acidic(pH 2.50-2.65) to maintain the activity of the bacteria and to clear the emission standard, the drainage was controlled by adding powdered limestone, the average grain size of which was 1.1 micrometer, to maintain pH at about 5 before the mixing. The condition of the influent is shown in table 4.

Table 3.--Chemical characteristics of mine drainage from the Yanahara Mine used for the pilot-plant scale test.

Component	Value
temperature	20°C
pH	2.5-2.65
Fe <sup>2+</sup>	171-303 mg/L
Total-Fe	693-815 mg/L
Zn <sup>2+</sup>	41.5-52.1mg/L
Cu <sup>2+</sup>	9.4-16.8mg/L
Cd <sup>2+</sup>	0.18mg/L
Mn <sup>2+</sup>	7.4 mg/L
Total-As	0.21mg/L
Acidity as CaCO <sub>3</sub>	1,991-2,442mg/L

Table 4.--Condition of the feed water used throughout the pilot plant-scale test.

Temperature (°C)	Inflow (L/day)	Yeast extract (g/L)	Na- lactate (g/L)	pH	Period (days)
24.7-30.2	180	0.5	3.5	6	16
25.8-28.8	360	0.25	3.5	6	2
22.2-27.7	360	0	3.5	5	12
19.0-25.6	720	0	3.5	5	13
18.1-22.8	720	0	3	5	8
14.8-19.8	720	0	2.5	5	10
10.0-16.2	720	0	2	5	20
10.8-14.9	720	0	1.5	5	18
9.0-14.2	720	0	1	5	14

The test was made increasing feed volume at three steps, 180, 360 and 720 L/day and was performed for 113 days continuously.

Sodium lactate and yeast extract were reduced from 3.5g/L to 1.0g/L and 0.5g/L to zero, respectively. Temperature in the reactor ranged between 9.0°C and 30.2°C, depending on the weather, because the test plant was not housed in order to test the procedure in practical use too.

Excess hydrogen sulfide except for the volume, which was bubbled into the reactor, was vented.

The COD in the effluent was measured by the dichromate titration.

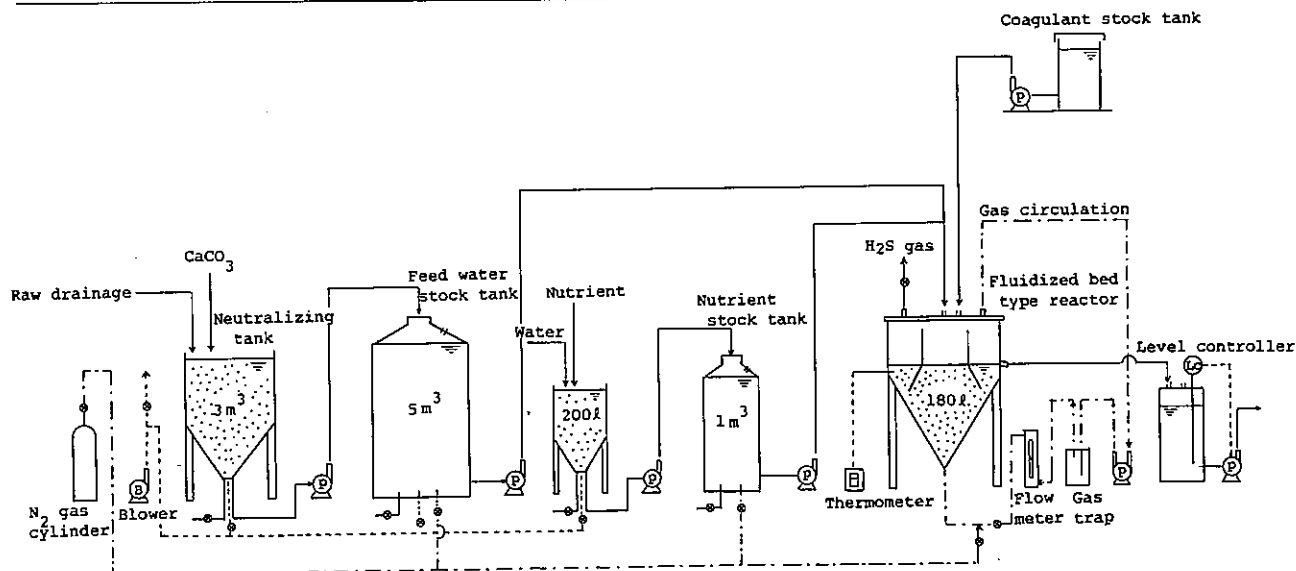


Figure 2.--Diagram of drainage treatment system used in the pilot plant-scale test.

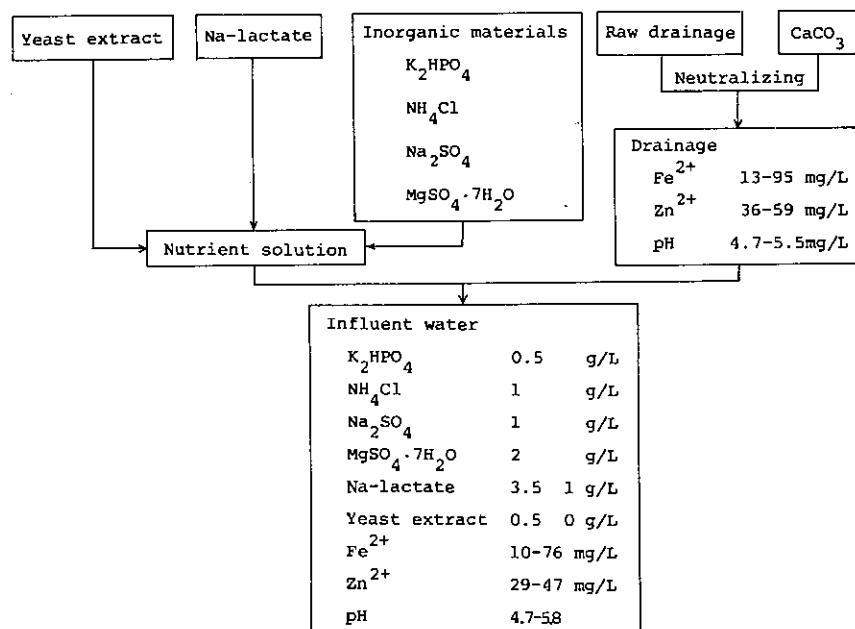


Figure 3.--Flow diagram of feed water in the pilot plant-scale test.

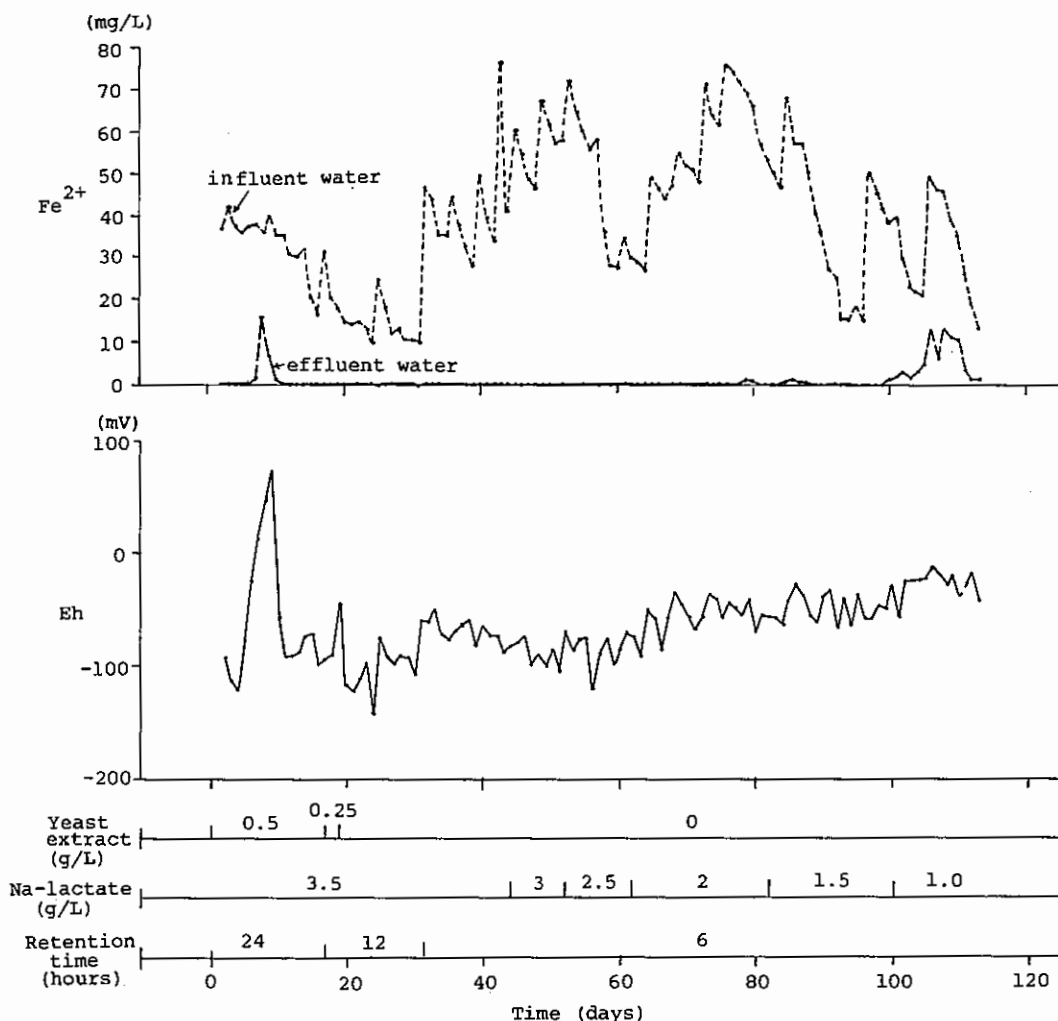


Figure 4.--Result of pilot plant-scale test.

### Results

Most of the metals in the mine drainage were separated into precipitate of sulfides consistently as was the case in the laboratory experiment. The results of the test are shown in figure 4. The  $\text{Fe}^{2+}$  and  $\text{Zn}^{2+}$  ions in the feed water were reduced to  $\text{Fe}^{2+} < 1.0 \text{ mg/L}$ ,  $\text{Zn}^{2+} < 0.2 \text{ mg/L}$ , below the emission standards in Japan, except for the period of the start-up and the very last stage. The cause of the increasing  $\text{Fe}^{2+}$  in the effluent at the start-up is not known. The same phenomenon at the end of the experiment was clearly due to the decreasing of Na-lactate. The COD in the effluent, however, was 800-1,000 mg/L throughout the test, which is over the standard.

The followings was concluded from the results of the pilot plant-scale test.

- (1) A cut in the amount of yeast extract was possible
- (2) Mine drainage with chemical quality of  $\text{Fe}^{2+} 40 \text{ mg/L}$ ,  $\text{Zn}^{2+} 40 \text{ mg/L}$  and  $\text{pH} 4.7$  was treated successfully at  $12^\circ\text{C}$ , sodium lactate level at  $1.5 \text{ g/L}$ , and retention time of 6 hours (720 L/day inflow)

- (3) Concentration of the bacteria remained at from  $4-5 \times 10^9$  cells/mL as same as was in the laboratory experiment.

### CONCLUSIONS

A continuous flow test of a biological method for treatment of mine drainage at Yanahara Mine showed that most of the metals in the mine drainage can be reduced in a pilot plant-scale process when suitable organic carbon for sulfate-reducing bacteria is supplied, and inflow and temperature of the reactions are properly controlled.

However, the following problems must be solved to complete a system for practical use:

- (1) The COD value in the effluent must be decreased to within the emission standard
- (2) Cheaper nutrients must be selected in order to hold cost down.

#### ACKNOWLEDGEMENT

This work was performed as one of the projects of research and development for mining-related pollution control by the Metal Mining Agency of Japan. The author wishes to thank all the members concerned with the project, particularly Dr. Imai, a professor of Okayama College of Commerce, for his presenting useful bacteria, Desulfovibrio vulgaris.

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[http://dx.doi.org/10.1016/0043-1354\(85\)90346-X](http://dx.doi.org/10.1016/0043-1354(85)90346-X)

[http://dx.doi.org/10.1016/0043-1354\(87\)90042-X](http://dx.doi.org/10.1016/0043-1354(87)90042-X)