Long-term evaluation of acid rock drainage mitigation measures in large lysimeters

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Abstract

In the course of a six year project in the framework of a German-Romanian scientific cooperation, large lysimeters with a volume of up to 65 m^3 were used for up to three years long experiments on controlling acid rock drainage formation in three different types of pyritic mine waste: (A) weathered six years old waste rock material, (B) sorted, freshly broken, low-grade ore, and (C) unweathered tailings material. An addition of sodiumdodecylsulphate (SDS) partly reduced the activity and number of metal sulphide oxidizing bacteria of the type *Acidithiobacillus ferrooxidans* but did not kill the bacteria. Consequently, the release of metals and sulphate from mine waste was not significantly reduced. The application of the biocide isothiazolinone (Kathon RH 886) reduced the release of Cu, Zn, Fe and sulphate between 5 and 50 % for different types of mine waste. A cover of organic material only achieved a moderate reduction of the heavy metal and sulphate release from the weathered six years old waste rock material: Cu was reduced by 50 %, Zn by 30 %, Fe by 90 %, and sulphate by 40%.

Keywords: Acid mine drainage, Acid rock drainage, Biocides, Microcalorimetry, Mine waste, Pyrite oxidation

1. Introduction

Abatement of acid rock drainage (ARD) is of increasing scientific and technical interest because of stringent regulations regarding environmental pollution. Acidity and dissolved heavy metals released from different sources of mine waste including underground and open pit mines, mine waste rock deposits, and tailings heaps and ponds result in a deterioration of soil and water quality. For new mine waste deposits high safety standards have been established in several countries to prevent ARD generation. Substantial costs are required for mine waste remediation. Measures for ARD prevention and safe storage of sulphidic mine waste have been thoroughly evaluated. Results of large scale experiments using control of water- and oxygen-flow, under water storage, or application of pH-regulating or inhibitory substances are well known (Dugan and Apel, 1983; Onysko et al., 1984; Erickson et al., 1985; Backes et al., 1988; Watzlaf et al., 1988; Evangelou, 1995; Rastogi, 1995; Schüring et al., 1997; Schippers et al., 2000; Mudder and Olson, 2004), but little information is available about the long term effect of mitigation measures on metal sulphide oxidizing, bioleaching bacteria (Ledin and Pedersen, 1996).

In the course of a long-term-project in the framework of a German-Romanian scientific cooperation, large mine waste lysimeters were constructed and used for experiments on ARD abatement (Cosma et al., 1999; Jozsa et al., 1999; Schippers et al., 2001). Geochemical and microbiological data were collected over a period of several years. Additionally, a technique based on microcalorimetry was applied to rapidly evaluate the effects of abatement measures. This paper summarizes the most relevant findings of the six year project on mitigation of ARD.

2. Materials and methods

The following subsections describe the experimental setup, the material used, and how the experiments were run and samples analyzed.

The experiments were carried out in the Cu-, Pb-, and Zn-mine Ilba near Baia Mare, Maramures, in North-West Romania. The underground mine has been in operation for more than 40 years. The main waste dump consisted of sulphidic sedimentary rock, was about 5 m high and was located on the bottom of a valley surrounded by about 500 m high mountains. The climate in that area is humid. During the summer moderate temperatures prevail and in the winter there are freezing temperatures and snow.

2.1. Lysimeters

The lysimeters were installed into the main waste dump to ensure natural climatic conditions. The four-chamber percolator (4CP) was constructed of reinforced concrete and was protected against acid attack and mechanical stress by spray-application of CORODUR 2K® (Lehmann&Voss&Co., Hamburg, Germany). It consisted of four separate lysimeters with a volume of 65 m^3, each. Approximately the same amount of waste material (ca. 120 t) was excavated in the vicinity of the 4CP for two additional chambers each. A HDPE-liner was put in these chambers for isolation and the waste rock was replaced to fill the excavations. The six column percolator (6CP) was made of prefabricated concrete pipe segments and contained 2.3 m^3 waste material in each lysimeter column. The overall height of the waste in all devices was approximately 3 m. Sampling of solids from different depths in the lysimeters through hollow PVC pipes with a diameter of 100 mm, and a separate, quantitative collection of the effluents was possible.

2.2. Mine waste and tailings material

The waste material used for the experiments in the 4CP and the excavations consisted of weathered six year old run of mine waste rock material from the main waste dump. The entire material was of brownish colour due to iron precipitates. The rocky material showed a lot of cracks and cleavage surfaces, which were also partly brown, indicating that iron precipitation also occurred at inner surfaces inside the rocks. In the 6CP three materials of different grain size were tested: (A) weathered waste rock from the main waste dump (grain size <100 mm), (B) the 4-30 mm fraction selected by screening from freshly crushed waste rock, and (C) an unweathered, coarse flotation tailings material (grain size less than 1 mm, with 10 % addition of fresh flotation pyrite concentrate to increase the amount of leached ions). The composition of the materials in the different lysimeters is given in Table 1.

2.3. Experiment description

Two different biocides and a cover of organic material were tested for their efficiency to reduce bacterial metal sulphide oxidation (bioleaching) activity and release of metals and sulphate from the waste. Each experiment was run with a control in which the same type of mine waste was used and no inhibitory measures were applied. The experiments lasted between two and three years. For a balance of metal- and sulphate-output, all effluents were collected at least once a week and measured for volume, pH, concentration of Cu, Zn, Fe, Mn, Mg, Ca, Al, Si, and sulphate. The cumulative release for each compound was calculated by summarizing the products of volume and concentration for each effluent sample.

The experiments with biocide addition were run either with sodiumdodecylsulphate (SDS) or with isothiazolinone (Kathon RH 886 of Rohm & Haas). SDS was applied by spraying 100 L SDS-solution in a concentration of ca. 50 g/L on the surface of the waste in one chamber of the 4CP, followed by recirculation of the SDS-containing effluent several times, to achieve a thorough distribution. Two SDS-applications of 5 kg, each, were performed between day 130 and 200 and between day 330 and 365, respectively. Seven additional applications of 2 kg SDS, each, were performed later on (between days 484 and 680). Altogether 24 kg of SDS were applied until day 680 corresponding to a dosage of about 200 g/t of waste rock. The SDS concentration in the effluent fluctuated between 0 and 100 mg/L (Jozsa et al. 1999). At the 720th day of the SDS-experiment an addition of 15 kg solid SDS was combined with the application of a similar cover of organic rich material and crushed limestone as described below.

Isothiazolinone was applied in a concentration of 100 mg/L on the surface of the three different types of waste in the 6CP, followed by recirculation of the isothiazolinone-containing effluent several times to achieve a thorough distribution. After each recirculation, biocide was added to maintain its concentration at 100 mg/L.

In one excavation an 1.2 m thick cover of organic material (as recommended by SAARMontan GmbH, Saarbrücken, Germany) consisted of a mixture of organic rich material such as top soil, fermented pig manure and saw-dust (2:1:1 by volume). It was separated from the mine waste below by a 15 cm thick intermediate layer of crushed limestone (grain size 7-20 mm).

During the experiments air temperatures ranged from - 20°C to 40°C but simultaneous temperature measurements inside the lysimeters at different depths showed only a temperature fluctuation between 0°C and 25°C.

2.4. Analysis

Chemical analysis of the effluents was performed by ICP, AAS, and IC. Additionally, the content of water soluble species was determined in water extracts of waste material from different depths at the end of the experiments. Isothiazolinone and SDS were determined spectrophotometrically, the latter via chloroform extraction in the presence of methylene blue. Approximately every six weeks, solid samples were taken from different depths of each lysimeter for microbiological analysis as described by Schippers et al. (1995). Briefly, cell numbers of metal sulphide oxidizing, *Acidithiobacillus ferrooxidans*-like bacteria were determined by the most-probable-number (MPN) technique using a selective culture medium for acidophilic Fe(II)-oxidizers. The bioleaching activity (biological pyrite oxidation) was measured at 30°C by microcalorimetry based on the heat output due to the exothermic pyrite oxidation (Rohwerder et al., 1998; 1999; Elberling et al., 2000).

3. Results and discussion

In the effluents of the control experiment in the 4CP, a release of Cu, Zn, Mn, and Fe from the waste was observed (Fig. 1a). Cu and Zn showed an on-going release during the experiment, whereas Fe increased only during the first 630 days. Afterwards the increase of the Fe output became less significant. The release of Fe and Zn after SDS-addition increased over the values of the control experiment (Fig. 1b). The cumulative release of Mg, Ca, Al, and sulphate is not significantly different for the control- and SDS-experiments (Fig. 2). It is apparent that all elements and sulphate continue to be further mobilized during the experiment. The pH was measured at four different depths of the waste rock. The values fluctuate mainly between 3 and 5 for both experiments, as given in Fig. 3.

For the control experiment, the cell numbers of *Acidithiobacillus ferrooxidans*-like bacteria per g waste rock (ww) are shown in Fig. 4a. The values ranged in general between 10^4 cells/g and 10^8 cells/g waste rock. For the SDS-experiment the cell numbers of *Acidithiobacillus ferrooxidans*-like bacteria (Fig. 4b) were generally lower than those of the control experiment at all depths. After the second SDS-addition at about 350 days the cell numbers declined and at several time points cells were not detectable. The strong fluctuation of the cell numbers might be explained by the strong variation of the SDS concentrations between 0 and 100 mg/L in the effluents (Jozsa et al. 1999). Overall, the SDS concentration was not high enough to kill the bioleaching bacteria at all depths of the waste rock. The bioleaching bacteria could grow again, whenever the SDS concentration decreased due to bacterial degradation or washing out.

The interpretations based on the cell numbers are corroborated by the measurements of bioleaching activity (biological metal sulphide oxidation) using microcalorimetry (Fig. 5). An overall decrease of the bioleaching activity over the time of the control experiment was found. Whereas most values fell around 10 µW/g waste rock indicating moderate bioleaching activity, at a depth of 1.3 m the values generally ranged around 20 µW/g waste rock and included several even higher readings with up to 55µW/g waste rock. SDS caused a considerable reduction of the bioleaching activity as shown in Fig. 5b. Most values were below 10 µW/g waste rock indicating a low bioleaching activity.

On the one hand, the inhibition of the bioleaching activity was obviously not strong enough to cause a significant reduction of the metal and sulphate release (Fig. 1 and 2). On the other hand, due to the improved wetting of the waste rock by the detergent SDS, wash-out of metal salts might be increased, causing a considerable increase in metal release from SDS treated waste even without further bioleaching. After pyrite oxidation some sulphate remained in the waste as precipitates presumably as metal sulphates, gypsum and jarosite. The analysis of water-soluble compounds in the waste (waste-resident) at the end of the experiment presented in Table 2 showed that significantly more water-soluble metal species remained waste-resident in the control- than in the SDS-experiment. Improved wash-out in the SDS-experiment (due to recirculation and surfactant effects of SDS) is the most likely reason. The waste-resident species remained probably in the cracks and crevices of the solid waste and may become mobilized by flooding. Conclusively, a reduction of the bioleaching activity could have been compensated by the increased wash-out of metal salts, which could explain the similar cumulative metal and sulphate release for the control- and for the SDS-experiment.

The data for the second biocide isothiazolinone (Kathon) with three different types of waste material in the 6CP are summarized in Fig. 6. Although in laboratory experiments the biocide has been previously shown to completely inhibit bioleaching (Schippers et al., 2000), the data presented here indicate that at best a reduction of 50 % cumulative metal and sulphate release could be achieved. Bioleaching bacteria and their activity in the 6CP lysimeters could still be detected (data not shown). Obviously the biocide concentration of 100 mg/L after several times recirculation in the 6CP was too low to kill the bioleaching bacteria probably due to poor biocide distribution. The material specific biocide consumption to achieve the steady-state concentration of 100 mg/L was 1.12 g/t for the weathered waste rock, 3.73 g/t for the freshly crushed waste rock, and 0.75 g/t for the flotation tailings.

Results for the experiment with a cover of organic material in the excavations are summarized in Fig. 7. For the metals Cu, Zn, Fe, Mg, Ca, and sulphate the results indicate that after 900 days the cumulative leaching output was reduced under the organic cover, but may still be considerably high: Cu was reduced by 50 %, Zn by 30 %, Fe by 90 %, Mg by only 10 %, Ca not at all, and sulphate by 40%. Thus, the cover of organic material only achieved a moderate reduction of the heavy metal and sulphate release. Since a limestone layer was applied together with the cover of organic material, Mg and Ca release from the limestone may have superimposed the effect of the cover on Mg and Ca release from the waste rock. Bioleaching bacteria and their activity in the excavations could still be detected (data not shown). Obviously the cover did not completely inhibit the bioleaching bacteria in the underlying mine waste.

Another reason for a high metal release in the SDS- and the organic cover-experiments may be the formation of water soluble metal-organic complexes via biodegradation of the added organic substrates.

4. Conclusions

The results of this study show that the tested measures to prevent ARD formation in pyritic mine waste only partly reduced the activity and number of metal sulfide oxidizing bacteria and the release of heavy metals and sulphate. SDS only transiently reduced the number and activity of bioleaching bacteria. Isothiazolinone and the cover of organic material and crushed limestone on mine waste reduced the release of metals and sulphate to some extent.

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Fig. 1. Cumulative release of heavy metals (kg) and pH of the effluent of the a) control- and b) SDS-experiment with weathered waste rock material. Mn was only measured until the 560th day due to technical problems.

Fig. 2. Cumulative release of other metals and sulphate (kg) of the effluent of the a) control- and b) SDS-experiment with weathered waste rock material. Si was measured for a limited time for the control-experiment only, since its release was low.

Fig. 3. pH in various depths of the mine waste of the a) control- and b) SDS-experiment with weathered waste rock material.

Fig. 4. Cell numbers of *Acidithiobacillus ferrooxidans*-like bacteria determined by the most-probable-number method (log [MPN/g (ww)]) in various depths of the mine waste of the a) control- and b) SDS-experiment with weathered waste rock material.

Fig. 5. Bioleaching activity measured as heat output (µW/g (ww)) by microcalorimetry in various depths of the mine waste of the a) control- and b) SDS-experiment with weathered waste rock material.

Fig. 6. Influence of biocide application (isothiazolinone) on the cumulative metal and sulphate release after 420 d of bioleaching for three types of mine waste in comparison to the different control-experiments (100 % efficiency).

Fig. 7. Effect of a cover of organic material and crushed limestone on the release of metals and sulphate (kg) from weathered waste rock material in the excavations within 900 days of bioleaching compared to the control-experiment.

Table 1

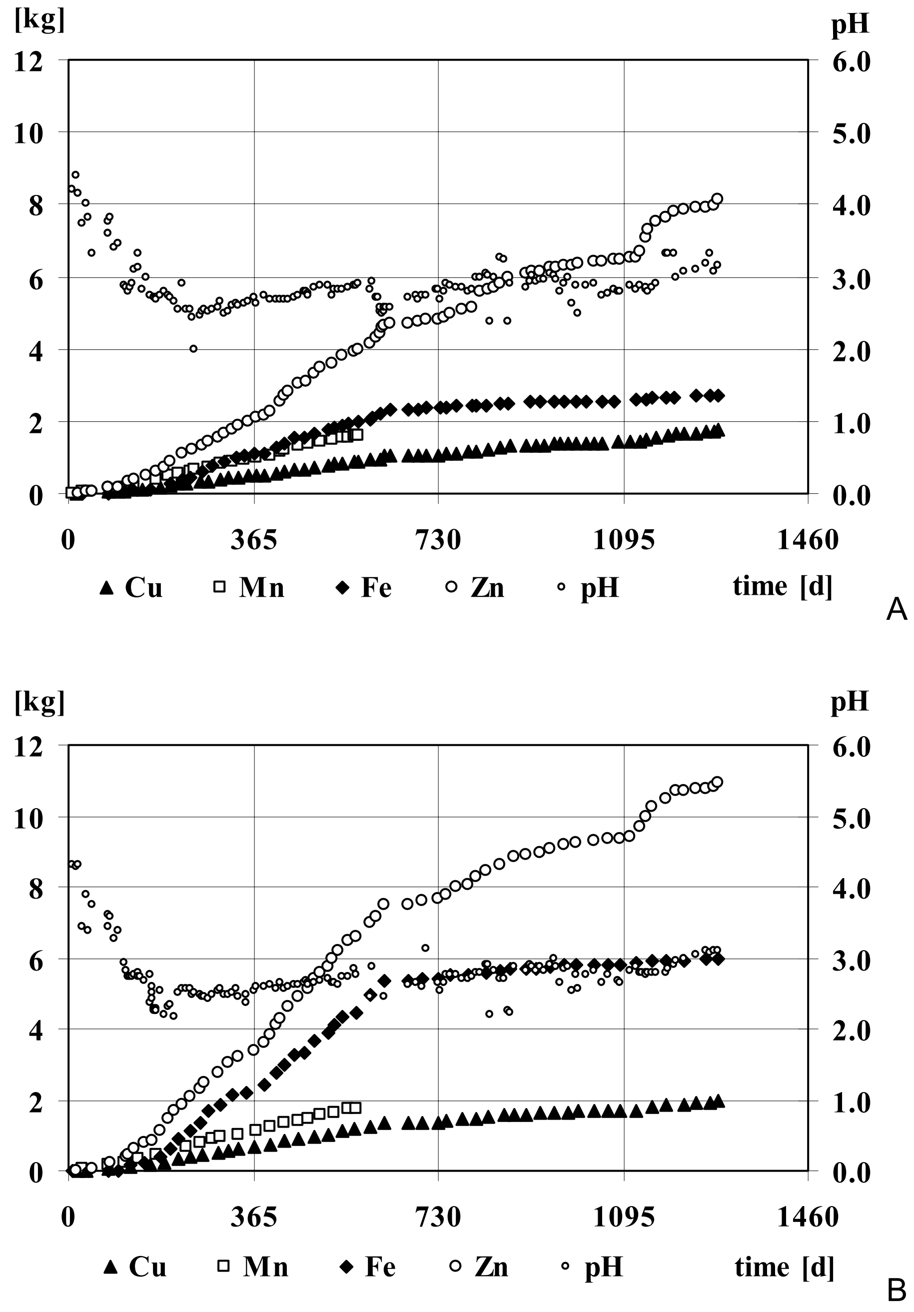
Chemical composition of the mine waste used for lysimeter experiments (mean values of at least three samples; 4CP = four-chamber-percolator; Excav. = excavations; 6CP = six column-percolator, SDS = sodiumdodecylsulphate; *Pyrite content was calculated from Fe and S values; nd = not detected)

Experiment	Grain size	Total amount [% (w/w)]												
	[mm]	Al_2O_3	CaO	MgO	Na_2O	K_2O	SiO_2	Cu	Zn	Mn	Pb	Fe	S	*Pyrite
4CP (control)	<400	15.00	3.75	1.78	1.33	4.38	64.50	0.05	0.14	nd	0.20	4.38	2.03	3.79
4CP (SDS)	<400	12.06	0.85	1.03	0.77	4.57	65.15	0.07	0.05	nd	0.42	4.76	1.87	3.50
Excav. (control)	<400	12.06	0.85	1.03	0.77	4.57	65.15	0.07	0.05	nd	0.42	4.76	1.87	3.50
Excav. (cover)	<400	12.08	0.86	1.13	0.80	4.60	64.58	0.06	0.06	nd	0.46	5.60	1.78	3.34
6CP (weathered)	<100	12.49	2.34	2.43	2.00	3.72	61.33	0.03	0.09	0.07	0.07	6.04	2.22	4.16
6CP (crushed)	4-30	2.44	3.77	2.65	3.86	8.17	61.05	0.05	0.17	0.10	0.09	4.73	2.19	4.09
6CP (tailings)	<1	1.11	6.35	1.69	3.20	4.85	59.41	0.04	0.40	0.21	0.11	7.71	8.74	16.37

Table 2

Content of water-soluble species in the waste material (waste-resident) at different depths at the end of the 3.5 years long control- and SDS-experiments in the 4CP (SDS = sodiumdodecylsulphate, nd = not detected)

Depth	Sulphate [g/t]		Cu [g/t]		Zn [g/t]		Mn [g/t]		Fe [g/t]	
[m]	Control	SDS	Control	SDS	Control	SDS	Control	SDS	Control	SDS
0.0	161	nd	0.18	nd	1.85	nd	0.95	nd	1.24	nd
0.2	74	242	0.92	nd	4.67	0.46	1.69	0.27	0.73	0.01
0.4	198	311	1.08	0.50	3.16	2.65	1.98	1.31	0.60	nd
0.6	594	460	1.59	0.42	5.52	2.00	2.60	1.14	0.57	0.04
0.9	757	433	3.08	0.44	8.84	6.12	3.34	2.03	1.65	nd
1.2	784	548	3.46	nd	12.46	3.69	3.15	1.19	1.87	nd
1.5	725	643	2.71	nd	12.53	2.68	3.09	1.08	1.61	nd
1.8	820	624	3.35	nd	10.66	1.88	3.41	0.78	1.36	nd
2.1	1023	666	8.41	nd	25.10	0.71	6.49	0.03	0.61	nd
2.4	861	672	4.51	0.45	13.71	8.69	4.65	2.89	0.35	nd
2.7	1102	927	4.74	1.54	14.02	29.02	4.81	3.23	0.16	0.03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4
2
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6.0
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4.0
3.0
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0.0
0
365
730
1095
1460
▲ Cu
□ Mn
◆ Fe
○ Zn
∘ pH
time [d]
A
[kg]
pH
12
10
8
6
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2
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▲ Cu
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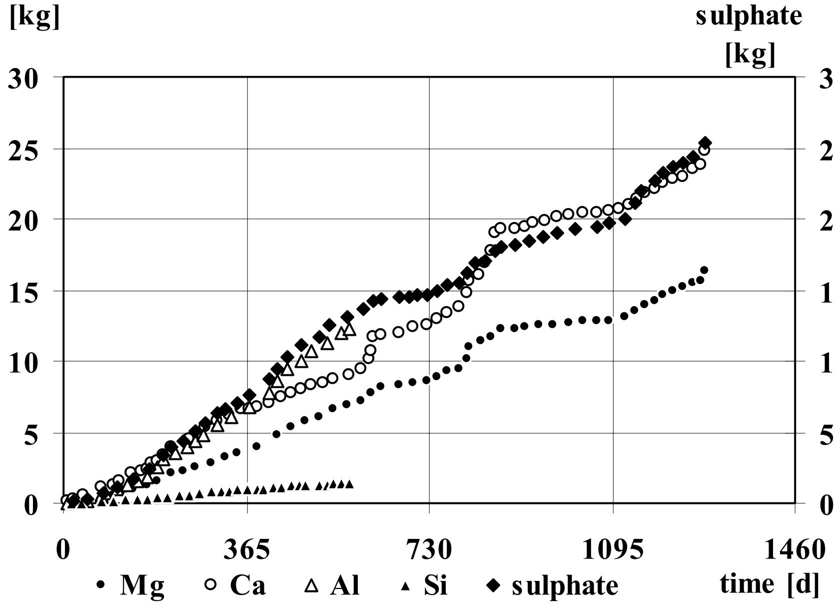
∘ pH
time [d]
B

FIG. 1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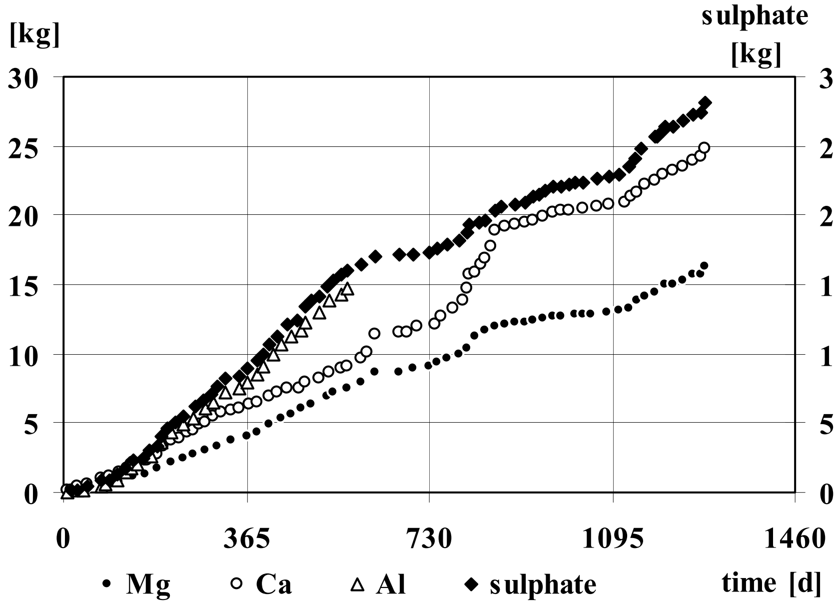
▲ Si
◆ sulphate
time [d]

A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300
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730
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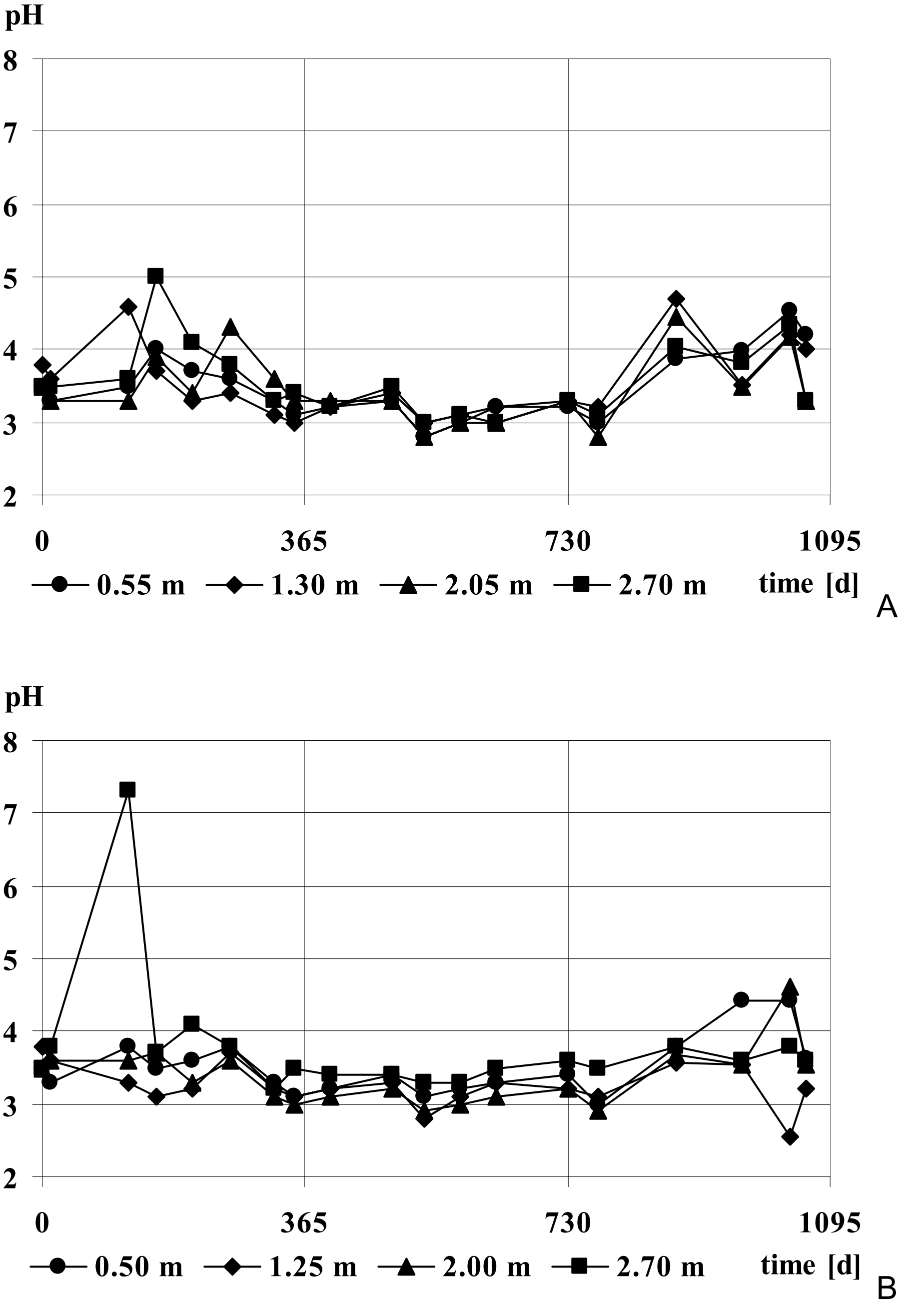
1460
• Mg
○ Ca
△ Al
◆ sulphate
time [d]

B

FIG.2


pH
8
7
6
5
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3
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365
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0.55 m
1.30 m
2.05 m
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time [d]
A
pH
8
7
6
5
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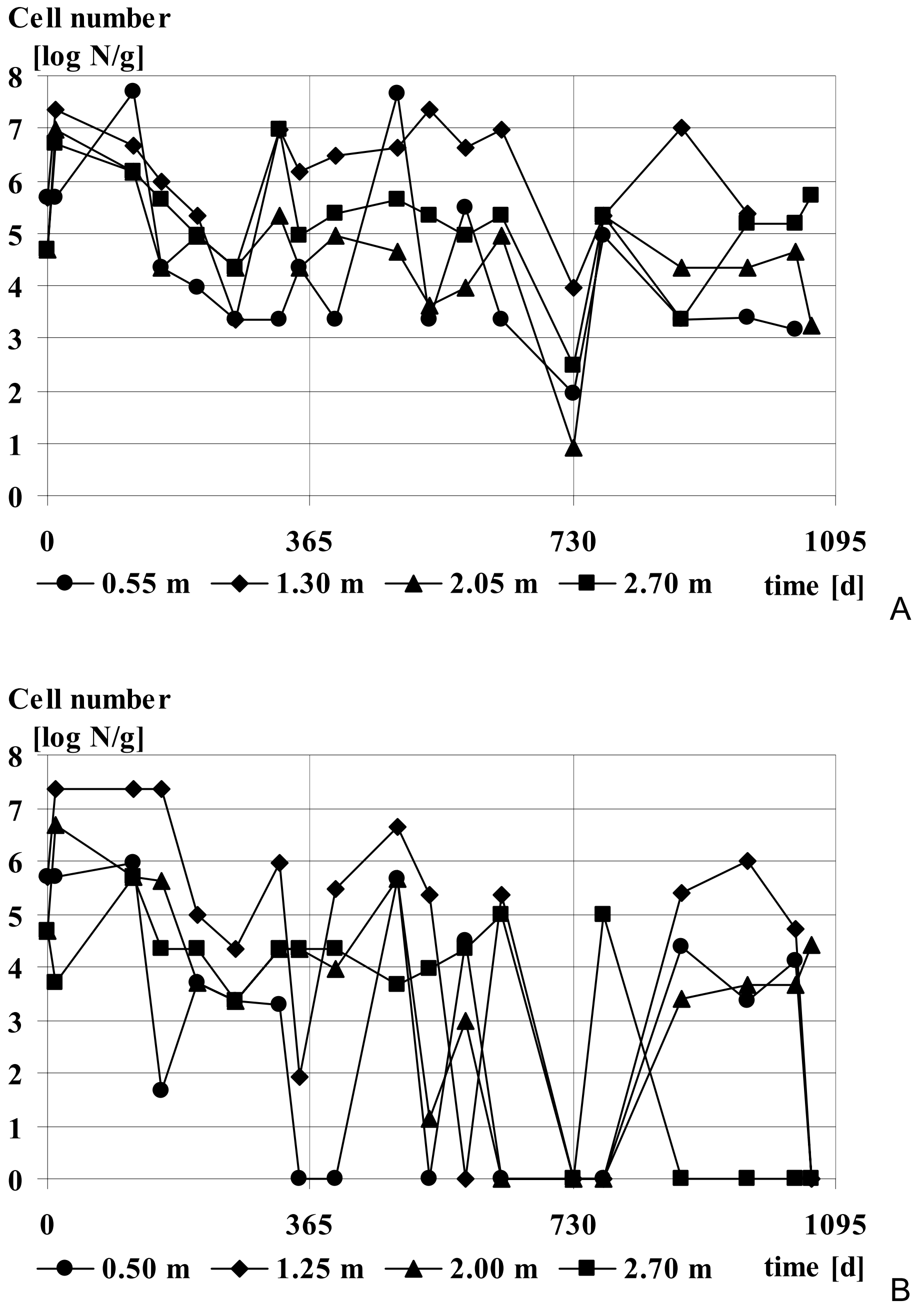
1095
0.50 m
1.25 m
2.00 m
2.70 m
time [d]
B

FIG. 3


Cell number
[log N/g]
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7
6
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4
3
2
1
0
0
365
730
1095
0.55 m
1.30 m
2.05 m
2.70 m
time [d]
A
Cell number
[log N/g]
8
7
6
5
4
3
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365
730
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0.50 m
1.25 m
2.00 m
2.70 m
time [d]
B

FIG.4

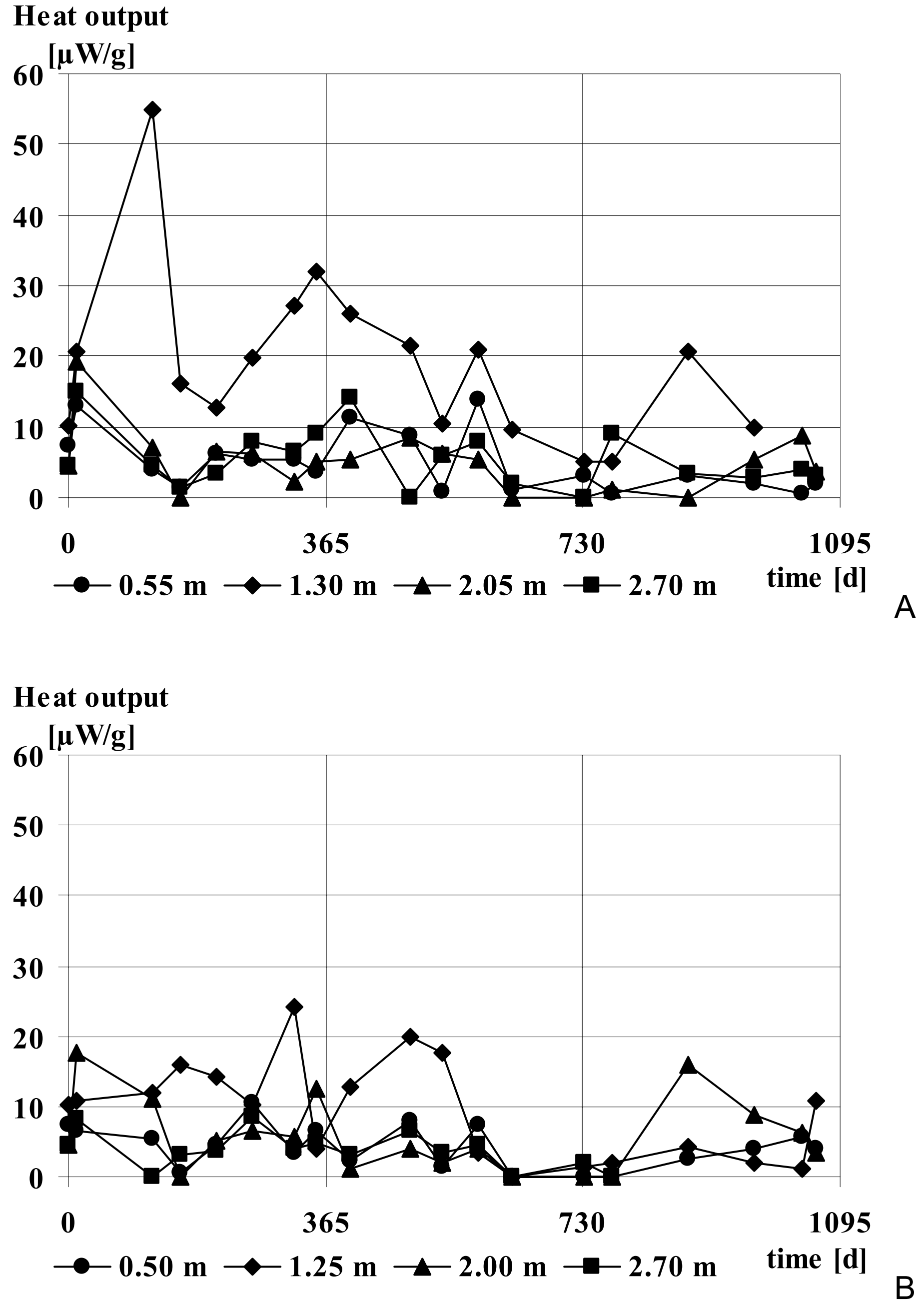
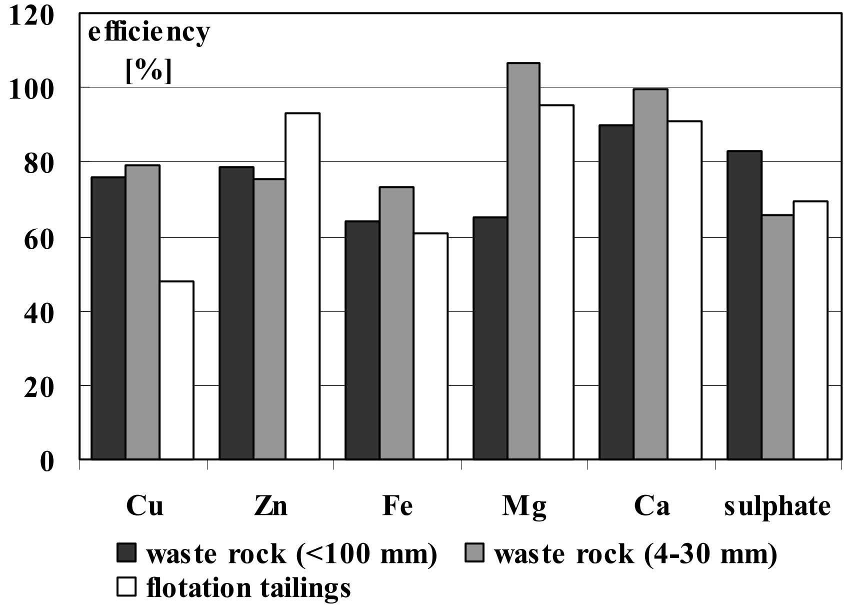

Heat output
[µW/g]
60
50
40
30
20
10
0
0
365
730
1095
time [d]
0.55 m
1.30 m
2.05 m
2.70 m
A
Heat output
[µW/g]
60
50
40
30
20
10
0
0
365
730
1095
time [d]
0.50 m
1.25 m
2.00 m
2.70 m
B

FIG.5



120
100
80
60
40
20
0
efficiency [%]
Cu
Zn
Fe
Mg
Ca
sulphate
waste rock (<100 mm)
waste rock (4-30 mm)
flotation tailings

FIG.6

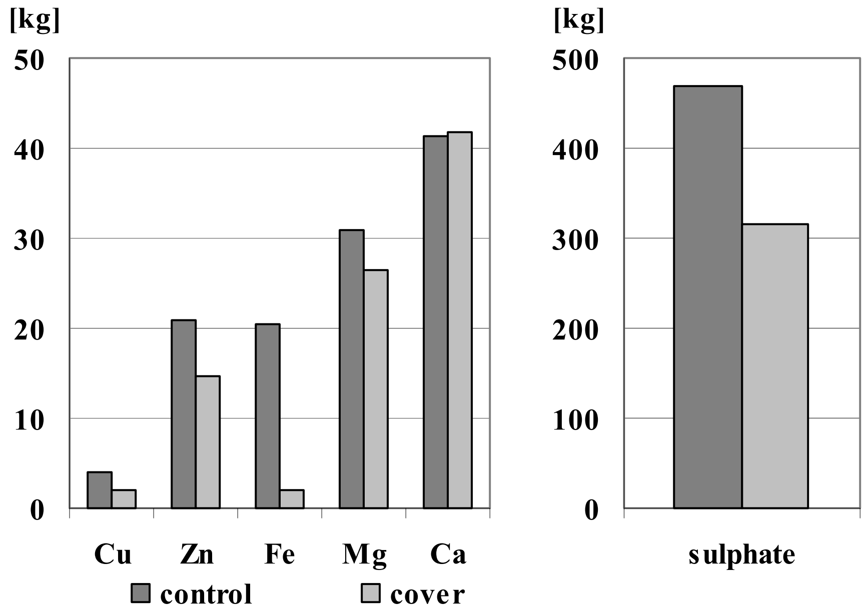

[kg]
50
40
30
20
10
0
Cu
Zn
Fe
Mg
Ca
control
cover
[kg]
500
400
300
200
100
0
sulphate

FIG.7