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Possibilities of acid mine drainage treatment in Sokolovská uhelná, Czech Republic

Introduction

One of the main tasks in exploitation of mineral deposits in open-pit mines is also cause the contamination of ground and surface waters with toxic chemicals and metals, which are presented in acid mine water [11].

Acid mine water (AMD), spoil heaps and sludge beds are typical remains of mining activities, which regeneration and cleaning to eliminate and minimize environmental impacts are costly from technical and technological point of view.

Mine waters belong to unique waters defined by the §40, Directive No. 254/2001 (Water Low) and by §40, Directive No. 44/1988 (Mining Low). These waters are all underground, surface and precipitation waters, that penetrate into strip or underground mine areas from overburden, subsoil, walls or simply like inflow of precipitation waters until their connection with others surface or ground waters.
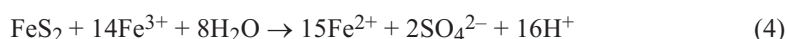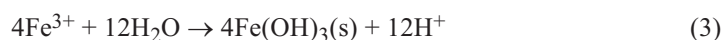
Mining activities at the open pit Jiří and depth Jiří generated AMD, which results as a consequence of a complex series of geochemical reactions that occur when sulphide minerals are exposed to the atmosphere under an oxidizing environment, producing polluted waters strongly acidic with high concentrations of iron, manganese and sulphate, responsible for the resulting damage to health of aquatic flora and fauna. Although AMD is naturally occurring, underground and open pit mining activities may greatly accelerate its production because they expose fresh iron and sulphide surfaces to oxygen. Predictions of the future

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loading of dissolved metals from inactive mine sites suggest that sulphide oxidation and the release of dissolved metals will continue for decades to centuries. Iron sulphide minerals, especially pyrite (FeS_2), contribute the most to formation of AMD. The pyrite oxidation process has been extensively studied and can be summarized by the following reaction sequence [8]:

$$FeS_2 + 3.5O_2 + H_2O \rightarrow Fe^{2+} + 2SO_4{}^{2-} + 2H^+ \tag{1}$$

$$4Fe^{2+} + O_2 + 4H^+ \rightarrow 4Fe^{3+} + 2H_2O \tag{2}$$

$$4Fe^{3+} + 12H_2O \rightarrow 4Fe(OH)_3(s) + 12H^+ \tag{3}$$

$$FeS_2 + 14Fe^{3+} + 8H_2O \rightarrow 15Fe^{2+} + 2SO_4{}^{2-} + 16H^+ \tag{4}$$

During the first step, pyrite reacts with oxygen and water, producing Fe^{2+} and sulphuric acid by reaction (1). The second step involves the oxidation of Fe^{2+} to Fe^{3+} by reaction (2), which is pH-dependent. The third step corresponds to the hydrolysis of Fe^{3+} with water to form a ferric hydroxide precipitate (ferrihydrite) and the release of additional acidity, which is promoted by the pH-dependent reaction (3). The fourth step is defined as the oxidation of additional pyrite by Fe^{3+} according to reaction (4). The pyrite oxidation is controlled by bacterial species that have definite pH growth range and pH growth optimum [8]. The conversion of Fe^{2+} to Fe^{3+} in the overall pyrite reaction sequence has been described as the rate determining step [16], which can be greatly accelerated by bacterial action. In AMD there are many mineral-degrading acidophiles involved in the metabolism of pyrite and the metabolic pathways involved in this process are pyrite dissolution, iron oxidation, iron reduction, and sulphur oxidation [8]. The most common pyrite-oxidizing bacterium is *Acidithiobacillus ferrooxidans*, which is of great practical importance due to the extensive acid and metal pollution generated when this species releases metals from acid mine waters [13]. Once pyrite oxidation and acid production has begun, conditions are favorable for bacteria to further accelerate the reaction rate. At pH values of about 6 and above, bacterial activity is thought to be insignificant or comparable to abiotic reaction rates. The knowledge of the mineralogical processes occur during the atmospheric oxidation of pyrite and other sulphide minerals in the presence of oxidizing bacteria and any other product generated as a consequence of oxidation reactions (e.g., heavy metals solubilised by acid solutions) is very useful in both the prediction of AMD and its treatment. Traditionally, the treatment of AMDs consists in the neutralization with limestone ($CaCO_3$) or similar materials, resulting in the precipitation of Fe and other metal hydroxides as well as also gypsum ($CaSO_4$). AMD treatment with limestone can increase the pH to 6.0–7.5 [12], allowing the metals to be removed from solution. Several procedures for AMD treatment have been developed [1, 2, 3, 4, 5, 7, 9, 10, 15, 17, 18], although adsorption has been the preferred method for heavy metal removal, because it is considered a particularly effective technique. There are two general

categories of AMD treatment which are well investigated: (1) active treatment, which requires the use of chemical treatment system to buffer acidity, and (2) passive treatment, which allows naturally occurring chemical and biological processes to do the work in a controlled system outside of the receiving polluted effluent. However, AMD remediation can be very costly and difficult due to the high costs of activated carbon's production and regeneration for water treatment [14].

The local AMD are characterized by high content of sulphates, iron, manganese and low pH. Before their releasing into the water body the mine waters are processed in the Water Preparing Plant Svatava (WPPS), where mainly high concentration of iron and suspended solids are removed. Also low pH rates are conditioned. The ways and conditions for discharging of mine waters into the Svatava River are given by the Resolution of The Water Right Office (Environmental section of Karlovy Vary Area Regional Council, reference number 655/ZZ/10-5). This Resolution is valid till 30.4.2014. After processing in the WPPS mine waters are discharged into Svatava River. The amount of these waters is $Q_{average}$= = 400 $dm^3 \cdot s^{-1}$, Q_{max}= 500 $dm^3 \cdot s^{-1}$, Q_{year} = 12,615,000 $m^3 \cdot year^{-1}$. The peak capacity of the discharged waters is given by the WPPS potential and in this time it is 400 $dm^3 \cdot s^{-1}$. Emission and specific limits of the discharged waters are given by the mentioned Resolution as follows (Table 1).

TABLE 1

Emission of some parameters and specific limits

TABELA 1

Dopuszczalne zawartości wybranych składników

Indicator	Emission limit [mg·dm^{-3}]		Specific limit [t·year^{-1}]
	p	m	
NL$_{105}$	20	40	252.3
Fe$_{total}$	3	8	37.8
Mn	1	4	12.6
SO$_4^{2-}$	1 200	1 500	15 138.0
pH		6–9	

Where: p – permissible value, m – maximum value

The conditioning of mine waters in WPPS consists in alkalinization by lime suspension, oxidation by air oxygen of bivalent iron Fe^{2+} to trivalent form Fe^{3+}, separation of sediments from the purified water inside two Seclar reactors and succesive gravitational and mechanical dewatering of sediments running in composite, equalizing and thickening tanks and subsequently in the Netzsch filter press.

1. Materials and methods

1.1. Sampling sites

AMD from the depth Jiří and also open pit Jiří, specifically from the pumping station (PS) Lomnice, which is very problematic from the point of their composition, inflow to the WPPS. With respect to fluctuating water flow from particular areas during the year, the separate samples were taken and on the basis of average pumping in 2010 the ratio was calculated, in which these samples were mixed. Particular and mixed samples were treated by input analyses. The sample of mixed AMD was further processed with regard to elimination of iron, manganese, sulphates and set-up of pH.

The ratio, in which the mixture of particular AMD was prepared, was calculated from quantity of incoming AMD from particular areas pumping station Lomnice and depth Jiří, see Table 2.

TABLE 2

Quantity and share in total of incoming AMD from particular areas

TABELA 2

Ilości i udział objętościowy wód kopalnianych z poszczególnych miejsc poboru

Year	PS LOMNICE [m^3]	Depth JIŘÍ [m^3]
2010	754 352	958 845
Share in total	44%	56%

Information about the water composition from the depth Jiří was extended by company Sokolovská uhelná. The results shown in the table 3 are the average value from year 2010. The samples of AMD from open pit Jiří, in the concrete from PS Lomnice, were chemically analyzed with the help of Merck high-speed testers straight in situ. The results are shown in the Table 3.

TABLE 3

Composition of AMD from depth Jiří and PS Lomnice

TABELA 3

Skład wód kopalnianych z kopalni Jiří i pompowni Lomnicy

Locality	pH	c(Fe) [mg·dm^{-3}]	c(Mn) [mg·dm^{-3}]	c(SO$_4^{2-}$) [mg·dm^{-3}]
Depth Jiří	6.8	26	2	332
PS Lomnice	2.5	135	8	1540

The samples of AMD were collected and immediately mixed in situ in ratio that was calculated on the basis of the above flows of AMD.

The basic chemical analysis was made, by the help of Merck high-speed testers, immediately after mixing in situ to avoid possible changes in chemical composition of mine waters, see Table 4. The waters were then transported to the Water Technology Laboratory of VŠB – Technical University of Ostrava.

TABLE 4

Composition of mixed AMD sample

TABELA 4

Skład próbki wód kopalnianych stosowanych w badaniach

pH	$c(Fe)$ [mg·dm^{-3}]	$c(Mn)$ [mg·dm^{-3}]	$c(SO_4^{2-})$ [mg·dm^{-3}]
3.5	40	8	1150

1.2. Iron, manganese and sulphate removal and pH adjustment methods

pH adjustment and iron removal was made by the dosing of calcium hydroxide in the form of suspension (10% whitewater).

As for the manganese removal, it was tested by combination of pH adjustment (calcium hydroxide) and oxidation (potassium permanganate) [6].

As for the sulphate removal, there was used the procedure of chemical precipitation of sulphates developed in the Institute of environmental engineering, VŠB – Technical University of Ostrava. This procedure using the combination of ions Ca^{2+}, OH^-, Al^{3+}, AlO^{2-}. The course of precipitation mechanisms is probably the following:

$$3SO_4^{2-} + 6Ca^{2+} + 2AlO^{2-} + 4OH^- + 29H_2O \rightarrow 3CaO \cdot Al_2O_3 \cdot 3CaSO_4 \cdot 31H_2O$$

Final product of the reaction is called ettringit. It is a complex compound of the relationship Al : SO$_4$: Ca = 2 : 3 : 6, which crystallizes in the form of characteristic needles. The formation of ettringit is supposed to be in water solutions containing $CaSO_4$, $Ca(OH)_2$ and aluminium compounds in the third stage of oxidation. Aluminium is in the process of precipitation added in the form of sodium aluminate [19].

1.3. Batch experiments and water analyses

All experiments were carried out on the stirring column MK6. pH adjustment, iron and manganese removal at a speed of 150 r·min^{-1}, sulphate removal at a speed of 250 r·min^{-1}.

Analysis of manganese was carried out by colorimetric test Microquant, Merck, and then according by ČSN 830540 part 16 [21].

Analysis of iron was carried out by colorimetric test Microquant, Merck, and then according by ČSN 830540 part 15 [21].

Sulphates were determined by using pre-programmed methods HACH No. 995 at a wavelength of 450 nm on a spectrophotometer HACH DR/2000 (Direct reading spectrophotometer) using reagent SulphaVer 4, which contains citric acid and barium chloride.

pH was determined by pH/mV Pocket Meter WTW pH 330i.

2. Results and discussion

2.1. pH adjustment and iron removal in raw AMD

pH of the raw AMD was 3.5 and the iron concentration was 40 $mg \cdot dm^{-3}$. The AMD was neutralized by means of $Ca(OH)_2$ in the form of 10% whitewater. Iron was transferred to insoluble form. Reaction time was 10 minutes, and then the suspension was filtrated. The pH value and concentrations of iron, manganese and sulphate were determined. Measured values are summarized in the Table 5.

TABLE 5

AMD composition after reaction with $Ca(OH)_2$ in the form of whitewater

TABELA 5

Skład wód po neutralizacji mlekiem wapiennym

Sample	$Ca(OH)_2$ (10% whitewater) [$g \cdot dm^{-3}$]	pH	c(Fe) [$mg \cdot dm^{-3}$]	c(Mn) [$mg \cdot dm^{-3}$]	c(SO_4^{2-}) [$mg \cdot dm^{-3}$]
1	0.088	5.85	<0.1	7.0	1 156
2	0.097	6.56	<0.1	6.7	1 162
3	0.106	7.09	<0.1	6.0	1 138
4	0.123	8.10	<0.1	3.0	1 122
5	0.141	8.37	<0.1	2.9	1 120

There is evident, that the optimal dosage for neutralization is 0.106 g. $Ca(OH)_2$ per 1 dm^3 of the raw AMD. And during the same process the iron was completely removed. The costs of the reagent – $Ca(OH)_2$ necessary to adjust 1 m^3 of AMD, are 0.37 CZK (0.013 Euro) at present.

### 2.2. Manganese removal in raw AMD

Demanganization was tested by combination of dosing of calcium hydroxide for pH adjustment and potassium permanganate for oxidation.

The input concentration of manganese in the raw AMD was 8 mg·dm^{-3}. After neutralization with calcium hydroxide CaOH)$_2$, the concentration of manganese, with the regard to the requirement of legislation,in the treated AMD wasstill high (about 6 mg·dm^{-3}), see Table 5, because a fraction of manganese precipitated in the form of hydroxides and hydrated oxides of manganese, another fraction stayed in a dissolved form. For the removal of manganese there was used a strong oxidizer – potassium permanganate KMnO$_4$. Due to the fact that the oxidation takes place preferably at neutral and slightly alkaline pH, the AMD is first neutralized with Ca(OH)$_2$. Experiments with potassium permanganate oxidation of manganese were carried out at pH 7 (procedure A) and pH 8 (procedure B).

2.2.1. The removal of manganese from AMD at pH 7 (A)

After neutralization, the concentration of manganese in AMD was about 6 mg·dm^{-3}.

The literature suggests [20] that according to the reaction:

$$3Mn^{2+} + 2MnO^-_4 + 2H_2O - 5MnO_2 + 4H^+$$
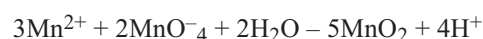
is necessary to dose 1.92 mg of oxidizing agent (KMnO$_4$) to convert 1 mg soluble divalent form of manganese into insoluble quadrivalent form. From this datathere was calculated the basic dose of potassium permanganate (11.52 mg·dm^{-3} KMnO$_4$), from which was derived multiples of this dose (0.6, 0.8, 0.9, 1.0, 1.1, 1.2;), which were then used for manganese removal (demanganization).

First, the stock sample (6 dm^3) of neutralized AMD was prepared according to the procedure above. Multiplicands of the basic dose of 0.1% solution of KMnO$_4$ were then added into each 1 dm^3 of neutralized water. Suspension was after reaction filtrated and pH, c(Fe), c(Mn), c(SO$_4$) were defined in the filtrate. The results are shown in Table 6.

TABLE 6

Results of AMD demanganization (pH 7) with KMnO$_4$

TABELA 6

Skład wód po usunięciu manganu (pH 7) przy użyciu KMnO$_4$

Sample	KMnO$_4$ (0.1%) [ml·dm^{-3}]	pH	c(Mn) [mg·dm^{-3}]	c(Fe) [mg·dm^{-3}]	c(SO$_4{}^{2-}$) [mg·dm^{-3}]
1	6.912	7.02	2.0	<0.1	1 090
2	9.216	6.99	1.1	<0.1	1 116
3	10.37	6.96	0.9	<0.1	1 186
4	11.52	7.02	0.4	<0.1	1 118
5	12.67	7.06	0.2	<0.1	1 120
6	13.82	6.98	0.0	<0.1	1 122

The costs of the reagent $Ca(OH)_2$ necessary to adjust 1 m^3 of AMD, are 0.37 CZK (0.013 Euro). The costs of potassium permanganate achieving the residual concentration of manganese in the treated AMD 0.4 mg·dm^{-3} per 1 m^3 are 0.86 CZK (0.035 Euro). The total costs of reagents are 1.23 CZK (0.048 Euro) at present.

2.2.2. The removal of manganese from AMD at pH 8 (B)

The procedure was similar to manganese removal as in A, with the difference that in the basic dose of potassium permanganate were considered the residual concentrations of manganese in the alkaline AMD (at pH 8, c(Mn) was 3 mg·dm^{-3}). The basic dose is 5.76 mg·dm^{-3} of KMnO$_4$, from which were derived multiples of this dose (0.8, 0.9, 1.0, 1.1, 1.2, 1.3), which were then used for manganese removal, results are shown in Table 7.

TABLE 7

Results of AMD demanganization (pH 8) with KMnO$_4$

TABELA 7

Skład wód po usunięciu manganu (pH 8) przy użyciu KMnO$_4$

Sample	KMnO$_4$ (0.1%) [ml·dm^{-3}]	pH	c(Mn) [mg·dm^{-3}]	c(Fe) [mg·dm^{-3}]	c(SO$_4^{2-}$) [mg·dm^{-3}]
1	4.61	7.83	2.0	<0.1	1 104
2	5.18	7.73	1.6	<0.1	1 118
3	5.76	7.75	1.1	<0.1	1 126
4	6.34	7.73	0.8	<0.1	1 108
5	6.91	7.62	0.7	<0.1	1 134
6	7.49	7.44	0.4	<0.1	1 122

The costs of the reagent $Ca(OH)_2$ necessary to adjust 1 m^3 of AMD, are 0.43 CZK (0.017 Euro). The costs of potassium permanganate achieving residual concentration of manganese in the treated AMD 0.4 mg·dm^{-3} per 1 m^3 are 0.56 CZK (0.02 Euro). The total costs of reagents are 0.99 CZK (0.038 Euro) by present day.

The results show that to achieve residual concentrations of manganese below 0.5 mg·dm^{-3} it is necessary to use 0.9 times the basic dose for the neutralized AMD and 1.3 time the basic dose for slightly alkaline water. From the costs calculations of response to each of the procedures A and B is evident, that even if there is a higher level of basic dosage in B that from an economic point of view the B is economically more preferable.

2.3. Removal of sulfates in the raw mine water

The results in Table 6 and 7 shows that the AMD will be adjusted with required efficiency with regard to the elimination of iron and manganese. The sulfate concentration is complying with the requirement of the Karlovy Vary Region Regional Office decision. However, according to the Czech Government Regulation No. 229/2007., that establishing indicators and values of permissible pollution, permitted for water-supply streams the maximum permissible concentration of sulfate ions is 250 mg·dm^{-3} and for other surface water is 300 mg·dm^{-3}. Therefore, subsequent experiments were focused on reducing of sulphate concentration.

The input concentration of sulfate in the raw AMD was 1150 mg·dm^{-3}. From the above table (see Table 5, 6, 7) is evident, that there is no significant reduction of sulphate concentration in the AMD treated with calcium hydroxide, respectively calcium hydroxide and potassium permanganate. That is why the desulphating method, developed at the IEI VŠB – TU of Ostrava, is described in the text above.

Whereas the basic condition for precipitation of sulphates with sodium aluminate is highly alkaline environment (pH 12.4 to 12.6), it was first necessary to determine the dose. The dose of Ca(OH)$_2$ for 1 dm^3 AMD was determined experimentally on the 1.8 g·dm^{-3}.

Subsequent experiments tested the optimal dose of reagent AlR-F (concentrated solution of sodium aluminate). The stock sample of high alkalized AMD was prepared (6 dm^3). The increasing doses of sodium aluminate solution were added into each 1 dm^3 of sample, followed by mixing and filtering of the suspension and then determined pH value and sulphate concentration in the filtrate.

The table 8 shows that the dose needed to reduce the sulfate concentration less than 300 mg·dm^{-3} in the treated AMD, is 0.9 ml AlR-F. In the subsequent Table 9 there was carried out verification of the dose of sodium aluminate solution It also was determined in the filtrate concentration of manganese and iron.

TABLE 8

Results of sulphate removal with AlR-F

TABELA 8

Zawartość siarczanów po użyciu AlR-F

Sample	AlR-F [ml]	pH	$c(SO_4^{2-})$ [mg·dm^{-3}]
1	0.7	12.55	606.0
2	0.8	12.57	366.5
3	0.9	12.58	286.0
4	1.0	12.63	164.0
5	1.1	12.70	132.4
6	1.2	12.62	102.6

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TABLE 9

The dosage attestation of AIR-F

TABELA 9

Zaświadczenie dawki AIR-F

Sample	AlR-F [ml]	pH	$c(SO_4^{2-})$ [mg·dm^{-3}]	c(Fe) [mg·dm^{-3}]	c(Mn) [mg·dm^{-3}]
1	0.9	12.56	271.2	<0.1	<0.1
2	0.9	12.58	284.0	<0.1	<0.1
3	0.9	12.62	272.6	<0.1	<0.1

At this dose of sodium aluminate, sulfates are removed to the desired concentration. At the same time iron and manganese ions were completely removed from the treated water.

The costs of the reagent $Ca(OH)_2$ necessary to adjust pH to 12.4–12.6 per 1 m^3 of AMD, are 6.30 CZK (0.26 Euro). The costs of concentrated solution of sodium aluminate (AlR-F) necessary to reduce the sulfates concentration below 300 mg·dm^{-3} per 1 m^3 f treated AMD are 12.24 CZK (0.5 Euro). The total costs of reagents are 18.54 CZK (0.76 Euro) at present.

It is clear that the costs associated with using this technology are high. It is necessary to take into account the fact that this treated water has a significant alkalinity and therefore it is not possible without further treatment discharged to thereby involve additional costs.

Conclusions

Current technology performed at WPPS discharging AMD adjusted at pH value and the iron concentration. By this technology it is solved partial removal of manganese. Carried out laboratory experiments were aimed at assessing the possibility of extending the current technology by level of manganese removal, respectively desulphation.

Manganese removal method with $KMnO_4$ dosage is effective and easy in terms of technical equipment in a plant. Results of laboratory tests provide information on both doses of reagents needed and the costs associated with their acquisition, which is compared to the cost of the reagents at the current technology more than doubled.

Sulphate removal technology with $Ca(OH)_2$ and $NaAlO_2$ dosage, developed at Institute of Environmental Engineering, VŠB – Technical University of Ostrava, is effective, but it is demanding with regard to the further technical and technological equipment and also from an economic point of view, this adjustment is very expensive.

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**MOŻLIWOŚCI ZAGOSPODAROWANIA KWAŚNYCH WÓD KOPALNIANYCH
NA PRZYKŁADZIE KOPALNI SOKOLOWSKA**

Słowa kluczowe

Wody kopalniane, żelazo, mangan, siarczany, usuwanie

Streszczenie

Kwaśne wody kopalniane są szeroko rozpowszechnionym zagrożeniem środowiska związanym z zarówno czynnymi jak i wyeksploatowanymi kopalniami, a wywodzącym się z mikrobowego utleniania pirytu w obecności wody i powietrza. W wyniku tego procesu powstaje kwaśny roztwór zawierający jony metali. Obecne prace mają na celu skorygowanie niskiego odczynu pH, usunięcie żelaza, manganu i siarczanów z kwaśnych wód kopalnianych, które są generowane w czynnym szybie Jiri w kopalni Sokolowska w Czechach (Jiří, Sokolovská uhelná). Lokalne kwaśne wody kopalniane stanowią poważny problem ze względu na swój skład oraz na proces uzdatniania wody w oczyszczalni ścieków w Svatawie (WPPS), gdzie koryguje się jedynie odczyn pH i usuwa głównie z zawiesiny ciała stałe oraz najbardziej skoncentrowane żelazo.

POSSIBILITIES OF ACID MINE DRAINAGE TREATMENT IN SOKOLOVSKÁ UHELNÁ, CZECH REPUBLIC

Key words

Acid mine water, iron, manganese, sulphate, removal

Abstract

Acid mine drainage (AMD) is widespread environmental problem associated with both working and abandoned mining operation, resulting from the microbial oxidation of pyrite in presence of water and air, to form an acidic solution containing metal ions. The present study aims to adjust low pH, remove iron, manganese and sulphate from AMD generated at open pit Jiří and depth Jiří, Sokolovská uhelná, Czech Republic. The local AMD is very problematic due to its composition and process taking place in the Water Preparing Plant Svatava (WPPS), where only pH value is adjusted and mainly high concentration of iron and suspended solids are removed.