Calcium Carbonate as an Agent in Acid Mine Water Neutralization

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Summary

Neutralization of acid mine water is the fundamental step in acid mine water treatment, accompanied by the elimination of iron, partial elimination of manganese and dissolved components of heavy metals, before its discharge into the receiving body. Most frequently lime hydrate, in the form of milk of lime, is used in the neutralization process. The negative outcome of lime hydrate application is an increase in the treated water hardness and a formation of sludge difficult to drain, which could partially be solved using calcium carbonate or combining both agents. The paper observes the effects of calcium carbonate in the neutralization of acid mine water in the locality of Jiří Mine in the Sokolov Region. Attention is paid to testing neutralization methods that are both economically favourable as well as technologically effective. Samples were studied for the time variations in pH values, initial and final concentrations of iron and manganese. Last but not least, it includes an economic appraisal considering the current costs of the agents. As for the neutralization agents, the cost savings are as much as 30% when combining the two agents; under the given conditions the sole application of calcium carbonate proved as economically unprofitable.

Keywords: mine water, calcium carbonate, neutralization

1. Introduction

Acid mine water mainly originates from the oxidation of pyrite and other sulphidic minerals, which represents an environmental problem in many countries. The pH value of acid mine water usually ranges around 3, it often contains high concentrations of metals such as Fe, Mn, Al and anions, e.g. SO_4^{2-}, as well as increased concentrations of Zn, Co, Pb, Cr, and Cu. The source conditions of mine water are very varied and thus it is not always possible to unambiguously specify ‘typical mine water’ for the individual deposits. Even in one geological structure it is possible to identify incomparable hydrochemical water mixtures. The types and consequently the chemism of mine water within one mining company often vary in time and space. It also changes along with the advance of winning operations deeper, it changes in dependence on the quantity and composition of natural mine water sources in the given mining period, etc. In active mines the mine water chemism changes along with different mine water types getting mixed, due to pollution by various process and refuse substances, products of weathering, products of bacterial and biological decomposition, etc. In addition, termination of mining activities alters the mine water regime a great deal too. In this case, one of the sources, i.e. the service and technological water, is cut off and the mine water gets the character of old workings water (Grmela, 1999).

The impact of such water is predominantly connected to the majority of water organisms being adapted to neutral water, and low pH values are thus lethal. As each locality affected by mining contains mine water characteristic for its composition and chemism, it is necessary to deal with water treatment individually and to select suitable technological procedures on the grounds of sample analyses. Therefore, effective methods must be used not only to increase pH to be neutral but also to remove harmful ions and suspended solids. Mine water treatment must result in such water quality indicators that sustain life conditions for organisms in rivers as well as which shall make possible further uses of water, e.g. service water (Heviánková et al., 2011).

World-wide,neutralization of mine water by various agents, such as calcium hydroxide, calcium carbonate, caustic soda, has been studied in numerous localities with acid mine water occurrence (Maree, du Plessis, 1994, Maree et al., 1998, Geldenhuys at al., 2001, Potgieter-Vermaak et al., 2006). The works are grounded in the assumption that neutralization of acid mine water by finely ground calcium carbonate or its mixing with calcium hydroxide may be equally effective but less expensive than neutralization by sole calcium hydroxide in the form of milk of lime. Calcium carbonate in the neutralization of acid mine water may be preferred to calcium hydroxide or caustic soda due toits lower costs, production of higher density sludge easier to drain, more effective control of pH and lower probability of an overdose. Nevertheless, compared with calcium hydroxide, calcium carbonate manifests longer reaction time (Maree et al., 1992, 1994, 1996). Considering the stated disadvantages, even less expensive and more effective agents are still searched for, including possible combinations of neutralization agents (Potgieter-Vermaak et al., 2006, Feng et al., 2004, Petrik et al., 2003, Xenidis et al., 2000).

2. Materials and Methods

2.1. Chemicals

The applied technical neutralization agents of $Ca(OH)_2$ (calcium hydroxide) and $CaCO_3$ (calcium carbonate) come from the production of Kotouč Štramberk, s.r.o. All the other chemicals used for various analyses were in ‘pro analysis’ quality.

2.2. Mine water samples

The acid mine water samples were drawn from the locality of Jiří Mine in the Sokolov Region, to be more specific, from the Pumping Station (PS) Lomnice, where acid mine drainage from PS Lomnice 2B and PS J3 is conducted to. Before being discharged into the receiving body, mine water is treated in the Mine Wastewater Treatment Plant (MWTP) Svatava, which focuses on the elimination of high iron contents, lowering the concentration of manganese and suspended solids and the control of mine water low pH values (Heviánková et al., 2011).

The method and conditions of mine water discharge into the Svatava River are set by the Decree of Water Management Office (Section of the Environment and the Agriculture, Regional Office of Karlovy Vary,filed under No. 655/ZZ/10-5). The decree is valid till 30 April 2014. After treatment in MWTP Svatava the water is discharged into the Svatava River in the quantity of $Q_{average} = 120\ l \cdot s^{-1}$, $Q_{max} = 380\ l \cdot s^{-1}$, $Q_{year} = 4{,}500{,}000\ m^3 \cdot year^{-1}$. The maximum quantity of the discharged mine water is given by the capacity of MWTP Svatava and currently it is about $400\ l \cdot s^{-1}$. The emission and weight limits of the discharged mine water is set by the above stated Water Management OfficeDecree as follows (see Table 1).

Mine water treatment in MWTP Svatava is grounded in the alkalinization in a tank containing lime suspension, air oxidation of bivalent iron Fe^{2+} into the trivalent form Fe^{3+}, separation of sludge from the treated water in two reactors of Seclar type and subsequent gravity and mechanical dewatering of sludge that is carried out in receiving, equalizing and thickening tanks and finally, a Netzsch sludge press is used. Based on the data on the tank volumes provided by the company of Sokolovská uhelná, právní nástupce, a.s., the operator of MWTP Svatava, a theoretical holding time was calculated, namely in the alkalinization tank it is about 15 minutes and in the downstream reactors, where sludge coagulation, flocculation and separation take place, it is further 2 hours. The real holding time in the rectangle tanks of MWTP Svatava will, though, be shorter. The relation $t_{real} = 0.4$ to $0.6 \cdot t_{theoretical}$ is applied for the real holding time in an oblong tank (Dohányos et al., 1998). The information on the holding time is very important with respect to considering potential

Table 1. Emission and weight limits

Tabela 1. Limity emisji oraz limit emisji rocznej

Indicator	Emission limits ($mg \cdot l^{-1}$)		Weight limit ($t \cdot year^{-1}$)
	p[a)]	m[b)]	
SS_{105}	10	20	45
Fe_{total}	3	5	13.5
Mn	1	3	4.5
SO_4^{2-}	800	1200	3600
pH	6–9		

SS – suspended solids

[a)] permissible concentration values

[b)] maximum permissible concentration values neutralization using calcium carbonate as it requires longer reaction time.

Mine water was sampled into 5 l polyethylene barrels that had been pre-washed with 2% solution of nitric acid and thoroughly washed with mine water before own sampling. The samples were kept cool and transported into the laboratory. The mine water in question is characteristic for high contents of sulphates, iron, manganese, low pH value and occurrence of heavy metals. Table 2 states the concentrations of the observed indicators in the mine water sample.

2.3. Equipment

The pH value was measured using a pH 330i metre by WTW with pH electrode Sen Tix 41. The method of atomic absorption spectrometry (Varian AA 280FS) was used to determine the concentration of total iron and manganese. The concentration of sulphates was determined spectrophotometrically at the wavelength of 450 nm using a UV-VIS spectrophotometer (HACH 2000/DR).

2.4. Methods

A Lovibond ET 720 column was used to study the effects of various neutralization agents to modify the pH value and concentrations of iron, manganese and sulphates. For each test, 500ml of acid mine water (AMW) in 1000 ml polyethylene beakers were used. The amount of the dosed agents differed in dependence on the undertaken experiment. The suspension was stirred at a constant speed of 150 rev·min^{-1}, at concurrent measuring of pH at the interval of 1 minute. The samples were left to settle, filtered through a filter paper for a qualitative analysis KA 4 at middle filtration speed. The filtrate was analyzed for pH value, concentration of iron and manganese using the AAS method and the concentration of sulphates using the UV-VIS spectrophotometric method.

3. Results and Discussion

The efficiency of dose and type of neutralization agents depends on several factors. The choice is usually restricted by reaction rate, sludge production, total costs, handling safety and methods, health risks and effect of an excessive dose (consequences of a possible overdose).

The paper compares the course of neutralization of acid mine water using powdered calcium carbonate, calcium hydroxide in the form of lime suspension and combination of both the agents. It compares the rate of the neutralization reaction and selected indicators of the treated mine water quality. The target pH value was selected as pH 7.

3.1. Neutralization of acid mine water using calcium hydroxide

In practice it is $Ca(OH)_2$ in the form of so-called milk of lime that is the most common agent to increase the pH value of acid mine water. As a solution it reacts with acids under the formation of calcium salt precipitates.

The objective at this stage was to identify an optimal dose of $Ca(OH)_2$ to reach the pH value of 7(see Table 3). The reaction time was 30 minutes. The pH value was read continuously.

A significant increase in pH occurs in the first minutes of dosing milk of lime. The optimal dose to reach pH 7 is set as 150 mg.l^{-1}of $Ca(OH)_2$.

Table 2. Values of the observed acid mine water indicators

Tabela 2. Wartości wskaźników dla kwaśnych wody kopalnianej

Indicator	Value
SO_4^{2-}	1368 mg.l^{-1}
Fe_{total}	52.6 mg.l^{-1}
Mn	19.7 mg.l^{-1}
pH	3.25

Table 3. The pH value of mine water treated by calcium hydroxide

Tabela 3. Wartości pH dla wody kopalnianej po zadaniu zasadą wapniową

Dose of $Ca(OH)_2$ (mg.l^{-1})	pH
120	5.9
140	6.64
150	7.05
160	7.75
180	7.98

3.2. *Course of acid mine water neutralization by finely-ground $CaCO_3$*

Literature (Maree, du Plessis, 1994, Potgieter-Vermaak et al., 2006, Maree et al., 1992, 1998b) reports successful applications of calcium carbonate as a substitute for more expensive calcium hydroxide. The doses in the literature range in tens of grams per one litre of treated mine water and the required contact time is several hours. This paper works with the option of possible substitution of calcium hydroxide by calcium carbonate maintaining the doses of agents and thus the quantities of formed sludge. Calcium carbonate used in the neutralization processes is a white crystalline substance, insoluble in water but soluble in acids, found in nature as limestone.

The rate of neutralization reactions was observed via monitoring changes in pH values of the acid mine water in dependence on the dose of neutralization agent within 30 minutes, where the time was determined with respect to technological effectiveness. The doses of calcium carbonate were as follows: 150 (dose based on the dose of $Ca(OH)_2$ considering the quantities of formed sludge); 180; 200, 500; 1000 and 2000 $mg \cdot l^{-1}$.

Chart 1 outlines the results depicting the dependence of neutralization rate on the dose.

The applied doses of $CaCO_3$, except for the dose of 2000 $mg.l^{-1}$, were not sufficient to reach the pH value of around 7 in the set time. A significant increase in the pH value to 4.55 at doses of 150 $mg \cdot l^{-1}$, or 4.89 at the dose of 180 $mg \cdot l^{-1}$ and 5.29 at the dose of 200 $mg \cdot l^{-1}$, occurred during the first 8 minutes. Further growth was rather slow andafter 30 minutes the pH value was 5.01, 5.78 and 5.91 respectively for the doses above.

When adding higher doses of $CaCO_3$the pH value was 6 at the dose of 500 $mg \cdot l^{-1}$ in 8 minutes, at the dose of 1000 $mg \cdot l^{-1}$ in 4 minutes and at the dose of 2000 $mg \cdot l^{-1}$ in 2 minutes. Further growth of pH was rather slowin all cases. After 30 minutes the pH value was 6.60, 6.81 and 6.95 respectively for the doses above.

It is clear from the stated dependence of pH on time that for the individual doses of $CaCO_3$ there is a decrease in time required to improve the pH along with a rising dose of $CaCO_3$. Nevertheless, even higher doses in selected limits do not cause significant differences in the maximum pH values in given time. The results confirm vitally longer reaction times of mine water and calcium carbonate.

3.3. *Neutralization of acid mine water using $CaCO_3$ and $Ca(OH)_2$*

Considering the fact it was not possible to obtain the required pH values in the previous set of experiments due to own dosing of $CaCO_3$ and as the required dose of $CaCO_3$ is economically unacceptable

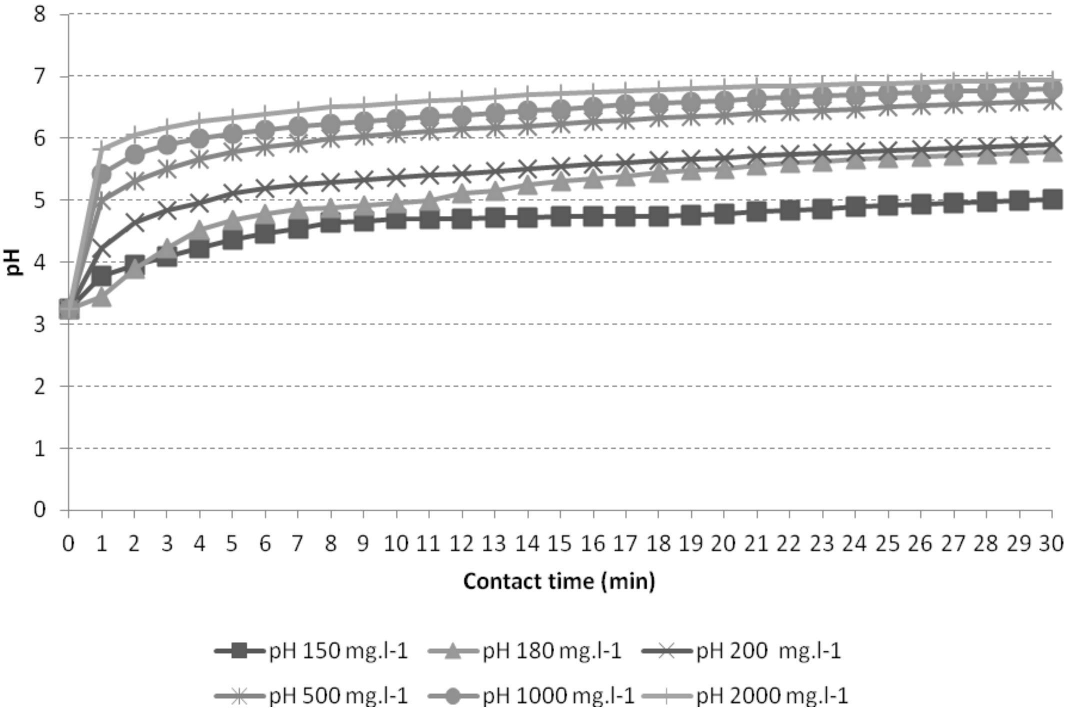

Chart 1. Effect of the dose and contact time on pH value

Wykres 1. Wpływ dawki i czasu reakcji na wartość pH (see Chapter 3.4) the combination of $CaCO_3$ and $Ca(OH)_2$ was selected for further tests.

The objective of using both the neutralization agents was to substitute a certain proportion of $Ca(OH)_2$ by cheaper $CaCO_3$. The contact time of calcium carbonate and mine water (i.e. before calcium hydroxide is dosed) and the doses of calcium carbonate were determined based on the results stated above, namely the contact time as approx. 4 minutes, when the increase in pH value is prominent (see Chart 1) and the doses as 150; 180; 200 and 500 mg·l^{-1} of $CaCO_3$. The selected doses of calcium carbonate are grounded in the expected costs of the agent, when the tested dose over 500 mg·l^{-1} would be more economically demanding than the application of sole calcium hydroxide, as apparent from Table 5. Therefore, the doses of 1000 mg·l^{-1} and 2000 mg·l^{-1} were not tested in combination with calcium hydroxide. The doses of $Ca(OH)_2$ were derived from the dose required for neutralization towards pH 7, i.e. 150 mg·l^{-1} of $Ca(OH)_2$. Doses ranging from 1/3 to 1/2 of the original dose were selected, specifically 42; 50; 62; 74 and 78 mg·l^{-1}, and were dosed approximately 4 minutes after having dosed $CaCO_3$ under simultaneous stirring in the stirring column. Having added $Ca(OH)_2$ in the form of lime suspension, the contact time was further 11 minutes. The total stirring time of mine water and agents was thus 15 minutes. Contrary to calcium carbonate, the reaction of milk of lime and mine water requires considerably shorter time.

Apart from the course of neutralization, the experiments also observed the concentrations of manganese and iron in the neutralized mine water. The concentrations of both the observed metals dropped in dependence on the treated water pH value. Chart 2 summarizes the obtained results.

The results in Table 4 imply that the required pH around 7 was obtained in case of dosing 150 mg·l^{-1} of $CaCO_3$ and 78 mg·l^{-1} of $Ca(OH)_2$; 180 mg·l^{-1} of $CaCO_3$ and 62 mg·l^{-1} of $Ca(OH)_2$; 200 mg·l^{-1} of $CaCO_3$ and 50 mg·l^{-1} of $Ca(OH)_2$; 500 mg·l^{-1} of $CaCO_3$ and 42 mg·l^{-1} of $Ca(OH)_2$. There is an apparent trend in the fall of the required amount of $Ca(OH)_2$ along with the rise in the dose of $CaCO_3$, which suggests possible reductions in the costs of chemical neutralization agents. The stated combinations of doses were used to evaluate the economic costs of the applied agents.

3.4. Costs of neutralization agents in selected neutralization methods

Apart from the above stated combinations of both agents, the economic costs appraisal of neutralization agents includes all individually tested doses of calcium carbonate (150; 180; 500; 1000 and 2000 mg·l^{-1}) and the dose of calcium hydroxide required to reach pH around 7 (150 mg·l^{-1}). Table 5 summarizes the results below.

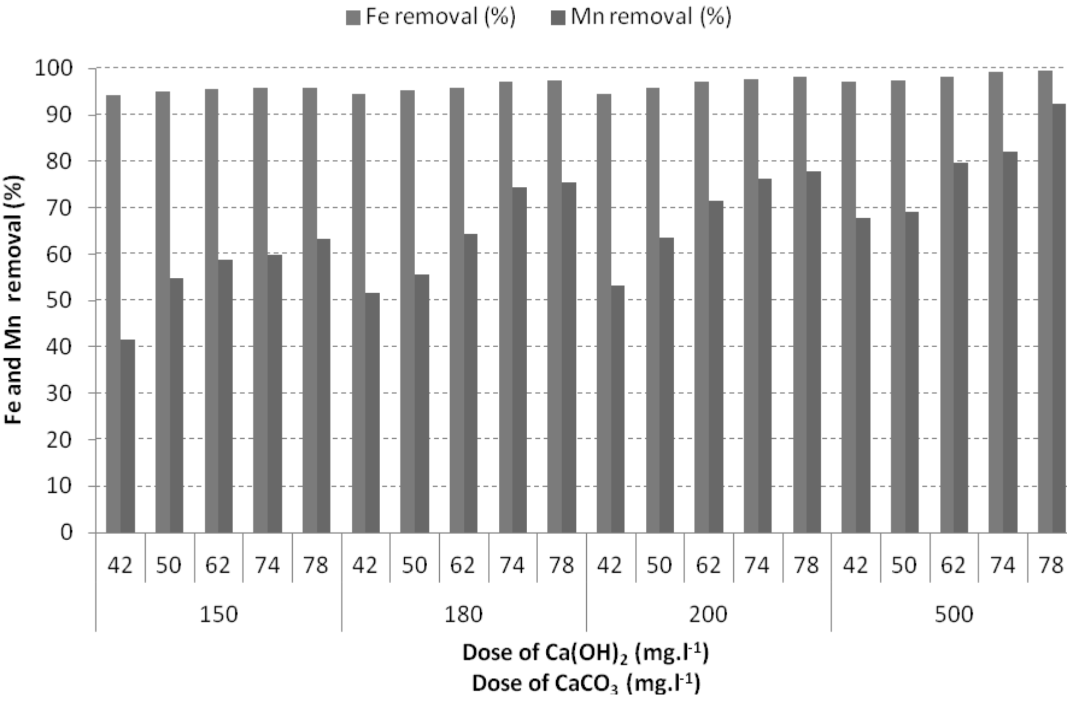

Chart 2. Removal of iron and manganese from treated mine water (in per cents)

Wykres 2. Stopień usunięcia żelaza i manganu w wodzie

Table 4. Observed indicators of treated mine water in dependence on the doses of $CaCO_3$ and $Ca(OH)_2$

Tabela 4. Wartości wskaźników w wodzie w zależności od dawki $CaCO_3$ i $Ca(OH)_2$

$CaCO_3$ ($mg.l^{-1}$)	$Ca(OH)_2$ ($mg.l^{-1}$)	pH	c(Fe) ($mg.l^{-1}$)	c(Mn) ($mg.l^{-1}$)
150	42	5.91	3.01	11.5
	50	6.71	2.63	8.9
	62	6.81	2.37	8.11
	74	6.85	2.23	7.93
	78	**6.92**	**2.19**	**7.21**
180	42	6.52	2.95	9.55
	50	6.77	2.52	8.74
	62	**6.98**	**2.18**	**7.03**
	74	7.71	1.45	5.06
	78	7.82	1.37	4.83
200	42	6.67	2.83	9.21
	50	**6.93**	**2.21**	**7.19**
	62	7.69	1.53	5.61
	74	7.91	1.18	4.67
	78	8.01	0.95	4.38
500	**42**	**7.28**	**1.43**	**6.37**
	50	7.52	1.31	6.11
	62	8.26	0.87	4.01
	74	8.52	0.36	3.52
	78	8.59	0.19	1.5

Table 5. Evaluation of costs per used agents to reach the final observed indicator values

Tabela 5. Określenie kosztów uzyskania założonych wskaźników jakości wody

Agent	$CaCO_3$						$CaCO_3$				$Ca(OH)_2$
							$Ca(OH)_2$				
Cost of agent ($CZK{\cdot}t^{-1}$)	890						890				3300
							3300				
Dose ($mg{\cdot}l^{-1}$)	150	180	200	500	1000	2000	150	180	200	1000	150
							78	62	50	42	
Costs of agents ($CZK{\cdot}m^{-3}$)	0.134	0.16	0.178	0.445	0.89	1.78	0.391	0.365	0.343	1.029	0.495
Contact time (min)	30	30	30	30	30	30	15	15	15	15	15
Final pH	5.01	5.78	5.91	6.6	6.81	6.95	6.92	6.98	6.93	7.28	7.05
Fe_{total} ($mg{\cdot}l^{-1}$)	6.76	5.34	3.88	2.66	2.38	2.2	2.19	2.18	2.21	1.43	1.65
Mn ($mg{\cdot}l^{-1}$)	15.92	15.45	13.13	8.98	7.15	7.06	7.21	7.03	7.19	6.37	6.73

Using sole calcium carbonate to neutralize acid mine water, the costs for the required agent doses are rather high and thus this pretreatment method cannot be recommended from the economic point of view. Combining both agents to neutralize acid mine water at the dose 200 $mg{\cdot}l^{-1}$ of $CaCO_3$ and 50 $mg{\cdot}l^{-1}$ of $Ca(OH)_2$ the costs per agents are 0.343 $CZK{\cdot}m^{-3}$. These costs are about 30% lower than using sole calcium hydroxide in acid mine water neutralization.

4. Conclusion

Lime hydrate precipitation method, also used in MWTP Svatava, is relatively costly (3300 $CZK{\cdot}t^{-1}$, in 2012) and produces sludge of low density, which brings about further costs of the operation of downstream technology of sludge treatment. The total costs of alkalinization could be reduced if calcium hydroxide was partially substituted by calcium carbonate (890 $CZK{\cdot}t^{-1}$, in 2012). Apart others, this alternative has also other advantages, such as easier handling of the agents, production of higher density sludge, more effective control of pH and lower risk of a possible overdose.

The results of this study show that for the acid mine water from Jiří Mine in the Sokolov Region the dose of sole calcium carbonate to reach neutral pH in 30 minutes is about 2000 $mg \cdot l^{-1}$, which in acid mine water treatment represents costs of agents amounting to 0.89 $CZK \cdot m^{-3}$. Combining both the agents the lowest costs of agents are at the dose of 200 $mg \cdot l^{-1}$ of $CaCO_3$ and 50 $mg \cdot l^{-1}$ of $Ca(OH)_2$, i.e. 0.343 $CZK \cdot m^{-3}$. However, the cost of sole calcium hydroxideis 0.495 $CZK \cdot m^{-3}$. Therefore, cost savings for neutralization agents amount to 30% if both agents are combined.

The assumption of sole application of calcium carbonate as a neutralization agent was confirmed for the mine water in question, but this neutralization method did not prove as economically effective. When considering upgrading theacid mine water treatment technology using doses of calcium carbonate, both its advantages and disadvantages must be taken into account. The advantages are lower costs of the agents, production of higher density sludge, better sedimentation characteristics, including better drainability of the sludge, dosing of loose agent without preparing suspensions, lower demands on the preparation of milk of lime whose dose is about 2/3 lower than in its sole application in acid mine water neutralization. The disadvantages can be seen as incorporation of another batcher, providing space for storing the chemicals, higher costs in connection with the sludge increase and longer reaction time.

Within the carried out experiments, the mine water pH values were modified to neutral ones. In MWTP Svatava the mine water is alkalized towards pH of 8–9, and thus it is crucial to consider other series of experiments if the agents should be combined, under concurrent utilization of the results stated above.

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Zastosowanie węglanu wapnia, jako środka neutralizującego kwaśne wody kopalniane

Podstawowym krokiem przy uzdatnianiu kwaśnych wód kopalnianych, przed ich zrzutem do odbiornika, jest ich neutralizacja, której towarzyszy usuwanie zwłaszcza żelaza, części manganu oraz części metali ciężkich obecnych w formie rozpuszczonej. Najczęściej do neutralizacji stosowany jest wodorotlenek wapnia w postaci mleka wapiennego. Negatywnym skutkiem zastosowania wodorotlenku wapnia jest podwyższenie twardości uzdatnionej wodyiwytworzenie bardzo trudnego do odwodnienia osadu, co mogłoby być częściowo rozwiązane poprzez zastosowanie węglanu wapnia, ewentualnie kombinując obydwa środki. W niniejszej pracy badano wpływ węglanu wapnia na proces neutralizacji kwaśnych wód kopalnianych z kopalnio odkrywkowej Jiří w Sokolovskim Zagłębiu Węglowym. Uwaga skierowana była na testowanie ekonomicznie opłacalnej technologicznie skutecznej neutralizacji. W próbkach badany był przebieg zmian pH w czasie, początkowe i końcowe koncentracje żelaza i manganu. Również wykonana została ocena strony ekonomicznej przy uwzględnieniu aktualnych cen środków neutralizujących. Oszczędności kosztów środków neutralizujących w kombinacji obydwu środków wynosząca 30%, samodzielne zastosowanie węglanu wapnia okazało się być, w danych warunkach, ekonomicznie nieopłacalne.

Słowa kluczowe: woda kopalniana, węglan wapnia, neutralizacja