

Purification of Mine Waters Using Lime and Aluminum Hydroxochloride

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ABSTRACT

Analysis of the scientific and technical literature shows that there are quite a few methods of mine water processing. Reagent methods can be considered as the most promising and economically expedient. Mine waters are characterized by a high content of hardness ions and sulfates. The concentration of sulfates varies between 5–35 mg-eq/dm³, hardness – 10–40 mg-eq/dm³. It has been established that effective purification of water from sulfates can be achieved with the use of lime and 5/6 aluminum hydroxochloride. The efficiency of the process depends on the doses and ratio of reagents. The degree of softening and purification of water from sulfates increases with an increase in the dose of aluminum coagulant within certain limits. When using 5/6 aluminum hydroxochloride, the efficiency of water purification from sulfates is quite high and small amounts of chlorides are introduced into the water with the coagulant.

Keywords: mine waters, softening, aluminum hydroxochloride, sulfates, hardness ions.

INTRODUCTION

Water is an important natural resource that play an exceptional role in metabolic processes, is of great importance in industrial production and agriculture. The growth of cities, the rapid development of industry, the intensification of agriculture, the significant expansion of the area of irrigated land, the improvement of cultural and household conditions and a number of other factors lead to an increase in the need for water, so the problems of water supply become more complicated (Brankov et al., 2012; Voza et al., 2015).

One of the most complicated problems is a rational use of water resources in the industrial regions of Ukraine, in particular, where several thousand large industrial enterprises that use significant volumes of natural resources are concentrated (Buzylo et al., 2019; Buzylo et al., 2020). The high concentration of industrial and agricultural production, transport infrastructure,

combined with high population density, created an extremely high technogenic and anthropogenic load on the biosphere (Tong et al., 2021; Remeshevska et al., 2021).

Water quality is deteriorating due to natural and anthropogenic factors (Chugai and Saf-ranov, 2020). Mine waters of coal enterprises are a large-scale source of pollution of water bodies in the regions (Boyacioglu, 2014; Pavlychenko et al., 2022). They are characterized by increased mineralization, general hardness, the content of suspended substances, sulfates, chlorides, phosphates, fluorides, iron, manganese, copper, petroleum products, and other substances are quite significant due to the intensive discharge of polluted water into water bodies without appropriate treatment (Gavrishin, 2018). As a result, a significant number of natural water bodies in the industrialized regions of Ukraine are characterized by a high level of mineralization.

As it known, a distinctive technological feature of mining enterprises is not only the consumption of water to ensure production processes, but also the release of significant volumes of mine and quarry water to the surface of the earth. They are formed due to underground and surface waters, which penetrate into underground mine operations (Rambabu et al., 2020; Trus et al., 2020b).

Mine water is contaminated with finely dispersed carbon-rock mixture (suspended substances), mineral salts, including heavy metals (lead, cobalt, mercury, etc.), bacterial impurities, etc. Quarry waters are formed due to surface and underground waters. Water that has entered mining operations, like mine water, is polluted and must be purified before discharge into reservoirs or during technical water use.

The wastewater of many enterprises contains an increased amount of sulfate ions, the MPC of which in drinking and household water is 500 mg/dm³, and for fishing ponds – 100 mg/dm³. Large volumes of wastewater contaminated with sulfate ions are produced during reverse osmosis treatment.

Unfortunately, there are currently no economically feasible methods for processing concentrates that contain a large amount of hardness salts and sulfate ions, and their storage in sludge storage facilities worsens the already difficult ecological situation. Under such conditions, the development and implementation of water demineralization and softening technologies becomes vital.

In this work, the research was conducted on the processing and purification of mine waters with a high level of mineralization by reagent methods. As a rule, such waters are characterized by an increased content of hardness ions and sulfates (Runtti et al., 2017).

Sulfates lead to increased mineralization, which leads to corrosion (Dou et al., 2017; Vasyliov et al., 2020). In drinking water, the recommended level of sulfate is 250 mg/L (WHO, 2022), but some countries have set stricter requirements for sulfate content to prevent pipe corrosion, for example, in Finland the value of the mentioned parameter is less than 150 mg/L (Yurasov et al., 2022). Sulfate discharge limits for mine waters and other industrial effluents typically range from 250 to 1000 mg/L. A high content of hardness ions leads to salinization of water bodies and their siltation.

Among the well-known technologies for water desalination and purification from sulfates, baromembrane, ion exchange, biological and reagent water treatment technologies are the most common recently (Kinnunen et al., 2018). Those methods that allow not only to remove sulfates from the water environment, but also to obtain valuable components in the treatment of sulfate effluents are appropriate.

Ion exchange is used to remove sulfates. Sulfate ions can usually be removed using weakly basic anion resins (Guimaraes and Leao 2014; Trus, 2022b). The maximum efficiency of removing sulfates from the solution is observed at a dose of ion exchange resin of 1000 mg/100 cm³. The ion exchange process depends on the flow rate, the height of the ionite layer, and the initial ion concentration (Range and Hawboldt, 2019). However, due to limited capabilities and selectivity, ion exchange may not be suitable as a primary method, but rather to be used after chemical precipitation. In addition, after exhausting the exchange capacity of ionite, it is subject to regeneration. After regeneration, large volumes of concentrated solutions are usually generated, which must be properly processed (Trus et al., 2021).

Sulfates removal by adsorption has been studied with a wide range of materials: activated carbon, modified zeolites, modified geopolymers, limestone, modified coconut fibers, modified rice straw, alumina, iron sand, and waste pulp and paper (Sadeghalvad et al., 2021). In general, adsorption can be an efficient and economical method. However, in industrial application, there are many factors affecting technical and economic feasibility. First, the adsorbent must have a high adsorption capacity and selectivity to sulfate. Currently, there is a shortage of commercially available adsorbents specifically for the removal of sulfates from mine effluents. Raw materials and adsorbent preparation (including possible chemical modification) should be inexpensive, simple and as environmentally friendly as possible.

Biological wastewater treatment has become widely used in the treatment of wastewater with a high concentration of sulfate ions (Bhuyan et al., 2020). Biological destruction of sulfates in wastewater occurs with the help of sulfate-reducing bacteria. However, the use of bacteria to extract sulfate ions requires certain conditions for their growth and life, which creates some inconveniences in their use.

The electrocoagulation method is quite effective, but its main disadvantage is that it is quite energy-consuming (Mamelkina et al., 2017; Zhu et al., 2019).

The use of membrane methods makes it possible to purify wastewater and reuse it in technological processes (Bodzek, 2019). Nanofiltration and reverse osmosis are most suitable for removing sulfates from water. In the process of nanofiltration, sulfates, chlorides, hardness ions and heavy metals are effectively retained (Trus et al., 2020a). The efficiency of removal of hardness ions and sulfates increases with increasing pH, and the selectivity of the nanofiltration membrane increases with increasing sulfate concentration. However, the accumulation of suspended and dissolved solids on the surface of the membrane creates a layer of fouling and scaling, which causes increased operating costs due to the need for chemical cleaning and the use of anti-scaling agents, reduced membrane life, and reduced water treatment efficiency (Trus et al., 2022a; Trus et al., 2022b).

Thus, most of the methods are inefficient, costly or unsustainable (Tang et al., 2017; Amaral Filho et al., 2016).

The main advantages of the reagent water softening method are the cheapness of the process (Öztürk and Ekmekçi, 2020; Dou et al., 2017). In addition, there is a possibility of hardness salts release in the form of non-toxic sediments that can be disposed of (Gomelya et al., 2014).

MATERIALS AND METHODS

Water from Tarnovska mine and Zahidno-Donbaska mine were used as initial solutions for investigation. To the solution, the composition of which is given in Table 1, with a volume of 100 cm³, CaO suspension was added and thoroughly mixed until CaO dissolved in water. With intensive stirring, the calculated volume of 5/6 aluminum hydroxochloride (AHC) was added. The suspension was thermostated at a temperature of 40 °C for 2 hours. Then it was left at room temperature for 4 hours for settling, and the sample was filtered. In order to reduce hardness and alkalinity, CO₂ was blown through the solution, and the precipitate formed was filtered through a filter (Figure 1). The residual concentration of sulfate and chloride ions, final hardness and alkalinity, mineralization of the solution were determined.

The degree of purification from sulfate anions (A_1) and the degree of softening (A_2) were determined by the formulas:

$$A_{1(2)} = \frac{C_{in(1,2)} - C_{f(1,2)}}{C_{in(1,2)}} \quad (1)$$

where: C_{in} – initial concentration of hardness and sulfate anions in the solution, mg-eq/dm³;

C_f – final concentration of hardness and sulfate anions in the solution, mg-eq/dm³.

Table 1. Characteristics of solutions

Physico-chemical parameters	Units	Tarnovska mine	Zahidno-Donbaska mine
Total hardness	mg-eq/dm ³	17.0	31.0
Calcium	mg/dm ³	200.4	1835.7
Magnium	mg/dm ³	85.1	454.8
Sulfates	mg/dm ³	290.0	1210.0
Coloration	degree	31.4	41.2
Turbidity	NUT	92.6	112.7
pH	–	7.89	7.27
Total alkalinity	mg-eq/dm ³	5.0	16.0
Chlorides	mg/dm ³	992.7	105.0
Permanganate oxidizability	mg/dm ³	11.6	32.3
Nitrates (NO ₃ ⁻)	mg/dm ³	2.2	10.6
Nitrites (NO ₂ ⁻)	mg/dm ³	3.39	6.42
Total mineralization (TDS)	mg/dm ³	2042.0	30294
Hydrocarbonates	mg/dm ³	305.0	213.5
Dry residue (general mineralization)	mg/dm ³	2210.0	32900.0
Suspended substances	mg/dm ³	124.0	178.0

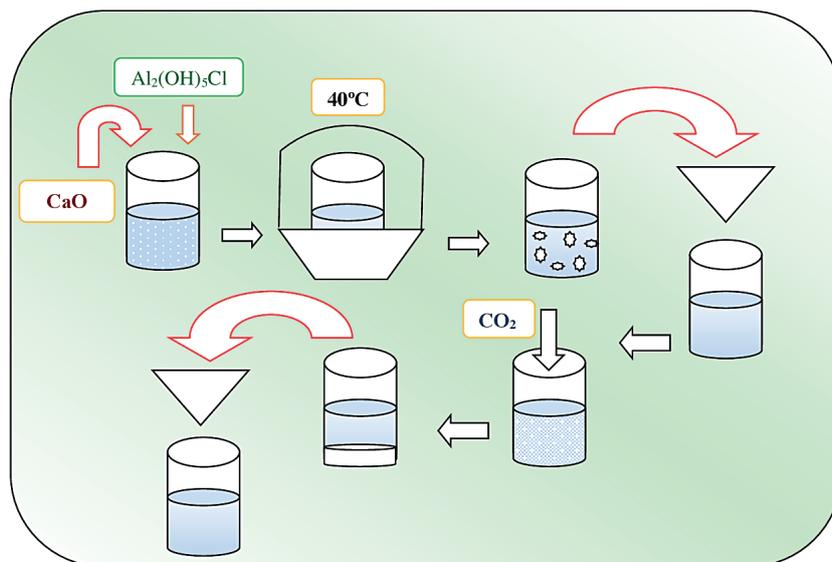


Figure 1. Process diagram

RESULTS AND DISCUSSION

In reagent purification processes such reagents as $\text{BaCl}_2 \cdot 2\text{H}_2\text{O}$ i Na_3PO_4 is often used, however, this reagent is quite toxic (Bustos-Flores et al., 2021). Lime is suitable for pretreatment processes (Fernando et al., 2018; Vu et al., 2021). However, the use of gypsum settling as a single process is often not effective enough to meet environmental restrictions when soluble metal sulfates such as sodium sulfate are present in the water. To increase the efficiency of the process, flotation is used together with sedimentation (Santander-Muñoz et al., 2021).

The sulfate removal process occurs by sedimentation of ettringite (Tian et al., 2019). Aluminum coagulants are quite widely used to purify

water from sulfates (Gomelya et al., 2014; Vasiichuk et al., 2022). Calcium aluminate is also used (Guerrero-Flores et al., 2018). When sodium aluminate is used with lime during the removal of sulfates, significant alkalinity of water occurs (Trus and Gomelya, 2021). The widespread use of 2/3 AHC is hindered by the introduction of a significant amount of chlorides into the solution compared to 5/6 AHC (Trus et al., 2019). The choice of 5/6 AHC connected with its main advantages, which are presented in Figure 2.

The results obtained with the application of 5/6 AHC are shown in the Table 2 and 3. As can be seen from Table 2, the efficiency of water purification from sulfates depends on the consumption of aluminum coagulant and lime. At low consumption, the effectiveness of lime for purifying water

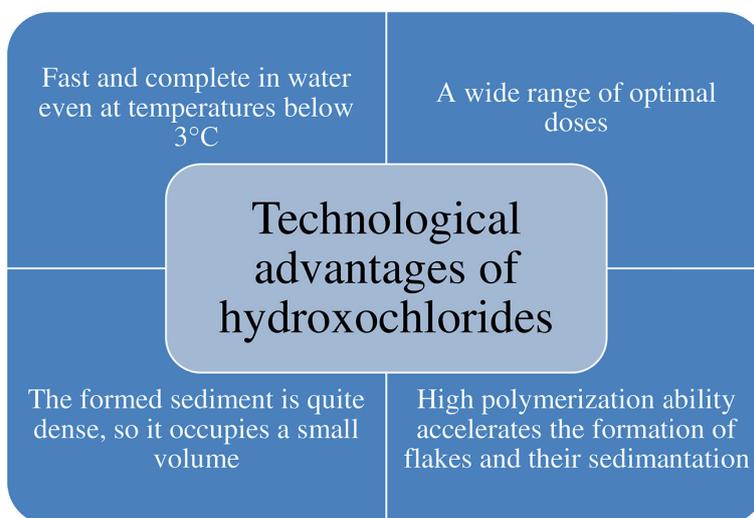


Figure 2. Technological advantages of hydroxochlorides application

from sulfates was low with a significant increase in the concentration of chlorides in water. When the doses of lime are increased, the degree of removal of sulfates increases significantly, which makes it possible to reduce the concentration of sulfates in some cases to 105–110 mg/dm³. It should be noted that the residual water hardness in most cases was 2.2–3.1 mg-eq/dm³, and the residual alkalinity was 8.1–8.7 mg-eq/dm³. The efficiency of sulfate

removal increases with an increase in the consumption of lime and AHC, although with optimal lime consumption, an increase in the dose of AHC does not lead to an increase in the efficiency of sulfate removal, but only leads to an increase in the content of chlorides in water.

The use of 5/6 AHC was also effective in removal sulfates from solutions with a concentration of 25.2 mg-eq/dm³. As in the previous

Table 2. Dependence of the removal efficiency of sulfate ions from the Tarnovsk Mine on the consumption of lime and 5/6 AHC

Dose CaO, mg-eq/dm ³	Dose 5/6 AHC by Al ₂ O ₃ , mg-mol/dm ³	SO ₄ ²⁻ , mg/dm ³	Hardness, mg-eq/dm ³	Alkalinity, mg-eq/dm ³	A1	A2
29.0	1.00	145	5.7	10.7	50.00	66.47
29.0	1.20	140	5.4	9.2	51.72	68.24
29.0	1.40	135	3.1	9.2	53.45	81.76
29.0	1.60	130	2.7	9.0	55.17	84.12
29.0	2.00	120	2.5	5.5	58.62	85.29
31.9	1.00	135	5.6	11.5	53.45	67.06
31.9	1.20	130	5.3	9.4	55.17	68.82
31.9	1.40	120	3.0	9.1	58.62	82.35
31.9	1.60	105	2.5	8.7	63.79	85.29
31.9	2.00	100	2.4	6.5	65.52	85.88
34.8	1.00	130	5.6	11.7	55.17	67.06
34.8	1.20	125	5.2	9.6	56.90	69.41
34.8	1.40	110	3.1	9.5	62.07	81.76
34.8	1.60	105	2.4	8.7	63.79	85.88
34.8	2.00	105	2.2	8.1	63.79	87.06

Table 3. Dependence of the removal efficiency of sulfate ions from the Zahidno-Donbaska Mine on the consumption of lime and 5/6 AHC

Dose CaO, mg-eq/dm ³	Dose 5/6 AHC by Al ₂ O ₃ , mg-mol/dm ³	SO ₄ ²⁻ , mg/dm ³	Hardness, mg-eq/dm ³	Alkalinity, mg-eq/dm ³	A1	A2
81.4	4.20	470	9.9	10.7	61.16	68.06
81.4	5.04	420	5.6	9.2	65.29	81.94
81.4	5.88	380	3.3	9.2	68.60	89.35
81.4	6.72	364	3.2	9.0	69.92	89.68
81.4	8.40	343	3.2	5.5	71.65	89.68
89.5	4.20	417	9.7	11.5	65.54	68.71
89.5	5.04	395	5.9	9.4	67.36	80.97
89.5	5.88	265	2.5	9.1	78.10	91.94
89.5	6.72	205	2.7	8.7	83.06	91.29
89.5	8.40	190	2.6	6.5	84.30	91.61
97.7	4.20	410	9.3	11.7	66.12	70.00
97.7	5.04	385	2.7	9.6	68.18	91.29
97.7	5.88	270	2.5	9.5	77.69	91.94
97.7	6.72	202	2.0	8.7	83.31	93.55
97.7	8.40	165	1.8	8.1	86.36	94.19

cases, the efficiency of water purification from sulfates depends on the consumption of lime and the dose of 5/6 AHC (Table 3). Purification was ineffective when doses were less than the stoichiometric amount. With a stoichiometric amount of lime, the degree of removal of sulfates reaches 61.16–71.65%, and in general it slightly increases with an increase in the dose of 5/6 AHC. In this case, the efficiency of purification increases with an increase in the consumption of AHC with sufficient doses of lime. The best results were obtained with a 20% excess of lime and with an aluminate dose of 8.40 mg-mol/dm³ based on Al₂O₃. In this case, the residual concentration of sulfates decreases to 165 mg/dm³, with a water hardness of 1.8 mg-eq/dm³, the concentration of chlorides at the level of 245 mg/dm³, and alkalinity at the same time reached 8.1 mg-eq/dm³. Thus, water purification from sulfates with simultaneous softening can be carried out with the help of lime and 5/6 AHC.

CONCLUSIONS

Application of Al₂(OH)₅Cl and CaO ensures high efficiency of sulfate removal. The efficiency of sulfate removal increases with an increase in the coagulant dose. The same can be said about the effectiveness of water softening. It should be noted that with an increase in the dose of coagulant, not only the effectiveness of water softening increases, but also the residual alkalinity of the water decreases significantly.

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