Metallurgical and Materials Data 1, no. 2 (2023): 57-63

Publisher: Association of Metallurgical Engineers of Serbia

Metallurgical and Materials Data

www.metall-mater-data.com

Stability of solid residue after integral treatment of acid mine drainage

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A R T I C L E I N F O R M A T I O N :

https://doi.org/10.30544/MMD6

Received: 21 June 2023

Accepted: 30 June 2023

Type of paper: Research paper

A B S T R A C T

Acid mine drainage is a challenging environmental problem caused by mining activities. Long-term copper ore exploitation and vast amounts of mining waste lead to severe soil contamination as well as river and groundwater pollution. In this paper, research is focused on utilizing waste mine waters and off-balance parts of the mine deposit (such as flotation tailings) and converting their negative characteristics to an environmentally friendly one. In the authors' previous work, acid mine water from Robule, which is part of the Bor copper mining and smelting complex in Serbia, was successfully treated using flotation tailings, and the resulting solution was suitable for safe discharge. This paper investigates the characteristics of the solid residue that remained after the treatment of the modified flotation tailings in order to evaluate the possibility of its safe disposal back into the environment. The solid residue was subjected to TCLP and EN 12457-4 tests as standard procedures that assess the leaching characteristics of a material. The solid residue was also exposed to long-term leaching with melted snow to simulate environmental conditions for one year. This drainage water was analyzed and compared with the results of the standard leaching procedures. The investigated treatments lead to improved economic and environmental effects, while the process itself is adjusted to preserve the environment.

Keywords: Acidic Mine Drainage, Flotation Tailings, Circular Economy, Fly Ash.

1. Introduction

During the exploitation and flotation processes, approximately 99% of the ore is converted into flotation tailings (Brierley & Brierley 2013, Dold 2008). Flotation tailings are natural or artificial lagoons for disposing of liquid flotation tailings deposits (crushed comminuted material and chemically contaminated water) (Lekovski, Mikić & Kržanović 2013). Namely, flotation tailings were identified as one of the primary sources of heavy metals in the environment (Qin et al. 2017). Previous researchers have concluded that the abandoned flotation tailings could increase the concentration of heavy metals in the surrounding areas, affecting soil fertility (Huang et al. 2013, Milićević et al. 2013), surface water and groundwater (Milićević et al. 2013, Concas et al. 2006), sediments, and plants (Lilić et al. 2008, Randjelovic et al. 2020), causing a significant health risk to the residents (Colin-Torres et al. 2014, Pereira, Ribeiro & Goncalves 2004). Disposing of the flotation tailings in an inappropriate and environmentally unsafe way is the source of one of the main problems for the mining industry, which leads to the formation of Acid Mine Water (AMD). The formation of AMD occurs as a result of the spontaneous oxidation of sulfide minerals, which are also part of flotation tailings (Kebede, Titus & Bhekie 2017). During exploitation, sulfide minerals are exposed to various mechanisms of oxidation (i.e., contact with water or oxygen from the atmosphere or in the presence of microorganisms) and become chemically unstable (Lottermoser B.G. 2010). For a long time, oxidation of sulfide minerals (galena, sphalerite, chalcopyrite, and pyrite) has been systematically studied on a global and local scale (Bogdanovićet al. 2020), and oxidation of pyrite (FeS_2) is considered the primary source of acid mine water (Kebede, Titus, & Bhekie 2017). Once the formation of acidic mine water starts, it is a highly intense and accelerated process that is challenging to control. According to previous research (Bell et al. 2001; Pelo et al. 2009; Evangelou & Zhang 1995; Smičiklas et al. 2021), the acidity and toxic metal content of acid mine waters have the most severe and negative impact on people and the environment. Acid mine water and the dissolved toxic metals therein can lead to the contamination of surface and groundwater through run-off or filtration processes that can last centuries. Along with the water flow transport of the dissolved metals, the dispersion of fine particles through wind-borne transport from the surface of flotation tailings can also occur. These two processes are simultaneously classified as powerful sources of contamination that

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can cause problems in ecosystems and nearby inhabitation. (Kebede, Titus & Bhekie 2017, Navarro et al. 2008, Xian-De et al. 2013). The acidic solution is rapidly enriched with high concentrations of metals, which can last for centuries and require adequate treatment (Baruah & Khare 2010, Aguiar et al. 2016, Gray 1997). Treatment of AMD refers to reducing the concentration of toxic elements below the maximum allowed concentrations for discharging into the recipient, which will not negatively impact the environment.

There are two main strategies for AMD remediation: prevention and treatment of acid mine waters. Prevention implies the inhabitation of oxidation of sulfide minerals (Kebede, Titus & Bhekie 2017, Aubertin et al. 2016), and the treatment of already formed acid mine waters involves the collection and neutralization of AMD through active or passive methods. Active methods include the continuous addition of alkali to the required level of neutralization of acidic waters, with the possibility of metal extraction at relevant pH values (Rajaković et al. 2013). The main disadvantage of this method is the formation of chemically unfavorable solid residue, formed after neutralization of acidic mine waters, which is unstable in the environment and has no economic value (Gardić et al. 2017). Contrariwise, passive methods depend on natural processes to neutralize AMD acidity, oxidation, and metal deposition. As compared with active methods, the main advantage of these systems is that they are more economical, do not pollute the environment, and do not generate additional waste (Skousen et al. 2017). Besides various chemical agents such as hydrated lime ($Ca(OH)_2$), calcium oxide (CaO), and sodium hydroxide (NaOH) for increasing pH and removing metals from AMD, an innovative approach involves alternative neutralization materials. Alternative neutralization materials are high-alkaline materials generated as waste products from the same or other major industries. For example, fly ash, waste from dairies, paper mills, steel mills, wine industries, tires, seafood, eggshells, etc., are used worldwide. Due to their diversity, alternative materials are gaining more and more attention every day (Klain et al. 2005).

Previous research (Petronijević et al. 2020a, Sokić et al. 2019) has confirmed that, due to their favorable characteristics, the flotation tailings from the copper Majdanpek Mine (Serbia) have the ability to neutralize the acidic mine water of Robule Lake. Besides their neutralization potential, the potential economic and environmental benefits to the environment were also determined (Petronijević et al. 2020b). The main characteristic of the acid mine water from Robule Lake is a high daily flow that requires cheap, simple, fast, but effective treatment of the polluted water. For this purpose, the mechanism of neutralization of AMD Robule with Majdanpek flotation tailings was examined and proven successful (Petronijević et al. 2020a). However, the aim of this paper is to define the characteristics and stability of solid residue (SR) after the neutralization treatment of acid mine water in Robule. The impact of flotation tailings before treatment on the environment was tested, as was the impact of SR after the neutralization process. The main advantage of the flotation tailings is the high content of carbonates, which provides a high degree of neutralizing capacity.

According to Petronijević et al. 2020a, XRD results of flotation tailings after the treatment (SR) showed a high concentration of carbonates 15-20% and an increasing buffering capacity of SR as a result of the precipitation of hydroxides. According to what was previously mentioned, this paper focuses on the classification of mining waste and the investigation of possible harmful effects of flotation tailings on the environment before and after the treatment. The utilization of flotation tailings as off-balance parts of the mine's deposits For this purpose, the SR was subjected to TCLP and EN 12457-4 tests as standard procedures to assess the leaching characteristics of a material. In addition, the stability of the SR was investigated by using melted snow collected in the vicinity of the study site as a leaching agent. The aim was to simulate environmental conditions at a laboratory scale for one year. The obtained drainage water was analyzed and compared with the results of standard leaching procedures.

2. Materials and methods

2.1. Study Area

It is estimated that more than 780×10^6 t of mining waste (overburden and flotation tailings) has been deposited in Bor and its environment in the last century. It is assumed that this deposition contains more than 1,140,000 t of copper (Cu), and approximately 290-350 t of copper is irretrievably lost annually by the leaching of mining waste, additionally polluting surface flows (Milićević et al. 2013). Although mining waste in the vicinity of Bor has been studied in great detail, such tests have not been performed on samples of flotation tailings from Majdanpek so far, which is also part of Zijin Mining Group (the new owner of the copper mining and smelting complex in Bor, Serbia). Since 1961, this mine has been operating, and approximately 378 million tons of flotation tailings have been produced and disposed of. According to the data obtained from Zijin management, the average metal content in the flotation tailings is 770 gt^{-1} Cu, 0.155 gt^{-1} Au, and 1.066 gt^{-1} Ag. Using this data, a simple calculation shows that the flotation tailings contain 292,000 tons of copper, 59 tons of gold, and 403 tons of silver. The exact area of interest was presented on the map during the previous investigation (Petronijević et al. 2020a).

2.2. Sampling

Flotation tailings were sampled in a checkerboard pattern with a sterile sampling trowel and placed in separate sterile bags at 25 equidistant spatial locations. Field spacing was 50 m, and excavation volumes were 0.2 x 0.2 m at about 1 m depth. In laboratory conditions, composite samples were prepared and analyzed by the quartering method. The acid mine water sample was collected from draining pipes at Acid Lake Robule in sterile polyethylene containers in a refrigerator at 4 °C. Temperature, pH, and Eh parameters were measured on location before the samples were transported to the laboratory.

2.3. Characterization of Samples

Previous work (Petronijević et al. 2020a, Sokić et al. 2019) specified the physicochemical characteristics of acid mine waters of Robule Lake before and after treatment with flotation tailings and X-Ray Diffraction (XRD) and Acid Neutralization Capacity (ANC) analyses of the flotation sample before and after treatment of acid mine waters of Robule with 15% of flotation pulp. As previously mentioned, XRD analyses conducted in previous research presented a high concentration of carbonates in flotation tailing, which was confirmed with ANC tests.

The results of XRD and ANC tests point out a high capacity for neutralization and emphasize the possibility of multi-time tailings usage for the neutralization process and the increasing buffering capacity of SR due to the precipitation of metal hydroxides. The same ratio, 15 % of flotation pulp, was used in these experiments in order to prepare enough amounts of SR for leachability testing. Particle size distribution was also analyzed on tailings samples before and after the treatment with bulk density parameters.

The chemical compositions of the liquid and solid samples were measured by the Inductively Coupled Plasma Optical Emission Spectroscopy (ICP-OES) method using an iCAP 6500 Duo spectrometer (Thermo Fischer Scientific, Bremen, Germany). The sulfate content was determined by gravimetric analysis of a substance by selective precipitation in an aqueous solution and then filtrated and measured by the Atomic Absorption Spectroscopy (AAS) method using Perkin Elmer Analyst 300 (PerkinElmer, Inc., Norwalk, CT, USA).

2.4. Experiment

After the successful treatment of acid mine water Robule, further research was focused on defining the characteristics of the solid residue that remained after the neutralization process for the purpose of its safe disposal.

AMD Robule + Flotation Tailings → treated solution safe for environment + Solid residue SR

According to Petronijević et al. 2020a, the average pH value of AMD is 2.47, and from the analyzed ions, sulfate (7.5 g/l), Al2+ (1017.62 mg/l), Fe ions (287 mg/l), manganese (66 mg/l), and zinc (17.6 mg/l) predominate, while hydrogen carbonates ($HCO3^-$), nickel, arsenic, lead, boron, silver, and beryllium are less abundant with concentrations of 2.44 mg/l, 0.6 mg/l, <0.007 mg/l, 0.188 mg/l, 0.02 mg/l, 0.013 mg/l, and 0.013 mg/l, respectively. Also, removal efficiency for Fe, Al, and Cu was 99%, 98% removal of Pb, and 92% removal of Zn in treated solution, respectively.

According to the legislation of the Republic of Serbia [30], this solid residue was characterized by the **T**oxicity **C**haracteristic **L**eaching **P**rocedure (**TCLP**) and EN 12457-4 test as standard procedures to define the leaching characteristics of the material. TCLP is a standard method defined by USEPA to determine the mobility of both organic and inorganic substances present in liquid, solid, and multiphase wastes.

The procedure is as follows [31]: Solid waste is leached with a solution of glacial acid in a solid/liquid ratio of 1:20. This is an analytical method that represents a worst-case scenario, and based on its results, waste is classified as hazardous or not hazardous. For the long-term prediction of metal ion mobility, the EN 12457-4 method is used [32]. The test procedure is similar to the TCLP test, except that distilled water is used as an extraction fluid and the solid-liquid ratio is 1:10. The experiment is conducted for 24 hours at a mixing speed of 0,5 rpm, and the particle size has to be less than 1 cm. Depending on the results, waste and drainage water are classified as inert, non-hazardous, or hazardous waste.

A leaching test with melted snow was performed to investigate the leachability of heavy metals from the tailings under more realistic field conditions (compared to the TCLP and EN 12457-4 tests).

In the vicinity of the study site, sufficient amounts of snowfall were collected, which, after melting, were used as an extraction fluid in the leaching test. The snow was collected in two clean plastic containers during winter 2018.

Experiments consisted of two samples: one was a mixture of flotation tailings before treatment and melted snow, and the second was SR after neutralization treatment with melted snow. Both samples were mixed at the same ratio as the EN 12457-4 test prescribes: L/S = 10.

In accordance with the conditions in the field and previous research (Wang et al. 2015, Radovanović et al. 2017), a long-term test of tailings leaching was performed by exposing the samples of flotation tailings to the influence of the precipitation (melted snow) in an orbital shaker. The shaking was performed for three months at 54 rpm at an average temperature of 16 °C in order to simulate one year's leaching period in field conditions. Every day, the L/S ratio was observed, and if necessary, the melted snow was added to obtain the initial 1:10 ratio.

3. Results and discussion

3.1. Characterization of the Flotation Tailings' Sample from Copper Mine Majdanpek before the treatment of AMD Robule and SR after the treatment

Results of the chemical composition of flotation tailing before and after the neutralization treatment of the AMD present a high concentration of sulfur (8.16 and 8.8%, respectively), which indicates a high potential for the generation of AMD. The results also represent a slight increase in Cu (0.066–0.089%), Ni (24.95–30 ppm), Zn (0.0918–0.101%), and Fe (10.03–11.4%) in SR after the treatment due to precipitation of these metals from the AMD in contact with carbonates and other neutralizing components from the flotation tailings. The chemical composition of the flotation tailings from copper mine Majdanpek before and after the treatment of acid mine water Robule is given in Table 1.

Table 1. Chemical composition of the flotation tailings from Copper Mine Majdanpek before and SR obtained after the treatment of acid mine water Robule (%)

Elements	Flotation tailings [%]	SR [%]
pH	8.48*	7.49*
Antimony (Sb)	0.0055	0.035
Copper (Cu)	0.066	0.089
Vanadium (V)	0.0085	0.0115
Cadmium (Cd)	5 ppm	5.49 ppm
Nickel (Ni)	24.95 ppm	30 ppm
Lead (Pb)	0.009	0.0115
Chromium total (Cr)	34.43 ppm	35.5 ppm
Zinc (Zn)	0.0918	0.101
Sulfur (S)	8.16	8.80
Aluminium (Al)	0.69	0.72
Iron (Fe)	10.03	11.4
Boron (B)	0.063	0.168
Cobalt (Co)	21.46 ppm	25.5 ppm
Manganese (Mn)	0.126	0.127
Arsenic (As)	24.9 ppm	25.1
Barium (Ba)	20.3 ppm	22
Molybdenum (Mo)	6.35 ppm	6.56
Selenium (Se)	<2.3 ppm	<2.3 ppm

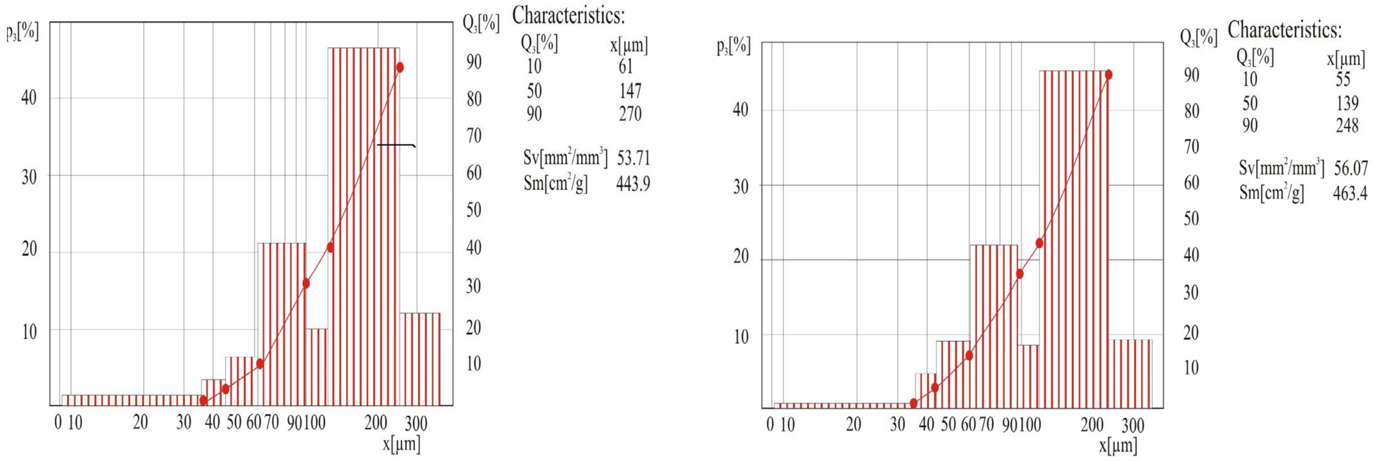
*unitless

As it is presented in Figure 1, solid residue after the treatment has ~4 % more powder particles (size less than 63 µm) while the content of fractions > 250 µm decreased in SR, probably due to fragmentation of larger fractions of carbonates and braking apart during the leaching experiment. According to (Hansen, Yainatos & Ottosen 2005), for example, the leaching of copper is a function of pH for different particle sizes. Copper from the smallest fraction (below 53 µm) can be dissolved up to 70%, and from a bigger fraction (<212 µm) up to 35% at pH below 4. These results result from increasing copper sulfide and decreasing oxide and carbonate copper content with increasing particle size. Particle size distribution is one of the key parameters that should be studied in detail because it determines the treatment and waste disposal processes.

The bulk density of the tailings sample before and after the treatment was also determined. The measured value for tailings before treatment is 1.21 g/cm³, while tailings after treatment are 1.17 g/cm³, indicating a decrease in fraction size, i.e., reducing surface.

3.2. TCLP and EN Test results of Flotation Tailings' Sample from Copper Mine Majdanpek before and SR obtained after the treatment of AMD Robule

The TCLP test results are presented in Table 2, along with the maximum allowed concentration values (MAC). All values are below the regulation level, which classified flotation tailings and solid residue as non-hazardous waste. The leaching test also showed higher concentrations of Cu, Ni, Sb, Se, and Pb in the leached solution of SR than in the initial sample of tailings due to its higher content in SR (as shown by the chemical analysis of samples before the TCLP test in Table 1).



The particle size distribution of flotation tailings before treatment

The particle size distribution of SR

Fig. 1. Particle size distribution of flotation tailings before and SR obtained after the treatment

According to the EN test data presented in Table 3, flotation tailings before treatment are classified as **hazardous waste** based on high values of Antimony (Sb), while solid residue after treatment is classified as **non-hazardous waste**. Based on (Lindsay et al. 2015), some elements, such as Sb, show greater mobility at approximately neutral pH conditions. Selenium (Se) values are slightly elevated for both samples, but not enough to categorize the waste as hazardous. It is characteristic that the values of sulfate are significantly more leachable in the SR, but also within the limits of non-hazardous waste. This is probably due to the presence of the water-soluble sulfates generated and precipitated through AMD neutralization.

Since these two tests, TCLP and EN 12457-4, obtained different results (worst-case scenario and leaching with deionized water), an additional long-term leaching test was performed. This was carried out considering the fact that one of the long-term exploitation consequences is the release of SO_2 into the atmosphere and the formation of slightly acid precipitation (which has not been sufficiently tested). According to Nyborg et al. 1997, rainfall usually has an approximately five times higher sulfuric (S) concentration than snowfall, so in this paper, melted snow is used for the leaching test in order not to bring additional sulfur into the leaching mixture.

Results of the chemical characterization of melted snow and leaching solution after the long-term leaching test are given in Table 4.

Table 2. Results of TCLP tests of flotation sample before and SR obtained after the treatment

Parameter [mgL^{-1}]	Sb	As [μgL^{-1}]	Ba	Cu	Cd	Mo	Ni	Pb	Se [μgL^{-1}]	Ag	Cr	Zn
Flotation tailings	2	<0.01	<0.01	4.1	0.2	<0.01	0.3	<0.1	0.02	<0.01	<0.01	7.3
SR	2.4	<0.01	<0.01	6.02	0.2	<0.01	0.4	0.5	0.04	<0.01	<0.01	4.46
MAC	15	5	100	25	1	350	20	5	1	5	5	250

Table 3. Results of EN 12457-4 test of flotation sampling before and after treatment

Parameter	Flotation tailings	SR	Inert waste	Non-hazardous waste	Hazardous waste
pH	7.76	7.2		> 6	
Units	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg
Antimony	24	2.6	0.06	0.7	5
Arsenic	0.0001	0.0001	5	2	25
Copper	0.1	0.1	2	50	100
Barium	0.1	0.1	20	100	300
Cadmium	0.01	0.01	0.04	1	5
Molybdenum	1.93	0.1	0.5	10	30
Nickel	1	5	0.4	10	40
Lead	1	1	0.5	10	50
Selene	0.12	0.26	0.1	0.5	7
Chromium	0.1	0.1	0.5	10	70
Zinc	0.1	0.1	4	50	200
Sulphate	1000	17600	1000	20000	50000
Chloride	100	0.1	800	15000	25000
solid residue after evaporation	125	2410			

Table 4. *Chemical characterization of melted snow and leaching solution after extended leaching test*

Parameter	Melted snow - [mg/l]	Flotation tailings + snow [mg/l]	SR + snow [mg/l]	Regulative level TCLP [mg/l]	EN 12457-4 tests [mg/kg]		
					Inert waste	Non-hazardous	Hazardous waste
pH	6.32	6.8	6.6	/		>6	
Sb	<0.05	3.1	0.24	15	0.06	0.7	5
As	<0.02	0.0001	0.0001	5	5	2	25
Cu	0.06	<0.01	0.01	25	2	50	100
Ba	0.056	0.01	0.01	100	20	100	300
Cd	<0.005	0.01	0.01	1	0.04	1	5
Mo	<0.01	0.23	0.01	350	0.5	10	30
Ni	0.05	<0.1	< 0.1	20	0.1	0.5	7
Pb	0.05	0.5	1.1	5	0.5	10	50
Zn	0.16	0.01	0.01	250	4	50	200
Se	<0.02	0.07	0.015	1	0.1	0.5	7
S	<0.02	1050	1970	/	1000	20000	50000
Cr	<0.01	<0.01	0.01	5	0.5	10	70
/				Not defined			

A long-term leaching test showed that flotation tailing is not safe for disposal into the environment before the treatment step due to the leaching of antimony, which has a much higher concentration than the regulatory level of the EN 12457-4 test allowed, and classified flotation tailing as hazardous waste. Other element concentrations in the leaching solution are significantly below the regulatory levels of the TCLP and EN 12457-4 tests. Also, the results presented in Table 4 are very similar to those obtained by the EN 12457-4 test, showing that flotation tailings are not suitable for deposition without any previous treatment.

After all three leaching tests were performed, the obtained results of SR showed a concentration well below regulation limits. Based on these results, the described treatment methodology can be implemented in a scale-up system to utilize both liquid and solid mining waste and preserve the environment. According to previous results, a proposed line system of treatment is presented in Figure 2. This idea will be the topic of further research, which should include optimization of the flow and amount of AMD and tailings and sizing of the required equipment to achieve the necessary process efficiency.

The definition of an economical and environmentally friendly type of waste integral treatment that is based on sustainable development principles, utilization of already disposed waste, reducing present and future pollution, reducing remediation costs, and converting toxic waste to a free resource should be imperative for the present mining management and is supported by the data presented in this paper.

Instead of individual flotation tailings treatment, this paper presents research on the use of the tailings as an agent for the treatment of AMD accumulated in Lake Robule. After this simultaneous treatment of AMD and the flotation tailings, the resulting treated water is safe for discharge (as shown in the previous paper). Also, according to the results of the applied leaching tests, the obtained SR (treated flotation tailings) is safe for disposal, with all parameters far below the regulation limits. Modern scientific achievements and modern technologies have enabled the economic revaluation of valuable elements from solid and liquid mining waste with the improvement of environmental standards. Treatment of flotation tailings is usually expensive, although it is essential for preserving the environment. Some future perspectives for research on

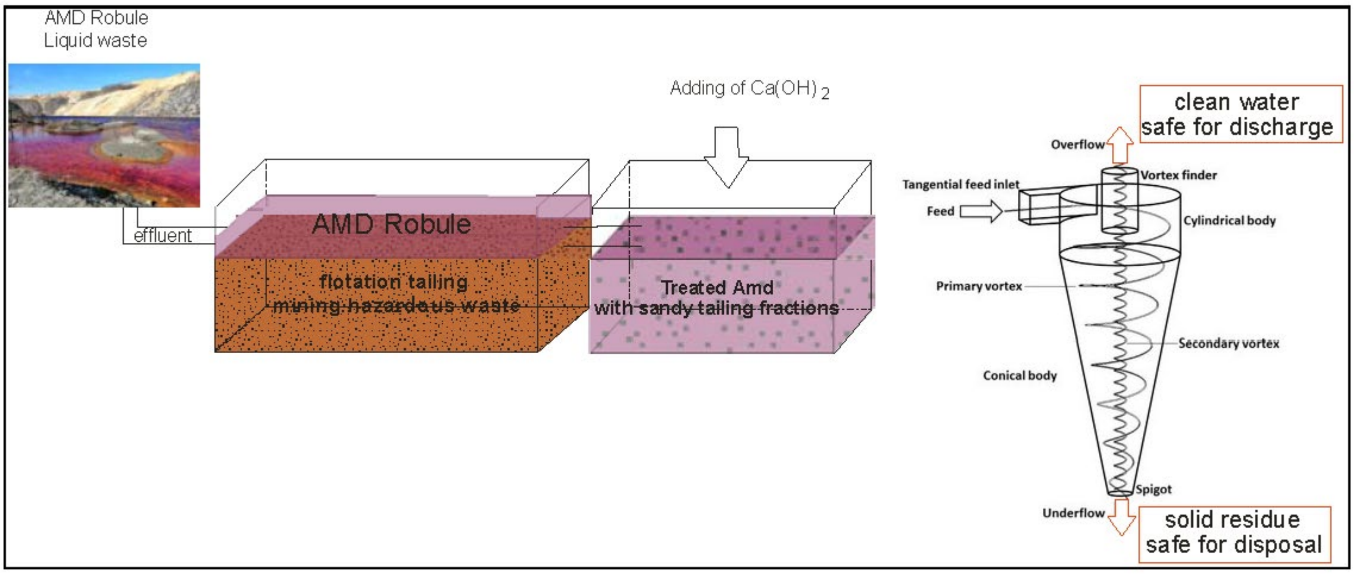


Fig. 2. Schematic proposition of active treatment of AMD Robule with safe disposal of SR after treatment

this topic could cover calculating the Environmental Risk, applying the BCR procedure to top soil pollution, analyzing fumigation to investigate the impact on surface waters, or airborne pollution dispersion.

4. Conclusions

Treatment methodology, defined in this paper, presents an environmentally friendly type of integral treatment based on sustainable development principles: utilization of already disposed waste, reducing present and future pollution, reducing remediation costs, and converting toxic waste to a free resource, which should be imperative for present mining management. Incorporating previous work results with this one, an environmentally friendly, inexpensive (due to utilization of already disposed of unsafe waste), and effective treatment is suggested, based on the following conclusions:

- The solid residue SR of flotation tailings after AMD neutralization treatment retains its high sulfur concentration with a slight increase (from 8.16 to 8.80 mas%) and has a higher content of heavy metals. These results come from the precipitation of those metals.
- After the AMD treatment of flotation tailings, the solid residue has an about 4% increase in powder particles of size less than 63 μm, while the content of particles greater than 250 μm decreases. This is supported by the measured values of bulk density before (1.21 g/cm^3) and after (1.17 g/cm^3) AMD treatment. This is due to carbonate degradation and the breaking apart of particles during the leaching experiment.
- For the TCLP test results, all values are below the regulation level, which classifies both the flotation tailings and solid residue as non-hazardous waste.
- Due to the concentration of Antimony (Sb), according to the EN 12457 test, the flotation tailing before AMD treatment (24 mg/kg) is classified as hazardous waste above 5 mg/kg for the maximum allowed concentrations, while the solid residue after AMD treatment (2.6 mg/kg) is classified as non-hazardous waste (for the maximum allowed concentration of 0.7 to 5 mg/kg).
- Based on all three leaching tests (TCLP test, EN 12457-4 test, and long-term leaching test), the results of SR show a concentration of hazardous elements well below the regulation limits.

Acknowledgments

This research was supported by the Ministry of Science, Technological Developments and Innovation of the Republic of Serbia (Contract number: 451-03-47/2023-01/200023; 451-03-47/2023-01/200135).

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