

ACID MINE DRAINAGE PREDICTION FOR TAILINGS IN THE BAIA MARE AND SOUTHERN APUSENI MINING AREAS, ROMANIA

Erika LEVEI¹, Marius ROMAN¹, Mirela MICLEAN¹, Gheorghe BORODI² & Marin SENILA¹

¹INCDO-INOE 2000 Research Institute for Analytical Instrumentation, 67 Donath Street, 400293 Cluj-Napoca, Romania, e-mail: erika.levai@icia.ro

²National Institute for Research and Development of Isotopic and Molecular Technologies, 65-103 Donath, 400293 Cluj-Napoca, Romania

Abstract: To predict the acid mine drainage potential of tailings from Baia Mare (Bozanta and Sasar) and Southern Apuseni (Ribita and Mialu) mining areas, acid base accounting test, pH, sulphur, total carbon and metal contents of wastes were determined. The results revealed high Cu, Pb, Zn, Cd, As, Fe concentrations in all studied tailings. The highest metal concentrations were determined in the Bozanta tailing impoundment. The net neutralization potential and neutralization potential ratio revealed that tailings stored in the Mialu, Sasar and Bozanta impoundments are possible acid mine drainage sources and thus can pose contamination risk for the environment. The tailings from Ribita deposit have low sulphur content and high neutralization potential and thus very low acid mine drainage potential.

Key words: acid mine drainage, tailings, metals, Baia Mare mining area, Southern Apuseni mining area

1. INTRODUCTION

Mining contributes to economic growth but has also important negative impact on the environment (Dold, 2008; Nordstrom, 2011). During mining and ore processing operations, huge amounts of tailings consisting of ground rock and process effluents are generated and disposed as slurry in surface tailings management facilities (TMF) (Grangeia et al., 2011).

Presently, the challenges associated with tailings disposal and storage are increasing on the one hand, due to the advance of technology that allow the exploitation of low grade ores but generates higher volumes of waste and, on the other hand, due to numerous tailings related environmental accidents. The most important tailings dam failures in Europe were reviewed by Rico et al., (2008). From the most severe accidents it can be mentioned those from Aznalcollar, Spain in 1998 (Grimalt et al., 1999; Simon et al., 1999) and Baia Mare, Romania in 2000 (Cordos et al., 2003; Gurzau et al., 2012), when millions of tons of tailings carrying dissolved metals and reactive particles were discharged into the environment causing considerable damages. In addition, to such

catastrophic events, the transport of tailings into the adjacent environment by wind and water erosion along with ground and surface waters contamination represent other risks associated to TMF for decades after closure (Navarro et al., 2008).

In the presence of atmospheric oxygen and water, acid release can occur by oxidation of the sulphide minerals (pyrite, arsenopyrite, galena, chalcopyrite, sphalerite) and dissolution of iron sulphates and other sulphate minerals with lower solubility (Smuda et al., 2008; Lapakko, 2002, Edelev, 2013). This phenomenon producing acid to neutral waters containing high loads of dissolved metals is commonly known as Acid Mine Drainage (AMD) (Garcia et al., 2005).

The oxidation of sulphide minerals in unaltered rock is a naturally occurring process with slow reaction rates. During mining, the exposure of large surfaces of sulphide rich ores to water and air increase the oxidation rate and generate high amounts of acidic waters (Akabaza et al., 2007).

The AMD generating processes appear during the operational stage of TMF, determined by management deficiencies, geotechnical construction or climatic conditions. However, these processes reach the maximum after the cease of tailings

storage, when the water level in TMF decrease and appear unsaturated areas, containing oxygenated waters and atmospheric oxygen (Dold & Fontboté, 2001; Sima et al., 2011). The main environmental impact of AMD is associated with the high contents of metals that are mobilized by the acidic waters.

There are several tests for the determination of tailings AMD generation potential. Static tests are based on the geochemical characterization of tailings and consist in the determination of acid production and neutralization capacity of tailings, while kinetic tests provide information on the tailings geochemical changes in time and consist in modeling the acid production and its neutralization rates in different time spans (Lapakko, 2002, Garcia et al., 2005). The most used static prediction test is Acid Base Accounting (ABA), based on the balance between acid producing processes expressed as Acid Producing Potential (APP) and acid consuming processes expressed as Neutralization Potential (NP). There are several variations of these tests depending on the method used to determine the APP and NP in tailings (Plante, 2012; Lapakko, 2002). A comparative assessment of different static tests used for the determination of tailings AMD generation potential is presented by Plante et al. (2012).

The standard ABA test developed by Sobek et al. (1978) considers that all sulphur is present as pyrite and will be totally transformed in sulphuric acid, while the NP is determined by digestion of tailings with an excess of HCl under boiling conditions and titration of the excess acid with NaOH. This approach will overestimate the produced acid because in natural condition not all sulphur species will produce sulphuric acid. Another approach for APP estimation is based on the sulphide sulphur content of tailings. However, this method will underestimate the actual APP of samples with high content of acid producing sulphate minerals (jarosite). Thus the selection of the appropriate APP calculation method can be made based on the tailings mineralogy that indicates if acid producing sulphate minerals are present in tailings (Lapakko, 2002). The NP can be calculated from the total or inorganic carbonate content, considering that the carbon is present in tailings only as calcium carbonate. Alternatively, it can be determined by digestion of tailings at boiling point or at room temperature, with different volumes of acids and back titration of the excess acid (Sobek et al., 1978; Lapakko, 2002; Lawrence & Scheske, 1997; Kossof et al., 2012).

The kinetic tests such as the leach or humidity tests, give better estimation of the AMD risk but they are time consuming (needs months to years) and are not cost effective for high number of samples.

Romania has a long tradition of base (Cu, Cd, Pb, Zn) and precious metals (Au, Ag) mining, most centres and tailing impoundments being located in the north of the country (Maramures County) or in the west, in the Apuseni Mountains (Golden quadrangle). The environmental impact of AMD in different mining sites was studied all over the world (Stevanovic et al., 2013; Rashed, 2010; Romero et al., 2010; Grangeia et al., 2011; Navarro et al., 2008). In Romania although there are a high number of mining sites, most of them in conservation or abandoned, there are only a few published data on the AMD generation potential of tailings stored in TMF (Zobrist et al., 2009; Dogaru et al., 2009; Sima et al., 2011; Stumbea, 2010; Damian & Damian, 2006; Nagy-Korodi et al., 2011; Balaban et al., 2011; Levei et al., 2013.)

The objective of this study was to assess the acid drainage potential and the risk of environmental contamination with metals released from tailings resulted from nonferrous and precious metals mining, stored in four TMF from Baia Mare and Southern Apuseni mining areas, Romania.

2. MATERIALS AND METHODS

2.1. Site description and sample collection

Baia Mare and Southern Apuseni are two of the major metallogenic provinces in Romania, located in the eastern part of the Carpathian belt and in southern part of the Apuseni Mountains, respectively.

The ore deposits of the two studied areas are related to Miocene–Pleistocene calc-alkaline volcanic activity. In the Baia Mare area the main ore types are epithermal Pb–Zn(–Cu–Au–Ag) vein deposits. The Southern Apuseni area is famous for associated world-class low-sulphidation and subordinate high-sulphidation epithermal veins, breccia-hosted gold and associated porphyry copper deposits (Neubauer et al., 2005). The mineralogical features of gold and base metal deposits from the Apuseni mining area indicates their belonging to the low sulfidation epithermal type, similar to the polymetallic mineralization of the Baia Mare area (Marcoux et al., 2002).

In this two areas (Fig. 1) the main industrial activities was based on mining and processing of nonferrous ores containing base and precious metals (Levei et al., 2009; Damian et al., 2008; Zobrist et al., 2009; Sima et al., 2011).

The Sasar TMF is located on the alluvial plane of Sasar River, covers about 36 ha and was used for tailing storage between 1970 and 1982. The Bozanta TMF is placed on the alluvial plane of Lapus River, covers 102 ha and was functional

between 1976 and 2006. Both TMF were used for the storage of tailings resulted from the mining and processing of Au-Ag deposits (Baita, Sasar, Valea Rosie, Suior), complex Pb-Zn (-Cu-Au-Ag) sulphide deposits (Cavnic, Baiut) and cupriferous deposits (Nistru, Baia Sprie). Currently they are under conservation, have no impermeable liner underneath but a drainage system secures their structural stability (Gurzau et al., 2012; Senila et al., 2012).



Figure 1. Location of the Baia Mare and Southern Apuseni mining areas, Romania

Two of the most important waste deposits in the Southern Apuseni mining area are the Ribita and Mialu TMF. The Ribita TMF is situated in the Crisul Alb river basin, covers about 60 ha and was used between 1986 and 2006 for storage of tailings resulted from the exploitation of epithermal gold veins and porphyry-copper ores from Ruda-Barza and Valea Morii and Valea Arsului deposits. The Mialu TMF is placed in the Certej river basin, covers 43 ha and between 1983 and 2006 stored the tailings resulted from Au-Ag-Te epithermal ore (Sacaramb) and polymetallic epithermal ore (Coranda-Hondol) exploitation. Presently both TMF are under conservation (Sima et al., 2011).

Five composite samples from the Bozanta, Sasar, Ribita and Mialu TMF were randomly collected in August 2012 from 0-25 cm depth using a polypropylene shovel. For each composite sample 6 subsamples were collected from an area of 100 m² and mixed to obtain a composite sample that was subjected to analysis.

2.2. Sample preparation and analysis

The mineralogical composition was determined by powder X-ray diffraction (XRD) using the high-resolution Bruker D8 Advance diffractometer (Bruker-AXS, Karlsruhe, Germany) at Cu K α radiation. The diffraction patterns were processed using MATCH! 2.0 software and the

ICDD PDF-2 database. Semi-quantitative analysis was made by the Reference Intensity Ratio (RIR) method.

For the determination of metal content 1 g of dried tailings (105 \pm 5 $^{\circ}$ C) ground and sieved to <250 μ m was digested with 28 mL aqua regia according to the standardized procedure described in ISO 11466 (1995). The filtrate was diluted to 100 ml with ultrapure water and the contents of metals were determined by inductively coupled plasma optical emission spectrometry (ICP-OES) using OPTIMA 5300DV multichannel spectrometer (Perkin-Elmer, Norwalk, USA).

Sulphur content in tailings was determined by X ray fluorescence using the spectrometer α 6500 (INNOV-X, Woburn, MA, US).

The pH of tailings was measured in 1/5 (m/v) aqueous extract with the 350i multiparameter (WTW, Wilhelm, Germany).

Total carbon content (TC) was determined by combustion using the Multi N/C 2100S Analyser (Analytik Jena, Jena, Germany).

The Acid Base Accounting (ABA) test was conducted according to modified Sobek method (Lawrence & Wang, 1996) assuming that sulphur is present only in the form of pyrite, while carbonate as calcite (CaCO₃).

To calculate the NP, a volume of 90 ml of distilled water and various volumes of 1N HCl were added to 2 g tailings. The samples were shaken at room temperature for 2 hours, then another volume of 1N HCl was added to samples and shaken for 24 hours. The final volume of the sample was made up to approximately 125 ml with water and the pH was measured. If the pH was around 2, the sample was titrated with 0.5N NaOH. If the pH was higher than 2.5, the sample was discarded and reanalysed with higher amounts of added acid. The NP expressed as the content of CaCO₃ (kg/t) was calculated according to equation 1:

$$NP = \frac{50x(1xV_{HCl} - 0.5xV_{NaOH})}{w} \quad (1)$$

Where I is the normality of HCl used, V_{HCl} is the volume of added acid (ml), 0.5 is the normality of NaOH used for titration, V_{NaOH} is the volume of base consumed (ml), and w is the tailings weight (g).

The fizz ratings were evaluated by observing the reaction between pulverized tailings and a few drops of HCl and classified as: none, slight, moderate or strong (Lawrence & Wang, 1996). The volume of 1 M HCl added for various fizz ratings are presented in table 1.

Table 1. Volume of 1N HCl added to tailings for different fizz ratings

Fizz rating	Added volume of 1M HCl (ml)	
	Initially	After 2 hours
None	1.0	1.0
Slight	2.0	1.0
Moderate	2.0	2.0
Strong	3.0	2.0

The APP level was calculated from the total sulphur content, based on the assumption that each mole of sulphur produces two moles of acid and one mole of carbonate will neutralize two moles of acid. Thus each S percent requires for neutralization 31.25 kg CaCO₃/t tailings.

The difference between NP and APP is named Net Neutralization Potential (NNP). The tailings is considered as AMD generating when the NNP value is lower than -20 kg CaCO₃/t, while a value higher than +20 kg CaCO₃/t indicates that the material is non- AMD generating. For NNP values between -20 and +20 kg CaCO₃/t the prediction about the AMD generation is uncertain (Lawrence & Wang, 1996; Song & Yanful, 2011).

Besides NNP, the Neutralization Potential Ratio (NPR) was calculated as the ratio between NP and APP. The tailings is considered as AMD producing if NPR values are lower than 1, non-AMD sources if NPR is higher than 2, and are uncertain AMD sources if NPR value ranges between 1 and 2 (Skousen et al., 2002, Plante et al., 2012).

3. RESULTS AND DISCUSSION

3.1. Mineralogical composition

The mineralogical composition of tailings is summarized in table 2. Primary minerals belonging to silicate group were found to be the most abundant in tailings from all TMFs. Minor or trace minerals involved in AMD generation (sulphides and sulphates) and carbonates with neutralization properties were identified as minor or trace constituents.

The identification of primary minerals (sulphides) and secondary minerals (sulphates) in the tailings from Mialu, Sasar and Bozanta TMFs, indicates that these tailings are still reactive exhibiting potential contamination risk through AMD generation. Minerals with neutralization potential (carbonates, albite, biotite) were also identified in traces in tailings from all TMFs.

Table 2 The mineralogical composition of tailings

Mineral group	Mineral species	TMF			
		Mialu	Ribita	Sasar	Bozanta
silicates	quartz	++ ^a	++	++	++
	albite	+	+	+	++
	muscovite	+	+	++	++
	biotite		±	±	±
	K-feldspar	+	+	+	+
sulphides	chalcopyrite	+	±	±	±
	pyrite	±			
	arsenopyrite	±		±	±
sulphates	jarosite	+		±	±
	gypsum	+		±	+
	anglesite			±	+
carbonates	calcite	±	±	±	+
	dolomite	±	+		±

^arelative abundance: major (++), minor (+), trace (±)

3.2. Chemical characterization of tailings

The range and average of metals concentration in tailings for each studied TMF are presented in table 3 and table 4. Although, strictly speaking, As is a metalloid, the term metal is used in this paper to include all the elements under discussion.

The high variability of metal concentration observed in tailings from each studied TMF revealed the inhomogeneous composition of tailings deposited in TMF. The chemical composition of tailings revealed high contents of metals in tailings.

The occurrence of major elements in tailings from Mialu, Ribita and Bozanta TMF was found to follow the order Fe>Ca>Al>Mg>K>Mn>Na>Ba. This order generally agrees with the tailings mineralogy, considering quartz and aluminosilicates as the dominant phases. In the tailings from Sasar TMF the major elements followed the order Fe>Al>Mg>K>Ca>Mn>Na>Ba.

The relative abundance of metals varied from tailings to tailings, some of the analyzed trace metals being found in higher contents than major elements. The average contents of Fe in tailings were similar in the four studied TMF, while the Al, Ca, Cu, Pb, Zn, Cd, As contents were of different orders of magnitude. The highest contents of As, Cd, Cu, Pb, Zn, Ni, Cr were found in tailings from Bozanta TMF, followed by those determined in tailings from Sasar TMF. In the tailings from Mialu and Ribita TMF the metal contents were lower than in tailings from Baia Mare mining area.

Table 3. The metal contents (mg/kg) in tailings from Sasar and Bozanta TMF, Baia Mare mining area

Metals	Range	Average ^a
Sasar TMF		
Al	6705-10705	7889
As	211-741	384
Ba	118-399	203
Ca	303-940	563
Cd	2.5-10.7	6.0
Co	1.2-15.4	8.2
Cr	7.8-65.1	23.3
Cu	138-527	312
Fe	29450-75400	43720
K	1430-2071	1775
Mg	2990-3620	3224
Mn	209-551	416
Na	87.3-472	284
Ni	1.0-12.6	7.3
Pb	354-802	527
Zn	137-698	452
Bozanta TMF		
Al	10950-16810	13232
As	550-2660	1446
Ba	68.8-97.3	85.8
Ca	1415-89900	23890
Cd	3.0-18.2	11.0
Co	5.0-27.4	15.6
Cr	123-556	303
Cu	149-13200	4980
Fe	39800-68500	57080
K	1767-2640	2061
Mg	1235-5810	3637
Mn	393-5300	2084
Na	204-448	306
Ni	13.6-68.0	37.2
Pb	630-878	749
Zn	221-3730	1201

^athe average of five sample;

The high Pb, Cu and As contents were confirmed by the presence of anglesite, chalcopyrite and arsenopyrite in the tailings. Despite the important contents of Ni, Zn, Cr and Cd determined in tailings no minerals containing these metals were identified by the XRD analysis. However it is known that sulphide minerals also contain traces of Cd, Co, Ni, Zn and As that may substitute the major metal or the sulphur (Koski, 2012). Minerals like albite, K-feldspar and muscovite are Al sources that act as a metal scavenger (Gilchrist et al., 2009). The total carbon contents were 0.5-4.1 wt % in Ribita, 0.2-1.0 wt % in Mialu, 0.3-0.8 wt % in Sasar and 0.1-3.4 wt % in Bozanta tailings.

3.3. AMD drainage potential

Tailings characteristics related to AMD

generation potential are presented in table 5.

Table 4. The metal contents (mg/kg) in tailings from Ribita and Mialu TMF, Southern Apuseni mining Area

Metals	Range	Average ^a
Ribita TMF		
Al	15645-30400	21464
As	66.8-482	194
Ba	17.6-137	69.7
Ca	15430-52350	32431
Cd	0.4-2.4	1.2
Co	12.6-16.0	14.7
Cr	18.4-32.3	28.3
Cu	64.4-376	156
Fe	33450-72500	47638
K	2092-2630	2339
Mg	8800-9700	9399
Mn	159-3300	1059
Na	401-609	492
Ni	22.1-38.4	29.8
Pb	15.1-79.0	48.5
Zn	32.3-228	116
Mialu TMF		
Al	1816-4385	2737
As	241-496	375
Ba	17.6-37.4	25.4
Ca	5165-18335	12063
Cd	0.8-3.7	2.3
Co	1.2-5.6	2.3
Cr	7.6-21.8	13.7
Cu	19.2-34.0	24.2
Fe	27950-42800	35570
K	956-2269	1599
Mg	246-1635	807
Mn	24.1-217	102
Na	76.7-163	103
Ni	6.2-18.7	10.4
Pb	86.5-212	149
Zn	48.0-173	109

^athe average of five sample;

Table 5. Tailings characteristics related to AMD generation potential

TMF	pH	S (%)	kg CaCO ₃ /t			
			APP ^d	NP ^b	NNP ^c	NRP ^c
Bozanta	6.79	0.035	1.09	1.80	0.71	1.65
Sasar	4.98	0.059	1.84	3.30	1.46	1.79
Ribita	8.16	0.057	1.78	26.9	25.1	15.1
Mialu	5.62	0.294	9.19	6.05	-3.14	0.66

^aAPP-acid producing potential; ^bNP-neutralization potential; ^cNNP- net neutralization potential (NP-APP); ^dNPR-neutralization potential ratio (NP/APP).

Generally, the total sulphur concentration was low, 0.03-0.14 wt% in tailings from Ribita TMF, 0.17-0.66 wt% in those from Mialu TMF, 0.03-0.12

wt % in Sasar TMF and 0.03-0.05 wt% in Bozanta TMF. The high sulphur contents, determined in tailings from Mialu TMF, suggest that this impoundment can act as AMD producer.

The pH of tailings ranged from acidic to circum neutral in the Mialu (3.1-7.6), Sasar (4.0-6.6) and Bozanta (4.8-8.3) TMF, while in Ribita TMF the pH was in the neutral range (7.6-8.6).

The NNP values showed that the tailings from Mialu, Sasar and Bozanta TMF are uncertain as AMD sources, while those from Ribita are non-AMD sources. The NPR values confirmed the AMD prediction based on the NNP values for Sasar, Bozanta and Ribita TMF, while tailings from Mialu TMF were classified as AMD generators. These findings are also confirmed by the pH values determined in tailings.

The classification of Sasar and Bozanta tailings as uncertain AMD sources and those from Ribita as non-AMD source by both NNP and NRP values is also confirmed by the mineralogical composition of the tailings. The Ribita deposit showed similar sulphur content with the other studied tailings, but had much higher neutralization potential and thus very low AMD potential. This fact is confirmed also by the high pH of tailings and the presence of dolomite as minor mineral (5-20%).

The classification of the Mialu tailings as uncertain AMD source by the NNP value and as AMD producing by the NRP value suggest that this tailings has the highest AMD generation potential, among the studied tailings. This finding is confirmed by XRD analysis that revealed the presence of chalcopyrite, jarosite and gypsum as minor minerals (5-20%). To confirm these findings an investigation on a larger number of samples is necessary.

Our results are in accordance with those reported by Sima et al., (2011) for the NNP of recent tailings stored in the Ribita TMF. Our data showed an average NNP in the uncertain range (-3.14 kg CaCO₃/t) for tailings from Mialu TMF, while Sima et al., (2011) classified these tailings as certain AMD sources, as resulted also from the NPR ratio in our study.

4. CONCLUSIONS

The studied mine tailings were highly contaminated with As, Cu, Pb, Zn, Cd, Fe, although the metals concentrations varied largely between the studied TMFs. The NNP and NPR values revealed that tailings stored in the Mialu, Sasar and Bozanta TMFs are possible AMD sources and thus can pose contamination risk for the environment, while tailings from Ribita TMF had have low sulphur

content and high neutralization potential and thus very low AMD potential. These findings were also confirmed by the mineralogical composition of the tailings.

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