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# An integrated management strategy for acid mine drainage control of sulfidic tailings

Carolina Mafra <sup>a,\*</sup>, Hassan Bouzahzah <sup>a</sup>, Lachezar Stamenov <sup>b</sup>, Stoyan Gaydardzhiev <sup>a</sup>

- <sup>a</sup> University of Liege, GeMMe Minerals Engineering and Recycling, Sart-Tilman Campus-B52, 4000 Liège, Belgium
- <sup>b</sup> Dundee Precious Metals Chelopech, 2087 Village of Chelopech, Bulgaria

#### ABSTRACT

The oxidation of sulfide minerals releases acidic leachate enriched in sulfate, iron, and toxic metals and metalloids (e.g., As, Sb, Cu, Pb, Cd, Zn, Hg, etc.) known as acid mine drainage (AMD). This eco-hydrological hazard is imminent when mineralogical and geochemical analysis indicates that acid-generating (sulfides) overcome the acid-neutralizing (carbonate) minerals in the ore and mining waste. AMD represents a multifaceted problem faced by the mining industry. The most widespread management technique is to store the tailings into engineered impoundments and actively neutralize the acidity produced by the oxidation of the sulfide minerals by adding alkaline chemical agents. However, this method involves a constant and long-term commitment, elevated operational costs, and several geotechnical and environmental drawbacks. In a pre-mining stage, geological, geochemical, mineralogical, and textural characterization of the mine waste could guide proper and site-specific mining waste management.

In this paper, we present an integrated strategy to reduce the acid mine drainage (AMD) potential of the Dundee Precious Metals Chelopech (DPM-Ch) tailings through a combination of environmental desulfurization and cemented paste backfill (CPB) – the latter one being already practiced on-site. In the proposed scenario, a desulfurization plant would be installed downstream to re-float the non-valuable reactive pyrite. The reactive pyrite concentrate would be combined with the CPB material, whereas the remaining tailings would be discharged at the tailings management facility (TMF). The lack of neutralizing gangue (NP of untreated tailings < 8 kg CaCO<sub>3</sub>/t) precludes converting the tailings into non-acid generating. However, reducing nearly six times the amount of lime required to neutralize the acidity produced by pyrite oxidation was feasible without substantially increasing operational costs. Supporting the environmental benefits of combining desulfurization and backfill, an X-ray mapping of the CPB material revealed that this approach effectively encapsulates the reactive gangue, ultimately avoiding pyrite oxidation and leaching of heavy metals.

#### 1. Introduction

Mining is an essential activity for both economic prosperity and social development. However, mining operations inevitably generate a large volume of waste material (e.g., the volume of waste-to-product ratio can vary from 100:1 to 1000000:1) (Qi and Fourie, 2019). Disposing of non-economic material (e.g., waste rock and tailings) is among the most sensitive challenges the mining industry faces. The accommodation of the rejected product from the beneficiation process into tailings dams leads to intrinsic technical, environmental, economic, and social threats. The growth of social and environmental liabilities has forced the mining industry to reduce its environmental footprint. In this framework, predictive and proactive measures for mine waste management in an early stage of mining operations are replacing the ancient reactive conduct (Broadhurst and Harrison, 2015; Aubertin et al., 2016; Lottermoser, 2017; Skousen et al., 2019). The best strategies are sitespecific and should include environmental characterization of the mining waste, implementation of protocols to prevent any environmental and social hazards, and adoption of guidelines to minimize the risks (Slack and Voulvoulis, 2006; Brough et al., 2013; Anawar, 2015; Fox et al., 2017; Lottermoser, 2017; Cracknell et al., 2018; Elghali et al., 2018; Parbhakar-Fox et al., 2018; Mafra et al., 2020; Corzo Remigio et al., 2021; Rubinos et al., 2021; Vaziri et al., 2022).

Traditionally, process tailings are stored in surface impoundments and actively treated by the addition of alkaline chemical agents (e.g., Ca (OH)<sub>2</sub>, CaO, NaOH, Na<sub>2</sub>CO<sub>3</sub>, and NH<sub>3</sub>) (Park et al., 2019; Skousen et al., 2019; Acharya and Kharel, 2020). Although this approach appears economical during mine operation, it requires long-term infrastructure, maintenance, expenditures, and commitment upon closure (Motsi, 2010; Sahoo et al., 2013; Aubertin et al., 2016; Park et al., 2019; Skousen et al., 2019). Additionally, this disposal method poses several geotechnical and environmental risks. The deficiency in controlling water balance and geotechnical risk assessments causes tailings dam failures. Over the past decade, more than 40 tailings dam failures have been reported worldwide, destroying thousands of square kilometers of land downstream, affecting the wildlife, and killing hundreds of people

E-mail address: carolina.nogueiramafra@research.uwa.edu.au (C. Mafra).

<sup>\*</sup> Corresponding author.

(http://www.wise-uranium.org/mdaf.html). The reactive mineralogy, fine particle size distribution, and high liberation degree induce a high chance of heavy metal leaching and groundwater pollution (Benzaazoua et al., 2008; Fall et al., 2010; do Carmo et al., 2017; Owen et al., 2020; Amar et al., 2020). These social and environmental impacts have contributed to the turmoil around tailings management (Rubinos et al., 2021)

When dealing with sulfide-bearing ores, acid mine drainage (AMD) becomes a daunting challenge because the intrinsic acid-neutralizing potential (NP) often does not exceed the acid-generating potential (AP) (Parbhakar-Fox et al., 2009; Dold, 2014; Lindsay et al., 2015; Parbhakar-Fox and Lottermoser, 2017; Elghali et al., 2018; Park et al., 2019). The generation of AMD depends not only on the mineralogy of the mining wastes (Blowes et al., 2003, 2005, Paktunc, 1999a,b, Paktunc and Dave, 2000b, 2000; Skousen et al., 1998; Blowes et al., 1994; Blowes et al., 2013; Paktunc and Dave, 2000a; Paktunc and Dave, 2000b; Dold, 2010; Mermillod-Blondin et al., 2011; Rubinos et al., 2021), but also on their physicochemical properties, presence of oxygen, water and reaction catalysts (Blowes et al., 2003; Erguler and Erguler, 2015; Parbhakar-Fox and Lottermoser, 2015; Jamieson et al., 2015; Elghali et al., 2018, 2019; Mafra et al., 2020; Amar et al., 2021). Although the fine particle size distribution and high liberation degree bring special attention to tailings, underground excavations and waste rock piles also generate acidic leachates Park et al., 2019; Ait-Khouia et al., 2021).

There are several alternatives to mitigate the risks associated with AMD generation, and the most appropriate is site-specific (Acharya and Kharel, 2020). Environmental desulfurization and cemented paste backfill (CPB) are amongst the most studied methods. The former increases the net neutralization potential of the tailings (Benzaazoua et al., 1999, 2004, 2008; Nuorivaara et al., 2019; Nakhaei and Irannajad, 2017; Amar et al., 2021), whereas the latter reduces the amount of sulfidic material to be stored under oxidizing conditions while providing ground support and increasing the ore extraction ratios (Yilmaz et al., 2013, 2020; Benzaazoua et al., 2008; Qi and Fourie, 2019).

Environmental desulfurization of the tailings can be achieved by several concentration methods (Broadhurst and Harrison, 2015; Wills and Finch, 2015; Nakhaei and Irannajad, 2017; Amar et al., 2021; Ait-Khouia et al., 2021). Froth flotation is a well-established concentration process to recover sulfide-bearing minerals (Wills and Finch, 2016) and can be applied downstream to recover the non-valuable pyrite. Following that, the low pyrite tailings fraction can be discharged at the TMF at a reduced neutralization cost, whilst the pyrite concentrate could be used in applications with lower environmental risk, such as backfilling (Benzaazoua et al., 2004b, 2008; Edraki et al., 2014; Kotsiopoulos and Harrison, 2017) or could be valorized (e.g., sulfuric acid) (Broadhurst and Harrison, 2015; Park et al., 2019; Ait-Khouia et al., 2021).

CPB is an engineered material composed of a blend of process tailings, water, and a hydraulic binder. Each component is added in a calculated proportion to achieve a solid percentage between 70% and 80% (Benzaazoua et al., 2002; Qi and Fourie, 2019). The ratio between each ingredient affects the transport, emplacement, consistency, short and long-term strength acquisition, and stability (Benzaazoua et al., 2002, 2004, 2008; Qi and Fourie, 2019). Most of the studies performed on CPB aim to investigate their geomechanical behavior (Belem et al., 2000; Benzaazoua et al., 2002, 2004, 2008; Fall et al., 2005; Qi et al., 2018; Qi and Fourie, 2019; Zhao et al., 2020; Ouffa et al., 2022). However, although the CPB technique promises to decrease the reactivity of the AMD forming minerals by encapsulating the reactive gangue into a matrix and decreasing the contact with oxygen and water, care must be taken to not simply displace the environmental impact from the surface to the underground (Benzaazoua et al., 2004a,b, 2008; Coussy et al., 2011; Kwong, 2004; Ouellet et al., 2003, 2006; Yılmaz et al., 2020; Hamberg et al., 2015; Hamberg, 2018).

Dundee Precious Metals Chelopech (DPM-Ch) mining complex processes approximate 2.2 Mt of ore per year to produce copper and pyrite concentrates. The final tailings are either sent to the paste plant to

backfill the stopes or discharged at the TMF (Todorova et al., 2017; O'Connor et al., 2020). The non-existent intrinsic neutralizing potential and the enrichment in pyrite of the tailings confer the material discharged at the TMF a high potential to generate acidity. Although lime addition and seepage control prevent effluent release into the environment, a new strategy to prevent AMD and ensure post-mining safety is proposed by this study. This paper: (1) evaluated the feasibility of applying environmental desulfurization to the DPM-Ch tailings; and (2) the environmental behavior of the cemented paste backfill material when in contact with the mine water; (3) proposed an integrated management strategy for the tailings produced at DPM-Ch.

#### 2. Dundee Precious metals Chelopech

## 2.1. Local geology and mineralization

The Chelopech high-sulfidation Cu-Au deposit is part of the Apuseni–Banat–Timok–Srednogorie (ABTS) belt, Europe's most extensive Cu-Au metallogenic belt (Chambefort, 2005; von Quadt et al., 2007). The Panagyurishte metallogenic district, located in the Central Srednogorie magmatic zone, is formed as a result of successive accretion of island arcs during the subduction of the Tethyan ocean beneath the Eurasian plate, generating intense calc-alkaline magmatism throughout the Late Cretaceous (Stoykov et al., 2002; Chambefort, 2005; von Quadt et al., 2007; Georgieva, 2017).

The stratigraphic sequence of the Panagyurishte metallogenic district consists of metamorphic basement rocks intruded by Paleozoic granitoids. These units are overlaid by a Cretaceous volcanosedimentary sequence and Palaeogene/Neogene foreland sediments (Chambefort, 2005; Chambefort and Moritz, 2014).

The Chelopech mineralization is hosted by the Chelopech Formation, part of the Upper Cretaceous volcano-sedimentary sequences. The principal mineralization host, the Lower Chelopech Unit, consists of a basal sequence of siltstones and calcareous argillites with subordinate terrigenous sandstones and angular conglomerates. Upwards these sediments become intercalated with volcanic sequences. The Upper Chelopech Unit consists of andesitic and dacitic lavas and tuffs with siliciclastic, volcaniclastic, and argillaceous sediments intruded by subvolcanic bodies of porphyritic andesites (O'Connor et al., 2020). The mineralization is lithologically controlled and occurs in a broad range of morphologies. Some ore types include massive sulfide bodies within enveloping haloes of pervasive silica-sericite alteration, stockwork zones of fine sulfide veinlets, mineralized breccia, discontinuous sulfide veins, and disseminated ore in altered wall-rock (Chambefort and Moritz, 2014; O'Connor et al., 2020).

Three mineralization stages formed the Chelopech deposit: an early iron-sulfur stage, a second copper-arsenic-sulfur stage encompassing the economic copper and gold stage, and a late lead–zinc stage (Chambefort, 2005; Chambefort and Moritz, 2006). Alteration zones, typical of high-sulfidation mineralization, encompass an innermost advanced argillic alteration zone followed by a quartz-sericite zone and an external propylitic alteration (Georgieva, 2017). The mineralization is restricted to the advanced argillic alteration halo, which is characterized by the presence of "vuggy silica," massive silica, alunite, and clay minerals (kaolinite/dickite) (Chambefort, 2005; Chambefort and Moritz, 2014; Georgieva and Hikov, 2016).

## 2.2. Dundee Precious metals Chelopech mining complex

Dundee Precious Metals Chelopech (DPM-Ch) mining complex encompasses an underground mine, a processing plant, a tailings management facility, and a paste fill plant. The main target copper minerals are chalcopyrite (50%) and copper sulfosalts (enargite, luzonite, and tennantite) (45%), with minor copper oxides (5%). Gold occurs as free grains (10%) and refractory gold (90%). As native metal, gold is found associated within silver (Au<sub>3</sub>Ag) and in auriferous tellurides, whereas

refractory gold can be found intergrown with pyrite, chalcopyrite, sphalerite, copper sulfosalts, and with chalcedonic silica (O'Connor et al., 2020).

Although the pyrite circuit recovers part of the pyrite due to its association with gold (O'Connor et al., 2020), the pyrite yield is controlled to maintain an optimal Au-pyrite blend to match the metallurgical requirement for the Au grade in the concentrate. Thus, around 20% of the pyrite present in the ore is still rejected in the final tailings. The rejected pyrite inevitably oxidizes and releases acidity under oxidizing conditions, as shown by the kinetic tests performed by Mafra et al. (2020).

The concentration plant in DPM-Ch has a throughput capacity of 2.2 million tonnes (Mt) per annum (O'Connor et al., 2020). The ore treatment flowsheet includes crushing run-of-mine (ROM) ore in an underground primary jaw crusher, followed by a SAG grinding circuit in a close circuit with cyclone classification. Subsequent rougher, scavenger, and three-stage cleaner flotation circuits and concentrate dewatering produce the copper–gold concentrate. A pyrite concentrate enriched in gold is produced from the copper circuit cleaner tails (O'Connor et al., 2020).

The final tailings, a combination of bulk flotation tailings and pyrite tailings, are thickened and, depending on the paste plant capacity, either directed to the paste fill plant or towards the TMF (O'Connor et al., 2020). Currently, approximately 40% of the tailings are used as backfill material, whereas 60% are discharged at the TMF (Todorova et al., 2017).

The TMF occupies a surface area of about 110 ha and receives about 1.2 Mt of tailings material annually. The tailings are delivered as a slurry at 50% solids by pipes, and the deposition site rotates throughout the year (O'Connor et al., 2018; Plamen and Valova, 2017). The acidic water that percolates the stopes is also pumped into the TMF. In order to avoid heavy metals and arsenic release, lime is added to the TMF to keep the pH at a setpoint of 8. The TMF's water is recycled and recirculated in the process, thus decreasing the requirements for freshwater supply (Plamen and Valova, 2017).

At the paste backfill plant, the tailings are filtered, mixed with water and sulfate-resistant cement, strength class 42.5 at the appropriate percentage solids (50–60 wt% solid) (Todorova et al., 2017). Boreholes and pipes are responsible for transporting and delivering the paste into the stopes (O'Connor et al., 2020). The amount of added cement depends on the strength required to support the artificial massif, which depends on the stope's width, length, and height (Todorova et al., 2017). Generally, the percentage of pyrite present in the feeding material affects the density of the final CPB. Consequently, it plays a role in the

amount of binder required and the final strength of the paste (Benzaazoua et al., 2004a).

## 3. Integrated mine tailings management

There are several approaches to controlling and treating AMD. Mineralogy, ore texture, formation and settling of metal hydroxides, and available infrastructure should be considered when defining the most efficient treatment method (Mermillod-Blondin et al., 2011; Skousen et al., 2019; Rubinos et al., 2021). Integrated approaches combine different methods to prevent AMD (Anawar, 2015; Acharya and Kharel, 2020). Tailings desulfurization followed by backfilling has been regarded as an attractive method to improve the environmental sustainability of sulfide-rich and coal processing operations by reducing the volume of acid-generating tailings to be managed at the surface while providing advantages to the underground operations (Benzaazoua et al., 1999; Benzaazoua et al., 2008; Benzaazoua et al., 2000; Bois et al., 2005; Hesketh et al.; 2010; Edraki et al., 2014; Broadhurst and Harrison, 2015; Ait-Khouia et al., 2021).

Fig. 1 displays the integrated mine tailings management strategy proposed as an alternative disposal method to minimize the risks associated with the AMD potential of DPM-Ch tailings. Mafra et al. (2020) showed that the active treatment applied at DPM-Ch's tailing dam could not avoid AMD generation. As a result, the effluent has an elevated concentration of heavy metals (e.g., As) at a low pH. The proposed method suggests a desulfurization flotation circuit downstream of the concentration process, aiming to recover the reactive (liberated and partially liberated) non-economic pyrite, whereas the locked pyrite fraction remains in the tailings. The desulfurization concentrate would be mixed with the CPB material and placed underground, whereas the partially desulfurized tailings would be neutralized at the TMF. Mafra et al. (2020) defined in detail the pyrite liberation concept used in this study. In summary, all grains with a liberation degree greater than 20% are considered fully reactive, whereas pyrite grains with a liberation degree lower than 20% are regarded as non-reactive.

The existing infrastructure could facilitate the application of the proposed waste management flowsheet. The new tailings management approach would require only minor adjustments on the current operational one. Limited capital cost would be required since the pyrite circuit and paste plant facility infrastructure are already present at the mining complex.

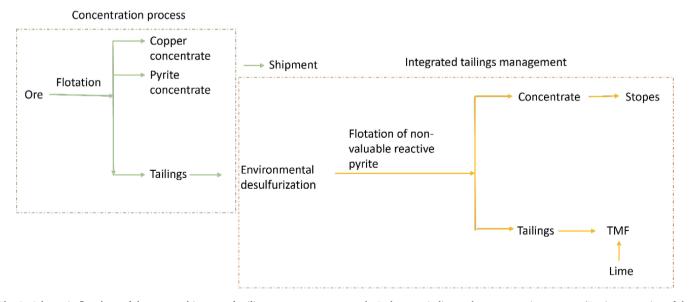


Fig. 1. Schematic flowsheet of the proposed integrated tailings management approach. Red square indicates the concentration process. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

#### 4. Material and methods

#### 4.1. Samples

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#### 4.1.1. Tailings

A composite sample (herein referred to as untreated tailings) was collected close to the main embankment wall of the DPM-Ch tailings management facility (Fig. 2). The sample was immediately dried at 60  $^{\circ}\text{C}$  for 24 h and kept in a refrigerated room to avoid further oxidation. Details about sampling, mineral characterization and acid mine drainage assessment can be found in Mafra et al. (2020). This paper focuses only on what is therein referred to as composite sample 1 because of the better performance in selectively recovering the reactive pyrite grains.

## 4.1.2. Cemented paste backfill

Freshly CPB samples are taken routinely from the paste plant after the mixing tank to define their mechanical properties through the uniaxial compression test (UCS test). The fresh CPB is placed inside a 20 cm height and 10 cm diameter cylinders made of PVC and kept inside a controlled atmosphere chamber that mimics the underground conditions for a pre-defined period (56 or 90 days). After the pre-defined curing time, the strength and moisture content of the sample is determined (Todorova et al., 2017). Two samples used for the UCS test (Table 1) were sent to the University of Liege in Belgium (ULg) for mineralogical characterization and monolithic leaching test.

#### 4.2. Flotation

Three flotation tests were performed at slightly variable regimes to define the conditions leading to an optimum flotation yield of the

**Table 1**Cemented paste backfill samples.

	Curing time (days)	Cement (%)	UCS result (KPa)
CPB - 56	56	2	504
CPB – 90	92	5.7	411

reactive pyrite while the locked fraction remained in the final tailings. Two flotation tests were performed at ULg and one at the mine site (DPM-Ch). Before flotation, the untreated tailing was dry sieved using a Ro-Tap sieve shaker to eliminate the 150  $\mu m$  oversize fraction. No attempt was made to refresh particle surfaces.

The flotation tests were performed in a 2-L Denver laboratory machine, using 500 g of feed each time. The desulfurization circuit used the same reagents as the pyrite circuit at the concentration plant. Therefore PAX (potassium amyl xanthate), supplied at different dosages, was used as a collector with neither frother nor pH modifier added. Table 2 depicts the process parameters used during each batch flotation test. The flotation trial performed at the DPM-Ch lab was undertaken using process water to mimic industrial conditions, whereas the tests at the ULg lab were performed using tap water. The difference in water composition led to a slight difference in pH value, as seen in Table 2.

**Table 2**Process parameters used for batch pyrite flotation.

Flotation test	pН	PAX (g/ ton)	Flotation time (min)	Sodium silicate (g/ton)
ULg 1	3.1	90	11	_
ULg 2	3	40	10	120
DPM-Ch	3.35	50	9	_







Fig. 2. Dundee Precious Metals Chelopech mining complex location (Mafra et al., 2020).

#### 4.3. Monolithic leaching test

The monolithic leaching test (MLT) evaluates the leachability of potential contaminants over time. Cemented paste backfill samples collected after the UCS test were cut to 2.5 cm in height. A nylon wire was placed onto the sample to enable manipulation throughout the MLT.

The experiment followed the Dutch standard test (EA NEN 7375:2004) (Netherlands Normalisation Institute Standard, 2004). The samples were placed inside an inert beaker, ensuring they were completely immersed in the leachate. 600 mL (V  $_{\mbox{\scriptsize Liq}}/\mbox{V}$   $_{\mbox{\scriptsize specimen}}=4)$  of mine water was used as a leaching agent instead of deionized water to mimic the conditions in the stopes. The beaker was placed on an agitator with a rotating speed that allowed the leaching agent to maintain satisfactory homogenization without destabilizing the sample (speed around 350 rpm). The leaching reagent was renewed at 8 intervals (0.25 days; 1 day; 56 h; 4 days; 9 days; 16 days; 36 days, and 64 days). Each eluate was analyzed for pH and conductivity. After the last leaching interval (64 days), the monolith samples were dried at 40  $^{\circ}$ C until stabilization of their mass.

A section perpendicular to the leaching surface was cut from each sample and analyzed by a Field Emission Gun Scanning Electron Microscope (FEG-SEM, Zeiss Sigma 300). Backscattered (BSE) images and X-ray elemental mapping were performed from the border to about 5 mm depth across the CPB section to evaluate composition changes and increase in porosity. Observations were conducted at a working distance of 8.5 mm with a magnification of 342 and an acceleration voltage of 21 keV. Fig. 3 depicts the procedure followed to characterize the environmental behavior of the cemented paste backfill material.

## 5. Results and discussion

## 5.1. Tailings characteristics

The physicochemical characteristics of the untreated and partially desulfurized tailings are summarised in Table 3. Their detailed description can be found in Mafra et al. (2020). The neutralization potential (NP) was determined using the sequential HCl addition protocol proposed by Bouzahzah et al. (2015), the acid-generating potential (AP) was calculated based on the total sulfur content (AP = 31.25\* %  $S_{total\ sulfur}$ ) (Sobek et al., 1978), and the effective AP (Effective AP =  $X_i \times L_p \times 0.5345$ ) was calculated considering the pyrite content ( $X_i$ ), and the percentage of reactive pyrite in the sample ( $L_p$ ) acquired by automated mineralogy (Mafra et al., 2020).

The ore mineralogy of the DPM-Ch deposit is dominated primarily by sulfides and copper sulfosalts (O'Connor et al., 2020). The mineralogical

study performed by Mafra et al. (2020) identified only a trace amount of sulfosalts in the untreated tailings, whereas the first flush of the kinetic test released elevated As content. Based on these findings, we assume that the high arsenic content evidenced in Table 3 is most likely related to secondary phases and not to primary minerals.

Secondary As-bearing phases are stable under oxidizing and mildly acidic conditions (e.g., Fe-oxides and scorodite) (Anawar, 2015; Hamberg et al., 2015; Corzo Remigio et al., 2021), and active measures in the tailings management facility during mine operation mitigate the release of arsenic-rich water downstream of the mine. However, the kinetic test in Mafra et al. (2020) highlighted the remarkably acidic conditions expected in a long-term forecast. Under these extreme conditions, the Asbearing secondary phases are destabilized, generating a high environmental risk (Hamberg et al., 2015; Hamberg, 2018). Whatever the case, an effort is required to maintain the remaining arsenic-bearing phases stable, and an effective way to decrease the reactivity of the gangue minerals is by limiting the contact with oxygen and water.

A size-by-size liberation degree analysis highlighted that flotation was inefficient in recovering the grains finer than 75  $\mu m$ . Table 4 shows that 75.6% of pyrite grains smaller than this fraction remained reactive in the partially desulfurized tailings. A selective flocculant could be added to form particles of a suitable size for flotation (Yu et al., 2017; Ait-Khouia et al., 2021). Overall, although the AP of the tailings after environmental desulfurization is still very high (104 kg CaCO\_3/t), the desulfurization process decreased the effective AP to 44 kg CaCO\_3/t. Therefore, in practice, this sample should produce lower acidity when compared to the theoretical value calculated by the Sobek test (Sobek et al., 1978).

## 5.2. Flotation performance

Flotation efficiency is, to a large extent, affected by particle size, surface oxidation degree, type and concentration of reagents, water chemistry, pulp conditions (pH and Eh), and flotation time (Wills and Finch, 2016). At DPM-Ch, flotation parameters have been optimized by the flowsheet test program completed in 2005 (O'Connor et al., 2020). Therefore, in our study, the choice of reagents was guided by the pyrite circuit scheme operational at the DPM-Ch site (O'Connor et al., 2020).

In order to guarantee the complete removal of the fully reactive pyrite, the first flotation trial (ULg 1) was performed under the addition of a collector in an amount higher than the usual one (90 g/ton), and the flotation was run until froth loading became empty. The optical microscope observation of the concentrate revealed the presence of both reactive (free and partially locked) and non-reactive (locked) pyrite, whereas a limited amount of very fine, generally completely liberated

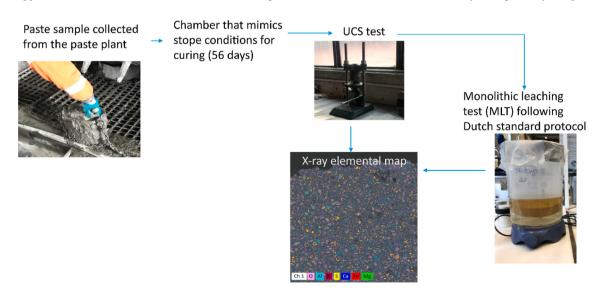


Fig. 3. Procedure to characterize the cemented paste backfill material.

Table 3 Composition, granulometry, and mineralogy of composite sample and flotation products. \*Reactive pyrite = (100% - locked pyrite).

Characterisation	Parameters	Units	Untreated tailings	Concentrate	Tailing
Chemical assay	S	(wt. %)	8.41	38.3	3.34
	С		0.004	_	0.011
	Fe		7.85	32.8	2.58
	Cu		0.13	0.5	0.05
	As	(ppm)	243	751	140
Bulk mineralogy by AM	Quartz	wt. %	49.7	13.7	63.1
	Kaolinite		26.2	6.8	24.4
	Muscovite		3.8	0.7	3.7
	Other silicates		3.4	0.9	1.4
	Carbonates		0.002	0.00	0.01
	Pyrite		16.1	77.4	6.3
	Other sulfides		0.08	0.21	0.02
	Rutile		0.19	0.08	0.16
	Barite		0.45	0.15	0.75
	Traces		0.10	0.01	0.22
Granulometry	$d_{20}$	μm	14		7
	$d_{50}$		38		13
	$d_{80}$		99		46
	$d_{95}$		191		67
Pyrite liberation degree	Liberated	%	58.0	76.1	23.0
	Partially liberated		21.0	21.8	19.0
	Locked		21.0	2.0	58.0
	Reactive pyrite (Lp)*		79.0	97.9	42.0
Static test results	Volume HCl	mL	1.5		1.1
	NP	kg CaCO <sub>3</sub> /t	7.5		0.1
	AP		263		104
	Effective AP		208		44
	NP/AP		0.03		0.0001

**Table 4**Pyrite liberation degree after sieving.

	After sieving and with AP correction based on liberation degree									
	Untreated tailings			Partially d	ially desulfurized tailings					
	+ 125	+75	+38	-38	Reconstructed	+ 125	+75	+38	-38	Reconstructed
Liberated	29.6	63.2	82.9	92.9	58.0	6.3	19.5	65.3	86.5	23.0
Partially liberated	27.4	23.5	12.6	5.9	21.0	21.8	23.0	10.3	8.5	19.0
Locked	43.0	13.3	4.5	1.1	21.0	72.0	57.5	24.4	5.0	58.0
Reactive pyrite (Lp)	57.0	86.7	95.5	98.8	79.0	28.1	42.5	75.6	95.0	42.0

grains remained in the tailings fraction. Therefore, collector dosage and flotation time were reduced. However, despite the adjustments of collector dosage and flotation time, the optical observations of ULg 2 flotation products suggested that this flotation trial did not reach the desired selectivity. In fact, liberated pyrite was still reporting to the tailings, whereas gangue minerals floated.

The microscopic inspections of the DPM-Ch trial revealed that this flotation test achieved the highest selectivity. A large part of the non-reactive (locked) pyrite remained in the tailings, whereas most reactive fraction (free and partially locked) floated. The better efficiency achieved here could be explained by the availability of dissolved ions in the process water. It is known that copper ions activate pyrite at low pH (Zhang et al., 1997; Ejtemaei and Nguyen, 2017). The process water is enriched in copper and iron ions, and the Cu (II) present in the pulp could be responsible for activating pyrite.

Additionally, the iron (III) ions could act as an oxidizing agent for copper minerals, thus solubilizing the reminiscent Cu, activating pyrite surfaces, and stimulating flotation. Notwithstanding, despite the better efficiency, some fine-sized liberated grains were still lost in the tailings. The failure in floating these fine reactive grains is assumed to be due to the known difficulties in floating particles smaller than 50  $\mu m$  (Trahar and Warren, 1976; Miettinen et al., 2010) and by in-situ oxidation of the mineral assemblages.

## 5.3. Acid mine drainage assessment

Mafra et al. (2020) highlighted the almost non-existent intrinsic neutralization potential of the DPM-Ch tailings (<8 kg CaCO<sub>3</sub>/t). Hence, flotation alone could not shift the desulfurized tailings to the non-acid generating zone (Fig. 4 – yellow arrow). In order to avoid the release of acidity generated by pyrite oxidation, the tailings produced during the environmental desulfurization still need to be neutralized. Fig. 4 illustrates the estimated amount of lime (CaO) required to neutralize the solid tailings produced during the beneficiation process (black arrow) and the tailings after desulfurization (green arrow) (Mafra et al., 2020). Environmental desulfurization could reduce nearly six times the amount of neutralizing agent required to neutralize the tailings discharged to the TMF. Adding an adequate quantity of lime is particularly important to avoid destabilizing arsenic-bearing minerals. Thus, decreasing the environmental risk associated with the DPM-Ch tailings dam.

## 5.4. Geochemical behavior of cemented paste backfill material

The monolithic leaching test represents the leaching mechanism of monolithic materials in a diffusion-controlled environment. The test can be used to assess the long-term environmental behavior of the CPB in a mine closure scenario where the underground mine will be flooded with groundwater (Schafer, 2016; Yılmaz et al., 2021).

X-ray elemental mapping was done on the monolithic samples (before and after being subjected to the monolithic leaching test) to

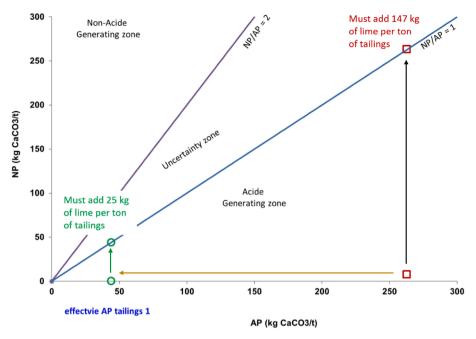


Fig. 4. Effort needed to neutralize the acid-generating tailings (NP of the lime determined by Sobek test equals 1736 kg CaCO3/t). Modified after Mafra et al. (2020). Yellow array – desulfurization approach to transform the tailings as non-acid generating; black array – addition of neutralizing agent to neutralize the tailings (NP = AP). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

witness any notable difference in the chemical composition of the samples after their contact with the acidic mining water. For example, pyrite oxidation can release sulfate (SO<sup>2</sup>-), whereas the dissolution of carbonate and silicate minerals can produce secondary Ca, Mg, and Mn sulfates and hydroxides (Yılmaz et al., 2021).

Fig. 5 illustrates the sample before the monolithic leaching test (MLT) and shows the tailings assemblage encapsulated inside a calciumrich matrix. During curing, calcium was dissolved from the cement matrix and formed a crust at the border of the cylinder (Fig. 5c).

Fig. 6 displays the monolithic samples after the 64-day leaching test. When comparing the results shown in Figs. 5 and 6, one could note the absence of the calcium-rich crust after the leaching test. It is presumed that this crust reacts with the mining water providing a slight buffering

effect. Moreover, no mineralogical variations were observed. The minerals remained intact and encapsulated inside the matrix, with no oxidation traces observed. The consistent mineralogy leads to the assumption that the encapsulation of pyrite grains inside the cement paste can effectively decrease the reactivity of pyrite grains. Ji et al. (2012) attributed the efficiency of cement in suppressing  $SO_4^{2r}$  release to the following reactions:

$$Ca^{2+} + SO_4^{2-} + 2H_2O = CaSO_4 \cdot 2H_2O.$$

$$3\text{CaO} \cdot \text{Al}_2\text{O}_3 + \text{CaSO}_4 \cdot 2\text{H}_2\text{O} + 10\text{H}_2\text{O} = 3\text{Ca} \cdot \text{Al}_2\text{O}_3 \cdot \text{CaSO}_4 \cdot 12\text{H}_2.$$

CPB has shown to be an effective approach to optimizing the mine ore extraction while improving the sustainability of the operation (Coussy et al., 2011). The method has proved to isolate the reactive gangue, which involves pyrite, sulfosalts, and metal-bearing sulfides.

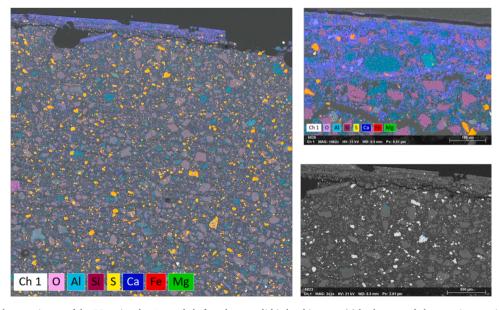
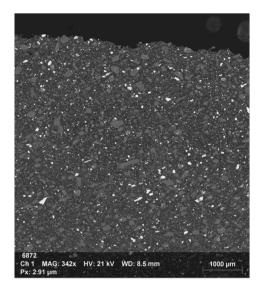


Fig. 5. Backscattered electrons image of the 56-curing days' sample before the monolithic leaching test. (a) backscattered electrons image with X-ray mapping of the sample, (b) zoom of the crust enriched in calcium at the boarder of the monolithic block, (c) backscattered electrons image of the sample.



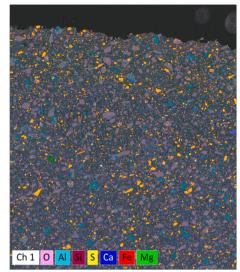


Fig. 6. (a) Backscattered electrons image of the 56-curing days' sample after the monolithic leaching test, (b) backscattered electrons image with X-ray mapping.

## 5.5. Evaluation of the integrated mine tailings management approach

The modal mineralogy of solid mine wastes should be used as a guide to determining the best tailings management strategy. DPM-Ch ore is enriched in sulfide minerals and potentially toxic metalloids (including As-bearing minerals), anticipating an elevated environmental concern to the waste material. The textural characteristics of the tailings (e.g., particle size distribution, liberation degree, and mineralogical associations) increase the pollution risk associated with this material.

The automated mineralogy analysis suggested that pyrite flotation was able to recover the majority of the reactive pyrite. The portion of reactive pyrite that remained in the tailings was finer than 75 µm and challenging to float. To overcome this issue, selective flocculation of pyrite grains to a suitable size could be applied (Yu et al., 2017; Ait-Khouia et al., 2021). Noteworthy is the significant decrease in the amount of lime required to neutralize the solid tailings, which would lead to a considerable economic benefit accompanying the reduced contamination potential downstream the mine. The capacity of the cemented paste backfill method to isolate reactive pyrite fractions highlights the environmental advantages of the technique. Both flotation and backfilling are already implemented at DPM-Ch, rendering the proposed integrated tailings management strategy relatively simple to implement. It could also be speculated that the lower expenditures on lime could compensate for any CAPEX required for implementing the desulfurization circuit.

Table 5 compares the conventional tailings management currently applied and the integrated one proposed. Table 5 suggests that a noticeable economic, technical, environmental, and social benefits in the long-term could be expected if an eventual tailings desulfurization campaign is undertaken. This new approach offers the possibility of reducing reclamation costs and improving the management of acid-generating tailings (Amar et al., 2020).

However, it is important to note that this study focused on the geochemical perspective of performing desulfurization. A detailed

**Table 5**Comparison between tailings disposal strategy used currently and the integrated approach proposed.

	Currently	Proposed
Tailings disposal strategy	60% Subaerial discharged at the TMF and 40% used to backfill the stopes	Desulfurization of the tailings, discharge of low sulfide fraction in the TMF, and add the desulfurization concentrate to the CPB material
Environment impact	Tailings discharged in the TMF generate AMD	Deposition of tailings with lower AMD potential covering old tailings richer in reactive pyrite
	The intrinsic neutralization potential of the tailings is close to 0	Most of the reactive pyrite will be removed by desulfurization
	No geomembrane underneath the TMF	Lower potential to release AMD
	Long-term commitment and a high amount of lime are required to neutralize the TMF	A lower amount of lime is required to neutralize the TMF
	CPB is capable of decreasing pyrite reactivity and release of acidity	CPB with higher strength and no change in the geochemical behavior*
Economy	High OPEX associated with the lime addition	OPEX associated with flotation reagent and energy
	Low energy expenditure: tailings are deposed by gravity	Higher energy expenditures associated with flotation
	CAPEX to increase the capacity of the TMF Closure plan is estimated at 14 M USD	CAPEX to build a desulfurization plant Reduction of the rehabilitation cost

<sup>\*</sup>Further studies are needed to investigate the impact of the increased pyrite content on CPB strength and geochemical behavior of the CPB.

investigation is needed to access the geotechnical aspects of using this low pyrite bearing tailings as a cover material and understand the geomechanical consequences of adding more pyrite to the CPB material. Similar studies have been done by Benzaazoua et al. (2008) for the Doyon mine. They showed that adding the pyrite concentrate to the paste did not affect the environmental properties and, at the same time, it increased the strength of the CPB and slightly decreased the amount of cement required.

#### 6. Conclusions

The presented study highlighted that prevention strategies for AMD management should be preferred over post-mining remediation. The integrated management approach shows potential economic, social, and environmental benefits. The main conclusions that can be drawn from this study could be summarized as follows:

- Environmental desulfurization using froth flotation is feasible for DPM-Ch tailings. Flocculant addition could potentially improve fines recovery.
- The tailings from DPM-Ch have a high environmental risk due to their mineralogy, particle size distribution, and textural characteristics.
- The tailings have elevated arsenic content, highlighting the importance of neutralizing the acid-generating potential of the material.
- The cost linked to the amount of alkaline agents for AMD neutralization directly impacts mine economics.
- Cemented Paste Backfill is an effective way of isolating the reactive pyrite accompanying polymetallic sulfide ores. As such, it represents a sustainable approach to managing mining wastes.

## CRediT authorship contribution statement

Carolina Mafra: Writing – original draft, Investigation. Hassan Bouzahzah: Supervision, Methodology. Lachezar Stamenov: Resources, Supervision. Stoyan Gaydardzhiev: Supervision.

## **Declaration of Competing Interest**

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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