

Performance of natural and residual materials for mine water treatment and mine sites rehabilitation

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Abstract

Several natural and residual materials are often available in the proximity of, or directly on mine sites. The use of these materials for mine water treatment and mine site rehabilitation can entail lower transportation costs, environmental footprints, and related effects. Wood waste, desulfurized tailings, peat, ash, dolomite, and contaminated solids from mine water treatment are a few examples of available natural and residual materials on mine sites. These materials can be used both as raw or modified or activated products, such as charred dolomite, chemically or physically activated biochar, and hydrothermally activated wood ash. The objective of the study is to document the performance of natural and residual materials for mine water treatment and mine site rehabilitation. Several case studies, conducted in various setting, from laboratory to field full-scale, are presented and discussed. Findings show that regardless of the materials used in the rehabilitation, the performance is dictated mostly by site-specific conditions for the same material. Finally, some research needs for further studies are identified.

Keywords: Mine drainage, desulfurized tailings, passive biofilters, mine water treatment residuals

Introduction

Mine water consists of mine drainage and mineral processing water. Mine drainage is runoff water contacted with mine waste (tailings, sterile rocks, underground and open pit mine workings), while mineral processing water is the effluents resulting after the solid-liquid separation of tailings downstream of mineral processing. Mine drainage is generally classified based on its pH as acid mine drainage (AMD), neutral mine drainage (NMD), alkaline drainage (AlkD), and saline drainage (Nordstrom et al. 2015). The main contaminants of mine drainage are the acidic or alkaline pH, and above criteria concentrations of metals/ metalloids and sulfate, as well as, sometimes, high concentrations of total suspended solids (TSS) and total dissolved solids (TDS). The dust generating TSS originates from multiple sources on mine sites (tailings erosion, ore milling, blasting, and transportation). TDS are sourced from processing chemicals, de-icing agents (used in cold climate for pursuing mining operation during winter), water recycling and reuse with an ambitious aim of minimal liquid discharge or zero liquid discharge (Tong & Elimelech 2016; Miller et al. 2022), and pockets of saline water from beneath the permafrost (Rotem et al. 2023). A less documented source of TDS is the porewater trapped in dry stack tailings. Dry stacking is a recent tailings management approach used for improved geotechnical stability of dewatered and filtered tailings that are placed and compacted in a mound, which avoids the need for a constructed dam (Furnell et al. 2022). For mine water, the TDS (dissolved salts and organic material) mainly consists of salinity (Na⁺, K⁺, Ca²⁺, Mg²⁺, Cl⁻, SO_4^{2-} , HCO_3^{-}), as the content of dissolved organic material is usually low. Other common contaminants in mining effluents include N-based contaminants (cyanide, cyanate, thiocyanide, ammonia, nitrite, and nitrate), thiosalts (meta-stable sulfur oxides in water), and phosphate.

Mine water treatment and rehabilitation of mine sites are a costly liability for the mining industry and governments worldwide. In an actual context of lower-grade ores mining for satisfying the increasing demand for mineral resources of the modern society, new and adaptive solutions are necessary for the efficient treatment of complex chemistries of mine water and for the large volumes of materials necessary for rehabilitation of mine sites. At the same time, several natural and residual materials are already available on the mine sites or in their proximity. Wood waste (Rakotonimaro et al. 2015; Keshvardoostchokami et al. 2023), desulfurized tailings (Magsoud et al. 2016; Ait-Khouia et al. 2021), physically and chemically activated biochar (Braghiroli et al. 2018a, 2018b, 2019, 2020; Robert & Braghiroli 2022), food waste and zeolites (Hwang & Neculita 2013; Hwang et al. 2012), peat (Rakotonimaro et al. 2019, 2021), ash (Calugaru et al. 2017), dolomite (Calugaru et al. 2016), and contaminated solids from mine water treatment (Rakotonimaro et al. 2017; Saint-Aimé et al. 2023) are examples of available natural and residual materials on mine sites. These materials can be used as amendments or covers for mine tailings (Rakotonimaro et al. 2017, 2018a). The use of these materials for mine water treatment and mine site rehabilitation can entail lower transportation costs, environmental footprints, and related effects. A life cycle assessment (LCA) of eight active and passive physicochemical and biochemical AMD treatment options showed that a 50% reduction in transportation distances resulted in the lowest LCA impacts for all scenarios (Hengen et al. 2014). These materials can be used both as raw or modified or activated products, such as charred dolomite, chemically or physically activated biochar, and hydrothermally activated wood ash.

Functions of amendments and covers for tailings

Mine water treatment, active (for operating mines) or passive (for closed and abandoned mines), using bio-geo-electrochemical methods, requires high quantities of products (chemicals or substrates). The large volumes and the often-high degree of contamination justify such high quantities of products necessary to treat the mining influenced water (Ryskie et al. 2021). Most of these products are transported from outside the mine sites. To avoid an increase in contamination over time and / or a long-term treatment of contaminated water, early prevention measures and control of porewater quality in mine tailings are necessary. To increase the likelihood of successful AMD prevention, prevention measures are achievable when applied in the first decade of the tailings discharge in tailings storage facilities (TSF). The prevention of AMD generated by weathered tailings is more complex and costlier. Experience shows that the flush-out the AMD already present in the contaminated porewater of highly oxidised acid-generating tailings may take at least 20 years (Genty et al. 2016) depending on the tailing's properties and climate.

Several mechanisms within amendments and covers made of organic or mineral, natural or residual materials can lead to geochemical and geotechnical stability of amended or covered tailings, including the following (Demers & Pabst 2021):

- Consumption of oxygen and creation of a reducing environment, favorable to the precipitation or preservation of metal biosulfides in the presence of Fe³⁺ to Fe²⁺ reducing bacteria, or sulfate to soluble sulfide reducing bacteria.
- Increase of pH and alkalinity of porewater, by addition of neutralizing materials, or by mineralized organic carbon produced by final degrader, such as sulfate-reducing bacteria.
- 3. Dilution of contaminated porewater, and improved efficiency of passive treatment systems.
- 4. Formation of a hardpan layer of secondary minerals by neo-formed

minerals precipitation playing a protecting role to prevent oxidation of unoxidized tailings.

- 5. Limitation of water infiltration in mine tailings by favoring horizontal runoff.
- 6. Improvement of geochemical and mechanical properties of tailings and development of a protective layer favorable to tailings revegetation.

Few case studies from the Quebec's Province of Canada are detailed in the next chapter.

Case study #1: East-Sullivan mine site.

The East-Sullivan mine, located at 6 km east of Val d'Or, Quebec, exploited Au, As, Cu, and Zn (1949-1966) from a massive sulfide orebody (approx. 50% pyrite and pyrrhotite), and left behind 15 Mt of tailings on a surface of 228 ha over a thickness of 2.1-13.5 m (Germain et al. 1994). AMD generated by the oxidized tailings had a pH of 2, and concentrations of Fe²⁺ and sulfate up to 18 g/L and 37 g/L, respectively (Germain et al. 1994, 2004). The rehabilitation of the site started with the construction of an experimental wood waste cover in the early 1980s to prevent sulfides oxidation. Following the experimental study, an accumulation of fresh and weathered wood-based organic residuals (bark fragments, logs, and sawdust of coniferous and deciduous species) was placed over the tailings, and then overlain by sewage and septic-tank municipal wastewater treatment sludge (Tassé et al. 1997). A seepage collection system around the covered tailings and the passive treatment in constructed wetlands was then implemented in the early 1990s (Germain et al. 2004). Due a high contamination with AMD, passive treatment was found inefficient. An innovative approach was later implemented (1998-2005), based on the recirculation of the water discharging from the covered tailings back through the organic cover, which served both as barrier for AMD prevention and as biofilter. This approach was successful, and water in wetlands surrounding the covered tailings was deemed of satisfactory quality (QME 2014). A 15-year assessment of monitoring data of a 12-sampling point network of water flowing from underneath the cover confirmed the compliance with regulation,

except for an uncovered part of the tailings, where Fe^{2+} concentrations maintained higher (Rakotonimaro et al. 2015). The cover was completed in the period 2019-2020. Moreover, based on the findings of a thorough study conducted by the Quebec's Ministry of Environment (QME) in July 2012, no associated risks for the avian fauna were found from the constructed wetlands (QME 2014). The importance of maintaining high moisture conditions in such a wood-waste cover (humid climate or saturated base of a TSF) and the replenishment of old by more recent wood-waste were underlined as main lessons learned from this case study (MEND 2004). New research is needed to confirm whether the maintenance of a high degree of saturation in tailings is enough for this approach or the AMD recirculation through the cover is still necessary.

Case study #2: Manitou mine site.

A former mine and custom mill processing Au, Ag, Cu, and Zn ores (1942–1979) left behind approximately 107 t of highly acid-generating tailings in a legacy impoundment (disposed in a valley, without control, and affecting 190 ha) (Aubertin et al. 1999; Maqsoud et al. 2016). The AMD was characterized by pH 2-3, and high concentrations of Cu (0.1-1 g/L), Zn (0.6-1 g/L), Fe (10-12 g/L), and sulfate (30-40 g/L) (Aubertin et al. 1999). In 2009, a reclamation approach consisting of the elevated water table (EWT) technique (control of the water table position to a depth maintains a high degree of saturation in the reactive tailings), combined with a monolayer (1.2–2.1 m) cover made of slightly alkaline low sulfide tailings from the nearby mine Goldex, was adopted (Ethier et al. 2020). The design objective of the reclamation strategy is to maintain a high degree of saturation in the tailings and the cover to limit the ingress of oxygen into the reactive tailings. A 3-year monitoring campaign indicated that the applied system meets the expected performance related to water table and oxygen flux. The mixing of alkaline water and AMD, from fresh (slurry) and weathered tailings, also generates an Fe-rich NMD (Maqsoud et al. 2016; Ethier et al. 2020). A final polishing step by a multi-unit passive treatment is necessary before the final release into the environment to reach the regulation.

Case study #3: Wood-Cadillac mine site.

A former Au and Ag mine (1939–1949), extracted from ores with As, generated 3.5 \times 10⁵t of tailings over 21 ha up to a depth of 3 m, and around 10⁵t of tailings spreading over 3 km along the Creek Pandora, in Abitibi, Quebec. The monitoring of As concentrations started in 1995 and showed up to 0.3 mg/L in natural underground and surface water, originating from As-NMD (Tassé et al. 2003; Germain & Cyr 2003). In 1999, a sand monolayer cover was applied on top of tailings, and a wood-waste based biofilter was constructed for As-NMD passive treatment. A detailed study conducted in 2007 proved the biofilter efficient for reaching target physicochemical criteria and toxicity (Libéro 2007). The historical physiochemical monitoring data (1999-2022), coupled to newest analysis (isotopic, microbiological, and mineralogical) showed the immobilization of As in stable mineral precipitates (Mehdaoui et al. 2023, 2024; Thevenot et al. 2024).

Case study #4: Lorraine mine site.

A former Au, Ag, Cu and Ni mine active for only 4 years (1964–1968), left behind around 6×105 t of acid generating tailings, disposed in a 15.5 ha pond (Genty et al. 2016). In 1998, a rehabilitation program was initiated, consisting of a multi-layer dry cover (CCBE) and four passive treatment units using one limestone and three dolomite drains to prevent further weathering of tailings and AMD generation. Reclamation strategy proved effective (Dagenais et al. 2005), but the Fe-rich AMD in the tailings pore water was progressively and slowly leached out over time (Genty et al. 2016). Total Fe concentrations decreased from approximately 11 g/L, before the construction of the cover, to around 2.5 g/L in 2015, while the pH increased from < 3to 5-6 over a 12-year period. The efficiency of one of the three dolomitic drains drastically deteriorated over time due to the built-up of Fe precipitates, which clogged the pore spaces, and limited the flow. In 2011, the replacement of this clogged dolomite drain by a three-unit passive biochemical treatment system (train), consisting of two biofilters separated by a

wood ash unit was achieved. The monitoring of the treatment system performed at various sampling points in the first 60 months of operation showed a substantial improvement of AMD quality discharged into the environment. Nevertheless, the loss of efficiency over time was observed, especially due to porosity clogging by Fe precipitates. A stabler long-term performance would be achieved by using a multi-step system of calcite-/dolomite-DAS (dispersed alkaline substrate, a mixture of coarse material, such as wood chips, and neutralizing agents) and passive biochemical reactors (Rakotonimaro et al. 2018b).

Newer applications: raw and modified, natural and residual materials.

Several other underused materials, both natural and residual, raw and modified showed promising performance in mine water treatment and mine site rehabilitation (Rakotonimaro et al. 2017, 2018; Calugaru et al. 2018). They include dolomite, biochar, wood ash, and mine water treatment residuals, such as Fe-rich AMD sludge, and N-rich biomass from ammonia removal, among others. The superior performance of half-charred dolomite (1 h, 750 °C), a mixture of MgO/CaCO3 vs raw dolomite $[CaMg(CO_2)_2]$ was found with synthetic and real mine drainage, including Ni-/Zn-rich NMD and AMD, in batch and column testing (Calugaru et al. 2016; Braghiroli et al. 2020). Similarly, porosity development in activated biochar, physically (at 900 °C) or chemically (using H₃PO₄, KOH or CO₂) led to better efficiency for AMD treatment. For instance, CO₂-activated biochar allowed for 99% Cu removal at 5-20 mg/L (Braghiroli et al. 2019). The improved textural properties of activated biochar, and higher micropore volume and proportion of oxygen carbonyl groups connected to its surface led to a twofold increase in sorption capacity vs raw biochar (303 vs 159 mg/g) for phenol, in synthetic and industrial effluents (Braghiroli et al. 2018b). The results on two types of wood ash (raw vs. modified) proved the faster kinetics and higher maximal sorption capacity of modified ash (activation of wood ash by alkaline fusion, prior to hydrothermal

synthesis) for Ni-/Zn-rich real NMD (Calugaru et al. 2017). The pH correction of final effluent might be necessary. In ongoing research, raw vs half-charred dolomite is tested for AMD prevention and porewater control of amended vs covered pyrrhotiterich acid-generating tailings.

Two recent studies also confirmed that N-rich biomass from NH₂-N treatment in mine water, using sorption on biochar or a MBBR (moving bed biofilm reactor) process is a promising potential amendment as substitute fertilizer for spruce production, forestry producers and horticultural nurseries (Robert & Braghiroli 2022) or for the revegetation with agronomic herbaceous plants of nonacid generating mine tailings (Saint-Aimé et al. 2023). In the first study, a biochar-based pellet substrate (a quarter of peat was replaced by bulk biochar impregnated with ammonium sulfate) yielded the highest biomass vs other tested scenarios. In the second study, the MBBR biomass-amended Goldex mine tailings (100 kg/ha of total N) yielded similar results for above-ground biomass as the fertilizer-amended tailings and commercial topsoil. Results also showed no trace metal contamination of the surface runoff, but an increased Se and Cr foliar concentration of the plants. New studies show promising results for mixture of peat with Fe-rich AMD sludge for the treatment of As-NMD (Thevenot et al. 2024), and satisfactory environmental stability of residues produced (Mehdaoui et al. 2023, 2024). More studies are needed with real effluents and in field settings (pilot and fullscale) for the evaluation of the influence of real conditions.

Conclusions

The valorization of natural and residual materials available on mine sites or in their proximity for mine water treatment and mine site rehabilitation reduces the transportation and usage costs, environmental footprint, and contributes to a circular economy. Several case studies were detailed as leading examples of successful application of natural and residual materials, combining prevention measures such as mono or multi-layer covers and passive treatment. New studies are necessary for a better understanding of mechanisms and processes explaining the successes obtained, and to adapt to other potential applications.

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