Recycling heavy metal mining waste into fired building and anti-acid bricks

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Mineral extraction leads to the generation of tailings that accumulates in the environment which is unsustainable (Lottermoser, 2007; Peša, 2021). Wind erosion and leaching can impact the environment due to the chemical and mineralogical composition of these wastes (Muimba-Kankolongo, 2021). The proximity of storage facilities to populated regions affects local residents' health. Mining nations, such as the Democratic Republic of Congo, are experiencing an increasing waste generation linked to key metal extraction. To conserve resources and reduce the environmental impact of these activities, materials science engineers must develop sustainable alternatives to storage. Mining tailings could serve as secondary resources, as they are still containing low concentrations of heavy metals that can be extracted through metallurgical processes. After removal, these wastes could be used in the fired and anti-acid brick industry (Jawadand & Randive, 2021) as it will be demonstrated hereafter.

The present study focuses on recycling mining tailings produced by the Kakanda concentrator, which are known to contain significant amounts of heavy metals such as copper, cobalt, and manganese. After recovering these metals, the treated tailings are used in the production of building and antiacid bricks. X-ray fluorescence and diffraction techniques, scanning electron microscopy were used to characterize the initial materials and the products at different processing stages. Thermogravimetries and differential thermal analysis were carried out to evaluate the thermal characteristics of the mixtures, and identifying firing profiles.

Malachite was identified as the main copper mineral, with small amounts of heterogenite as a cobalt mineral. Specific extraction and purification processes were applied to recover metal cations. This pre-treatment step involved acid leaching using H_2SO_4 at 2 mol/L. This led to the generation of solutions containing 4.5 g/L Cu, 0.85 g/L Co, 0.25 g/L Mn, and 0.80 g/L Fe that can be send to hydrometallurgical extractive plants. The main minerals (Table 1) in the pre-treated wastes are quartz and aluminosilicates.

Building bricks were prepared by adding clay to the pre-treated tailings. The manufacturing procedure involves a water/solid ratio of 0.20, a compaction pressure of 0.1 MPa and firing at 800°C for 2 hours. To prevent the formation of cracks linked to the β to α allotropic transformation of quartz, the bricks were slowly cooled down. Bricks fulfilling construction requirements (a water absorption below 20 wt.%, an opened porosity below 40%, and a compressive strength exceeding 10 MPa) were obtained by adding up to 70 wt.% of treated tailings The performance of these bricks can be attributed to the formation of high density phases, namely quartz (SiO_2) and mullite ($Al_6Si_2O_{13}$). The formation of mullite was promoted by iron while magnesium is responsible for the synthesis of spinel ($MgAl_2O_4$) and enstatite ($MgSiO_3$). The fine and uniform granulometry of the raw materials (D_{75} =16 µm and 14 µm for tailings and clay respectively) was very favorable for the sintering process (Samara et al., 2009).

Table 1. Mineralogical composition of depolluted wastes and clay by X-ray diffraction

Minerals	PDF	Mine wastes	Clay
Quartz SiO_2	00-048-1045	52±3	30±1
Clinochlore $Mg_5Al_2Si_3O_{13}(OH)_2.3H_2O$	00-012-0242	14±3	-
Muscovite $K(Al_4Si_2O_9(OH)_3)$	01-070-3754	8±1	10±1
Talc $Mg_3Si_4O_{10}(OH)_2$	00-013-0558	4±0	-
Vermiculite $Mg_3Al_2Si_2O_{10}(OH)2.4H_2O$	00-060-0341	-	4±2
Goethite FeO(OH)	00-029-0713	2±1	4±1
Kaolinite $Al_2Si_2O_5(OH)_4$	00-014-0164	-	24±1
Hematite Fe_2O_3	00-033-0664	1±0	-
Rutile TiO_2	00-021-1276	1±0	1±0
Gypsum $CaSO_4.2H_2O$	00-021-0816	1±0	-
Amorphous		17±1	26±1

The addition of waste glass particles smaller than 40 µm to a mix of tailings and clay and a higher firing temperature lead to the manufacturing of anti-acid bricks. To prevent brick distortion due to stress, a maximum of 20 wt.% of glass could be incorporated into the mix with a minimum of 20 wt.% of kaolinitic clay up to 70 wt.%

of treated tailings in water/solid ratio of 0.20. Bricks with suitable physico-chemical and mechanical properties, (water absorption below 6 wt.%, opened porosity below 12%, compressive strength above 17.5 MPa), were manufactured at 1100°C. After 65 days of immersion in 2 mol/L H_2SO_4, the overall mass loss was lower than 1 wt.%, as shown in Figure 1. The anti-acid characteristics are related to the vitrification of the external surface and the formation of crystalline phases characterized by strong chemical bonding. These phases are derived from the ternary systems SiO_2-Al_2O_3-Alkalis, SiO_2-Al_2O_3-MgO, and SiO_2-Al_2O_3-CaO quartz (SiO_2), cristobalite (SiO_2), mullite ($Al_6Si_2O_{13}$), spinel ($MgAl_2O_4$), wollastonite ($CaSiO_3$), and albite ($2NaAlSi_3O_8$)...

Mechanical performances similar to that of fibre-reinforced concrete were achieved, with a compressive strength of 80 MPa and an E modulus of 30 GPa. In addition, a wear rate of 0.2 is acceptable for antiacid bricks. Due to these characteristics, the synthetized bricks are good candidates for copper electrolysis cells walls and floor coverings where high degree of strain and acid conditions are encountered, such as in hydrometallurgical plants.

The vitrification process that occurs during brick firing leads to the sealing of pores and a consolidation of the bricks (Mao et al., 2020) as well as to the encapsulation of components inside an insoluble matrix. These phenomena are responsible for the very limited leaching in acidic solutions, hence classifying the produced bricks as non-leachable materials. In addition, we demonstrated that these bricks can be recycled in new anti-acid bricks.

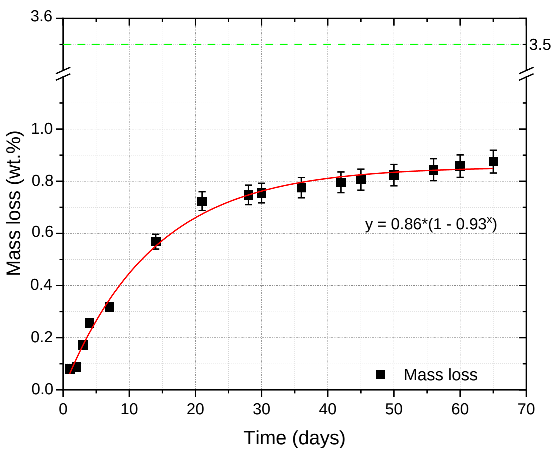

Figure 1. Mass loss of anti-acid bricks in 2 mol/L H_2SO_4 solutions over time

This study demonstrates that it is possible to recycle mining tailings in the manufacturing of construction and anti-acid bricks. We propose an integrating approach that allows to recover the heavy metals that they contain, to produce locally necessary building materials and to contribute to environmental remediation.

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