



Chemical Leaching of Sulfidic Mining Waste, Plombières Tailings Pond, Eastern Belgium: Insights from a Mineralogical Approach

Srećko Bevandić¹ · Panagiotis Xanthopoulos² · Philippe Muchez¹

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Abstract

Historic sulfidic mine waste material poses both an economic opportunity and an environmental threat (e.g., acid mine drainage). Due to low metal content in the mine waste and complex mineralogy, metal extraction of metal of interest is challenging. The Plombières tailings pond in eastern Belgium was selected as a case study to determine the potential for metal extraction of targeted metals from three different types of mine waste (metallurgical waste, brown, and yellow tailings). Material from this historic tailing originates from the exploitation and pyrometallurgical processing of the Mississippi Valley-type ores. The examined mine waste contains amounts of lead and zinc which can be potentially recovered by developing a hydro-metallurgical method, which is selective for zinc and lead with respect to iron. From the three selected chemical extraction techniques, i.e., sulfuric acid, methanesulfonic acid, and sodium hydroxide solutions, the best results were given by sodium hydroxide solution leaching, due to negligible concentration of Fe in pregnant leaching solution, and leached metals were not precipitated into new mineral phases. Pb and Zn showed, however, a leaching efficiency of only 22% (0.0–63.6%) and 14% (0.0–29.1%) in average respectively. Furthermore, this study investigated how the mineralogy of mine waste material influences the leaching efficiency of the samples. Regardless of the lixiviant and the material, the highest leaching efficiency of Pb and Zn was achieved for samples where abundant Pb and Zn minerals were present. In samples where no Pb- or Zn-bearing minerals were detected, leaching efficiency was lower or negligible.

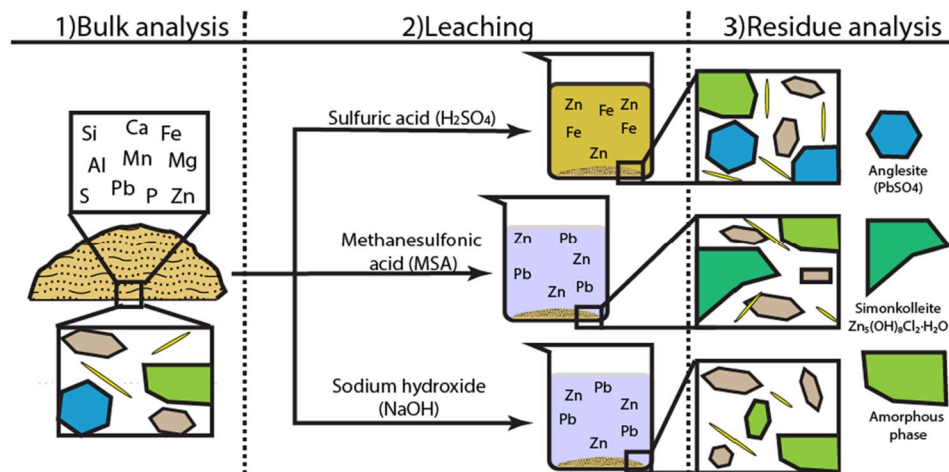
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✉ Srećko Bevandić
Sreko.bevandi@kuleuven.be

¹ Department of Earth and Environmental Sciences, KU Leuven, Celestijnenlaan 200E, 3001 Leuven, Belgium

² Department of Chemistry, KU Leuven, Celestijnenlaan 200F, 3001 Leuven, Belgium

Graphical Abstract



Keywords Leaching · Methanesulfonic acid · Mine waste · Sodium hydroxide · Sulfuric acid

Introduction

The mineral processing industries are generating a significant amount of mine waste material [1]. Depending on the leftover content of profitable metals (e.g., Cu, Ga, Au) content within the mine waste material, there is potential to consider them as a low-grade ore and could form an additional revenue for mining industries [2]. For this reason, recent studies are emphasizing the characterization of historic mine waste such as tailings, waste rock, and topsoil [3]. This type of waste often contains an attractive amount of profitable base (e.g., Cu, Pb, and Zn), critical (Ga, Ge, In) and precious (Au, Ag, Pt) metals as well as hazardous elements (As, Cd, Cr) [2–4]. Many of the elements which were previously not of economic interest are now of interest, for example, high-tech applications (e.g., Co, In, Ge, Ga), and they are often found in the mine waste material [5]. The mine waste resulting from mineral processing operations is usually stored in the form of tailing ponds. A single tailings pond or mine waste dump can reach up to 100,000 m³, but often there are multiple smaller mine waste deposits with similar geochemical and mineralogical characteristics around the historical mine site [6]. Taking into account these potentially vast quantities of material with their potential economic and negative environmental impact, it is important to do an in-depth characterization of the historical mining sites (e.g., geological, environmental, metallurgical) in order to assess the metal extraction of these wastes or their reusability for other purposes (e.g., construction materials) [2, 7, 8].

To date, mine waste material is mainly used as raw material in the ceramic industry for the production of roof tiles and bricks as well as an aggregate (e.g., for asphalts) and

less often as a secondary metal resource [7–12]. When mine waste is used for metal extraction, the investigation focus is on multiple metals, rather than on a single element. For instance, an investigation of the Pitcher mine district area (Oklahoma, US) established that recovery of Al, Ti, Pb, and Zn from mine waste was feasible at the prices when the study was done (2013) [13]. A more recent study focused on applying different mine waste material from an abandoned mine site at Gorno (Italy) in the metal industry as well in the agricultural sector as a soil additive. This approach is making deposits more sustainable and economically feasible [14].

Due to the increasing demand for base and precious (e.g., Au, Ag, Pt) metals and depletion of the ore grade, valorization and reprocessing of mine waste for secondary metal recovery are becoming more common in practice [15, 16]. For example, a recent study dealing with lead–zinc resources on a global scale estimated that the average grade of Pb and Zn (individual mineral deposits and mine waste) is 0.44% and 1.20%, respectively [16]. Therefore, recent studies focus on the recovery of metals from low-grade secondary resources such as tailings, furnace dust, metallurgical slags, and sludges. Metal extraction from these materials is usually performed by pyro- or hydrometallurgical processes or a combination of both [17–20]. Hydrometallurgically metal extraction routes are usually cheaper and can treat low-grade resources more efficiently and with better selectivity than pyrometallurgical ones [17].

As mentioned above, mine waste material often contains hazardous elements such as arsenic (As), chromium (Cr), cadmium (Cd), mercury (Hg), etc. [3]. This is especially relevant for mine waste that originated from sulfidic ores due to

their propensity to generate acid mine drainage (AMD) and can cause severe environmental accidents [21]. Therefore, a solution to mitigate the potential environmental impact of sulfidic tailings is to remediate the material containing hazardous elements, recover the metal value content, and re-use the matrix material for other purposes (e.g., ceramics, cement). Nevertheless, information about historical mine wastes is limited or missing. So the material has first to be characterized before the metal extraction potential is assessed at laboratory scale [22–25]. The optimal technique for reprocessing mine waste is the subject of many different studies [17]. Based on the available literature, leaching is considered to be a suitable method for extracting zinc and lead from secondary sources [26–29]. Studies that focus on the extraction of Pb and Zn are using lixiviants such as ammonia–ammonium mixtures, sodium hydroxide, methanesulfonic acid, sulfuric acid, nitric acid, or ammonia solutions [15, 30, 31].

The Plombières tailings pond is located in a small town (Plombières) in eastern Belgium. The Plombières mine waste was produced by pyrometallurgical (smelting) processing of Mississippi Valley-type (MVT) Pb–Zn ore from the old mining district Plombières-Altenberg-Moresnet [32]. Both mining and processing of the ore were done in Plombières. This mining site was historically important for the production of Pb and Zn in Belgium since ore was mined and processed from the Roman age until 1922 [33, 34]. The tailings pond covers a surface area of 8000 m² and consists of 4 layers: soil, metallurgical waste, brown, and yellow tailings [32]. Metallurgical waste-containing slags are widely spread over the ancient mining site. In addition, pipes for the melting of the ore and building material (e.g., mortar, red bricks) are incorporated within this layer. The soil layer most likely developed on top of the metallurgical waste [32].

At present, most of the research in this field focuses on one specific type of material for one type of mine waste [32]. The purpose of this study is to assess the metal extraction potential for Pb and Zn from the different mine waste materials of this tailings pond (i.e., metallurgical waste, brown, and yellow tailings) by using sulfuric acid, sodium hydroxide, and methanesulfonic acid solutions as leaching agents. Furthermore, the aim of this study is to get a profound insight into how the mineralogy of the samples influences the leaching efficiency.

Materials and Methods

Materials

Representative samples of different mine waste materials from the Plombières tailings pond were used in this study. Samples were selected based on the mineral variability

and bulk Pb and Zn contents as determined in a previous study [32]. For this study, samples of metallurgical waste, brown, and yellow tailings were selected for investigation. From each type of material, 4 subsamples were selected for investigation. The aim was to select 12 samples that would represent high, low, and average contents of Pb and Zn. The coordinates of the locations where samples were taken can be found in supplementary material Table 1S.

Nitric acid (65 wt% a.r.) was purchased from Acros Organics (Geel, Belgium). Multi-element standard (100 mg L⁻¹ in 2–5% HNO₃), scandium standard (1000 mg L⁻¹ in 2–5% HNO₃), and sulfuric acid solution (95 wt%) were obtained from ChemLab (Zedelgem, Belgium). Methanesulfonic acid solution, MSA (99.5%), was purchased from Carl Roth (Karlsruhe, Germany). Hydrochloric acid solution (37 wt%) was purchased from VWR (Fontenay-sous-Bois, France). Water was always of ultrapure quality, deionized to a resistivity of 18.2 MΩ cm with a Millipore ultrapure water system (Milli-Q Water). All chemicals were used as received without any further purification.

Analytical Methods

Before any analysis, samples were dried at 40 °C, deagglomerated with a pestle and mortar, split using a riffle splitter to get a representative sample and finally grounded to a powder (< 64 μm) using a pulverizing mill (time = 5 min; speed = 650 rpm). Analysis (mineralogical and geochemical) and leaching experiments were done for the samples from the splitting batch, in order to have consistency in results.

X-ray diffraction analyses were performed at the Geoinstitute of KU Leuven. Powdered and split samples (1.8 g) were subsequently mixed with 0.2 g (90:10 ratio) of an internal standard (zincite). The mixture of sample and standard were mixed with ethanol, milled with a McCrone micronizing mill and dried. For powder measurements, a Philips PW 1380 diffractometer was used at a voltage of 45 kV and a current of 30 mA. The machine is equipped with a Cu anode, a graphite monochromator, a receiving slit width of 1 mm, and a continuous scanning mode. Interpretation of the analysis was done with Profex 4.0 software using the BGMN program Rietveld refinement.

As noted in the supplementary material (Tables 2S and 3S), the geochemical analyses of the samples were described in the study of Bevandić et al. [32].

The metal concentration in the pregnant leaching solutions (PLSs) was measured by inductively coupled plasma—optical emission spectroscopy (ICP-OES) by using a Perkin-Elmer Optima 8300 spectrometer equipped with an axial/radial dual plasma view, a GemTip Cross-Flow II nebulizer, a Scott double pass with an inert Ryton spray chamber and a demountable one-piece Hybrid XLT ceramic torch with a sapphire injector (2.0 mm internal diameter). Dilutions were

done with 5 vol% HNO₃ solutions, and all ICP-OES samples were measured in triplicate. The dilution factor was chosen so that the final concentration was lower than 25 mg L⁻¹ and dysprosium was used as an internal standard. A Heraeus D-6450 oven was employed to dry the solid residue samples.

Experimental Methods

Leaching of the examined samples was carried out in 20 mL glass vials. Thermoshakers from ThermoFisher Scientific were used to mix the vials at constant temperature and mixing speed. After leaching, the solid phase was separated from the liquid phase by vacuum filtration with 1.6 μm porous size filtration paper.

For the leaching experiments, 2 g of powdered sample was mixed with the lixiviant solution ([MSA] = 10 wt%, [H₂SO₄] = 10 wt%, [NaOH] = 10 wt%). The vial was sealed and then placed in the Thermoshaker for 4 h at 60 °C and 200 rpm. Immediately after filtration, aliquots of approximately 1 mL were withdrawn and diluted with HNO₃ (5 vol%) for ICP-OES analysis. The leaching efficiency E_L (%) was calculated according to Eq. (1):

$$E_L(\%) = \frac{C_{PLS} \times V_{LIX}}{m_s \times C_s} \times 100, \quad (1)$$

where C_{PLS} is the metal ion concentration in the PLS (mg L⁻¹), V_{LIX} is the volume of the lixiviant used for leaching (L), m_s is the mass of the solid material used for leaching (kg), and C_s is the concentration of the metal in the material before leaching (mg kg⁻¹). The pH of the PLS was also measured after filtration. The filtrated solid residue after

leaching was washed with Milli-Q Water 3 times after filtration, subsequently dried in an oven at 40 °C for 24 h and then characterized by XRD analysis.

Results and Discussion

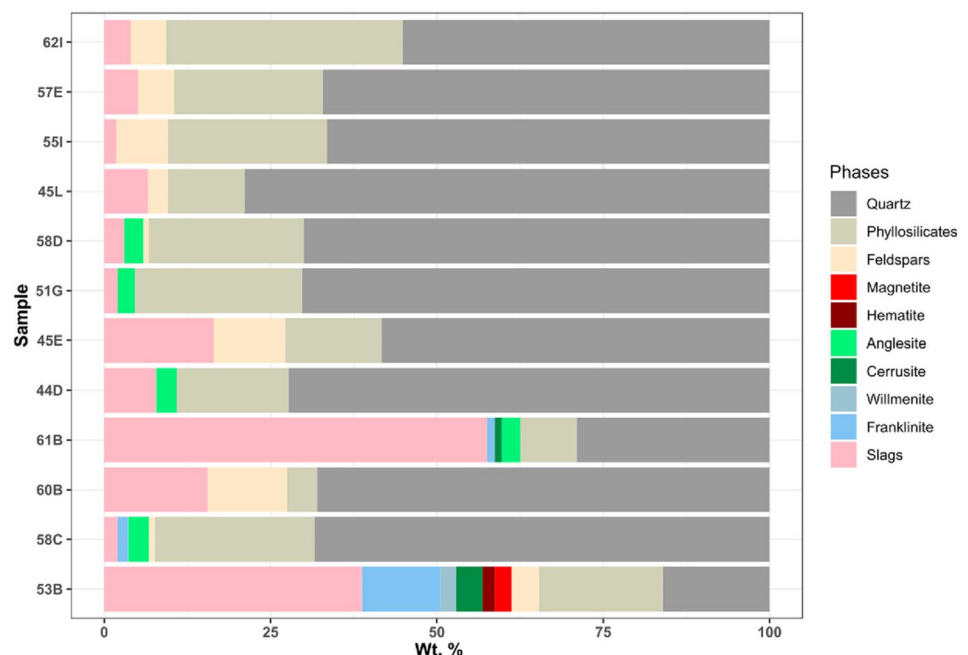
Characterization of Different Mine Waste

Mineralogy

Based on the mineralogical quantification (Fig. 1), more than 80 wt% of mine waste comprises quartz (SiO₂) (16–79 wt%), muscovite (KAl₂(AlSi₃)O₁₀(OH)₂) (4–35 wt%), and amorphous phases (1–60 wt%). An XRD and scanning electron microscopy (SEM–EDX) investigation of samples from the same site showed that these amorphous phases are pyrometallurgical slags [30]. All samples also contain feldspars (plagioclase [(NaAlSi₃O₈–CaAl₂Si₂O₈)] and K-feldspars [KAlSi₃O₈] (0–13 wt%). Depending on the type of material, Fe-, Pb-, and Zn-bearing minerals can be found. In the metallurgical waste layer, all listed metal-bearing minerals can be found. Iron minerals are magnetite (Fe²⁺Fe³⁺₂O₄) and hematite (Fe₂O₃) (Fig. 2). Lead phases found within the samples are anglesite (PbSO₄) and cerussite (PbCO₃) (Fig. 2). Franklinite (ZnFe₂O₄) and willemite (ZnSiO₄) are Zn-bearing minerals (Fig. 2). A mineral list based on the sample can be found in supplementary data Table 4S.

XRD results on the metallurgical waste samples revealed that Fe-, Pb-, and Zn-bearing minerals are magnetite, hematite, anglesite, cerussite, willemite, and franklinite. In the brown tailings, only anglesite was found. In samples of

Fig. 1 Mineralogical composition for different mine waste materials identified at Plombières



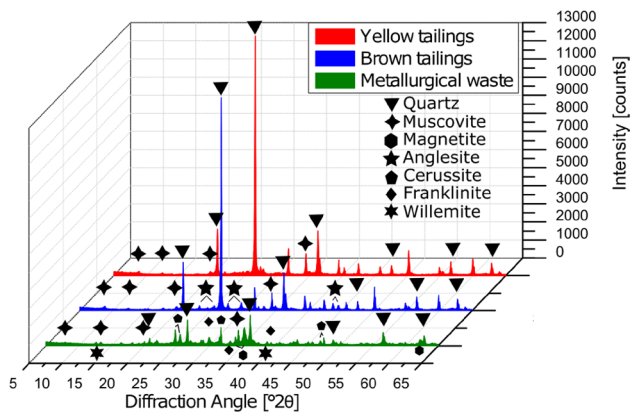
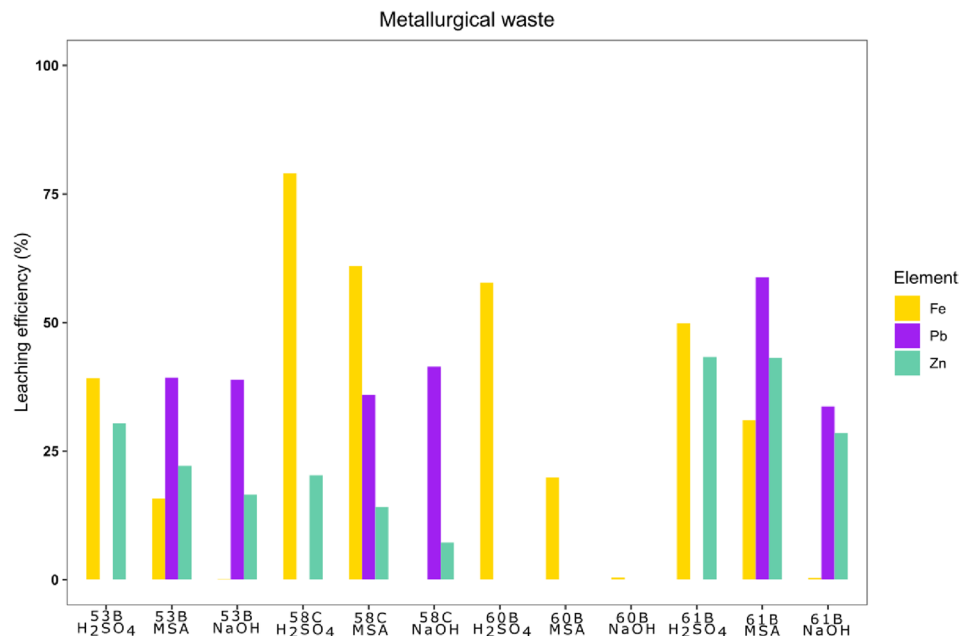


Fig. 2 XRD patterns of the different mine waste samples studied before leaching

the yellow tailings, no Fe-, Pb-, and Zn-bearing minerals were found. The presented results are consistent with the results of previous studies [32]. However, previous studies done on the same material from the same tailing pond using scanning electron microscopy (SEM), mineral liberation analyzer (MLA), electron probe micro-analyzer (EPMA), and ore microscopy did identify pyrite ($[\text{FeS}_2]$), Pb oxides, sphalerite (ZnS), and fraiponite ($[(\text{Zn},\text{Al})_3(\text{Si},\text{Al})_2\text{O}_5(\text{OH})_4]$) that were not detected by the current study [30, 33]. The phases not detected by XRD could be due to the low content of the minerals and, thus, low intensity, and due to high background caused by the amorphous phase present in the pyrometallurgical slags [35]. Furthermore, the same studies concluded that Pb and Zn are present in the pyrometallurgical slags [28, 36]

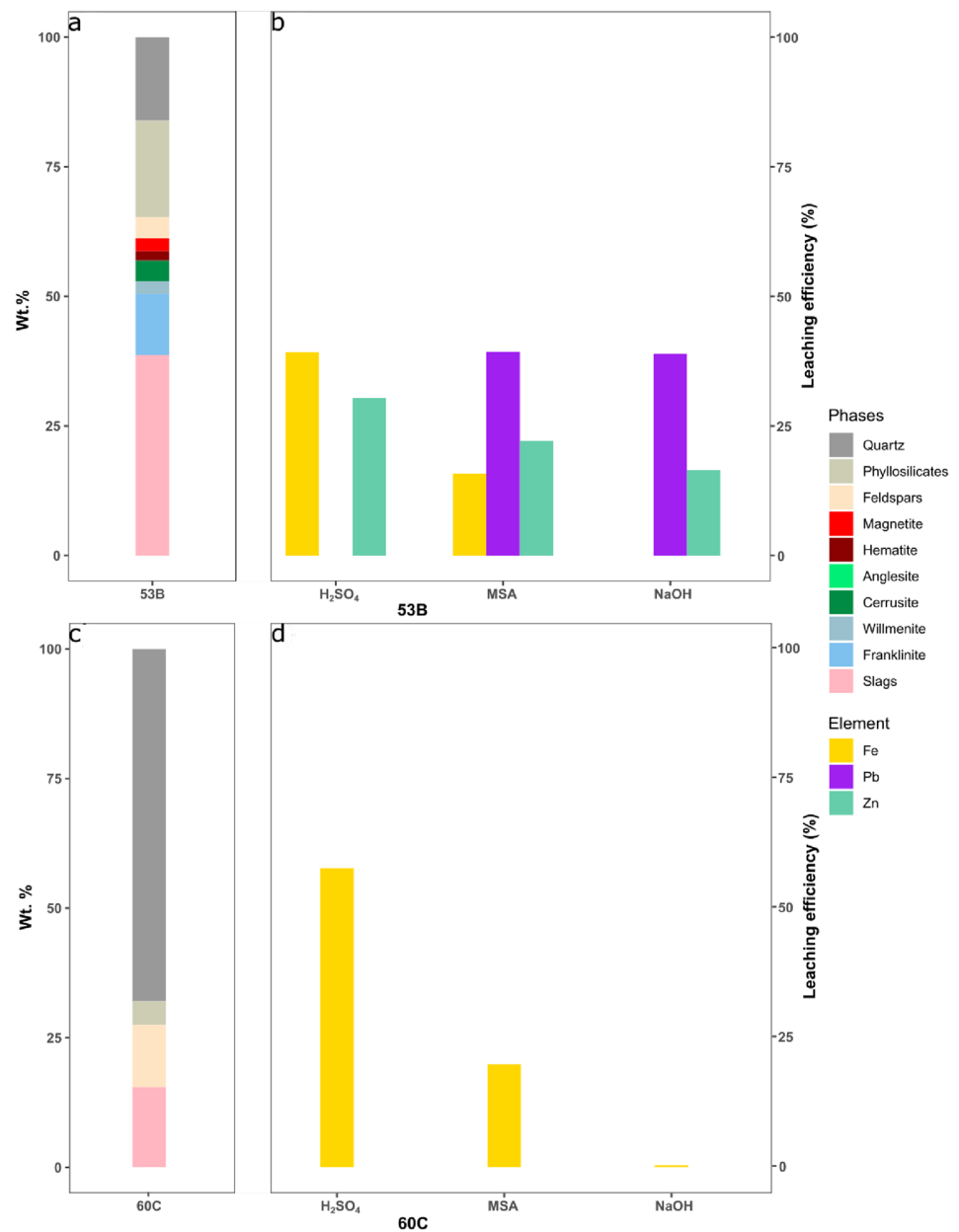
Fig. 3 Leaching efficiency results of Fe, Pb, and Zn of all metallurgical waste samples by using H_2SO_4 , MSA, and NaOH as lixivants. Conditions: lixiviant concentration = 10 wt%, $L/S = 10 \text{ mL g}^{-1}$, $T = 60 \text{ }^\circ\text{C}$, $t = 4 \text{ h}$



Leaching of Mine Waste Material

The leaching results of the metallurgical waste using MSA, NaOH, and H_2SO_4 are illustrated in Fig. 3. Results of leaching efficiency can also be found in Tables 6S, 7S, and 8S. The leaching of metallurgical waste material with MSA has the highest leaching efficiency for the targeted metals (Fig. 3). In particular, the averaged leachability for lead was 40% [range 0–63%] and that of zinc 33% [range 0–41%] (Fig. 3). However, the solubilization of iron (Fe) was also relatively high (2680 mg L^{-1}). Zinc was leached best with sulfuric acid (0–45%; average—23%), but the iron content remained high in the PLS (2261 mg L^{-1}) (Fig. 3). The leaching efficiency of lead was negligible. Based on similar studies, it would be plausible that Pb was solubilized during leaching and then precipitated as lead sulfate [37]. Lead and zinc were selectively leached (0–43%, average—38% and 0–29%, average—17%, respectively) with the rejection of iron by using sodium hydroxide. This behavior can be attributed to the amphoteric nature of Pb and Zn to form soluble hydroxide complexes, i.e., $[\text{Pb}(\text{OH})_3]^-$ and $[\text{Zn}(\text{OH})_4]^-$ at alkaline pH values [38–40]. Iron is known to be insoluble at the measured pH of the NaOH leachates (pH 13). A higher efficiency is noticed in the samples where Pb- and Zn-bearing minerals are found. To illustrate that compartment between two samples of metallurgical waste, the mineralogy of samples 53B and 60C is plotted in Fig. 4. In sample 53B, both Pb- and Zn-bearing minerals were detected, while in samples 60C, they were not (Fig. 4). Zinc was partly solubilized after each lixiviant (H_2SO_4 —30.4%; MSA—22.1% and NaOH—16.5%), while Pb was only partly solubilized after MSA (39.3%) and NaOH (38.9%). However, in sample

Fig. 4 Example of 2 samples of metallurgical waste illustrating that samples with detected Pb- and Zn-bearing phases have higher leaching efficiency (a, b), contrary to the sample where minerals of interest were not detected and leaching efficiency is negligible (c, d)



60C, neither Pb nor Zn was solubilized (Fig. 4). Moreover, sample 60C is the only sample of metallurgical waste where neither Pb nor Zn minerals were detected (Fig. 1), and it is the only sample where solubilization of metals of interest did not occur (Fig. 3).

Similar leaching yields were obtained for the brown tailings (Fig. 5). Lead leached best (40%) with MSA and NaOH and remained insoluble after leaching with H₂SO₄; however, anglesite was detected in the residue (Tables 8S, 9S, and 10S). Zinc showed the highest leaching efficiency (30–46%, average 36%) after leaching with H₂SO₄, followed by MSA leaching (27–41%, average 32%) and NaOH (5–30%, average 17%). Nevertheless, due to the low measured pH of the generated leachates with MSA and H₂SO₄,

iron solubilization was high (2050 mg L⁻¹ and 2728 mg L⁻¹ respectively). Similar to the metallurgical waste, leaching efficiency is higher in samples where minerals of interest (anglesite) are found. Sample 45E is the only sample where the leaching efficiency of Pb is 0% (Fig. 5). This is the only sample where anglesite was not observed (Fig. 1). Anglesite was detected in the other samples (Fig. 1), and the leaching efficiency of these samples are noticeable (Fig. 5). Contrary to the metallurgical waste, no Zn minerals were detected in the samples of the brown tailings investigated in this study.

Slightly different results were observed in the case of yellow tailings (Fig. 6). Leaching of yellow tailings with H₂SO₄ solubilized a significant amount of Fe (1376 mg L⁻¹) and leached 28% of Zn on average (0–50%). Similarly

Fig. 5 Leaching efficiency results of Fe, Pb, and Zn of brown tailings samples ($n=4$) by using H_2SO_4 , MSA, and NaOH as lixivants. Conditions: lixiviant concentration = 10 wt%, L/S = 10 mL g^{-1} , $T=60\text{ }^\circ\text{C}$, $t=4\text{ h}$

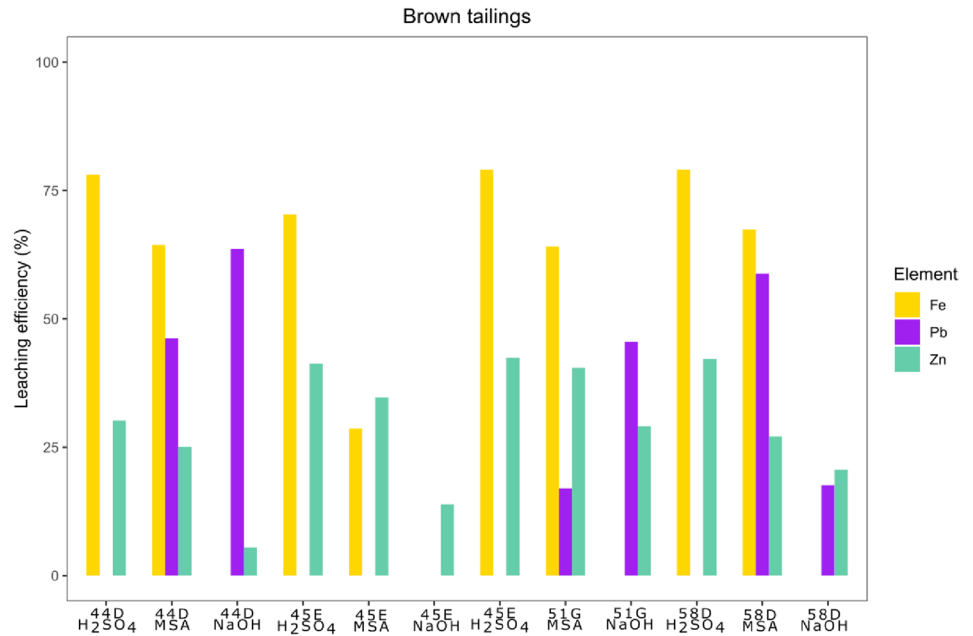
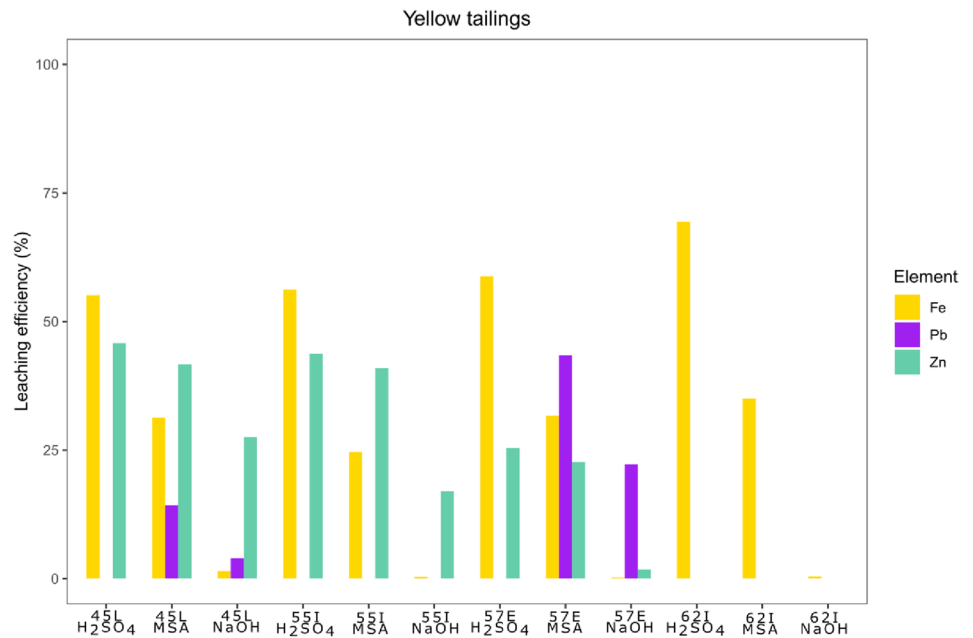


Fig. 6 Leaching efficiency results of Fe, Pb, and Zn of all yellow tailings samples by using H_2SO_4 , MSA, and NaOH as lixivants. Conditions: lixiviant concentration = 10 wt%, L/S = 10 mL g^{-1} , $T=60\text{ }^\circ\text{C}$, $t=4\text{ h}$



to the results of metallurgical wastes and brown tailings, leaching with sulfuric acid caused the formation of an anglesite precipitate in the solid residue (Table 10S). MSA leaching showed moderate leachabilities for the targeted metals. More specifically Zn and Pb were leached 27% (0–43%) and 14% (0–44%), respectively. No Pb- or Zn-bearing minerals were found, so the correlation between leaching efficiency and abundance of minerals of interest found earlier in the two other material types cannot be supported by the findings of the yellow tailings. However, both Pb- and Zn-bearing minerals were detected in

previous studies [32, 41]. The solubilization of iron in these leachates was 2261 $mg\text{ L}^{-1}$. The leaching efficiency for each material can be found in Tables 4S, 5S, and 6S. The sodium hydroxide solution selectively leached Pb and Zn at an average efficiency of 10% and 15%, respectively. The lower leaching efficiencies obtained from the leaching of yellow tailings compared to brown tailings and metallurgical slags could be associated with the low Zn and Pb contents in this material and the absence of Pb- and Zn-bearing minerals. The average content of both Pb and Zn is 0.2 wt%. This is low when compared with the average

content of Pb (2.8 wt%) and Zn (3.3 wt%) in the metallurgical waste layer [32].

Overall and based on the experimental results reported above, it can be concluded that Zn was extracted more efficiently with sulfuric acid or MSA but that also the iron concentration in the leachates was high in all examined samples. Pb was leached at moderate yields with sodium hydroxide and MSA, and it formed a lead sulfate precipitate after leaching with H_2SO_4 . Leaching of these mine wastes with sodium hydroxide was the most selective method to extract Pb and Zn with respect to iron.

Characterization of Solid Residue

Sulfuric Acid Leaching

As mentioned above, sulfuric acid showed a preferred leachability for Fe and Zn, rather than for Pb. A similar observation was made by Zhang et al. [37]. Moreover, it was noticed that samples richer in slags had lower leaching efficiency than samples with Zn-bearing minerals. Mineralogical analysis performed on the residue of metallurgical waste, brown, and yellow tailings samples revealed that all detected Zn minerals were partially dissolved (Fig. 6). As discussed above, sulfuric acid is not a suitable leaching solution for Pb, due to the formation of anglesite [37, 42]. This was confirmed by XRD analysis of the residue (Figs. 7, 8 and 9). In the original samples of yellow tailings, no Pb-mineral was observed but after leaching with sulfuric acid, and anglesite phases were detected at concentrations up to 2.5 wt% (Fig. 7).

The relative content of amorphous phases in the residue was higher than in the starting material (Figs. 7, 8, 9 and

Tables 8S, 9S, 10S). An increase in amorphous content is due to the major dissolution of mineral phases, which indicates slags could be partly dissolved.

Methanesulfonic Acid

Leaching with methanesulfonic acid resulted in the highest leaching yields not only for both Pb and Zn but also with high iron solubilization. Some interesting mineralogical results were observed on the solid residues when compared to the ones generated after leaching with sulfuric acid. More specifically, the Zn-bearing minerals were still present. For example in sample 53B (metallurgical waste), the franklinite content is 14.2 wt%, while in the residue, the content is 2.3 wt% (Fig. 7). Leaching experiments performed on material where franklinite is roasted before leaching, indicate that roasting increases the leachability of Zn [43, 44]. Furthermore, the formation of the simonkolleite ($Zn_5(OH)_8Cl_2 \cdot H_2O$) was noticed in the residue of samples measure after the MSA leaching (Figs. 7, 8, 9, 1S, 2S and 3S). This mineral is a result of the weathering of Zn slags [45]. As with the sulfuric acid, the content of the amorphous phase was higher, which is due to the important dissolution of the mineral phases.

Sodium Hydroxide (NaOH)

Leaching with sodium hydroxide gave more selective results for both Pb and Zn, with respect to Fe. Contrary to the previous two lixivants (sulfuric acid and methanesulfonic acid), no new by-product minerals were formed (Figs. 7, 8, 9, 1S, 2S and 3S) and a negligible concentration of Fe was leached

Fig. 7 Original mineralogy of sample 53B (metallurgical waste) and mineralogy of the residue after each type of leaching

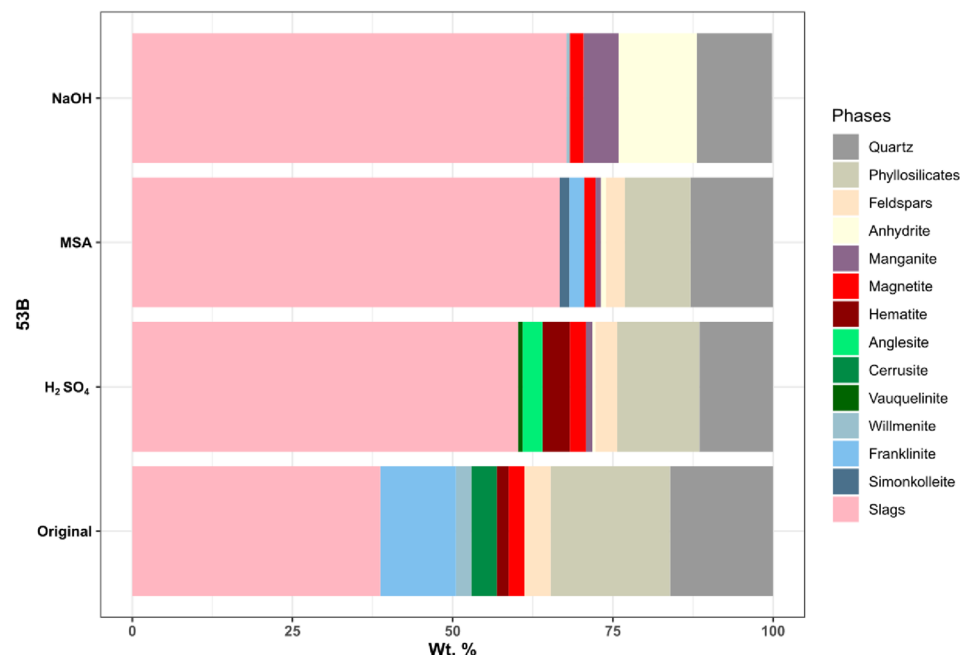


Fig. 8 Original mineralogy of sample 44D (brown tailings) and mineralogy of the residue after each type of leaching

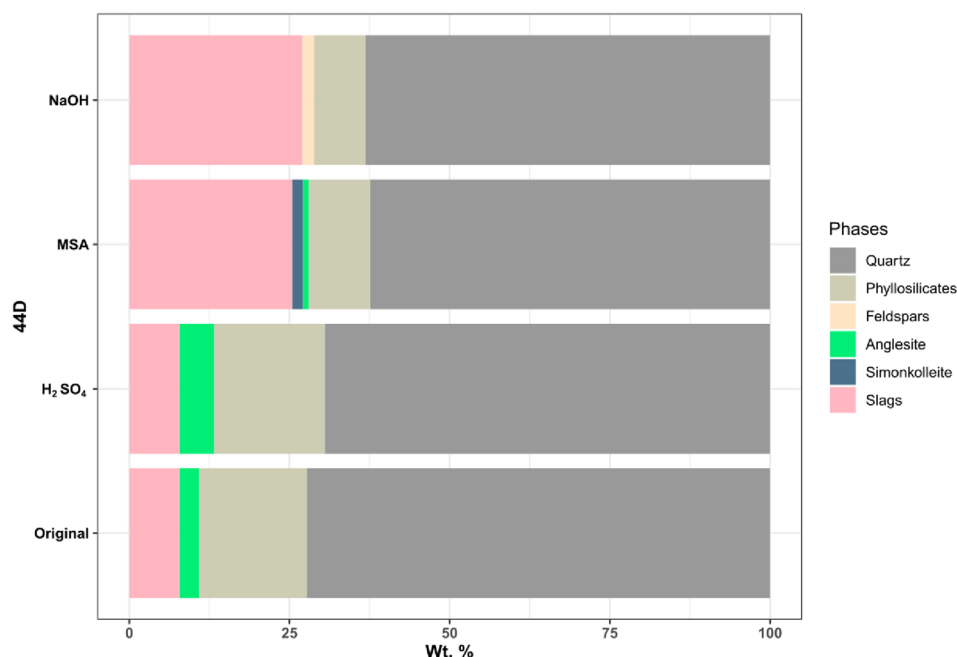
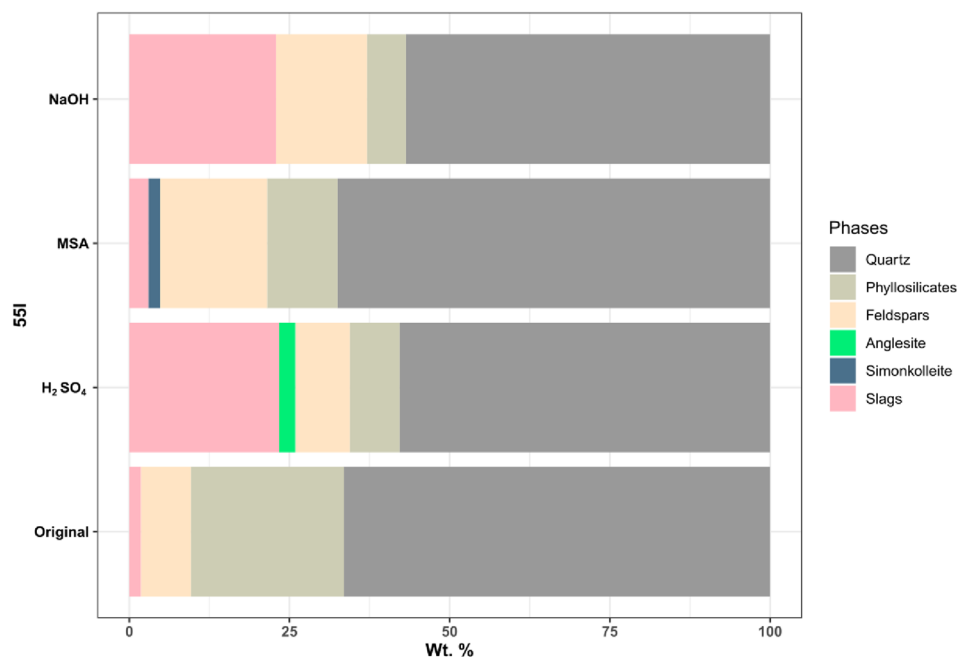


Fig. 9 Original mineralogy of sample 55I (yellow tailings) and mineralogy of the residue after each type of leaching



(Figs. 3, 4 and 5; Table 6S). Similar to the residue after the two other types of leaching, the content of the amorphous phase increased (Tables 8S, 9S, 10S). In the leaching residue of the metallurgical waste, franklinite was observed (Fig. 7). A study focusing on improving the solubility of franklinite in NaOH showed that franklinite decomposes if it is heated before submitting it to the lixiviant [46]. The same study demonstrated that the leaching efficiency of material consisting of similar phases (Zn-bearing minerals and slags) increased up to 95% if the material was heated up to a

temperature of 350 to 400 °C. This is related to the decomposition of franklinite at this specific temperature range [47].

Optimal Method and Material for Metal Extraction

Based on the previous sections, zinc showed the highest extraction efficiency (20–50%) after leaching with sulfuric acid in all three types of waste material. Nevertheless, the content of iron was high in these leachates (2261 mg L⁻¹). As a result, a post-treatment step would be required to

remove iron. This step usually involves the precipitation of iron as a hydroxide by raising the pH of the solution above 4 [48–50]. Leaching with MSA yielded extraction of both Pb and Zn but again the iron content was high and a Fe removal step is required. The addition of extra post-treatment steps for the removal of Fe increases the total cost. Leaching with sodium hydroxide appears to be a more attractive leaching method since Pb and Zn were selectively leached with respect to iron. Prior studies conducted on the leaching of Pb–Zn mine wastes with sodium hydroxide, showed that under optimized conditions, it was possible to leach 86% of Pb and 72% of Zn [51]. Hence, optimizing the NaOH-leaching conditions (e.g., temperature, L/S ratio, NaOH concentration, etc.) is expected to improve the leachabilities of Pb and Zn, but investigating these parameters was beyond the scope of this study. Based on the present data, the leaching efficiency is strongly controlled by the presence of the slag phases within the samples.

As mentioned above, the examined samples consisted of three types of materials: metallurgical waste, brown tailings, and yellow tailings. Having as criteria the obtained results after leaching with sodium hydroxide and the Pb and Zn contents in the solid samples, the most promising materials to be considered as a secondary low-grade ore are the metallurgical waste and the brown tailings [32]. Although the yellow tailings samples have low Pb and Zn contents, they could be reused as ceramic materials [52].

Conclusion

The lead and zinc extraction potential of the Plombières mine waste, after leaching with sulfuric acid, methanesulfonic acid and sodium hydroxide were investigated. Zinc was leached best (20–50%) with sulfuric acid and Pb (0–58%) with MSA, but both acids also leached a high amount of iron independently of the type of the material. Leaching with sulfuric acid yielded the formation of lead sulfate (anglesite) in the solid residues of all examined samples. Furthermore, MSA leaching led to the formation of simonkolleite in the solid residues. With both lixivants, additional steps of purifying the leachate by removing iron should be added. Moreover, an additional extraction step for lead or zinc present as newly formed phases in the residue is required. More selective Pb and Zn extraction were achieved by leaching with sodium hydroxide and, for this reason, were considered as the optimal leaching agent from the three considered. Leaching with NaOH did not cause the formation of new mineral phases, but franklinite (Zn spinel) did not dissolve completely. The too low leaching efficiency of Pb and Zn is due to the presence of these metals in the slags.

This study concluded that metals are dominantly leached from mineral phases (e.g., anglesite for Pb, gahnite for Zn),

while metals or not or only leached in low quantities from the slags present in all material types. This is confirmed by an increase in the relative slag content in the residue after leaching experiments. Based on this knowledge, future research should focus on increasing the leachability of slags by finding optimal conditions (e.g., increase of temperature) or by adding steps before leaching (e.g., roasting). A better understanding of the mineralogy is proven to be necessary for a profound insight in the leaching efficiency and recoverability of the metals of interest.

Supplementary Information The online version contains supplementary material available at <https://doi.org/10.1007/s40831-021-00445-0>.

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Declarations

Conflict of interest On behalf of all authors, the corresponding author states that there is no conflict of interest.

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