Master's Thesis Engineering Technology

2018-2019

Microwave assisted roasting of sulphidic tailings

Vincent Lemmens

Master of Chemical Engineering Technology

Introduction & objectives

VITO is an independent Flemish research organization in the area of cleantech and sustainable development. This thesis took place in the **Sustainable Materials Management unit**.

The **demand** for commodity metals, such as **copper (Cu)**, is increasing [1]. However, ore grades are dropping and novel technologies are needed to recover metals from such low grade ores. Mining practices produce large amounts of waste materials, such as **tailings**, that contain small amounts of **valuable metals** (Figure 1)**.** These tailings are a huge new stock of metals and minerals. In addition, sulphidic tailings that are poorly managed cause environmental concern as they are prone to form **acid mine drainage (AMD)** waters that contain heavy metals [2]. Therefore, **roasting** is performed as a pretreatment for both metal recovery as well as **desulfurization**, which reduces AMD if the residue is disposed but introduces the **release** of **SO_2** gas.

The aim is to **treat sulphidic tailings** with carbonate-rich waste rock (**marl**) in a **roasting** step to simultaneously oxidize sulphide minerals and **fixate the released SO_2**. The influence of this treatment on the Cu leachability in an ammoniacal solution was also tested.

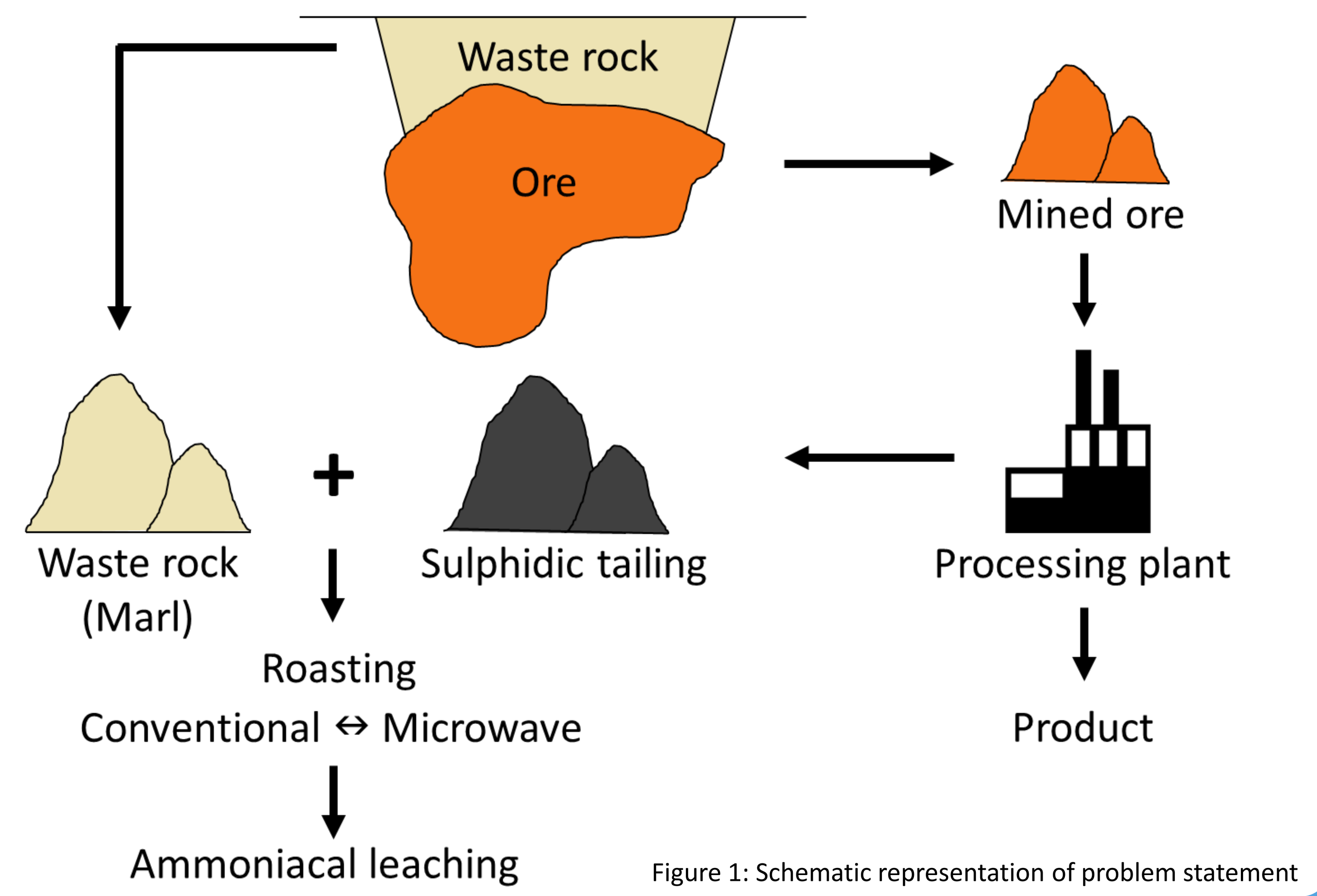

Figure 1: Schematic representation of problem statement

Materials & Methods

Conventional (CF) roasting

(*Nabertherm LH 60/14*)

- Samples (Table 1) roasted at **400 °C, 500 °C, 600 °C, 700 °C or 800 °C**
- Ramp up: 100 °C/h
- Dwell time: 1 hour

Microwave (MW) roasting

(*Milestone PYRO advanced microwave furnace*)

- Samples (Table 1) roasted at **300 °C, 400 °C or 500 °C**
- Ramp up: 1800 W (varies) 30 min
- Dwell time: **1 h**

- Samples (Table 1) roasted at **500 °C**
- Ramp up: 1800W (varies) for 30 min
- Dwell time: **15 min, 30 min or 1 h**

Table 1: Tailing to marl sample ratios

m% tailing	m% marl	Roasting furnace
100%	0%	CF, MW, *
80%	20%	CF, *
60%	40%	CF
50%	50%	CF, MW, *
35%	65%	CF, *
20%	80%	CF
0%	100%	CF, MW, *

Ammoniacal Leaching

- Leaching performed on roasted samples
- Leaching solution: **3.5 M NH_3 / 3 M $(NH_4)_2CO_3$**
- **4 hour** leaching at **60 °C** in a shaking water bath *(Grant GLS 400)*
- Leaching kinetics (50 % tailing/50 % marl; 500 °C; CF) at **60 °C; 30 min**, **1h**, **2h**, **3h** and **4h**

Analysis

- **XRF** (*Niton XL3T GOLDD+*) on roasted samples; determination of elemental content
- **XRD** (*PANalytical Empyrean*) on roasted samples; determination of crystalline phases
- **ICP-OES** (*Perkin Elmer AVIO 500+*) on filtered leachates; determination of **Cu**, S, Fe, Pb, Zn
- **TGA** (*Netzsch STA 449 C Jupiter*) on mixtures "*" (Table 1); mass loss relative to temperature

Results & discussion

Material composition

Tailing

XRF
- S - 35.5 %
- Fe - 27.7 %
- Ca - 2.1 %
- Cu - 0.5 %

Q-XRD
- Pyrite - 56.3 %
- Quartz - 27.9 %
- Others - 15.8 %

Marl

XRF
- Ca - 13.6 %
- Fe - 3.7%
- S - 0.4 %
- Cu - 0%

Q-XRD
- Muscovite - 37.5 %
- Calcite - 34 %
- Quartz - 14.1 %
- Others - 14.4 %

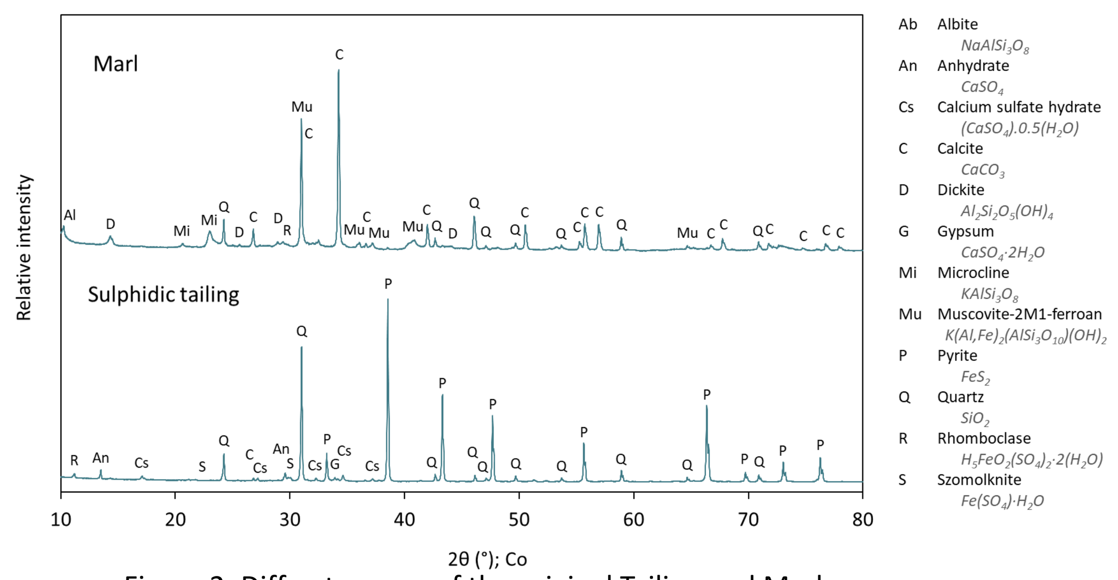

Figure 2: Diffractograms of the original Tailing and Marl

Conventional (CF) roasting

Microwave (MW) roasting

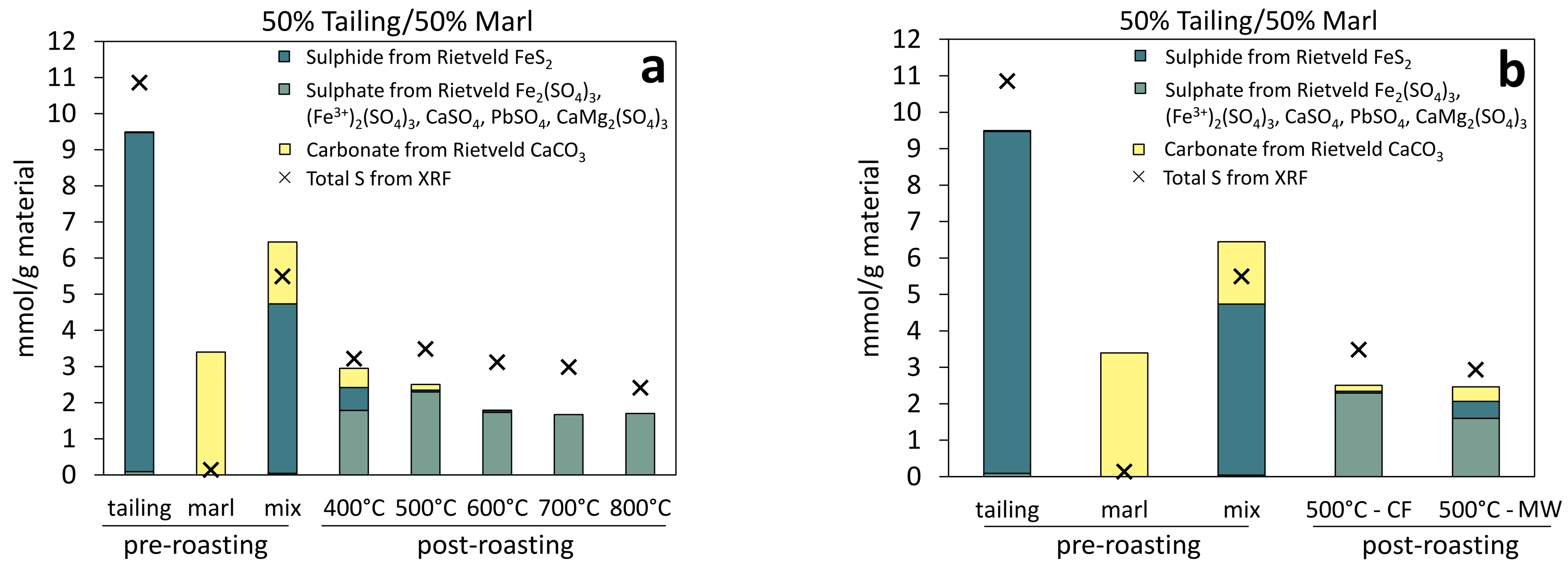

Figure 3: Sulphide, sulphate and carbonate concentration (mmol) per gram material; (a) CF and (b) MW

- At 400 °C FeS_2 is not fully oxidized
- Nearly complete FeS_2 oxidation at 500 °C
- Highest sulphate formation at 500 °C
- Minor sulphate and carbonate decomposition at 500 °C
- Decrease of sulphates ≥600 °C due to decomposition
- ≥600 °C all carbonate consumed (reaction or decomposition)

→ Optimal roasting temperature 500 °C

→ Marl content ↑, sulphate formed ↑, sulphate per amount of $CaCO_3$ consumed ↓

- At 500 °C FeS_2 is not fully oxidized

This might be due to:
- Sample temperature between 400 °C and 500 °C
- Less oxygen supply in the furnace
- Faster ramp up

Which causes:
- Less sulphate formation
- Less carbonate consumed

→ Potential of MW roasting is definitely conceivable with further optimization

Copper Leaching CF

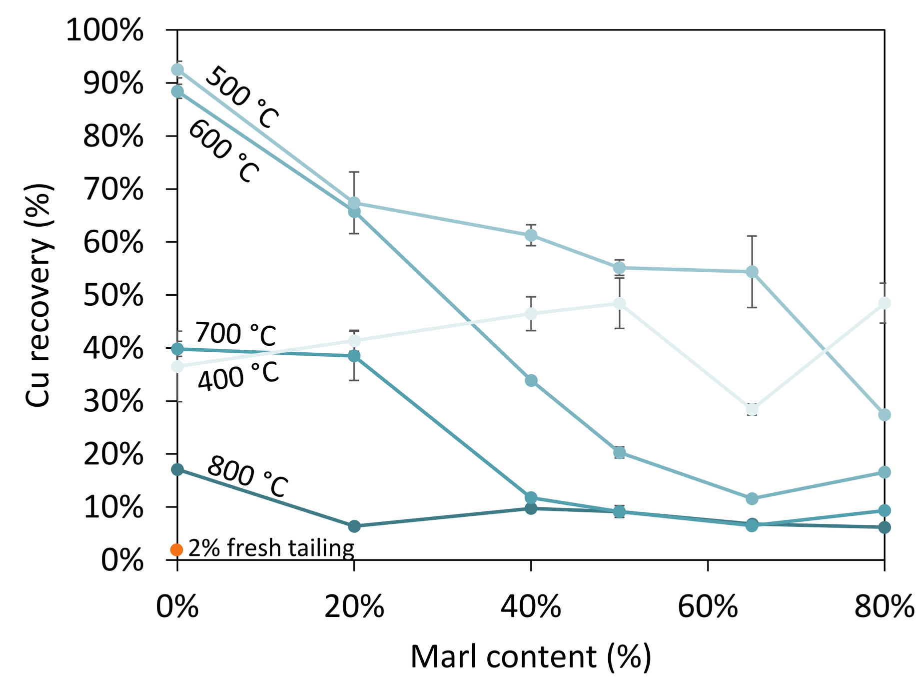

Figure 4: Copper recovery of conventional roasted samples

- Highest Cu recovery from samples roasted at 500 °C
- Decrease in Cu recovery with increasing marl content

→ Physical inhibition of Cu leaching or stable Cu phases

→ $CuSO_4$ likely the major Cu phase at 500 °C

Copper Leaching MW

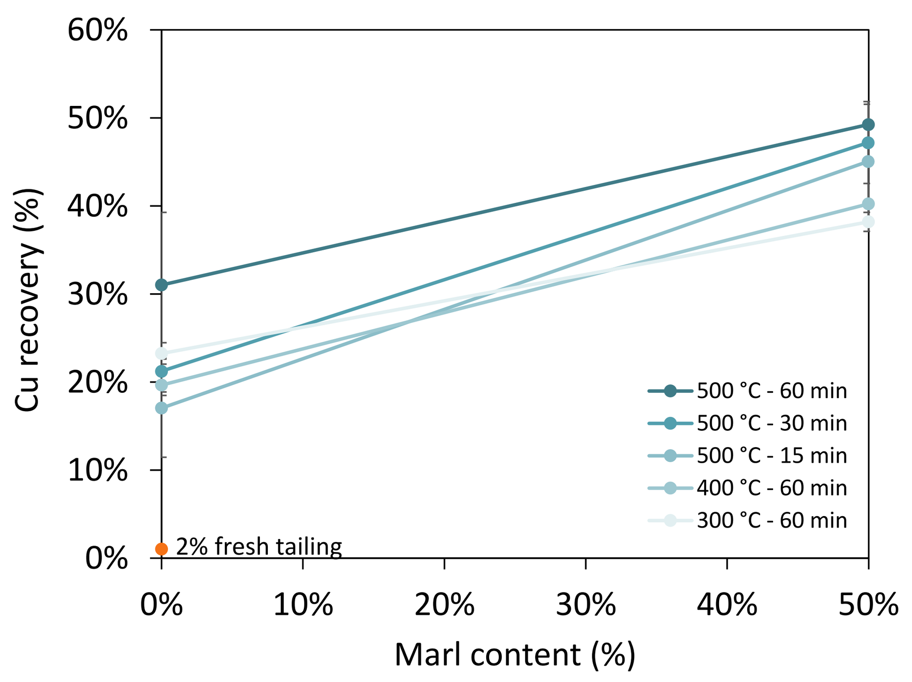

Figure 5: Copper recovery of microwave roasted samples

- Highest Cu recovery roasting at 500 °C for 60 min
- Increase in Cu recovery with increasing marl content

→ Same trends as leaching after CF roasting at 400 °C

→ Sample temperature likely between 400 °C - 500 °C

Conclusion

The sulphate formation (SO_2 fixation) is the highest for samples roasted at 500 °C in the CF. MW roasting performs slightly worse due to FeS_2 not being fully oxidized at 500 °C. However, MW roasting is definitely conceivable with further optimization. The addition of marl during the roast step increases SO_2 fixation, but less sulphates are formed per amount $CaCO_3$ consumed. Therefore, addition of the marl did not show an apparent ratio to be optimal. The highest Cu recovery is found in samples roasted at 500 °C. With the addition of marl during the roasting step, Cu recovery decreases (CF). However, for CF (400 °C) and MW (sample temperature 400 °C - 500 °C) roasting an increase is shown.

Supervisor / Co-supervisors: Prof. dr. ir. Leen Braeken - KU Leuven

Dr. Jeroen Spooren - VITO

Dr. ir. Maarten Everaert - VITO

Dr. Thomas Abo Atia - VITO

[1] M. Ozer, E. Acma and G. Atesok, "Sulfation roasting characteristics of copper-bearing materials," Asia-Pacific Journal of Chemical Engineering, no. 12, pp. 365-373, 2017.

[2] K. K. Kefeni, T. A. Msagati and B. B. Mamba, "Acid mine drainage: prevention, treatment options, and resource recovery: a review," *Journal of Cleaner productions,* vol. 151, pp. 475-493, 2017.

vito vision on technology

FLANDERS INNOVATION & ENTREPRENEURSHIP SIM Clusters for Growth

UHASSELT

KU LEUVEN