BIOHEAPLEACHING IN BOREAL CONDITIONS - TEMPERATURE PROFILE INSIDE THE HEAPS AND MICROBIOLOGY IN ELEVATED TEMPERATURES

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ABSTRACT

Terrafame operates bioleaching in two phases: In primary leaching the ore is leached in 8,5 - 9,5 m high 6,5 Mt heaps. After the primary phase the ore is reclaimed and stacked into secondary leaching.

The feed ore contains 5-15 % of pyrrhotite mineral which acts as an energy source when leached in the heap conditions. The pyrrhotite leaching dominates the beginning of the primary leaching. The oxidizing reaction is promoted by aeration and acidification of the ore. The reaction is vigorous and heats the heap shortly after the startup. Terrafame has utilized temperature sensing cables (DTS), which have been installed inside the heap along the stacking front. The cables have been in two or three layers and the cable measures the temperature in every 1 m. Results from different heaps are presented in the report. Temperatures up to 100 °C inside the heap are normal and in some areas also higher temperatures have been observed.

Microbial cell count and population structure has been analyzed in primary and secondary heaps. Samples have been taken from the discharge solutions of primary and secondary heaps. Sulfur and iron oxidizing bacteria were enumerated with the MPN-method. The cell count of iron oxidizing bacteria ranges between 0 and 10³ cfu/ml. Sulfur oxidizing bacteria tend to have higher numbers, staying mostly around 10³ and 10⁴ cfu/ml. In the secondary heap the corresponding numbers are 10³...10⁴ cfu/ml for both, iron and sulfur oxidizing bacteria. The microbial population was analyzed with DNA-fragment analysis and capillary electrophoresis. *A. ferrooxidans* was the only species that was consistently observed in both areas, whereas other species appear to prefer either location.

Therefore, it would appear that the secondary heap is more favorable for microbial growth in general. Temperature range and rapid changes in the conditions in primary leaching are the most probable reasons for the observed discrepancy.

Keywords: Terrafame, Nickel, Cobalt, bioheapleaching, pyrrhotite, temperature, DTS, microbiology, population, cell count, MPN, DNA fragment analysis, *Acidithiobacillus ferrooxidans, iron, sulfur*

Terrafame 1.

Terrafame operates a nickel mine in Sotkamo Northern Finland. The Terrafame deposit comprise two different polymetallic ore bodies hosted by a black schist, Kuusilampi and Kolmisoppi with 1550 million ton of classified resources. The mine district is 64 km2. The deposits are relatively easy to mine as an open pit.

Terrafame production process consists of open pit mining, four stage crushing, agglomeration, heap stacking and reclaiming, heap leaching in two stages and metals recovery. The production process is shown in Figure 1.

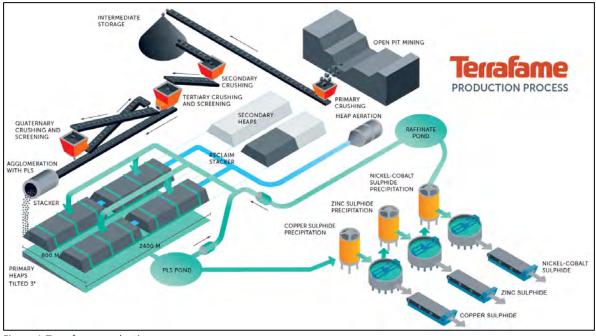


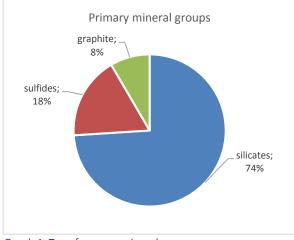
Figure 1 Terrafame production process

2. Ore

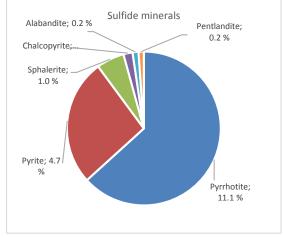
The ore is classified as a complex multi-metal black schist sulfide. The mineralogy and geochemistry of the deposit have been characterized in the literature.^{1,2}

The mineral composition of the sulfide component of the ore is 61.2 % pyrrhotite (FeS), 24.3 % pyrite (FeS₂), 5% pentlandite [(Fe_x/Ni_{9-x})₉S₈], 6.5 % alabandite (MnS) and 2.4 % chalcopyrite (CuFeS₂).

In the ore, pentlandite contains between 75-88 % of the contained nickel and pyrrhotite is the second most important mineral in terms of nickel content. Pyrite contains the main share (between 67-90 %) of contained cobalt while chalcopyrite is carrying copper and sphalerite zinc. The proportions of the minerals in the ore can be seen in Graphs 1 and 2.



Graph 1. Terrafame ore minerals



Graph 2. Terrafame sulfide minerals

3. Bioheapleaching process

Ore is stacked in a heap which is aerated and irrigated to oxidize sulfide minerals and dissolve valuable metals into solution as sulfates to be recovered by sulfide precipitation as separate metal sulfide products. The heaps are irrigated with an acidic (pH ~ 2.0) solution and aerated in order to enhance the chemical and biological oxidation process. Most of the acid required is generated in the heaps, as the mineral composition of the ore has the potential to produce more acid than is required to leach the sulfides. Metal-containing pregnant leach solution (PLS) is partly recirculated back to irrigate the primary pads and partly fed to the metals production plant, where the metals are extracted from the solution by precipitating them as sulfides.³

The ore is distinguished by a large quantity of sulfides, especially pyrrhotite (FerS₈), which dominates the chemistry during the early stages in leaching of valuable metals. A large amount of reductive sulfides, predominantly pyrrhotite, has to be leached before the valuable minerals start to react.⁴ This requires a significant degree of oxidation, which is achieved by feeding forced air through pipelines installed inside the heap. Minerals such as pyrrhotite and alabandite (MnS) will generate heat during the reaction.

4. Heat source

Sulfide oxidation generates heat and allows the Terrafame operation to continue heap leaching through the winter in Finland.

One of the reasons why small scale testing is not conventionally representative is the heat loss conditions which vary in a large heap significantly from a laboratory scale column. On the one hand, there is the significant insulation effect of the mass of a real heap and on the other, there is a significant cooling wind effect on the top of the heap, as can be seen in the graphs of the previous study.^{5,6}

Reactions

Iron and manganese oxidation reactions in main role (with reaction enthalpy):

Pyrrhotite oxidation

 $Fe_7S_8 + 3.5 O_2 + 7 H_2SO_4$ $-->7 FeSO_4 + 8 S + 7 H_2O$ -3132 kJ/mol $Fe_7S_8 + 14 O_2 + 0 H_2SO_4$ $-->7 FeSO_4 + S + 0 H_2O$ -6973 kJ/mol $Fe7S_8 + 15.5 O_2 + H_2O$ $-->7 FeSO_4 + H_2SO_4$ -7328 kJ/mol

Alabandite oxidation

MnS + 0.5 O₂ + H₂SO₄ --> MnSO₄ + S + H₂O - 324 kJ/mol

The potential heat generation in the Terrafame leaching heaps can be estimated to 70 - 140 W/t of ore derived from calculations presented in previous research.⁵ The calculation considers oxygen amount as limiting factor in the reactions. In the process the maximum amount of oxygen fed into the heaps is determined by the blower capacity.

This phenomena leads to an idea of temperature indicating leaching process state opening a possibility to follow up the leaching process more detailed.

5. Heap structure

A leaching heap is constructed in a field with bottom ground angle of ca. 3 degrees. One heap consists of three sections each of 350 meters wide and 400 m in length.

Drainage

The drainage structure constitutes of 110 outer diameter corrugated drainage pipes of ca. 350 m length every 5 meters delivering solution to a collection pipe and two sampling wells located 66 meters from each other. The drainage pipes are covered with a layer of finer gravel (12-32 mm) and ca. 20 cm of coarser gravel (12 -64 mm) is laid on top.

Ore

The feed material ore is crushed to p80 size of ca. 8 mm. The ore is agglomerated to ca. 3.5 %-w with PLS moisturizing. The agglomerated ore is stacked to 8,5 - 9,5 m high heaps.

Aeration

The aeration pipes of 110 mm outside diameter with perforation of 6 mm diameter in 2 m space are installed at three meter intervals in three layers. The aeration pipe installation scheme is to have an upper air pipe at height of 6 m from the heap bottom, the middle air pipe at height of 3,5 m from the heap bottom and the lower air pipe at 1 m height from the heap bottom.

Irrigation

The heap irrigation is managed with drip line irrigation. The irrigation grid is with 60 cm drop-todrop distance with designed irrigation rate of 5 L/m²h during summer and potential to rise to 7 L/m²h during winter. The sections are split into ten irrigation cells of ca. 40 m long and 330 m in width.

The heap structure is shown in Figures 2 and 3.

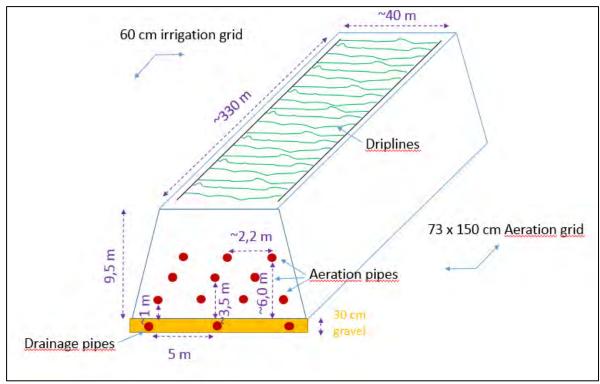


Figure 2. One irrigation cell

One heap consists of three sections with ten cells each.

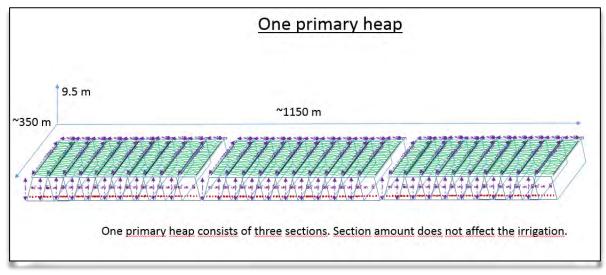


Figure 3. One primary leaching heap

6. Temperature measurement

Terrafame is using optical fibre cable (DTS cable, Distributed Temperature Sensor) for temperature sensing inside the heaps. The sampling resolution is 1 m.

DTS cable

The Distributed Temperature Sensor illuminates the glass core of the optical fibre with a laser pulse of 10 nanosecond duration (this corresponds to a 1 m pulse.) As the optical pulse propagates down the fibre, it undergoes scattering even in the absence of impurities and structural defects. Part of this scattered radiation is known as Raman scattering. Because this vibrational energy is a well-defined function of temperature, the ratio of the signals is also. It is this ratio, in conjunction with the time of flight of an optical pulse, which is used to determine the temperature of the fibre at a given point.⁷

Temperature measurement in summer 2018

Terrafame measured heap temperature profile inside a heap in summer 2018. The optical cable was made to optimal length for the heap structure. The idea was to measure the temperature in different heights inside the heap. Therefore, it was decided to loop the cable three times through the heap, which required 1200 m of cable.

The cable was assembled inside primary heap manually while the stacking was stopped for maintenance. The cable assembly in the heap is shown in Figure 4.

Reference measurement

Temperature measurement in outside conditions is always effected by natural temperature and therefore a reference temperature measurement is needed. In this case a reference measurement was assembled near the heap in a barrel of water. Both, the DTC cable and the reference probe (RBR Solo) are measuring the same temperature at that point, which can be used for calibration.

Aeration in heap

The temperature measurement was done in a heap where the aeration was installed in an experimental method. The aeration pipelines were assembled normally in three layers, but in the middle layer pipeline perforation was done only for half of the pipeline. Reason for this kind of assembly was to study the leaching performance of the heap. In the history it was observed that there is uneven behavior in heap temperature development when the blowers are situated only in one side of the heap. This structure is described in Figure 4.

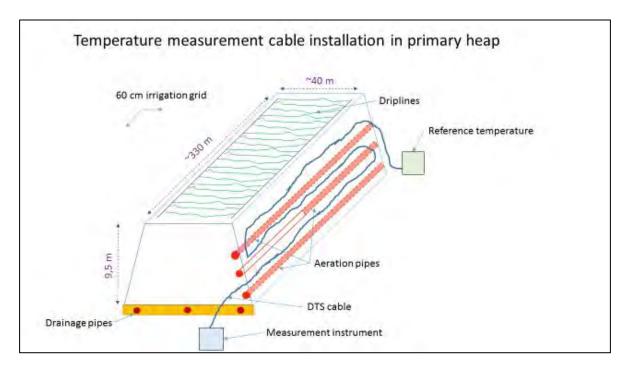


Figure 4. DTS cable inside the heap

7. Results

Temperature measurement was started in 17.5.2018 and continued until 25.7.2018. The measurement was stopped after a failure in the DTS cable most probably due to temperature increase over +185 °C in one slope of the heap. This phenomena occurs sometimes when the irrigation falls too low. However the data before the failure was taken out and the information has been analyzed and a temperature profile has been formed.

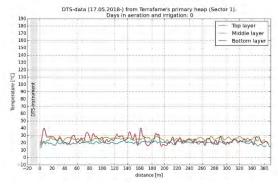
Temperature profile development

The development of the heap temperature can be seen in following Graphs 3-6.

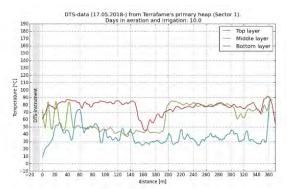
The cable was assembled in May and outside temperature is normally at level of 0 - +15 °C in Sotkamo area. However temperature inside the heap increases quite fast to 30 °C in all layers. This temperature increase is caused by spontaneous reactions without aeration or irrigation. This phenomena can be seen in Graph 3.

Effect of aeration into heap temperature

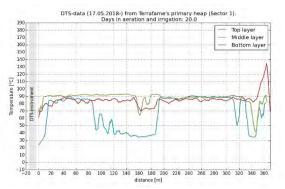
The oxidative heat generating reactions start already in the stacked heap but as the aeration is started the temperature starts rising faster. The point where the aeration starts inside the heap is easily observed in the temperature profile. In a few days the temperature rises up to 80 °C in the bottom layer and in the aerated part of the middle layer. In the Graphs 4. and 5. it can be seen that the heap is heating somewhat uneven and faster from the side where the aeration input is located. The delay thorough the heap distance is up to thirty days in most of the heap area. In this case the temperature is rising also in the second half of the heap due to the experimental set up with extra aeration in the middle layer. This observation will help planning of the aeration in the heaps.



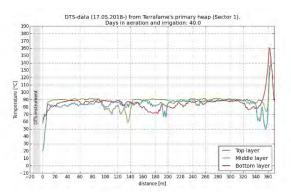
Graph 3. Temperature development inside a heap without forced aeration.



Graph 4. Temperature development inside a heap, 10 days in aeration.



Graph 5. Temperature development inside a heap, 20 days in aeration.



Graph 6. Temperature development inside a heap, 40 days in aeration.

The temperature seems to be limited to 80 $^{\circ}$ C at the normal irrigation area of the heap, presumably due to vapor pressure increasing in the solution and evaporation taking over. However in the fringe areas the irrigation is quite challenging and the air – solution ratio rises high. Also the natural aeration due to wind is oxidizing the slopes and higher temperatures can be observed which can be seen in Graphs 5 and 6.

As the temperatures inside the primary heap are quite high the conditions for microbial activity may not be optimal. At least the naturally active microbial populations found in the orebody are unlikely to be accustomed to the observed temperature range.

8. Microbiology in elevated temperatures

Samples were taken from the discharge solutions of primary and secondary heaps and the microbial population was analyzed with TRFLP (Terminal restriction fragment length polymorphism) during the past two years. As a part of routine analyzis carried out by Terrafame, the temperature of the liquid has also been measured and sulfur and iron oxidizing bacteria have

also been enumerated with the MPN method since 2008.

Relative abundance analyzed with TRFLP

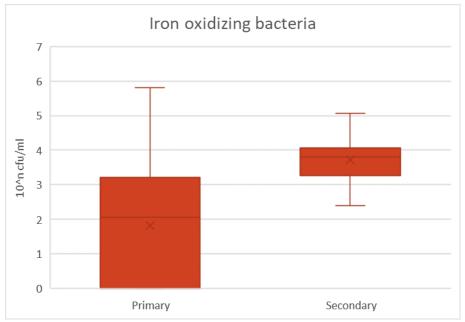
TRFLP is a semi-quantitative method for assaying the relative abundance of copies of DNA. Usually it's used to identify a family or genus, but in certain cases it can also identify an individual species based on the length of a certain DNA fragment. Unlike qPCR, it is particularly well suited for broad spectrum analyzis of an unknown population.

Acidithiobacillus ferrooxidans was the only species that was consistently observed in the discharge solution of primary and secondary heaps. However, two species seemed to show a preference for either the primary or secondary heap. In the primary heap, it was reasonably common to detect *A. caldus*, whereas in *A. thiooxidans* would appear to prefer the conditions of the secondary heap. Other species were also detected, but those detections were not considered consistent or reliable enough.

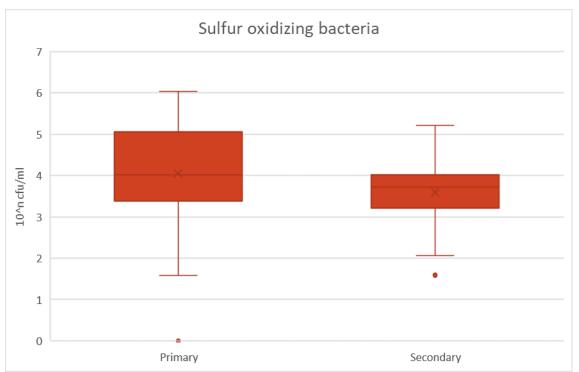
MPN, Sulfur and iron oxidizing microbes

Cell count of sulfur and iron oxidizing bacteria in the discharge solution of the heaps can be seen in Graphs 7 and 8. The samples were analyzed with the MPN method, which is able to enumerate only a certain fraction of all vegetative cells, whereas a corresponding qPCR result would be at least an order of magnitude higher. Sulfur oxidizing bacteria were incubated at 37 °C, and iron oxidizing bacteria at 25 °C. It is important to note that, the relatively low incubation temperature may limit the growth of thermophilic bacteria in the assay, which would further lower the perceived cell count.

In the primary heap, the cell count of iron oxidizing bacteria ranges between generally <11 and 103 cfu/ml, whereas sulfur oxidizing bacteria tend to have higher numbers, staying mostly between 103 and 105 cfu/ml. In the secondary heap the corresponding numbers are 103...104 cfu/ml for both, iron and sulfur oxidizing bacteria.



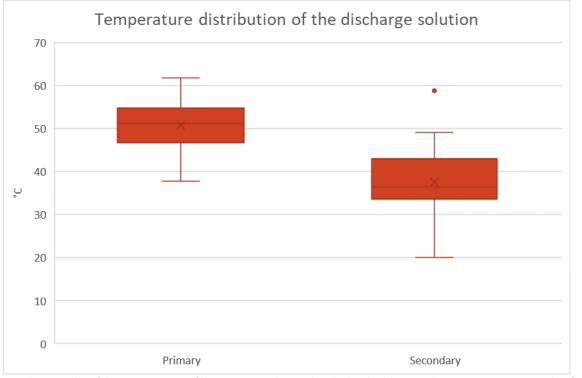
Graph 7. Concentration of iron oxisizing bacteria analyzied with the MPN method from the discharge solution. Sample size is 375 analyzis in the primary and 52 in the secondary heap. All samples were taken during 2018 and they represent every month of the year.



Graph 8. Concentration of sulfur oxisizing bacteria analyzied with the MPN method from the discharge solution. Sample size is 205 analyzis in the primary and 39 in the secondary heap. All samples were taken during 2018 and they represent every month of the year.

Temperature of the discharge solution

Due to the exothermic nature of pyrrhotite leaching, the temperature of the discharge solution of the primary heaps remains high above ambient throughout the year, but only rarely exceeds 60 °C (Graph 9.). The observed temperature difference between primary and secondary heaps is caused by the difference in pyrrhotite content of the ore. Because of the lack of pyrrhotite in the secondary heaps, the thermal output per unit of mass is considerably smaller, which causes a noticeable temperature difference between the two process phases.



Graph 9. Box plot of the temperatures of the primary and secondary bioleaching heaps. All measurements were taken from the discharge solution between 1.3.2017-1.3.2019.

9. Discussion

Temperature profile development in the heap indicates the percolation of the heap i.e. aeration and irrigation relationship. When increasing aeration the temperature development is faster up to certain level. When temperature increases to 80 °C the water evaporation starts limiting the increase.

In the history the heap aeration has been constructed with the same aeration grid in all the heap area. This measurement shows the uneven distribution of air when the feed is from one side only. The different layers of the heap are heated differently and for some areas the temperature increase takes up to thirty days. In the heap leaching process management of the aeration and irrigation ratio is important factor to be emphasized.

The tolerable temperature range of most species identified in TRFLP was significantly higher than the observed temperature of the discharge solution of the heaps. Therefore, it is likely that the microbes colonize hot areas of the heap and are later detached and carried to different areas by the irrigation solution. DTS results have confirmed that the temperature profile is temporally and spacially heterogenous, which would allow cooler liquid to contain species that are unable to effectively multiply in such temperatures.

Based on what is known about the tolerable temperature range of the observed species, it would appear that as long as the temperature remains between 20...60 °C, we could expect to see at least 10 different species colonizing the ore and process solutions. At around 80 °C, it can be expected that at least 3 species will be able to multiply efficiently.

Some identified bacterial species appear to prefer either the primary or secondary heap environment, which is probably caused by their preferred temperature, pH and other conditions. Based on the MPN results, it can be observed that the conditions of the primary leaching area are generally more favorable for sulfur, rather than, iron oxidizing bacteria. In the discharge solution of the secondary heap it has been observed that the concentration of iron and sulfur oxidizing bacteria are within the same order of magnitude. The cell concentration of iron oxidizing bacteria of the secondary heaps tends to be nearly three orders of magnitude greater than in the primary heaps.

10. Conclusions

The boreal climate sets up challenging environment for the process control of a heap leaching operation. The observed temperature profile inside the heap shows the reactivity of the ore when suitable conditions and reactants are available.

In this work a new heap aeration structure was tested and the results show us the importance of the aeration structure for the heap performance. This will help constructing aeration in a heap more detailed thus improving the heap performance in the future.

The observed difference between the microbial population of the primary and secondary heaps is most probably caused by their temperature difference. When the pyrrhotite content decreases there will no longer be rapid changes in temperature and the leaching process will also operate in lower temperatures. Due to these factors, the environment which the microbes inhabit, will be more favorable for different population structures in different process phases.

11. Research in the future

The tests done in the process heaps show clear relationship between heap temperature and aeration. These tests shall be continued to learn more about the heap behavior during the process. New installations are planned and also new procedures for the cable installations are to be developed.

Microbial assay methods will be further refined and an investigation on using NGS is ongoing.

12. Acknowledgments

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