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# RESEARCH CONCERNING THE OXIDATION DEGREE OF THE SULPHIDIC TAILINGS FROM THE NOVĂȚ TAILINGS STORAGE FACILITY

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**Abstact:** In this paper are presented the results of the research concerning the weathering degree of the sulphidic rich tailings deposited on the Novăț tailings storage facility (Novăț TSF) Maramureş county and considerations are made about the evolution of the acid drainage process in future. The stage of the oxidation differs as a result of the phenomena of bacterial catalysed oxidation, which occurred depending on the age of the deposited tailings and on the mineralogical, physical, chemical and biological factors. The phenomena of acid mine drainage, once begun will continue, which will lead to physical-chemical and mechanical changes of the tailings, with negative effects over the aquatic life downstream of the Novăț TSF.

**Key words:** sulphidic mine waste, Novăț tailings storage facility, biosolubilization, acid mine drainage, *Acidithiobacillus.*

## 1. INTRODUCTION

In the deposits of mining tailings take place, naturally, phenomena of bio-catalyzed oxidation of the sulphidic minerals. These phenomena take place under the catalyzing influence of the acidophilic iron- and sulphur-oxidizing bacteria, especially those from the *Acidithiobacillus*, species, naturally present in these sites, which determines the apparition of physical and chemical transformations in mine waste deposits, known as bacterial leaching process.
The bacterial leaching process consists in the oxidation of the iron and sulphur existing in metal sulfides (eg. pyrite) and the formation of sulfuric acid and metal solubilization. As a result of the circulation of rain waters or of the technological

waters among the mine waste deposits, solutions with an acid pH and high metal concentrations occur, especially of iron, copper and zinc, known in the literature under the name of acid mine waters or AMD - Acid Mine Drainage (Bond et al. 2000).
The natural phenomena of bacterial leaching of the metal sulpfides from the mining tailings are characteristic for all mining regions in which complex non-ferrous ores are being mined. They can take place over tens/hundreds years and can change the quality of the environment factors over a long period thus determining severe ecological disequilibrium (Jelea et al., 1998; Cosma et al., 1999; Jozsa et al., 1999; Jelea et al., 2001; Schippers et al., 2000; Jozsa et al., 2001; Schippers et al., 2001).

The analyses carried out after monitoring these deposits demonstrate that the phenomena of bio leaching accelerate in time, which has as a consequence the worsening of the impact on the environmental factors. The phenomena of bacterial leaching lead to the pollution of effluent seepage waters resulted from rainfalls/infiltrations which percolate the tailing mass and then reach the effluents with a high pH and with a charge of heavy metals.

The presence of the tailing deposits conduct not only the water pollution but also the air pollution, having as immediate effect the transformation, the degradation or the total destruction of the structures of the biocenoses from the adjacent zones, affecting both the terrestrial and aquatic flora and fauna.

Although the consequences of the acid drainage date back to the Roman era, the first reports about the influence of the bacteria in the process of leaching metals from sulfides were presented by Rudolfs and Helbronner in 1992 and only in 1947 did the American researchers Colmer and Hinkle (1947) isolate from an acid mine water and characterize the acidophilous species *Thiobacillus ferrooxidans* (Temple & Colmer, 1951) and *Thiobacillus thiooxidans* (Temple & Delchamps, 1953). The acidophilic species of the *Thiobacillus* genus have been recently re-classified in the *Acidithiobacillus* genus (Bond et al. 2000; Kelly & Wood, 2000).

The research begun at the middle of the last century (Temple and Colmer, 1951; Temple and Delchamps, 1953), were centered on improving the process of bacterial leaching for the industrial application of metal recovery from the poor ores. This leads to the through knowledge of bacterial oxidation of the mineral sulpfides, (Sampson et al., 2000; Edwards et al., 2001; Gehrke et al., 2001; Sand et al., 2001; Bevilaqua et al., 2002; Rawlings, 2002; Rimstidt & Vaughan, 2003).

For a long time, no adequate measures have been taken in the mining industry to control this phenomenon. Therefore the tailing deposits have been built without having in view the transformations that will occur in time. Nowadays, most of the deposits of mining and flotation tailings with sulfide contents older than 10 – 20 years undergo strong phenomena of degradation by bacterial oxidation.

The measures for mitigation of the acid mining draining have, in most cases, addressed to the consequences of these phenomena and not to the causes that generate them. Ignoring the phenomena of acid drainage lead to applying some inadequate measures for solving the environmental problems on the tailings resulted from mining the non-ferrous ores: planting some trees and bushes directly in the tailing or with

borrowed soil, building some small coast fences, etc., all these measures having only short-term favorable effects, especially for eliminating the hydraulic and wind erosion. But these measures are inefficient for preventing the oxidation and the acidulation of the deposited tailings.

The bio-catalyzed oxidation of the sub-layers determines transformations in the water permeability of the tailings, grain-size and cohesion transformation of the particles, and the formation of sub-layers with different filtration capacities, which cause sliding phenomena that lead to the decrease of the deposit stability and to the increase of the risk of some accidents with impact on the environment.

The most frequently met bacteria in the mining deposits are the species in the *Acidithiobacillus* genus. Despite having the same form and structure, the acidophilic bacterial species of the genus *Acidithiobacillus* are differentiated by some metabolic characteristics, such as: oxidation of the inorganic sulphur minerals, capacity of iron oxidation, formation of final products, adaptation to heterotrophy, anaerobic increase in the presence of the nitrate, the pH domain in which they live, etc.

Besides the bacteria from the *Acidithiobacillus* genus, in the same environment or in similar environments live other bacteria as well, which have a more or less important role in leaching metals and belong to other genus: *Leptospirillum*, *Sulfolobus, Thermothrix, Acidianus, Metallogenium, Metallosphaera, Sulfurisphaera*, *Sulfobacillus*), *Acidiphilium, Acidimicrobium*, *Ferroplasma*, *Acidisphaera*, *Ferromicrobium.*

The particular necessities of the bacterial species as compared to the mineral composition of the tailings and to the chemical form in which the chemical elements exist, to the environment pH and also to the physical factors (humidity, temperature, oxygen) and biological factors (resistance to the acid concentration, or to the metallic ion concentrations, etc.) leads to their installation or not, in the different waste layers. Depending on the physical and chemical characteristics of the mine waste layers (the mine wastes deposited at different depth levels in the rock dumps and settling ponds) at a certain moment; certain bacterial populations can be found in them.

By their metabolic activity the bacteria lead to physical and chemical transformations, specific as individual layer level, and the determining one is the layer acidulation. The decrease of the pH under the resistance limit for the bacterial species that has initiated the phenomenon has as effect its elimination, the new environment conditions becoming favorable to the installation of other more acidophilic bacterial species. Therefore, depending on the evolution of the physical and chemical parameters a succession of settling of the bacterial species takes place in the tailing, from the less acidophilic species (in the stage when the bacterial leach process is initiated) to more and more acidophilic species.

### 1.1. Objectives

Study of the oxidation degree of the sulphidic tailings deposited on the settling pond.

Considerations about the evolution of the acid mine drainage in the settling pond.

### 1.2. Object of study

The Novăţ tailings storage facility placed in an isolated mountainous region, north of the Borşa town, being placed on the administrative territory of the town Vişeul de Sus, Maramureş County. The access in the zone is carried out underground (Baia Borşa gallery), or on the industrial road Baia Borşa-Novăţ, which is 16 km long. The Novăţ TSF is located in the superior basin of the Novăţ river, upstream the confluence with the Ursului Stream.

It is a valley mine tailings dam and it ensures the settling of the flotation tailings and of the technological waters from the Baia Borşa Flotation.

## 2. MATERIALS AND METHODS

In order to carry out the analyses concerning the oxidation degree of the sulphidic tailings deposited on the settling pond the mine waste material has been sampled by carrying out four drillings: at the toe, in the middle and in the crest zone of the main embankment, as well as in the pond beach.

The chemical analyses of the tailing samples have been carried out for the chemical characterization of the tailings (table 1 and figures 1-4) as well as the chemical analyses of the watery extract 1/5 of the tailings (table 2).

The number of neutrophilic and acidophilic iron- and sulfur-oxidizing bacteria from the tailing samples has been determined for the microbiological characterization of the tailings (table 3).

## 3. RESULTS AND DISCUSSIONS

***Drilling 1 – at the toe of the dam***

The samples taken at the toe of the embankment, down to the depth of 3.0 m are very acid, the pH values being 2.6 – 4.4. The acidity of the samples decreases in depths.

The leaching process, mediated by the bacteria, lead to the decrease content of heavy metals in tailings: in the more acid samples from the superficial layers where the initial process of acid drainage occurred initially, the contents in heavy metals of the tailings are low. One notices an obvious leaching for Cu, Zn, Mn, Ca and Mg (table 1 and figure 1).

The acid pH and the leaching activity of the elements, mediated by the bacteria, leads to the leaching of the elements in the acid solutions that percolate through tailings; therefore in the solutions from the tailings are leached in different proportions Cu, Pb, Zn, Mn, Fe, Ca, Mg, depending both on the contents in elements of the tailings and on the pH of the solutions (table 2).

The presence in large number of the acidophilic iron- and sulfur-oxidant bacteria from the *Acidithiobacillus* genus will determine the maintenance and the amplification of the acid drainage phenomenon in this zone of the pond (table 3). The absence of the neutrophilic bacteria from the *Thiomonas* genus is determined by their disappearance as a result of the increased acidity.

**Table 1.** Chemical composition of the tailings samples

| Sample ID | Location of sample | Depth (cm) | Chemical composition of tailings samples (mg/kg of tailing d.s.) | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | Cu | Pb | Zn | Mn | Fe | Ca | Mg | $S^t$ | $SO_4^{2-}$ |
| 1. | Drilling 1 – at the toe of the dam | 0-20 | 1,125 | 2,000 | 1,875 | 65 | 108,750 | 6,750 | 2,625 | 137,097 | 22,633 |
| 2. | | 40-60 | 1,375 | 2,500 | 2,500 | 75 | 110,000 | 6,250 | 3,125 | 141,120 | 21,686 |
| 3. | | 60-80 | 1,500 | 2,380 | 3,500 | 100 | 107,500 | 8,375 | 3,250 | 134,400 | 20,164 |
| 4. | | 80-100 | 1,750 | 2,600 | 4,750 | 90 | 140,000 | 8,125 | 3,000 | 208,640 | 19,546 |
| 5. | | 150-200 | 2,000 | 2,030 | 5,125 | 125 | 97,500 | 8,375 | 3,875 | 137,600 | 24,690 |
| 6. | | 250-300 | 2,250 | 2,000 | 7,000 | 400 | 90,000 | 8,375 | 6,250 | 107,520 | 11,357 |
| 7. | Drilling 2 – in the middle of the dam | 0-20 | 625 | 1,720 | 270 | 65 | 105,000 | 6,750 | 2,750 | 105,600 | 31,727 |
| 8. | | 40-60 | 750 | 2,000 | 1,500 | 75 | 106,250 | 9,250 | 2,500 | 104,960 | 32,056 |
| 9. | | 60-80 | 750 | 1,880 | 2,500 | 110 | 81,250 | 9,125 | 3,125 | 102,400 | 32,920 |
| 10. | | 100-150 | 850 | 1,800 | 3,000 | 100 | 102,500 | 8,125 | 3,625 | 112,000 | 27,694 |
| 11. | | 250-300 | 1,375 | 1,700 | 4,750 | 150 | 105,000 | 7,500 | 5,000 | 131,200 | 24,402 |
| 12. | | 470-500 | 1,250 | 1,400 | 6,000 | 610 | 97,500 | 10,000 | 6,250 | 72,700 | 19,217 |
| 13. | Drilling 3 – the crest zone of the dam | 0-20 | 1,000 | 3,100 | 375 | 30 | 115,000 | 7,500 | 1,500 | 179,200 | 38,681 |
| 14. | | 20-40 | 750 | 2,450 | 375 | 50 | 117,500 | 8,375 | 1,875 | 160,000 | 39,792 |
| 15. | | 60-80 | 625 | 2,500 | 1,250 | 60 | 120,000 | 8,750 | 2,500 | 164,500 | 35,553 |
| 16. | | 100-150 | 1,375 | 2,000 | 3,500 | 100 | 100,000 | 10,500 | 3,375 | 128,000 | 30,328 |
| 17. | | 250-300 | 1,250 | 1,200 | 4,500 | 410 | 78,750 | 7,750 | 4,875 | 108,800 | 14,073 |
| 18. | | 450-500 | 2,375 | 3,200 | 7,250 | 260 | 177,500 | 6,250 | 4,000 | 342,400 | 10,041 |
| 19. | Drilling 4 – beach, 50 m from the dam crest | 5-20 | 2,500 | 4,200 | 10,000 | 170 | 195,000 | 4,750 | 3,375 | 329,600 | 6,954 |
| 20. | | 60-80 | 2,000 | 2,950 | 8,000 | 250 | 132,500 | 4,500 | 6,375 | 201,600 | 9,217 |
| 21. | | 150-200 | 2,000 | 2,400 | 7,500 | 385 | 132,500 | 9,500 | 6,875 | 243,200 | 10,452 |

**Table 2.** Chemical composition of watery extract of the tailings samples

| Sample ID | Place of sample | Depth (cm) | Chemical composition of watery extract 1/5 of the flotation tailing sample (ppm tailing d.s.) | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|
| | | | Cu | Pb | Zn | Mn | Fe | Ca | Mg | $SO_4^{2-}$ |
| 1. | Drilling 1 – at the toe of the dam | 0-20 | 14 | 14 | 345 | 4 | 1,150 | 4,644 | 57 | 13,284 |
| 2. | | 40-60 | 12 | 11 | 100 | 0.3 | 860 | 4,643 | 72 | 11,946 |
| 3. | | 60-80 | 13 | 9 | 57 | 1 | 154 | 4,612 | 43 | 9,480 |
| 4. | | 80-100 | 42 | 12 | 144 | 6 | 866 | 4,260 | 115 | 11,688 |
| 5. | | 150-200 | 58 | 12 | 300 | 16 | 1,169 | 4,255 | 248 | 14,576 |
| 6. | | 250-300 | 5 | 18 | 619 | 104 | 1,192 | 1,699 | 477 | 9,694 |
| 7. | Drilling 2 – in the middle of the dam | 0-20 | 8 | 6 | 49 | 4 | 1,417 | 4,369 | 126 | 16,595 |
| 8. | | 40-60 | 14 | 9 | 236 | 4 | 1,802 | 4,297 | 152 | 18,581 |
| 9. | | 60-80 | 28 | 11 | 126 | 3 | 716 | 4,240 | 84 | 11,456 |
| 10. | | 100-150 | 41 | 13 | 475 | 5 | 1,356 | 3,716 | 122 | 16,515 |
| 11. | | 250-300 | 120 | 10 | 127 | 9 | 422 | 3,714 | 127 | 12,600 |
| 12. | | 470-500 | < 0.3 | 6 | 1,418 | 163 | <0,6 | 3,801 | 482 | 12,952 |
| 13. | Drilling 3 – the crest zone of the dam | 0-20 | 7 | 10 | 25 | 2 | 1,077 | 4,297 | 43 | 14,145 |
| 14. | | 20-40 | 12 | 6 | 37 | 4 | 1,893 | 3,781 | 126 | 18,068 |
| 15. | | 60-80 | 12 | 6 | 40 | 4 | 1,458 | 3,907 | 139 | 15,691 |
| 16. | | 100-150 | 54 | 8 | 177 | 2 | 343 | 3,951 | 54 | 10,402 |
| 17. | | 250-300 | 2 | 3 | 462 | 35 | < 0.6 | 2,898 | 177 | 8,054 |
| 18. | | 450-500 | < 0.3 | 9 | 309 | 40 | < 0.6 | 1,480 | 229 | 5,317 |
| 19. | Drilling 4 – beach, 50 m from the dam crest | 5-20 | < 0.3 | 5 | 82 | 6 | < 0.6 | 300 | 55 | 1,254 |
| 20. | | 60-80 | < 0.3 | 6 | 164 | 18 | < 0.6 | 476 | 134 | 2,505 |
| 21. | | 150-200 | < 0.3 | < 0.6 | 44 | 24 | < 0.6 | 710 | 30 | 1,628 |

***Drilling 2 – in the middle of the dam***

In the D2 drilling location, as well as in the F1 drilling location, the samples are very acid, the pH being 2.5 – 3.5 down to the depth of 4.50 m; when the depth is over 4.70 m, the pH increases to the value of 6. Leaching of the elements in the tailing occurs as a result of the high acidity (table 1 and figure 2).

Like in case of the drilling at the toe of the embankment, the presence of the acidophilic iron- and sulfur-oxidant bacteria (table 3) will amplify the phenomenon of acid drainage in this zone of the tailings pond. The sulphur and iron content in the mine tailing is high, which will determine the maintenance of the bacterial processes of acid mine drainage for a long time.

***Drilling 3 – the crest zone of the dam***

The acid sub-layer, with high activity of acid drainage is from 0 to 2.50 m. But the oxidation process is also present in the tailings from 5.0 m, where the pH is 6. In these samples there is a large number of iron- and sulfur-oxidant bacteria, premises for some intense activities that go on (table 3).

***Table 3.*** Bacterial density from the tailings samples

| Sample ID | Location of sample | Depth (cm) | Water content (%) | pH | Bacterial density (bact.x$10^2$/g of tailing d.s.) | | |
|---|---|---|---|---|---|---|---|
| | | | | | *At. ferro-oxidans* | *At. thio-oxidans* | *Th.intermedia Th.novellus* |
| 1. | Drilling 1 – at the toe of the dam | 0-20 | 13 | 2.6 | 5,270 | 275,000 | 0 |
| 2. | | 40-60 | 13 | 2.7 | 1,230 | 1,230 | 0 |
| 3. | | 60-80 | 12 | 3.1 | 5,080 | 2,200 | 0 |
| 4. | | 80-100 | 13 | 3.2 | 1,220 | 8,340 | 0 |
| 5. | | 150-200 | 14 | 3.4 | 8,740 | 3,150 | 0 |
| 6. | | 250-300 | 16 | 4.4 | 2.83 | 2,360 | 0 |
| 7. | Drilling 2 – in the middle of the dam | 0-20 | 11 | 2.5 | 514 | 2,240 | 0 |
| 8. | | 40-60 | 10 | 2.5 | 29.3 | 2,920 | 0 |
| 9. | | 60-80 | 11 | 2.9 | 26,700 | 267,000 | 0 |
| 10. | | 100-150 | 8 | 2.6 | 256 | 3,630 | 0 |
| 11. | | 250-300 | 11 | 3.5 | 1,220 | 13,300 | 0 |
| 12. | | 470-500 | 12 | 6.0 | 1.2 | 23,800 | 0 |
| 13. | Drilling 3 – the crest zone of the dam | 0-20 | 13 | 2.6 | 1,200 | 262,000 | 0 |
| 14. | | 20-40 | 11 | 2.5 | 51.4 | 268,000 | 0 |
| 15. | | 60-80 | 10 | 2.6 | 80.3 | 4,750 | 0 |
| 16. | | 100-150 | 8 | 2.9 | 49,800 | 260,000 | 0 |
| 17. | | 250-300 | 8 | 6.1 | 11.9 | 2,600 | 0 |
| 18. | | 450-500 | 7 | 6.0 | 0.1 | 616 | 0 |
| 19. | Drilling 4 – beach | 5-20 | 8 | 5.8 | 0 | 3,040 | 25.2 |
| 20. | | 60-80 | 16 | 6.0 | 0.5 | 696 | 2.8 |
| 21. | | 150-200 | 16 | 7.2 | 0 | 1,830 | 126 |

***Drilling 4 – beach, 50 m from the dam crest***

The bio-oxidation process is also present on the surface of the pond beach, in the zones that are not covered by the water table.

The pH of the samples is mild-acid down to the depth of 1.50 m, the values being 5.8 – 6.

The number of iron-oxidant bacteria is low, but the presence of the neutrophilic bacteria and the occurrence of the sulfur-oxidant ones are the premises of some acceleration of the phenomena of acid drainage.

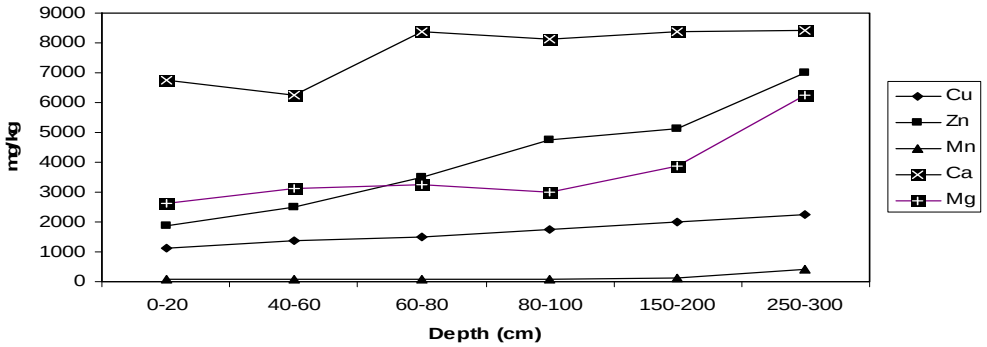


Fig. 1. Chemical composition at tailings samples – drilling 1

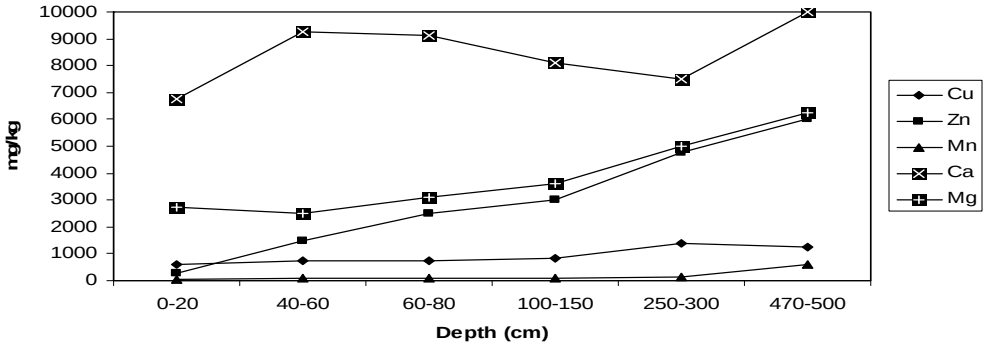


Fig. 2. Chemical composition at tailings samples – drilling 2

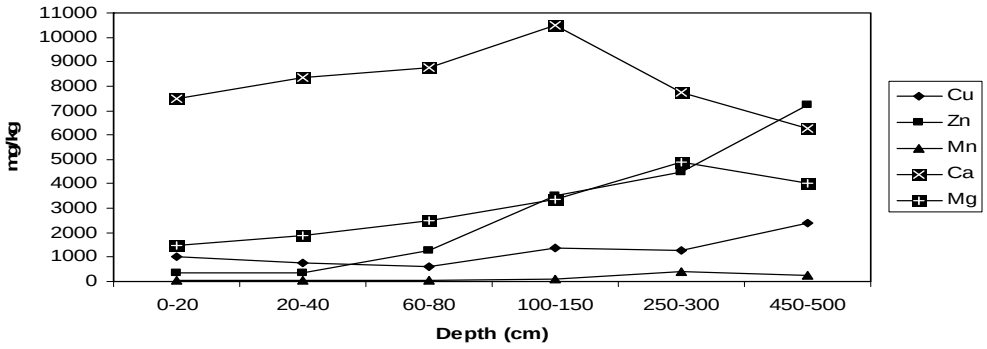


Fig. 3. Chemical composition at tailings samples – drilling 3

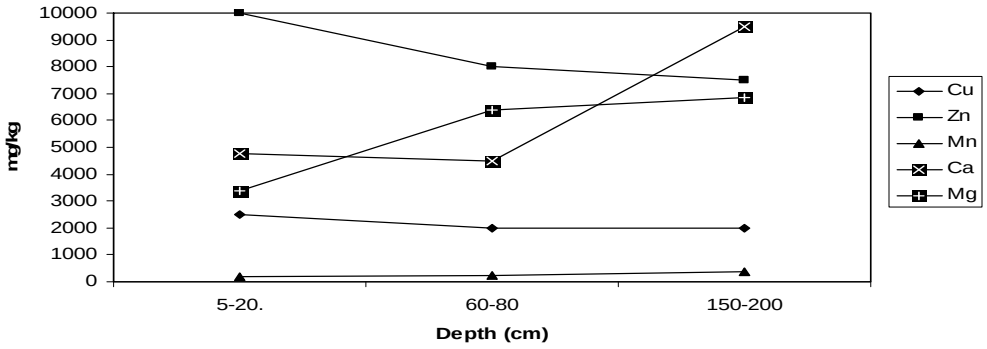


Fig. 4. Chemical composition at tailings samples – drilling 4

## CONCLUSIONS

The study of the physical, chemical and microbiological parameters carried out for the tailing samples taken from the dam and beach of the Novăț mine tailings pond leads to the following conclusions:

- The oxidation processes of the tailings have been pointed out in all four drillings done in the dam and the beach of the pond;
- The depth down to which the oxidation processes have determined the acidulation are different in the three drillings carried out in the pond dam:
  - Over the whole depth of the drilling D1 at the toe of the embankment (0 – 3.0 m)
  - Down to 3.0 m in the drilling D2 in the middle of the dam;
  - Down the 1.5 cm in the drilling D3 in the crest zone of the dam.
- In the beach of the dam, in the superficial layer, the oxidative processes are in an incipient stage, reaching 20 cm in depth;
- The phenomena of acid drainage have lead to the decrease of elements content, especially that of copper, zinc, manganese, magnesium. The calcium remains in the tailing, in an insoluble form of calcium sulfate;
- The contents of elements from the watery extracts are not proportional to the elements contents of the tailings or to their pH. These contents are influenced by many factors:
  - -Migration of the solutions determined by the climatic conditions (the ascent of the solutions determined by evaporation or their dilution by the rain waters);
  - Leaching or precipitation of elements at different pH values of the solutions;
  - Precipitation of the ions on the alkaline surfaces of the tailings grains;
- In the composition of the watery extracts, similarly to the mine waters, the metal with the biggest concentration is lead, followed by zinc and copper;
- The presence of the bacterial species in the tailings and their number has been influenced by the pH of the tailings and reflect their oxidation stage;
  - The species *At.ferrooxidans* is present in a large number in the media

with the pH between 2.5 – 3.5 (best conditions for this species);

- The species *At.thiooxidans* is present in a large number even in the samples with neutral pH, since these bacteria live at pH between 1.0 – 7.0;

The presence of the species *Thiomonas intermedia and T.novellus* only in the samples with mild alkaline and neutral pH from the pond beach indicate an incipient stage of the oxidation processes – the best pH for these bacteria is 6.7; the absence of these species from the acid samples taken from the pond dam is determined by their disappearance as a result of the increase of the acidity under the influence of the acidophilous species from the *Acidithiobacillus* species.

The oxidative processes in the pond dam slope are in an advanced stage. They will continue leading to the weathering of the deposited mine tailings.

The oxidative processes in the pond beach are initiated by the neutrophilous species. The presence of the *At.thiooxidans* species is the premise of the installation in future of the acidophilous species *At.ferrooxidans* which will determine the acceleration of the oxidative phenomena in the tailing from the pond beach.

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