

RESEARCH PAPER

Project of green recycling and disposal of spent copper heaps in a closed ecosystem

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ABSTRACT

The article presents the results of an environmental experiment conducted under the green utilisation project for mining waste from hydrometallurgical copper production. The primary objective of this project was to investigate the feasibility of safely conserving spent copper ore through the creation of soil and vegetation layers. The ore, after sulfuric acid leaching using iron-oxidising bacteria, which contributed to increased copper recovery and reduced sulfuric acid consumption, was used as the object of research. When more than 80% of copper was extracted from the ore, and the pH of the wastewater was subsequently increased from 2.0 to 4.0, a soil layer was applied to the surface of the mineral raw materials, and seeds of acidophilic herbs were planted. Subsequent observations of the plants over several months revealed their stable growth, the formation of a root system in the soil layer, and the capture of part of the ore dump layer. Additionally, there was a gradual decrease in wastewater acidity to a pH range of 6.0-6.5.

Keywords: mining waste, hydrometallurgy, copper heaps, ecological utilisation, landscaping

INTRODUCTION

The current state of ore deposits is characterised by a significant accumulation of mineral waste in the dumps, represented by off-balance ores from previous mining operations. In some cases, the dumps obtained during the processing of mineral raw materials using methods and technologies from the second half of the 20th century can significantly exceed the ores processed by factories at present in terms of the content of valuable metals [1-3]. Hydrometallurgical methods have been intensively developed to incorporate these raw materials into production, as they are significantly cheaper and more environmentally friendly compared to traditional pyrometallurgical melting technologies. At the same time, hydrometallurgical technologies also require compliance with important environmental protection measures, despite their ecological advantages over pyrometallurgy. In particular, the location of the dump and streams should prevent the ingress of sulfuric acid solutions into groundwater and soil, which is crucial for the hydrometallurgical technology of copper production. The environmental safety of hydrometallurgical copper production is achieved using sealed geomembranes in technological channels and ponds, the installation of monitoring wells to analyse groundwater around the perimeter of the deposit, and the recirculation of liquid extraction streams, thereby eliminating the need to dispose of waste [4].

However, the question arises about the environmentally safe disposal of waste materials at the final stage of heap development, after the maximum possible copper extraction has been achieved. While the spent streams of productive copper solutions can be utilised in other areas of hydrometallurgical production or used as a source of iron(III) sulfate production, the disposal of the depleted ore mass from the dump necessitates the adoption of measures to mitigate environmental risks. Simple storage of this type of waste or burying it in the ground is an unacceptable ecological solution, as it leads to pollution of the surrounding soil and groundwater, as well as the desertification of the area. It is impossible to consider spent copper ore as a raw material for construction and road materials, as well as certain types of mining waste, due to the chemical composition and structural changes that occur in minerals after sulfuric acid leaching. In this regard, there is a need to find a set of environmental protection methods for the disposal of hydrometallurgical waste [5].

The materials presented at the Mining in the Era of Green Solutions conference propose various rational sets of preventive measures to reduce environmental risks to an acceptable level, as well as study results on the impact of mine emissions on regional vegetation, including the development of copper ore deposits [1]. Often, artificial mineral formations can also be a potential source of heavy metal pollution. It contributes to their transfer to adjacent landscapes combined with the effects of acid precipitation, accompanying water, and

Aeolian flows [2]. The gradual natural leaching processes of these landfills, leading to the gradual spread of heavy metals into the environment, once again confirm the need and expediency of involving these mineral raw materials in integrated rational processing, followed by environmentally sound disposal. Thus, studies on the neutralisation of copper and other heavy metals using geopolymers have noted the high toxicity and carcinogenicity of these metals, as well as their tendency to accumulate in environmental components at levels that are often significantly higher than pollution rates, even compared to organic compounds [3]. Thus, hydrometallurgical processing of waste ores can be considered as the primary stage of mining waste disposal from off-balance ores in compliance with all environmental protection measures. Modern hydrometallurgy is recognised as the only real possibility for a more rational use of the copper deposit resources [4].

The next stage of environmental disposal of the waste dump is required after maximum extraction of copper and some other metals by sulfuric acid leaching. The equipment at the dump site features a system of monitoring wells, channels, and ponds with a geomembrane coating, allowing for the gradual stabilisation of acid and salt residues within an isolated ecosystem. The regulating process for the residual acidity and water-salt balance in the waste heap may also include the use of a previously equipped drip irrigation system, addition of a soil layer, alkalization and other reclamation methods to improve the environmental situation within 2-3 years [5]. Covering the landfill with a layer of soil may also involve the addition of fertilisers and lime until the optimal soil pH is achieved, followed by the seeding of acid-tolerant grasses. Further reclamation of the soil layer on the landfill may even include planting poplars [6-8]. In addition to planting poplars, modern research has also demonstrated the effectiveness of various rock dust compositions and lime additives on the growth of maple and pine trees in acidic soils [9-11].

MATERIAL AND METHODS

The spent copper ore from one of the off-balance deposits in the Karaganda region was used as the subject of research.

Copper was preliminarily leached using bacterial oxidation methods described in detail in previous studies [12-14]. In addition to intensifying copper extraction, activation of oxidative processes by the bacterial culture of *A. Ferrooxidans* reduced the consumption of sulfuric acid, with a gradual decrease in concentration to 0.5% at the final stage of the leaching process.

Studies on the material composition of the ore included X-ray fluorescence multi-element analysis and chemical analysis for copper content, both before and after leaching. Analysis of the change in copper content revealed a decrease from

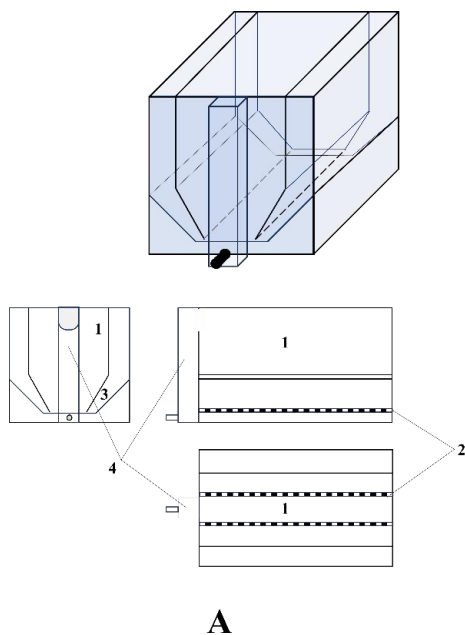
1,027% to 0.11%, corresponding to an extraction of 89.3%. The results of X-ray fluorescence of spent copper ore are presented in **Table 1**.

Table 1 Results of X-ray fluorescence analysis of a sample of spent copper ore

Element	Content, %	Element	Content, %	Element	Content, %
O	51.7	Cl	0.012	Cu	0.11
Na	1.66	K	0.967	Zn	0.013
Mg	1.082	Ca	1.165	Rb	0.006
Al	5.683	Ti	0.179	Sr	0.006
Si	36.12	Cr	0.011	Zr	0.007
P	0.071	Mn	0.09	Pb	0.020
S	0.065	Fe	1.06	-	-

A box with transparent walls and drainage systems was used as equipment for an experiment on environmental neutralisation and disposal of landfill ore, designed to collect water runoff from the upper and lower layers of the ore and soil mass. This equipment, shown in **Fig. 1**, in the form of a diagram (A) and a manufactured form (B), included four main technical elements: 1) a compartment for loading ore mass and soil layer; 2) drainage holes for common water runoff; 3) a compartment for accumulating common water runoff; 4) a compartment for collecting soil water from the upper layer.

The preparation of the material for the environmental experiment included loading spent ore into the central compartment of the equipment and achieving a stable pH of at least 4.0 in wastewater, followed by filling in a soil sample taken from the site next to the copper deposit with a layer of 3.0 – 3.5 cm (Figure 2). Endemic grasses growing in the region of the deposit, resistant to acidic environments, were considered for the selection of plant seeds. As a result, the following seeds of herbaceous plants were selected for sowing the soil layer: sedge, plantain and field mint.



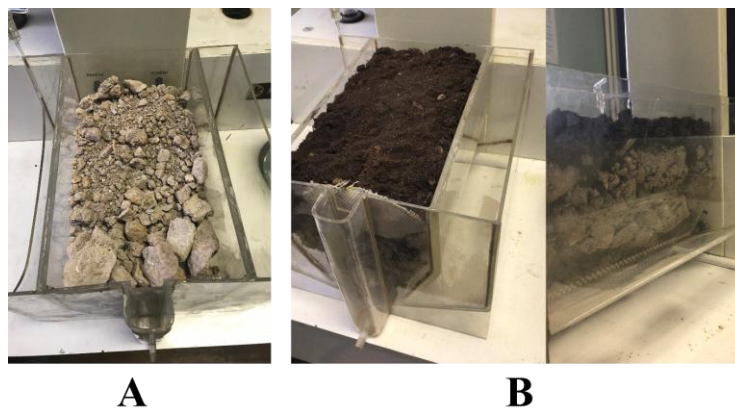
A

B

A - the diagram; B - the finished sample

1 - compartment for loading ore mass and soil layer; 2 - drainage holes for common water runoff; 3 - storage compartment for common water runoff; 4 - collection compartment for soil water of the upper layer.

Fig. 1 Equipment for collecting water from the neutralisation of spent copper ore.



A

B

A – loaded spent ore; B – the soil layer covering

Fig. 2 Preparation of ore material for an environmental disposal experiment.

Experimental part

The preliminary experimental section included hydrometallurgical leaching of copper from the source ore, using a full cycle of liquid extraction technology for copper production. During this stage of experimental work, the concentrations of copper, iron, and sulfuric acid in the circulating solutions of the process were analysed. However, the copper ore neutralisation stage, following sulfuric acid leaching, should be considered the main experimental part of this study. Following completion of the hydrometallurgical copper extraction step, the ore was subjected to several stages of water washing until acceptable residual copper, iron and acid concentrations were achieved. pH measurement and analysis for the content of residual concentrations of copper, iron and acid in the washing water were performed daily, with the calculation of the mass ratio of the consumed water to the weight of the tested ore. The pH was measured using a Consort 930 instrument, and the concentrations of copper and iron were determined by atomic absorption spectroscopy, with parallel verification by titration. The amount of residual acid was determined by the titrimetric method.

The further course of the experiment involved covering the ore material with a layer of soil and planting seeds of herbs resistant to acidic environments. The wastewater was collected separately from the soil and shared with the ore mass into the appropriate compartments of the equipment. At the same time, plant growth and wastewater indicators were monitored every week. The duration of the ecological section of the experiment, with the introduction of the plant factor into the system, was 11 weeks.

RESULTS AND DISCUSSION

Acceptable acidity values – pH of at least 4, for loading the soil layer and planting plants, were achieved after several cycles of water rinsing. The initial acidity index in the wastewater of the spent ore was pH = 1.8. After the mass ratio of rinsing water to the ore mass was 10%, the pH value increased to 4.2. The detailed dynamics of the pH change in the rinsing water are shown in Fig. 3.

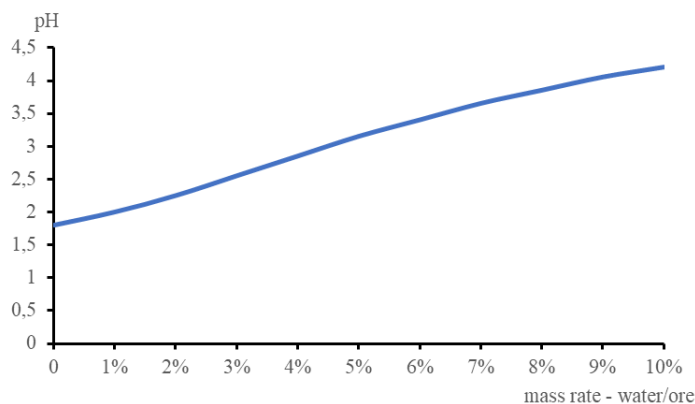


Fig. 3 Dynamics of pH changes in the rinsing water

The water from ore washing was also analyzed for residual concentrations of copper, iron, and sulfuric acid. The dynamics of changes in their concentrations, as determined by the quantitative ratio of water washings, are shown in Fig. 4.

The residual concentrations of copper in the waste solution were 0.125 g/L, iron – 3.27 g/L, and the concentration of sulfuric acid was 2.74 g/L, at a pH of 1.8 at the start of the water washing process. A significant decrease in the concentration of metals and sulfuric acid was observed, accompanied by a gradual increase in the amount of water to 10% of the ore mass, and a corresponding rise in pH to 4.2. The final concentration of copper was 0.015 g/l, iron – 0.1 g/l, sulfuric acid – 0.43 g/l. The rinsing water was removed from the system before loading the soil layer, and subsequent irrigation was performed with fresh water. Despite a significant decrease in the concentration of metals and acids, flushing runoffs of this composition should be considered as process water for other areas of hydrometallurgical production.

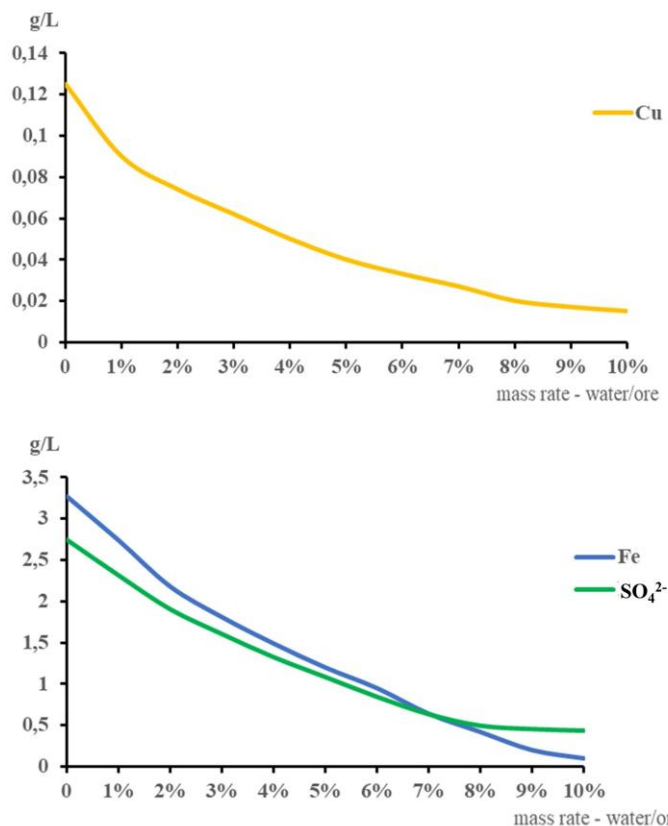


Fig. 4 Dynamics of changes in concentrations of copper, iron and sulfuric acid in the washing water

Wastewater was monitored once a week after sowing the seeds in the soil layer over the spent ore. Individual runoffs from the soil layer and water that also passed through the spent ore layer were analyzed. Samples of both types of water were analysed primarily for pH values (Fig. 5). The analysis of total runoff water also included the determination of the amounts of copper, iron, and sulfate ions (Fig. 6).

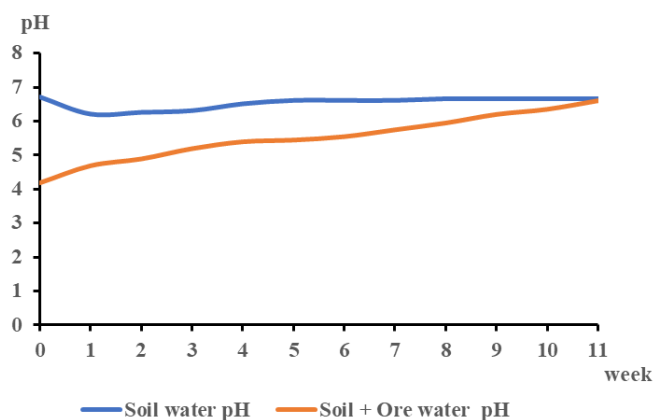


Fig. 5 Changes in pH values in water runoff from the soil layer and the common layer (soil + ore)

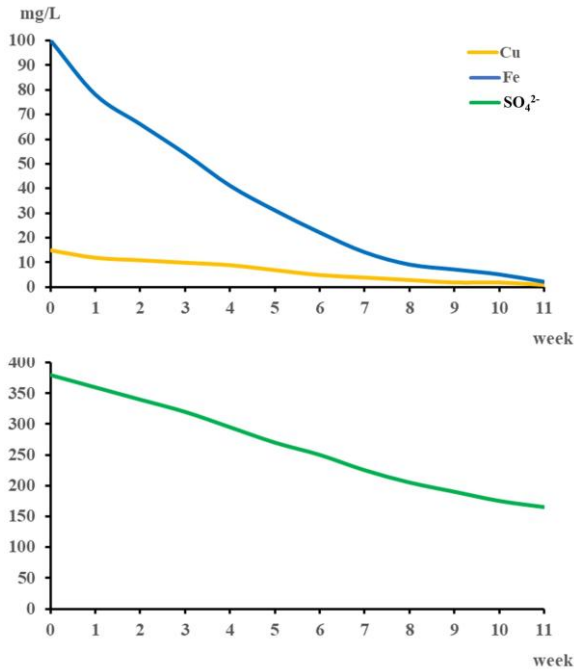


Fig. 6 Changes in concentrations of copper, iron, and sulfate ions in the water runoff of the common layer (soil + ore).

The first plant sprouts were observed on 5-7 days of the experiment. The first stage of active plant growth was noted in the fifth week of the experiment. The second stage of active growth was observed in 9 weeks of the experiment. The total follow-up period was 11 weeks. The formation of the root system of plants was noted at the final stage of the experiment, in the soil layer, with partial penetration into the ore mass layer (**Fig. 7**). This factor ensures the stability of the soil layer against weathering during the project implementation at the spent copper deposits. Plant growth was recorded weekly by photographing, as shown in **Fig. 8**.



Fig. 7 Formation of the root system in the soil and ore layers.

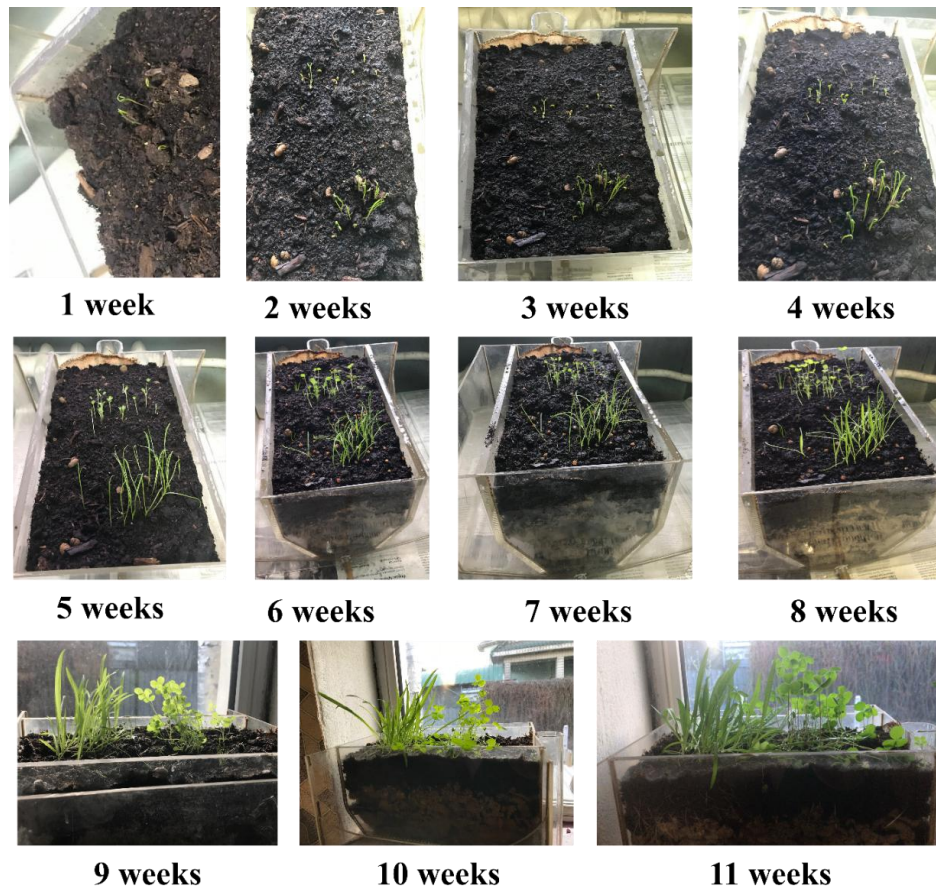


Fig. 8 Weekly observations of plant growth on the soil layer of spent ore

Analyses of the ore sample after a full cycle of ecological utilization, including preliminary bio-hydrometallurgical extraction of copper, followed by water washing and soil and plant reclamation, showed an environmentally acceptable copper content of 0.046%. The phase composition of the neutralized dump ore, shown in Table 2, corresponded to the parameters of ordinary rocks and soil.

Table 2 Phase composition of the neutralised ore sample

Name	Formula	Amount
Quartz, syn	SiO ₂	51.1%
Albite low	Na(AlSi ₃ O ₈)	29.1%
Muscovite-2M1	KAl ₂ (Si,Al) ₄ O ₁₀ (OH) ₂	13.5%
Clinochlore-IIb-2	(MgFe)((Si ₅ Al ₃)O ₂₀ (OH) ₁₆)	6.3%

It was established through experiments that it is possible to reduce the concentrations of harmful compounds in spent copper ore after sulfuric acid leaching is gradually applied, thereby restoring a balance favourable for plant life. The implementation of the laboratory experiment results may be feasible as a landscaping project for spent landfills of copper deposits, producing copper through the hydrometallurgical method. Taking into account the specifics of hydrometallurgical production, i.e. the isolation of technological flows and ponds, as well as the recycling of solutions, work to reduce environmental risks and restore the natural balance will be conducted in a closed system until acceptable indicators are achieved.

The analysis of the dry residue of plants showed the predominance of organic carbon fragments, as well as nitrogen and oxygen compounds in the composition. The spectra of these compounds and some light elements (Li, B, Be, N) are not recognised separately in exact quantitative terms. However, fluorescence showed the precise values of silicon, aluminium and their oxide compounds. Among the metal elements, the presence of alkali metals, such as sodium and potassium, and alkaline earth metals, including calcium and magnesium, was revealed. Iron up to 1.7% and zinc 0.08% were found in significant amounts of heavy and non-ferrous metals. A detailed analysis is presented in Table 3.

Table 3 The analysis of the sample of the dry residue of plants

Element	Amount, %	Element	Amount, %
SiO ₂	13,826	Cl	0,165
Si (other form)	6,464	Mg	0,105
Ca	6,094	Zn	0,08
K	3,106	Cu	0,005
Na	2,55	Zr	0,003
Fe	1,708	Cd	0,002
S	1,345	Sr	0,001
Al ₂ O ₃	0,684	C (organic)	54,117
Al	0,362	N, O (residues)	8,552
P	0,219	light elements (Li, B, Be)	0,612

Environmental Project Implementation Opportunities

By itself, hydrometallurgical processing of off-balance copper heaps is partly a method for the disposal of mining waste. At the same time, it enables the production of a metallurgical product - cathode copper of at least 99.99% purity. However, hydrometallurgy, like any production process, has its own potential environmental risks and production waste, despite its significant advantages over other mining and metallurgical technologies. In particular, the issue of disposing of spent ore piles after their full production can be addressed using the processes presented in this ecological experiment.

Two main phases can be distinguished – industrial and ecological ones. They are presented step by step in Figure 9 in the proposed project on green recycling and utilization of copper heaps. The industrial phase provides for the hydrometallurgical processing of the off-balance dump, with the maximum possible extraction of copper. The industrial phase of copper dump ore utilization includes three main stages:

A) The formation of a heap of landfill ore on the surface of a sealed geomembrane (or on a clay waterproof layer), the digging of isolated

technological channels and ponds intended to collect solution, the installation of an irrigation system (Fig. 9A);

B) A hydrometallurgical production process with recirculation of solutions, where the use of bio-oxidants from bacterial cultures will help reduce the consumption of sulfuric acid, as well as accelerate the process of lowering acidity at the final stage (Fig. 9B);

C) The final stage, when 80-85% of the dump site output is reached, the concentration of sulfuric acid is reduced. The technological solutions are gradually removed to other sites (Fig. 9C), and it is also possible to dispose of the spent solution as a source of iron III sulfate (Fig. 9C).

The ecological phase of heap recycling also includes three main stages, two of which (Fig. 9D, E) are performed in a closed, isolated ecosystem. At the final third stage (Fig. 9F), it becomes possible to dismantle insulating geomembranes and discharge wastewater into the environment when acceptable concentrations of copper, iron and sulfate ions are reached. A brief description of the processes of the successive stages of the ecological phase is presented below:

D) water flushing of the waste heap, involving an irrigation system and technological channels, using wastewater as process water for other production sites (Fig. 9D);

E) the possibility of including water flows in irrigation turnover, upon reaching the maximum permissible concentrations of metals and pH, laying the soil layer, planting seeds of endemic grasses resistant to acidic environments (Fig. 9E);

F) the final stage of the environmental recycling phase, with the possibility to remove the isolation of water flows and to release them into the environment, is performed when the water runoff parameters meet the sanitary and environmental standards of reservoir water; the composition of spent landfill ore is similar to the minerals of ordinary rocks and soil (Fig. 9 F).

CONCLUSION

The results of an environmental experiment aimed at utilising copper ore after sulfuric acid leaching have demonstrated the potential to significantly reduce the concentrations of harmful compounds in mineral raw materials, with the possibility of greening the surface of the spent waste rock. The use of a bio-oxidising factor at the leaching stage made it possible to reduce the consumption of sulfuric acid, maximise copper recovery, and recover the residual metal during water washing, with a gradual increase in pH to 4.0. The application of a soil layer to the surface of the washed ore, combined with the planting of seeds of endemic grasses, resulted in stable plant growth during 11 weeks of observation, accompanied by the formation of a root system. There was a stabilisation of pH – 6.5 acidity to the level of a neutral medium and a decrease in the concentration of copper, iron, and sulfate ions in wastewater during the vegetation growth on the surface of the soil layer. As a result, acceptable concentrations were achieved in the aquatic environment that meet the environmental standards of natural reservoirs, and the composition of the ore layer corresponded to the mineralogy of empty rocks and soil.

To date, spent copper heaps are desert rocks softened by long periods of sulfuric acid leaching, with an acidic pH environment and acidic salt residues. The wind transport of collapsing mineral rock particles from the spent heap, in turn, will negatively impact the ecological situation of the deposit region, leading to desertification of the adjacent territory and pollution of water bodies. Based on the above, the main idea of the project for green recycling of spent copper ores aims to solve this problem by introducing an ecological phase of landfill treatment. Proceeding from the fact that hydrometallurgical technology for copper extraction is partly a solution in the field of processing mining and metallurgical wastes from off-balance copper ore, the results obtained during the conducted experiments can be considered methods for subsequent full ecological utilisation of spent copper heaps. The infrastructure of the hydrometallurgical production site enables the environmental neutralisation of the copper heap through a system of isolated streams, allowing for maximum copper production. Thus, natural conservation of spent ore will be performed after achieving acceptable, slightly acidic indicators, the formation of a coating from the soil layer, and the growth of herbaceous plants with a well-developed root system. The subsequent stabilisation of the acidity indicators, the concentration of potentially harmful substances to acceptable standards, and the exclusion of environmental risks will make it possible to remove the insulation of the hydraulic system at the landfill site with access to the environment, as well as to plant shrubs and trees.

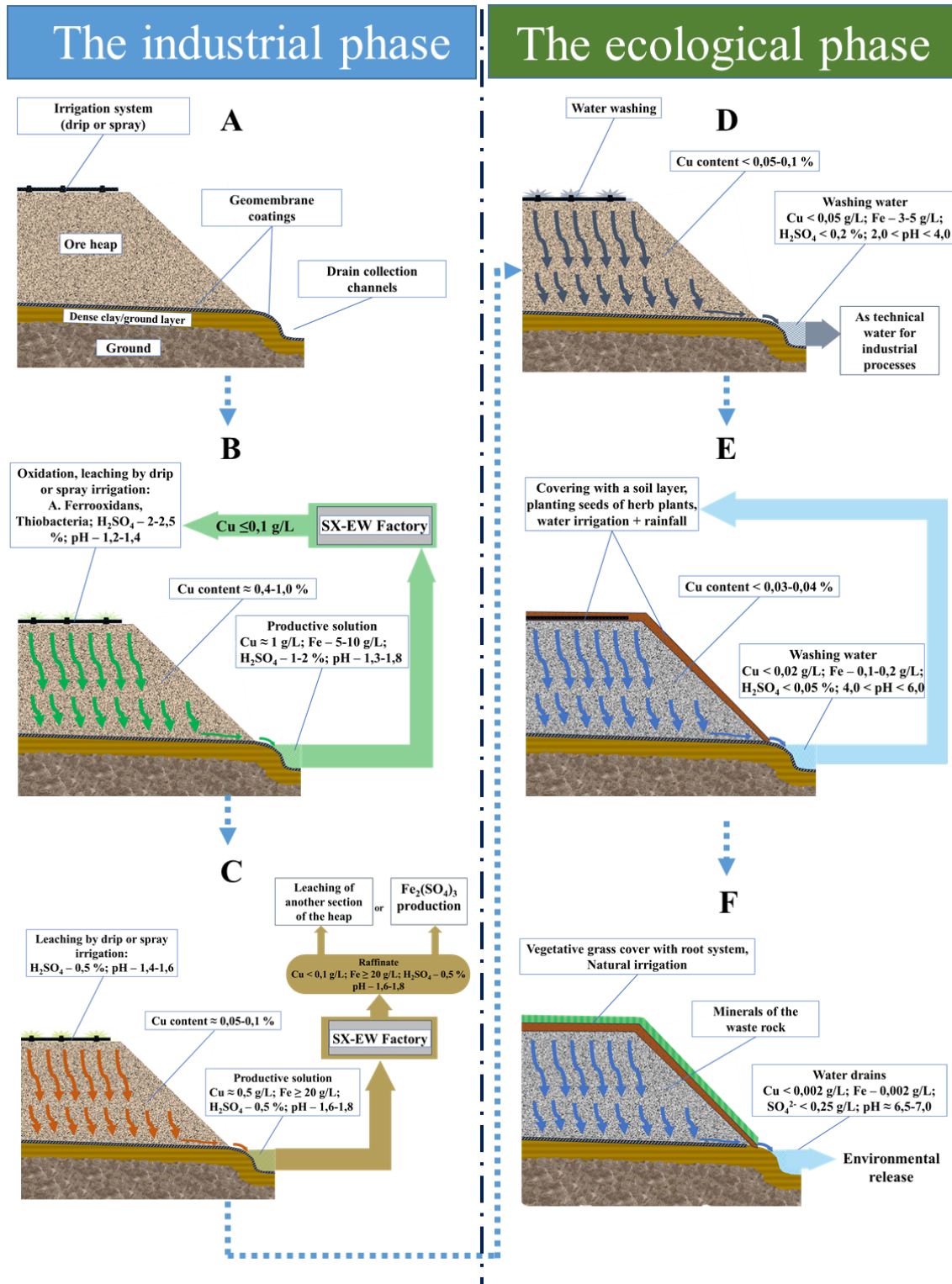


Fig. 9 Industrial and ecological phases of the project of green recycling and disposal of spent copper heaps

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