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The effect of biochemical oxidation on the hydrometallurgical production of copper

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ABSTRACT

The article presents the results of the use of the biochemical leaching method for copper-bearing non-commercial ore in a full hydrometallurgical cycle. The object of the study was the average copper-bearing ore of the Satbayev deposit, with an initial copper content of 0.26%. The experimental part of the full hydrometallurgical cycle included percolation leaching, extraction, and re-extraction tests, as well as electrodeposition tests and the production of finished cathode copper. Two variants of percolation leaching were performed as a comparison: a standard method using only sulfuric acid and a preliminary bacterial oxidation method for mineral raw materials with an adapted bacterial strain, Acidithiobacillus ferrooxidans. Percolation leaching experiments showed a significant reduction in sulfuric acid consumption when preliminary bacterial oxidation was used. Upon reaching the level of copper extraction from the ore of 86–87%, the final consumption of sulfuric acid for the standard leaching method was 15.5 kg per ton, while 9.4 kg per ton was required for the biochemical method. The productive solution obtained in the biochemical leaching process showed full suitability for all technological stages of hydrometallurgical copper production.

Keywords: Copper heaps, Bio-oxidation, Acidobacillus ferrooxidans bacteria, Leaching, Solvent extraction.

How to Cite:

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1. INTRODUCTION

The modern standard technology of heap leaching and liquid extraction of copper is described in detail in a number of works written by domestic and foreign researchers [1], [2]. To intensify the processes of leaching copper ores with acidic solutions, it is proposed to introduce various additives that accelerate the dissolution process and reduce the consumption of sulfuric acid. At present, the use of bacteria in the extraction of copper from ore is of great importance. The term "bacterial leaching" refers to the intensified process intended to leach metals from ores. A number of studies showed the economic benefits of the bacterial leaching process. It was established that the modern standard technology of heap leaching and liquid extraction of copper is described in detail in a number of works written by domestic and foreign researchers [1], [2]. To intensify the process of leaching copper ores with acidic solutions, it is proposed to introduce various additives that accelerate the dissolution process and reduce the consumption of sulfuric acid. At present, the use of bacteria in the extraction of copper from ore is of great importance. The term "bacterial leaching" refers to the intensified process intended to leach metals from ores. A number of studies have shown the economic benefits of the bacterial leaching process. It was established that the percentage of copper extraction increases by several orders of magnitude during the preliminary biotreatment of oxidized ores. It was shown that the adaptation of bacteria before bioleaching increases the process's efficiency. One of the promising new technologies is the technology of bacterial-chemical leaching, which does not require large material costs and does not create problems with atmospheric pollution. A number of studies have shown the economic advantages of the bacterial leaching process. It is shown that the adaptation of bacteria before bio-leaching increases the efficiency of the process [3], [4].

The role of the Acidithiobacillus ferrooxidans bacteria in the leaching of sulfide ores is widely known. Metals from water-insoluble sulfides pass into soluble sulfates during the biogeotechnological process. The A. ferrooxidans thionic bacteria oxidize all metal sulfides. Bacteria get the carbon they need to grow from carbon dioxide. These bacteria thrive in an acidic environment (pH in the range of 1.0–4.8) at temperatures between 3 and 40°C. The optimal parameters for the bacteria's development are a pH value in the range of 2-3 and a temperature of 28 °C. Thionic bacteria are found in reservoirs, soil, and deposits of sulfur and sulfide ores. But they show their activity in the presence of oxygen. Methods intended to use bacterial cultures as an oxidizing agent are known in domestic and international practice [5]. The main advantages of bacterial oxidation include the high efficiency of the conversion of ferrous iron into ferric iron as well as the low cost of this technology.

Adaptation and growth of the A. ferrooxidans bacterial culture are usually accompanied by certain changes in the solution parameters; in particular, there is an active decrease in the concentration of Fe2+ and an increase in Fe3+ ions. Often, copper compounds are toxic to the standard A. ferrooxidans strain, which requires additional microbiological selection with the cultivation of an adapted culture. A. ferrooxidans strains adapted to the conditions of copper raw materials enable the bioleaching of sulfides, with an emphasis on copper sulfides. A sample of the A. ferrooxidans-1333 strain is known. It was bred at the Korean Center for Culture Collection, and it showed high oxidation results for Fe2+ in the composition of chalcopyrite due to the high immobilization of bacteria to the specifics of this mineral [6].

The novelty lies in the use of bacteria as a factor catalyzing oxidative processes, which will significantly increase the degree of copper extraction into a productive solution. Previous studies determined the optimal conditions for growing A. ferrooxidans bacterial cultures adapted to the chemical composition of copper-bearing minerals [7]. Thus, it was established that it is advisable to pre-treat mineral raw materials with a bacterial solution with a pH of at least 1.5 for more efficient bacterial oxidation of mineral raw materials. At the same time, an increase in pulp acidity and a decrease in pH less than 1.2 have an extremely negative effect on the survival of bacterial cells. The optimal temperature regime for the growth and development of A. ferrooxidans culture is a temperature range of 20–30 °C. Application of the most favorable factors for the growth of bacterial cells (p - 2,3, t = 20 - 30 °) enables, as a result, to increase their concentration to 0,1 106 cells/cm3 before 2,8 106 cells/cm3. [8]–[11]ercentage of copper extraction increases by several orders of magnitude during the preliminary biotreatment of oxidized ores. It was shown that the adaptation of bacteria before bioleaching increases the process efficiency. One of the promising new technologies is the technology of bacterial-chemical leaching, which does not require large material costs, does not create problems of atmospheric pollution. A number of studies have shown the economic advantages of the bacterial leaching process. It is shown that the adaptation of bacteria before bio-leaching increases the efficiency of the process [3], [4].

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2. MATERIAL AND METHODS

An average sample of ore was taken as an object of research from the dump of the copper deposit in Satbayev town, Karaganda region. Analysis of the averaged sample showed an average copper content of 0.264%. Besides, fluorescence and X-ray phase analysis were used to analyze the ore sample. Their results are presented in Tables 1-2.

Table 1: Results of fluorescent analysis of satbayev deposit

Element	Content (%)	Element	Content ()%	Element	Content (%)
O	52.777	Cl	0.014	Cu	0.264
Na	1.44	К	1.126	Zn	0.012
Mg	1.13	Ca	2.718	Rb	0.007
Al	5.8	Ti	0.414	Sr	0.009
Si	27.041	V	0.007	Zr	0.01
P	0.06	Mn	0.116	Ba	0.04
S	0.076	Fe	2.58	Pb	0.0043

Table 2: Results of X-ray phase analysis of satbayev deposit

Component name	Formula	Content (%)
Quartz (syn)	SiO_2	54.50
Albite	$Na(AlSi_3O_8)$	18.60
Clinochlor-1MIIb, (ferroan)	$(Mg,Fe)_6(Si,Al)_4O_{10}(OH)_8$	9.40
Potassium Sulphite Hydrate	$K_2(S_3(SO_3)_2)(H_2O)_{1.5}$	6.40
Potassium hydrogen sulfate	$K_3H(SO_4)_2$	2.90
Potassium Sulphite Hydrate	$K_2(S_3(SO_3)_2)(H_2O)_{1.5}$	2.80
Muscovite	$H_2KAl_3Si_3O_{12}$	2.40
Hematite (syn)	Fe_2O_3	1.15
Pyrite	FeS_2	0.80
Malachite (syn)	$Cu_2(OH)_2CO_3$	0.60
Atakamit	$Cu_2Cl(OH)_3$	0.30
Chalcopyrite	$CuFeS_2$	0.15

The large-scale laboratory experiments included the leaching of copper in a full hydrometallurgical cycle, including the stages of extraction, re-extraction and electrowinning. This technology provides for closed circulation cycles of solutions according to the scheme shown in Figure 1.

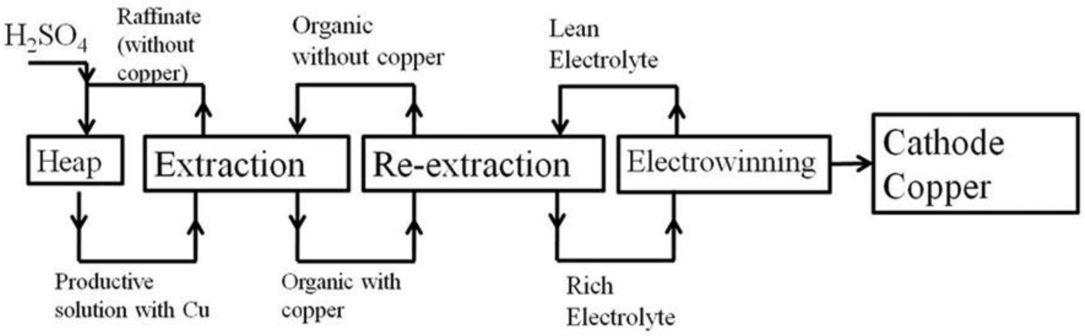
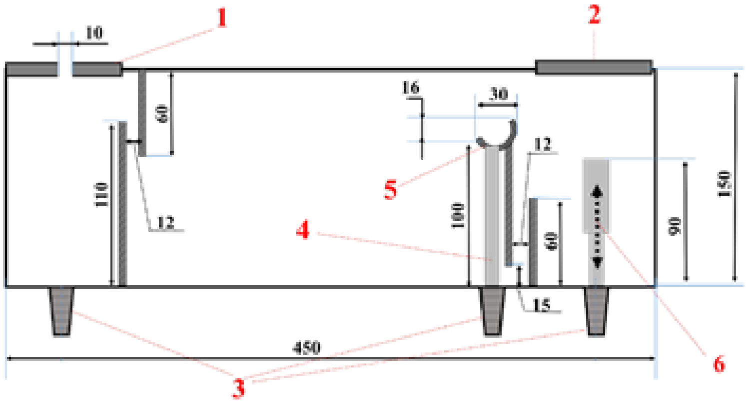


Figure 1: Scheme of hydrometallurgical production of cathode copper

225 kg of copper-bearing ore were loaded into 2 percolators from the dump of the Satbaev deposit to simulate the heap leaching process. The leaching solution was supplied using peristaltic pumps (Figure 2).

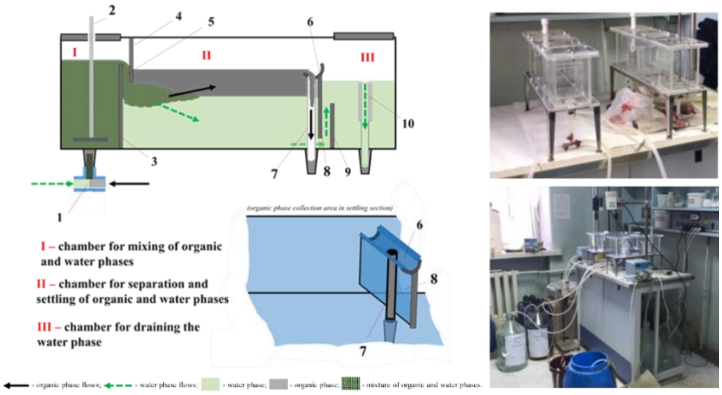
Figure 2: Percolators for heap leaching simulating experiments

An organic phase - 10% LIX 984 at a O:W flow ratio = 1:2 was used at the extraction stage. Specially equipped chambers were designed for the extraction and re-extraction section. They contained chambers for mixing, settling and draining of the water phase, as well as a chute for collection of organic matter. The extraction and re-extraction chambers have the same design and principles for the movement of water and organic flows. The structure of the chamber consists of (Figure 3): The flows in the chambers include (Figure 4).



1 - Cover with a hole for the stirrer about the mixing chamber; 2 – Cover of the draining chamber; 3 - Hose Fitting D = 10/8; 4 - Organics drain pipe D =10/8; 5 - Organic collection chute; 6 - Telescopic tube (max 90 mm; min 60 mm). 1 - Cover with a hole for the stirrer about the mixing chamber; 2 – Cover of the draining chamber; 3 - Hose Fitting D = 10/8; 4 - Organics drain pipe D =10/8; 5 - Organic collection chute; 6 - Telescopic tube (max 90 mm; min 60 mm).

Figure 3: Structure of the extraction and re-extraction chambers



1- tee for water and organic phase; 2 - stirrer of a mixer; 3 - lower partition of the mixing and settling chambers; 4 - upper partition of the mixing and settling chambers; 5 - flow of mixtures of organic and water phases; 6 - chute for collection of the organic phase; 7 - organic drain tube; 8 - intermediate partition of the chambers for settling and draining the water phase (the lower channel at the bottom, for the water phase); 9 - additional bottom partition in the chamber to contain the organic phase during shutdown; 10 - telescopic water phase drain pipe (phase level control).

Figure 4: Chambers and flows of organic and water phases

3. RESULTS AND DISCUSSION

3.1 Experimental results

Large-scale laboratory tests were launched when the percolators were loaded, and the extraction unit was installed. A standard solution of sulfuric acid with a concentration of 2.5% was supplied into Percolator 1. Pre-treatment with a bacterial solution was performed in percolator 2 at lower concentrations of sulfuric acid for the first 20 days, then it was leached with a 2.5% solution of sulfuric acid. The obtained productive solutions were analyzed daily for the content of copper and acid, then the necessary amount of sulfuric acid was added to achieve the required concentration, and the solution was reused in leaching. The productive solution was fed into the extraction unit with the subsequent leaching with already demineralized raffinate. The amounts of copper removed from the productive solution during the extraction process and the amount of copper in the current solution, including the raffinate, as well as residual copper in the organic phase, were summed up when the extraction was calculated. The results of the percolation leaching process presented in Table 3 included the calculation of sulfuric acid consumption per ton of ore and the final extraction of copper from the ore.

Table 3: Results of copper extraction and total consumption of sulfuric acid during percolation leaching of Satbayev deposit

Days	**Acid consumption (kg/t of ore)**		**Copper extraction (%)**	
	Standard	**Bio-leaching**	**Standard**	**Bio-leaching**
5	6.01	1.5	6.46	0.4
10	8.49	3.98	14.0	2.11
15	10.17	6.03	23.0	11.31
20	11.27	6.51	34.0	18.0
25	13.10	8.08	45.45	25.0
30	14.01	8.83	58.1	33.74
35	14.29	9.28	68.53	42.42
40	14.82	9.28	75.41	55.35
45	15.04	9.39	81.0	64.81
50	15.49	9.39	86.69	71.31
55	15.49	9.39	86.69	79.32
60	15.49	9.39	86.69	87,2

At first glance, the results of the large-scale percolation leaching experiments showed the dominance of the standard sulfuric acid method, compared with the results of bio-leaching. So, 86.7% of copper was extracted from the ore material within 50 days with standard leaching with sulfuric acid, while 71.3% was extracted over the same period with the use of the bacterial oxidation method. However, the leaching was extended for additional 10 days for the waste ore in percolator 2 due to the fact that the pre-treatment period was used for the bacterial oxidation method. As a result, the total extraction of copper was 87.2% in the biochemical leaching variant. The final dynamics of copper extraction by standard and biochemical methods is shown in the graphs of Figure 6.

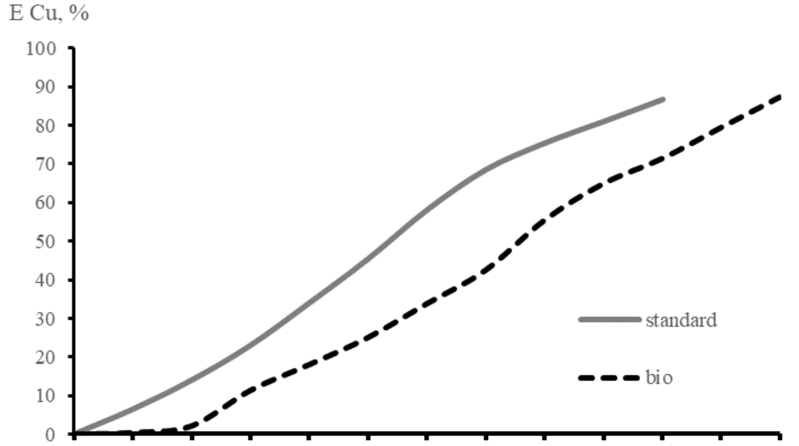


Figure 6: Dynamics of the total extraction of copper by standard and biochemical leaching methods

Another important factor in the large-scale tests was the sulfuric acid consumption rate. So, the total consumption of sulfuric acid for 50 days was 3484.2 g in a percolator with standard leaching. This figure was 15.49 kg of sulfuric acid when it was converted to a ton of ore. Additional amounts of sulfuric acid in percolator 2 with a biochemical leaching method were added less often, and the leaching was without additional strengthening during the last 20 days of the experiment resulted in the total consumption of sulfuric acid of 2112.4 g, that is 9.39 kg, in terms of a ton. Comparison of the dynamics of sulfuric acid consumption per ton of ore in standard and biochemical leaching methods is shown in the graphs of Figure. 7.

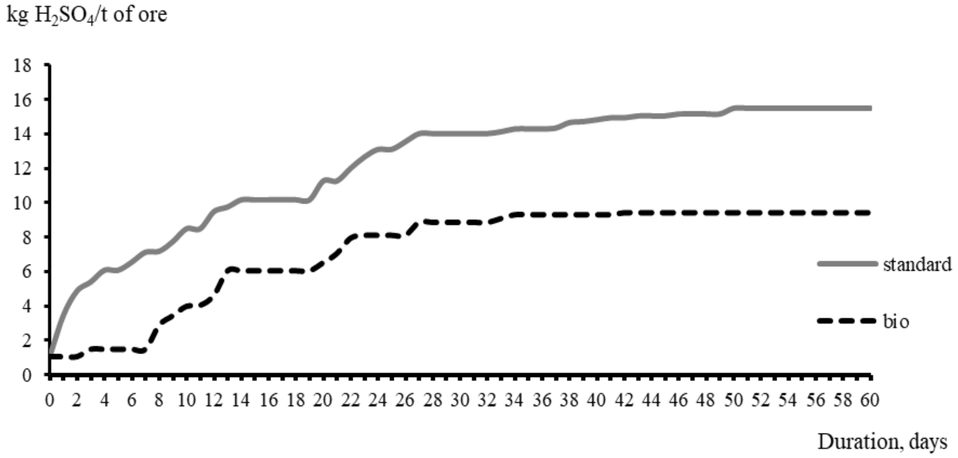


Figure 7: Dynamics of sulfuric acid consumption during standard and biochemical leaching

The resulting productive solutions of both options were processed in an extraction unit (Figure 8). Productive solution after copper extraction by organic phase (raffinate) was reused in leaching. Copper from the saturated organics passed into the re-extraction chamber and accumulated in the electrolyte. The final concentration of copper in the electrolyte obtained from the productive solution of standard leaching was 46.7 g/l, and from the biochemical leaching solution of - 47.12 g/l. These concentrations are acceptable for the further technological stage - electrowinning.

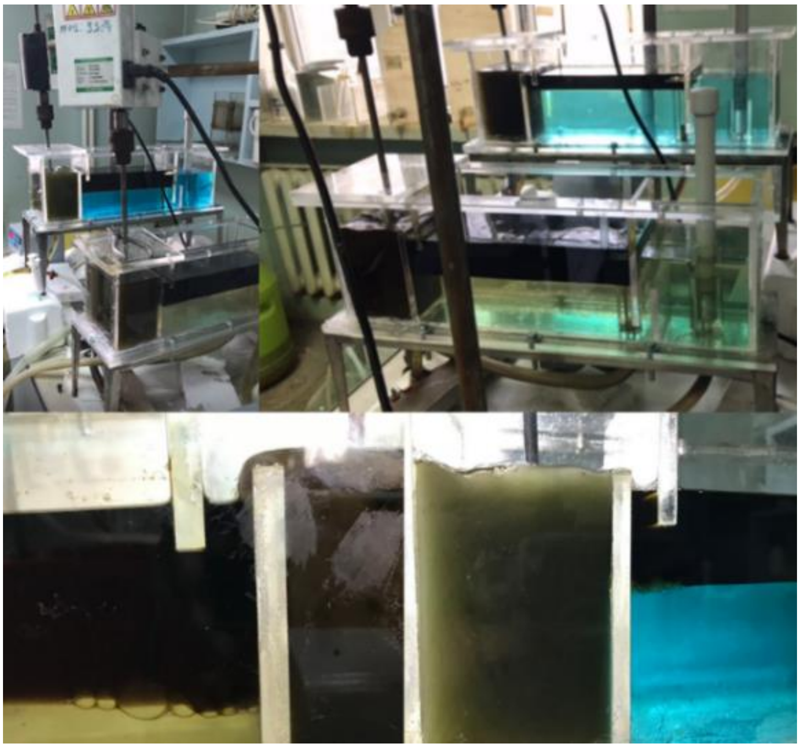


Figure 8: Processes of extraction and re-extraction during large-scale tests

Table 4: Final distribution of copper

Leaching method	Copper distribution balance, %			
	Residue in ore	**Productive solution**	**Organic**	**Electrolyte**
Standard	13.31	7.8	0.27	78.62
Biochemical	12.8	7.3	0.57	79.33

Electrolytes were fed into the electrowinning plant to complete the full cycle of hydrometallurgical production of cathode copper. Copper electrodeposition was performed in a 2-liter electrowinning bath equipped with 4 stainless steel cathodes and 5 titanium anodes (Figure 9). The copper content in the electrolyte should be in the range from 30 to 60 g/l in accordance with technological requirements. Therefore, electrodeposition was performed until the copper concentration in the electrolyte reached at least 30 g/l. As a result, 335.4 g of copper was deposited on 4 cathodes, the concentration of residual copper in the electrolyte was 30.15 g/l.

Figure 9: Copper electrowinning process from a saturated electrolyte

3.2 Discussion

The practice of use of oxidizing reagents is widespread in the field of hydrometallur-gical gold production. Methods of use of peroxides, chloroactive compounds, surfactants, bacterial cultures application for the decomposition of host gold sulfides, like pyrite, arse-nopyrite, etc., are known. [25-27]. However, the principle of oxidizing factors plays a dif-ferent role in contrast to the oxidation of gold-bearing minerals during the leaching process of copper deposits. Besides, the use of a number of oxidizing factors, especially containing active chlorine, is impossible and unprofitable for liquid-extraction technology of copper production.

Biotechnological copper leaching process involves irrigation of ore material or tech-nogenic waste containing metal sulfides with sulfuric acid solutions and iron salts, as well as introduction of viable thionic, iron-oxidizing bacteria. Conventional oxidation of sulfide minerals most commonly found in copper dumps, with the use of pyrite and chalcopyrite as examples, can be described by the following reactions (1-2):

$$2FeS_2 + 7O_2 + 2H_2O \rightarrow 2FeSO_4 + 2H_2SO_4 \quad (1)$$

$$CuFeS_2 + 4O_2 \rightarrow CuSO_4 + FeSO_4 \quad (2)$$

However, the presence of iron (II) and other metals that are less active than copper in solution may contribute to its precipitation if they are not in the maximum oxidation de-gree as exemplified by this reaction (3):

$$Cu^{2+} + 2Fe^{2+} \rightarrow Cu^0\downarrow + 2Fe^{3+} \quad (3)$$

Oxidative processes in sulfuric acid medium, with the participation of air oxygen will convert iron compounds from the oxidation degree +2 to +3 by the reaction (4):

$$4FeSO_4 + O_2 + 2H_2SO_4 \rightarrow 2H_2O + 2Fe_2(SO_4)_3 \quad (4)$$

The presence of a catalyzing factor in sulfuric acid medium during the leaching pro-cess helps to accelerate the transition of iron to the oxidized form +3 as an example of reac-tions (5-6):

$$2FeS_2 + 14H_2SO_4 \rightarrow 14H_2O + Fe_2(SO_4)_3 + 15SO_2 \quad (5)$$

$$2CuFeS_2 + 18H_2SO_4 \rightarrow 18H_2O + Fe_2(SO_4)_3 + 17SO_2 + 2CuSO_4 \quad (6)$$

Obtained compounds of iron (III) can act as an oxidizer. Iron ions transferring into the productive solution, in the maximum oxidation degree +3, contribute to further oxidation of acid-absorbing and noble copper containing minerals during the circulation of the pro-ductive solution after the extraction stage (leaching with raffinate). In practice the use of iron (III) sulfate is known as an oxidizing catalyst. This com-pound in the leaching process can contribute to dissolution of sulfide minerals of copper by reactions (7-8):

$$CuFeS_2 + 2Fe(SO_4)_3 + 2H_2O + 3O_2 \rightarrow CuSO_4 + 2H_2SO_4 + 5FeSO_4 \quad (7)$$

$$2CuS + 2Fe(SO_4)_3 + 2H_2O + O_2 \rightarrow 2CuSO_4 + 2H_2SO_4 + 4FeSO_4 \quad (8)$$

The practice of use of trivalent iron sulfate as an oxidizing agent is widespread at a number of copper deposits. At the same time, a widespread problem of hydrometallurgical facilities which develop copper deposits with high iron content is the accumulation of excessive concentrations of iron ions in the productive solution. The excessive concentration of trivalent iron ions, more than 10 g/l, negatively affects the extraction process, because it reduces the selectivity of the organic extractant for copper, which deteriorates the quality of the electrolyte and cop-per cathode obtained during electrowinning. It is inexpedient to consider iron (III) sulfate additive as an oxidizing reagent in such cases. Cultivation of iron-oxidizing microorgan-isms, in this case, will allow obtaining an optimum concentration of iron ions +3, from its initial content in the ore material.

4. CONCLUSION

Thus, the results of large-scale laboratory experiments in the full hydrometallurgical cycle showed that the consumption of sulfuric acid is significantly reduced with the preliminary bacterial oxidation method at the stage of copper leaching from mineral raw materials compared to the standard technology. Despite a slight increase in the process duration required for the preliminary adaptation of a bacterial culture on mineral raw materials, the final extraction of copper during biochemical leaching was almost on the same level with the standard technology with the lowest consumption of sulfuric acid. Tests of the complete technological chain showed the possibility of extraction and re-extraction of a productive bio-leaching solution with obtaining of an electrolyte that meets the electrowinning requirements. The results of both methods showed the copper extraction was at the same level of 86-87%. However, the effect of a significant reduction in the consumption of sulfuric acid was observed when the biochemical leaching method is projected to the production parameters. Thus, the total consumption of sulfuric acid for the standard leaching method was 15.5 kg per ton, while 9.4 kg per ton were consumed with the biochemical method. Acidophilic bacteria - Acidobacillus Ferrooxidans - were selected as microorganisms. The use of the most favorable factors for the growth of bacterial cells (pH – 2.3, t = 20-30 ° C), eventually allows to increase their concentration from $0,1 \times 10^6$ cl /cm^3 to $2,8 \times 10^6$ cl/cm^3.

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DECLARATIONS

Author contribution

Aigul Koizhanova: Project Administration, Methodology, Validation. David Magomedov: Investigation, Writing - Original Draft, Formal analysis. Nurgali Abdyldayev: Investigation, Resources, Supervision. Maria Yerdenova: Visualization, Investigation, Supervision. Akbota Barayeva: Conceptualization, Visualization, Investigation.

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Competing interest

The authors declare that we have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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