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Modeling of acid reagent moisture transfer during hydromining of sulfide copper

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Abstract

Purpose. To substantiate the selection of acidic reagents and to develop a mathematical description of moisture-transfer processes during the hydromining of sulfide copper, taking into account the physicochemical properties of the reagents, the parameters of the porous ore medium, and the biological factors involved in sulfide mineral oxidation.

Methods. The study employed a combination of theoretical analysis, mathematical modeling, and the generalization of experimental data. The selection of reagents, in particular sulfuric acid and iron salts, was substantiated using the Hammett acidity function (H_0) and the Hard and Soft Acids and Bases (HSAB) principle. Modified diffusion and filtration equations accounting for volumetric moisture content, permeability, and suction pressure were used to describe mass- and moisture-transfer processes. The hydrodynamic conditions of hydraulic-mixture flow were evaluated using the Froude and Reynolds criteria.

Findings. It was established that the efficiency of underground sulfide copper leaching is governed by the combined influence of ore granulometric composition, hydrodynamic parameters, reagent concentration, and biochemical oxidation factors. The rational process parameters were found to be an ore particle size of about 0.35 mm, a hydraulic erosion pressure of 1.6 MPa, an H_2SO_4 concentration in the range of 5-10%, a medium pH of 1.5-1.8, and a temperature of 25-32°C. The developed mathematical models enable the dynamics of moisture transfer in a porous medium to be described and the behavior of acid reagent transport within the ore mass to be predicted.

Originality. A comprehensive approach was developed to model moisture transfer of acidic reagents during hydromining of sulfide copper, combining physicochemical substantiation of reagent selection, description of the hydrodynamic conditions of hydraulic-mixture flow, and consideration of biotechnological factors in sulfide oxidation. Unlike existing approaches, the proposed model accounts for the volumetric moisture content, permeability, and suction pressure of the porous medium within a unified formulation.

Practical implications. The obtained results can be used to substantiate and design technological schemes for borehole hydromining and underground leaching of sulfide copper from low-grade and off-balance deposits. The integration of chemical, hydrodynamic, and biotechnological approaches creates the prerequisites for improving the completeness of copper recovery, promoting the rational use of mineral resources, and reducing the technogenic burden on the environment.

Keywords: *underground leaching; sulfide copper; borehole hydromining; acid reagent; moisture transfer; mass transfer*

1. Introduction

In the context of the growing global demand for nonferrous metals, the depletion of rich and easily accessible reserves, and increasingly stringent environmental requirements for mining operations, the development of resource-efficient and environmentally sound technologies for metal recovery from low-grade, off-balance, and technogenically disturbed deposits has become particularly important [1]-[3]. This is largely because the current mineral resource base is increasingly characterized by declining average grades of valuable components, more challenging mining and geological conditions, and a growing share of technogenically altered sites, for which conventional approaches often prove either insufficiently effective or economically burdensome [4]. Under these conditions, particular importance is attached to technological solutions capable of ensuring more complete utilization of mineral resources, reducing losses of valuable components, and mitigating the adverse environmental impacts of mining activities.

Conventional methods for the mining and processing of copper-bearing raw materials, based on surface or underground extraction followed by mineral beneficiation, are associated with substantial energy consumption, high capital investment, the generation of large volumes of waste, and, most importantly, a significant anthropogenic burden on the environment [5]-[8]. Another important component of conventional process flows is the thermal pretreatment and high-temperature processing of mineral and fuel raw materials, the

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efficiency of which depends strongly on material composition, the characteristics of thermochemical transformations, and the behavior of heat-treatment products [9]-[11]. In addition, where ore bodies exhibit complex morphology, low metal grades, or unfavorable hydrogeological conditions, the application of conventional process schemes may lead to higher production costs, lower recovery efficiency, and increased environmental risks [12]-[14]. For this reason, there is growing interest in alternative approaches that aim not only at metal extraction but also at aligning technological efficiency with the principles of sustainable resource management and environmental safety.

Accordingly, increasing attention has been given in international practice to geotechnological approaches to deposit development, particularly underground and heap leaching methods, which enable the use of raw materials that are unsuitable or only marginally viable for conventional mining [15], [16]. Unlike classical mining and metallurgical processing routes, these approaches rely on the direct interaction of reagents with the mineral matter either in situ or within a specially prepared medium, thereby creating opportunities to reduce stripping and haulage volumes, decrease the number of processing stages, and achieve more flexible control over valuable component recovery [17], [18]. At the same time, related areas of mining and technological research continue to advance approaches to the hydromechanical extraction of mineral raw materials, the control of process-flow parameters, and the prediction of the behavior of natural-technogenic systems [19]-[22].

Of particular interest is the use of acidic reagents in combination with oxidizing and biochemical factors that promote the intensification of sulfide copper mineral dissolution and increase the degree of copper recovery [23]-[26]. Contemporary research has also shown growing interest in electrochemical and membrane-based processes for the transformation and separation of metal-bearing systems, which broaden the understanding of possible mechanisms for enhancing the recovery of valuable components from mineral raw materials and pregnant solutions [27], [28]. This approach is regarded as a promising direction for improving the completeness of mineral resource utilization, reducing the extent of land disturbance, and mitigating the environmental risks associated with the accumulation of mining and metallurgical waste. At the same time, an important related area of current research is the minimization of the environmental consequences of mining operations and the management of technogenic formations and wastes [29], [30].

At the same time, the efficiency of underground leaching of sulfide copper is governed by the complex interplay of physicochemical, hydrodynamic, geomechanical, and biological processes occurring within the porous and fractured ore mass [31]-[34]. The choice of reagents, the conditions of their injection, the nature of flow and filtration through the rock medium, the kinetics of mineral dissolution, and the activity of microorganisms all have a substantial influence on mass-transfer intensity and the overall technological performance of the process. For this reason, one of the key scientific and practical challenges is the development of approaches to model moisture transfer and mass exchange of acidic reagents within the ore mass, enabling prediction of the behavior of the technological system and substantiation of rational operating parameters [35]-[37].

The complexity of sulfide copper leaching is primarily обусловлена the high chemical and structural stability of sulfide minerals, in which the transfer of metal into solution proceeds much more slowly than in the case of oxidized copper forms [38]. Unlike readily soluble mineral compounds, copper sulfides require not only an acidic medium but also a sufficiently high oxidizing potential to break down the mineral crystal lattice and convert the metal into a mobile form [39]. An additional complicating factor is the formation of secondary passivating films on grain surfaces, which hinder contact between the reagent and the mineral surface and reduce mass-transfer intensity [40]. Under natural conditions, process efficiency also depends strongly on ore granulometric composition, the porosity and permeability of the rock mass, the degree of fracturing, mineralogical heterogeneity, the solution filtration regime, and associated geochemical reactions, all of which complicate both the leaching process itself and its mathematical description [41]-[46]. Related studies [47]-[49] likewise emphasize the assessment of the rock mass structural state, raw-material preparation parameters, and processing conditions, further confirming the general importance of accounting for medium heterogeneity in modeling mining and geotechnical processes.

In the scientific literature, the leaching of copper-bearing raw materials is considered across several main research directions [50], [51]. In addition to leaching itself, studies on the processing of metal-bearing raw materials also address flotation, sulfidization, and other chemical and technological approaches aimed at improving the selectivity and completeness of valuable component recovery [52]-[55]. A substantial body of research is devoted to the chemical aspects of the process, particularly the selection of acidic and oxidizing reagents, the determination of their optimal concentrations, and the effects of solution acidity, temperature, redox potential, and contact time on the extent of mineral dissolution [56]-[59]. A separate line of research focuses on bioleaching, emphasizing the role of acidophilic microorganisms as catalysts for the oxidation of sulfide compounds and the regeneration of oxidizing agents in solution [60]. At the same time, ongoing studies seek to establish the governing patterns of filtration and mass transfer in porous media, as well as to develop mathematical models capable of describing reagent transport within the ore mass while accounting for its physical-mechanical and hydrogeological properties [61]. Several studies also highlight the value of integrating chemical, hydrodynamic, and biotechnological approaches to enhance copper recovery and intensify the overall process [62]-[64].

At the same time, an analysis of recent publications shows that the chemical, hydrodynamic, and biological aspects of underground sulfide copper leaching are most often investigated in isolation, without the development of an integrated model that accounts for the mass and moisture transfer of acidic reagents within a water-saturated or partially saturated ore mass [65], [66].

The influence of pore-space structure, suction pressure, variable moisture content, and nonlinear filtration parameters on the dynamics of moisture transfer under underground leaching conditions remains insufficiently studied [67]. In addition, the selection of reagents and the conditions of their application require further justification, taking into account not only their chemical activity but also their compatibility with the biochemical mechanisms of sulfide oxidation [68], [69]. For this reason, an important scientific objective is to develop an integrated approach to modeling moisture transfer of acidic reagents during the hydromining of sulfide copper, combining the physicochemical basis for solvent selection, the governing mass-transfer patterns in porous media, and the influence of biotechnological factors on metal recovery efficiency.

In Ukraine, sulfide copper deposits considered for underground leaching using borehole hydromining (BHM) are predominantly located within the Rivne-Volyn copper-bearing district. This district is situated in the southwestern part of the East European Platform (EEP), within the extensive Late Precambrian Volyn-Polissia trough. The area is characterized by the presence of copper-bearing trap complexes of the Lower Vendian, known as the Volyn Series, with a thickness exceeding 800 m. Geological exploration activities were concentrated within the Volyn ore district, where specialists of the State Northern Geological Enterprise (PDRGP "Pivnichgeologia") identified five ore-bearing fields: Tursko-Luhivske, Pivnichnohirnytske, Ratnivske, Katuske, and Bronnytske [70].

The key study object is the Kozlynychi sulfide copper deposit (13.97 km^2), located in the Volodymyrets district of the Rivne region, within the best-explored part of the Tursko-Luhivske ore-bearing field. Geological and mining analysis of the deposit structure showed that the host rocks for sulfide copper mineralization are predominantly siltstones and mudstones. This distinguishes the deposit from native copper occurrences, where the mineralization is typically hosted by basalts, basaltic tuffs, and lavaclastic breccias [71]-[73]. The thickness of the copper-bearing zone at the Kozlynychi deposit reaches several tens of meters, while the copper content ranges from 0.1 to 0.76%. The deposit is characterized by complex mining and geological conditions, including extremely high water abundance in both the productive and overlying strata. These conditions substantially complicate development by either open-pit or underground mining methods, making geotechnological approaches, particularly borehole hydromining combined with underground bacterial leaching, a promising option for its exploitation [74]-[78].

The geological map of the pre-Mesozoic surface of the Kozlynychi sulfide copper occurrence (Fig. 1) and the overview map showing the position of the Volyn copper ore district in the southwestern part of the East European Platform (Fig. 2) provide an important basis for understanding the geological structure of the area, the spatial setting of the study object, and the planning of technological operations.

The cartographic materials presented in Figure 2 and 1 illustrate the spatial and geological preconditions for the application of borehole hydromining and underground leaching within the Volyn copper ore district. This geological setting of the study area determines the heterogeneity of the filtration medium, the conditions governing the propagation of the acidic reagent through the rock mass, and, accordingly, the need for a mathematical description of mass- and moisture-transfer processes that accounts for the structural features of the ore-bearing rocks. Thus, an important scientific and practical task is to substantiate the selection of acidic reagents and to develop a mathematical description of moisture-transfer processes during the hydromining of sulfide copper.

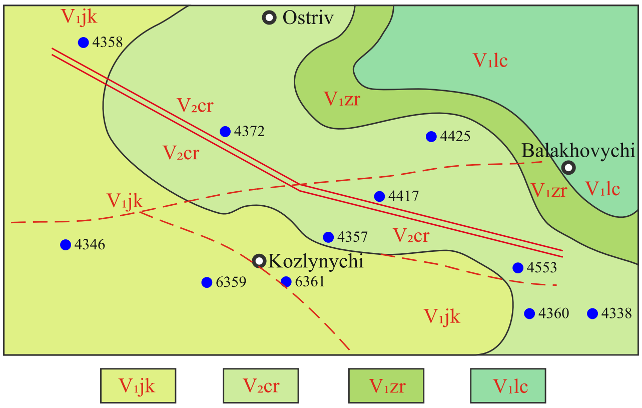
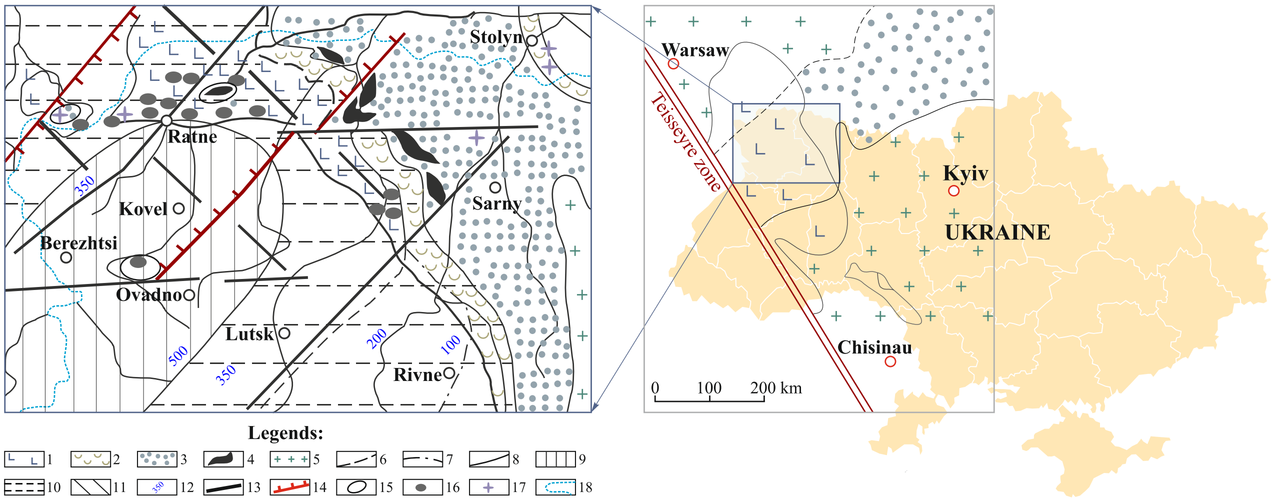


Figure 2. Geological map of the pre-Mesozoic surface in the area of the Kozlynychi sulfide copper occurrence



1 – predominantly basalts and lavaclastic breccias
2 – tuffs
3 – outcrops of Riphean deposits of the Polissya Series (Volyn-Orsha Aulacogen)
4 – outcrops of gabbro-dolerites
5 – Early Proterozoic crystalline basement
6-8 – distribution boundaries of Late Vendian trap complexes:
6 – numerous basalt flows; 7 – isolated basalt flows; 8 – top of the Volyn Series
9-11 – hydrothermal mineralization zones:
9 – prehnite;10 – zeolite; 11 – carbonate
12 – isopachs of the Volyn Series
13 – fault zones
14 – faults bounding the Central Belarusian suture zone of the crystalline basement
15 – cores of horst anticlines
16 – native copper occurrences
17 – copper sulfide occurrences
18 – state border of Ukraine

Figure 1. Schematic geological map of the Volyn copper ore district, showing its location within the southwestern part of the East European Platform and the outcrops of Lower Vendian rocks of the Volyn Series on the pre-Mesozoic surface

This study aims to model moisture transfer of acidic reagents in a porous ore medium, accounting for the reagents' physicochemical properties, mass-transfer parameters, and underground leaching conditions. To achieve this aim, the study analyzes the reagents used for sulfide copper leaching, characterizes the mining and geological conditions of the study site, and develops mathematical models of mass and moisture transfer.

2. Methods

The methodological framework of this study is based on an integrated analysis of the physicochemical, hydrodynamic, and biotechnological factors governing the efficiency of underground leaching of sulfide copper ores. The research combines theoretical analysis, mathematical modeling, and the generalization of experimental data obtained from studies of hydraulic erosion, leaching, and the biochemical oxidation of sulfide minerals. In developing the methodological approach, consideration was given to current research on in situ recovery schemes for sulfide copper ores, the modeling of coupled reactive transport processes, microbial transport in porous media, the identification of rate-limiting stages in bioleaching, the influence of ore-mass permeability on copper recovery efficiency, and the physicochemical features of chalcopyrite dissolution in ferric sulfate-sulfuric acid systems [79]-[84].

2.1. Technological scheme of the process

The overall technological scheme of underground sulfide copper leaching using borehole hydromining is shown in Figure 3. It involves injecting an acidic reagent through a system of injection wells into the ore-body zone, allowing it to flow through the ore mass and interact with the mineral matter, and recovering the pregnant solution through drainage wells. The process scheme also includes preparing and adjusting the working solution composition, reagent circulation, and subsequent copper recovery from the pregnant solutions.

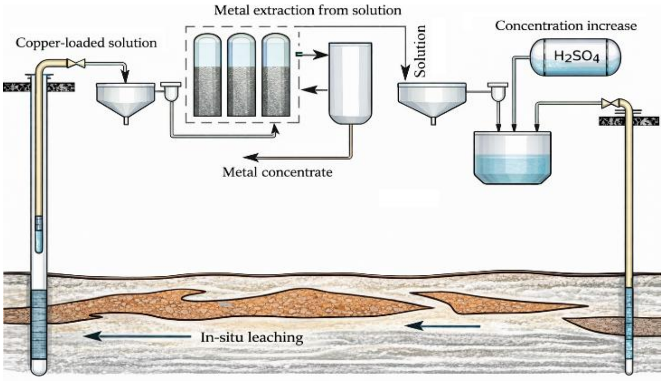


Figure 3. Schematic diagram of underground sulfide copper leaching using borehole hydromining

The efficiency of this technology is determined by the combined action of physical, chemical, hydrodynamic, and biological processes occurring in the porous ore medium. Accordingly, the methodological framework of this study includes a separate consideration of reagent selection, the mathematical description of moisture transfer in the rock mass, and the role of biotechnological factors in the oxidation of sulfide minerals.

2.2. Selection of reagents for sulfide copper leaching

The choice of reagents for underground sulfide copper leaching was substantiated with due regard to their chemical activity, selectivity toward ore minerals, oxidizing capacity, and compatibility with biochemical oxidation mechanisms. Particular attention was given to sulfuric acid, iron salts, and biological agents involved in the transformation of sulfide compounds into soluble forms.

To evaluate the acid-base properties of the medium, the Hammett acidity function, H_0, was used, as it allows characterization of the proton-donating ability of concentrated acidic solutions. It is defined as follows:

$$H_0 = -\lg\left(a_{H^+}\frac{f_{BH^+}}{f_B}\right) = -\lg\left(K_{BH^+}\frac{C_{BH^+}}{C_B}\right), \qquad (1)$$

where:

a_H – the proton activity in the given medium;

K_{BH}^+ – the ionization constant of the indicator;

C_{BH}^+, C_B – the concentrations of the ionized and non-ionized forms of the indicator, respectively;

f_{BH}^+, f_B – the activity coefficients of the fully ionized and non-ionized forms of the indicator, respectively.

The Hard and Soft Acids and Bases (HSAB) principle was also used to explain solubility behavior. In this framework, dissolution of an MX crystal is considered an acid-base reaction:

$$MX + a\,\mathrm{H_2O} \rightarrow M[aq]^+ + X[aq]^- . \qquad (2)$$

According to this approach, minimum solubility is observed when the crystalline salt is formed by either a hard cation-hard anion pair or a soft cation-soft anion pair.

The properties of the principal reagents involved in underground sulfide copper leaching were evaluated. It was established that using sulfuric acid as the primary reagent ensures intensive copper dissolution, particularly from oxidized minerals, owing to a decrease in solution pH and the resulting increase in hydrogen-ion activity. Humic substances, together with iron salts (Fe^{2+}, Fe^{3+}), catalyze redox reactions in solution, accelerate the transformation of sulfide compounds into soluble forms, and at the same time contribute to the stabilization of the chemical composition of the pregnant solution through the formation of complex compounds.

Thionic bacteria (*Thiobacillus ferrooxidans*, *T. thiooxidans*) act as biological catalysts in the oxidation of sulfide minerals in an acidic medium [85]. Their activity increases copper recovery by 10-15% compared with purely chemical leaching, which confirms the expediency of using a biochemical approach to intensify metal extraction processes.

The key chemical equations describing sulfide oxidation and the regeneration of the oxidizing agent Fe^{3+} are as follows:

1) pyrite oxidation, with pyrite being a typical associated mineral in sulfide copper ores, leads to the formation of sulfuric acid:

$$\mathrm{FeS_2 + 3.5\,O_2 + H_2O \rightarrow FeSO_4 + H_2SO_4}\,; \qquad (3)$$

2) a key stage of bioleaching is the biocatalytic oxidation of ferrous iron (Fe^{2+}) to ferric iron (Fe^{3+}) by thionic bacteria, primarily *T. ferrooxidans*:

$$\mathrm{4\,FeSO_4 + 2\,H_2SO_4 + O_2 \overset{Th.fer.}{\rightarrow} 2\,Fe_2(SO_4)_3 + 2\,H_2O}\,; \qquad (4)$$

3) ferric iron (Fe^{3+}) acts as the principal chemical oxidant responsible for the dissolution of copper sulfide (CuS):

$$2CuS + 2Fe_2(SO_4)_3 + 2H_2O + 3O_2 \rightarrow \\ \rightarrow 2CuSO_4 + 4FeSO_4 + H_2SO_4. \tag{5}$$

A comparative assessment of the main reagents used in underground sulfide copper leaching was conducted with respect to their functional roles, application conditions, and expected influence on process performance. The generalized characteristics of these reagents are presented in Table 1.

Table 1. Characteristics of reagents used for underground leaching of sulfide copper

Solvent/agent	Optimal parameters	Main effect on the process
Sulfuric acid (H_2SO_4)	Concentration: 5-10%	Intensive dissolution of oxidized minerals; reduction of solution pH
Iron salts (Fe^{2+}, Fe^{3+})	Fe^{2+} up to $0.1 \cdot 10^{-7}$ g-ion/m^3	Sulfide oxidation, reaction catalysis, stabilization of solution composition
Humic substances	$1\text{-}5 \cdot 10^7$ g/m^3	Activation of biochemical processes; improvement of conditions for bacterial growth
Thionic bacteria (*Thiobacillus*)	Concentration: 10^6-10^7 cells/mL; pH = 1.5-1.8; T = 25-32°C	Catalysis of sulfide mineral oxidation; increase in copper recovery by 10-15%

2.3. Mathematical modeling of moisture and mass transfer

The hydromining stage, which forms part of the overall process scheme, ensures the formation of an optimal granulometric composition of the disintegrated ore within the extraction chamber, with a characteristic particle size of approximately 0.35 mm. Achieving this size fraction maximizes the contact area between sulfide copper minerals and the working agent. Experimental studies on the disintegration of copper-bearing rock were conducted in the Department of Mineral Deposit Mining's laboratory at the National University of Water and Environmental Engineering using a dedicated test bench. In addition, field investigations of sulfide copper recovery using bioreagents were conducted in situ at the Kozlynychi deposit.

To describe mass-transfer processes in a porous medium, a modified diffusion equation was used that accounts for the effects of rock volumetric moisture content, permeability and filtration coefficients. The classical diffusion equation served as the basic framework:

$$\frac{dC}{dt} = D\frac{d^2c}{d^2x}, \tag{6}$$

where:

C – the concentration of the dissolved substance;

t – time;

D – the diffusion coefficient, which depends on the physical-mechanical properties of the rock and its moisture content.

In this study, the equation was further developed using relationships that account for the nonlinear dependence among volumetric moisture content, suction pressure, and moisture-transfer coefficients [86]. The resulting models enable the description of the actual behavior of solvent transport in porous rocks during underground leaching of sulfide copper.

To model the movement of the hydraulic mixture within the extraction chamber, dynamic similarity criteria were applied. The generalized dimensionless relationship for the process is expressed as follows:

$$\Phi(Fr, \mathrm{Re}) = 0, \tag{7}$$

where:

Fr – the Froude number;

Re – the Reynolds number.

Taking into account that, under laboratory conditions, the flow was considered within the turbulent regime, the Froude number was adopted as the governing similarity criterion:

$$Fr = \frac{v}{\sqrt{g \cdot l}} = idem, \tag{8}$$

where:

v – the average flow velocity;

g – the acceleration due to gravity;

l – the characteristic linear dimension.

To describe nonlinear moisture transfer in copper-bearing rock, a differential equation based on the law of mass conservation was used, in which the volumetric moisture content and the moisture-transfer coefficient were treated as functions of suction pore pressure:

$$\frac{\partial W_p(P_{BC})}{\partial t} = \frac{\partial}{\partial y}\left[\frac{K_p(P_{BC})}{\gamma_p}\frac{\partial P_{BC}}{\partial y}\right] + \\ + \frac{\partial}{\partial x}\left[\frac{K_p(P_{BC})}{\gamma_p}\frac{\partial P_{BC}}{\partial x}\right] + \frac{\partial K_p(P_{BC})}{\partial y} + I(t, x, y), \tag{9}$$

where:

v – $W_p(P_{BC})$ – the volumetric moisture content;

$K_p(P_{BC})$ – the moisture-transfer coefficient;

P_{BC} – the suction pore excess pressure of moisture;

γ_p – the specific weight of moisture;

$I(t, x, y)$ – the intensity of soil moisture exchange.

To account for the pronounced nonlinearity of the parameters, empirical relationships were used to describe the dependence of volumetric moisture content and the moisture-transfer coefficient on suction pressure (P_{BC}).

$$W_p(P_{BC}) = W_{\max}e^{-\alpha_{W1}P_{BC}}; \tag{10}$$

$$K_p(P_{BC}) = K_e e^{\alpha_e P_{BC}}, \tag{11}$$

where:

$W_{\max}$ – the maximum moisture content at $P_{BC} = 0$;

K_e – the constant coefficient under full saturation;

a_{W1}, a_e – constant dimensionless coefficients.

The developed mathematical models provide an integrated description of the principal processes governing underground sulfide copper leaching. In particular, the models enable the reproduction of the movement of the hydraulic mixture along the bottom of the erosion chamber, accounting for the influence of flow hydrodynamics, pore-space structure, and ore granulometric composition [87]. In addition, they allow quantification of the dependence of moisture-transfer coefficients on the physical-mechanical characteristics of the medium, such as permeability, porosity, density, and suction pressure, thereby enabling the prediction of the rock mass's filtration properties [88]. A separate modeling approach was also developed for mineral dissolution kinetics, incorporating the effects of particle size, reagent concentration, and biological factors, particularly the activity of thionic bacteria. The application of these models makes it possible to interpret mass- and moisture-transfer processes and to use the results for the further substantiation of the technological parameters of underground leaching.

2.4. Biotechnological component

The biotechnological component of the study examined the role of thiobacteria in oxidizing sulfide minerals and regenerating oxidizing agents in an acidic medium (pH 1.5-1.8) at 25-32°C. The analysis considered the operating conditions of acidophilic microorganisms, particularly the ranges of acidity and temperature characteristic of sulfide copper bioleaching processes. Within this framework, experimental data on bacterial activity under different environmental conditions were generalized, and their effect on intensifying the oxidation of sulfide compounds was evaluated. The biotechnological component was treated as part of an integrated system in which chemical reagents, mass-transfer parameters, and microbiological factors jointly determine leaching efficiency.

3. Results and discussion

The results of the study showed that the efficiency of underground leaching of sulfide copper is governed by the combined action of physical, chemical, hydrodynamic, and biological factors. The most significant influences on process performance are exerted by the granulometric composition of the disintegrated ore, the intensity of mass transfer at the solid-liquid interface, the concentration of the acidic reagent, and the conditions of biochemical oxidation of sulfide minerals.

3.1. Influence of ore granulometric composition

It was established that the rational particle size of the disintegrated ore is about 0.35 mm, since this size provides a sufficiently developed mineral-solvent contact surface and creates favorable conditions for intensifying mass transfer. Such a granulometric composition can be achieved by hydraulic erosion using a 25 mm nozzle at 1.6 MPa. Experimental results showed that under these conditions, and at an erosion rate of 1.4 m/s, the consistency of the hydraulic mixture at the outlet of the hydraulic elevator does not exceed 10-14%. This operating regime promotes more intensive mass exchange in the erosion chamber and creates the conditions for more uniform penetration of the reagent into the ore mass.

This result is of considerable technological importance, since excessively coarse particles reduce the specific surface area available for interaction between the minerals and the solvent and, accordingly, slow down the leaching process. In contrast, excessively fine particles may impair the medium's filtration characteristics and hinder the movement of pregnant solutions. Thus, a particle size of about 0.35 mm should be regarded as a compromise between intensifying dissolution and maintaining acceptable filtration conditions.

3.2. Dissolution kinetics

Analysis of mineral dissolution kinetics in the ore-solvent system showed that the character of the relationships differs substantially between oxidized and sulfide copper forms, reflecting the differences in their mechanisms of interaction with the reagent. For oxidized minerals, an almost linear relationship was observed between dissolution rate and pulp agitation intensity, indicating the predominance of a diffusion-controlled mass-transfer mechanism at the solid-liquid interface. As the solution flow velocity increases, the diffusion layer thickness decreases, thereby accelerating the transfer of copper ions into solution.

At the same time, the dependence of the dissolution rate of oxidized minerals on sulfuric acid concentration is logarithmic in nature. This indicates the existence of a concentration range beyond which a further increase in acidity no longer results in a proportional increase in the process rate. From a practical standpoint, this feature is important because it shows that an unlimited increase in reagent consumption is not justified in the absence of a corresponding gain in technological performance.

For sulfide copper minerals, particularly chalcopyrite, the kinetic behavior proved considerably more complex. As shown in Figures 4 and 5, the dissolution rate of chalcopyrite is described by a third-order polynomial relationship with pulp agitation intensity.

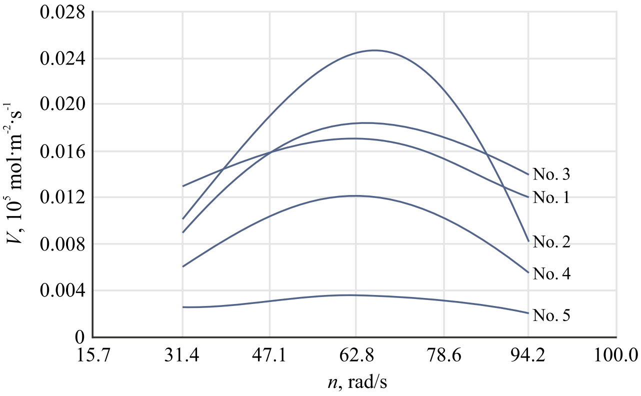


Figure 4. Dependence of the chalcopyrite dissolution rate on agitation intensity

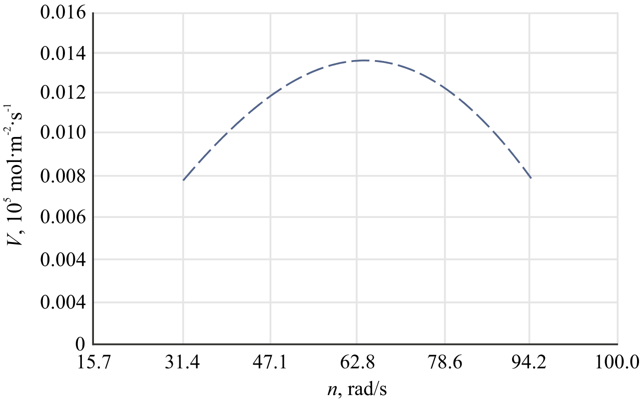


Figure 5. Generalized model of the dependence of the chalcopyrite dissolution rate on agitation intensity

This pattern reflects the multistage nature of the oxidation process in a porous medium, where chemical, diffusive, and microbiological limitations act simultaneously. One factor contributing to the complexity of the kinetics is the formation of passivating sulfur-bearing films on the mineral surface, which reduces the intensity of direct interaction between the reagent and the solid phase. Thus, comparing the obtained relationships shows that, for oxidized copper forms, the key factor in process intensification is the enhancement of external mass transfer, whereas, for sulfide minerals, a much more complex dissolution mechanism involving a combination of diffusive, chemical, and biochemical stages must be accounted for. This confirms the expediency of a differentiated approach to selecting leaching conditions based on the ore's mineralogical composition.

3.3. Mathematical modeling of moisture transfer

The developed mathematical model of solvent filtration in the rock mass accounts for the interrelationships among volumetric moisture content, suction pressure, and the porous medium parameters that govern the movement of the liquid phase. The model incorporates relationships that describe the temporal and spatial variation of filtration and moisture-transfer coefficients under the action of external hydrodynamic forcing. This makes it possible to evaluate the rate of reagent penetration into rocks of different permeability, analyze the influence of rock-mass structural heterogeneity and water saturation on leaching efficiency, and substantiate solution-injection regimes that ensure a more uniform distribution of the reagent within the leaching zone.

The generalized relationship obtained from the modeling results is as follows:

$$V = -5 \cdot 10^{-9} n^3 - 5 \cdot 10^{-6} n^2 + 0.009n - 0.009\,, \tag{12}$$

where:

V – the filtration rate or the rate of reagent movement in the porous medium;

η – the parameter characterizing the state of the medium or the process regime.

Analysis of the obtained relationship indicates a nonlinear variation in the filtration rate, suggesting complex interactions among capillary, filtration, and hydrodynamic factors within the ore mass. This behavior of the curve confirms that the intensity of reagent transport is governed not by a single parameter, but by their combined effect, including porosity, permeability, moisture content, and suction pressure.

Model verification by comparison with experimental data demonstrated its sufficient accuracy for practical application in the design calculations of borehole hydromining process schemes. The coefficient of determination ($R^2 = 0.997$) indicates a high degree of agreement between the calculated and experimental results, allowing the model to predict reagent propagation conditions within the rock mass and to assess the factors limiting mass-transfer intensity.

3.4. Mechanism of biochemical sulfide oxidation

The bacterial oxidation of sulfide minerals proceeds through a mechanism similar to electrochemical corrosion, in which microorganisms act as catalysts of oxidative processes. In this case, the sulfide mineral serves as an electron donor and performs the anodic function. In contrast, the bacterial cell participates in electron transfer and, in effect, performs the cathodic function. This interaction promotes oxidation and the gradual breakdown of the sulfide mineral.

The study showed that the process can proceed via two principal pathways. The first involves direct sorption of the bacterial cell onto the mineral surface, where oxidation occurs at the point of contact. The second is an indirect mechanism, in which bacteria catalyze the oxidation of ferrous iron to ferric iron, and Fe^{3+} ions subsequently oxidize the sulfide mineral chemically within the diffusion layer. It is the combination of these two mechanisms that determines the intensity of the biochemical degradation of the sulfide phase.

The electrochemical reactions accompanying the oxidation of arsenopyrite (FeAsS) lead to the formation of Fe^{2+}, As^{3+}, and sulfur compounds in the anodic zone. In the presence of bacteria, these products undergo further oxidation: Fe^{2+} is converted to Fe^{3+}, while sulfur-bearing compounds are oxidized to sulfate forms. This helps maintain the oxidizing potential of the system and intensifies the further dissolution of sulfide minerals (Fig. 6).

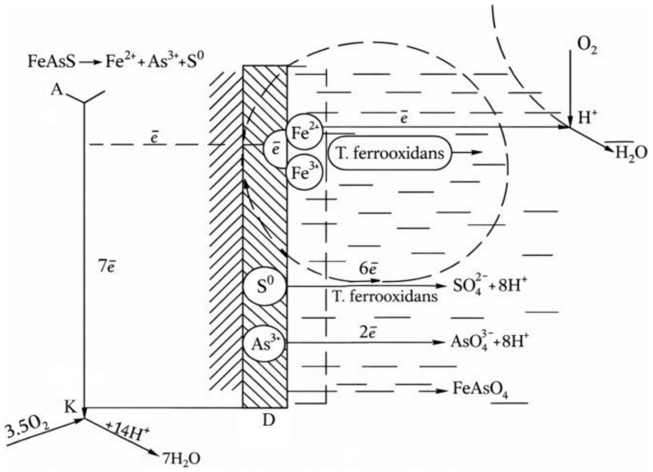


Figure 6. Scheme of bacterial oxidation of arsenopyrite: A – the anode; K – the cathode; D – the diffusion layer

From an energetic standpoint, the bacteria utilize the electrons released during the oxidation of sulfide matter or ferrous iron. In this process, the electron is accepted by the copper-containing protein rusticyanin, which then transfers it through the cytochrome system. Electron transport is accompanied by proton translocation across the membrane, generating the electrochemical gradient required for adenosine triphosphate synthesis. These processes are schematically illustrated in Figure 7.

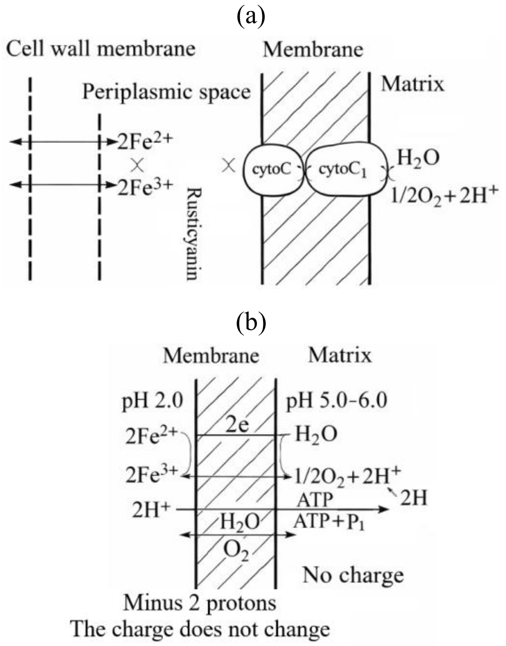


Figure 7. Oxidation of ferrous iron by T. ferrooxidans: (a) scheme of electron transport; (b) scheme of ATP generation

Thus, the role of thionic bacteria in underground leaching processes is not limited to accelerating individual oxidation reactions; they also help maintain the circulation of oxidizing agents within the system. This creates conditions for a more intensive biochemical oxidation of sulfide minerals and, accordingly, for improved copper recovery from sulfide raw materials.

3.5. Practical aspects

The synthesis and analysis of the results enabled the establishment of the roles of the principal reagents used in underground sulfide copper leaching and the assessment of their functional purpose, the ranges of appropriate operating parameters, and their influence on the technological performance of the process. To systematize these data, a comparative analysis of the reagents was carried out with due regard to their chemical activity, ability to sustain oxidation processes, compatibility with microbiological mechanisms, and influence on filtration conditions and solution penetration into the ore mass. The generalized results are presented in Table 2. As shown by the presented data, the most promising approach is the combined use of sulfuric acid, iron(III) sulfate, and thionic bacteria, as this combination provides an integrated chemical-biological mechanism for sulfide mineral dissolution. Sulfuric acid establishes the required acidity of the medium and promotes the transfer of copper into solution, iron(III) sulfate acts as a secondary oxidizing agent, and the bacteria sustain the regeneration of oxidizing agents while intensifying the oxidation of the sulfide phase.

Table 2. Optimal parameters of the principal reagents used in the underground leaching of sulfide copper

No.	Reagent type/name	Main purpose	Optimal application parameters	Effect on technological performance
1	Sulfuric acid (H_2SO_4)	Primary leaching reagent for transferring copper into solution	Concentration: 5-10%; pH = 1.5-2,0; Temperature: 20-35°C	Intensifies the dissolution of oxidized and, to some extent, sulfide minerals; increases Cu^{2+} recovery to 85-90%
2	Iron(III) sulfate ($Fe_2(SO_4)_3$)	Secondary oxidant; catalyzes reactions involving the Fe^{3+}/Fe^{2+} redox pair	Concentration: 1-3 g/L; pH = 1.8-2.5	Increases the oxidation rate of sulfide minerals and stabilizes the bioleaching process
3	Thionic bacteria (*Thiobacillus ferrooxidans*)	Biological catalyst of sulfide oxidation processes	Concentration: 10^6-10^7 cells/mL; Temperature: 25-32°C; pH = 1.5-2.0	Accelerate the biochemical oxidation of sulfides and increase Cu recovery by 10-15%
4	Humic substances (organic complexes)	Solution stabilizers and regulators of the redox state of the medium	Content: 0.1-0.3%; pH = 2-3	Reduce solution toxicity to microorganisms and improve the stability of the biocenosis
5	Oxidizing agents (H_2O_2, $NaClO_3$)	Auxiliary oxidants at the initial stage of leaching	Concentration: 0.5-2%; Contact time 2-4 h	Ensure rapid breakdown of passivating films on mineral surfaces
6	Surfactants	Improvement of wetting and permeability of the ore mass	Concentration: 0.01-0.05%; Temperature: 20-30°C	Promote more uniform reagent distribution and reduce hydrodynamic resistance
7	Wash water (H_2O)	Neutralization and removal of residual ions after leaching	Volume: 1.0-1.5 m^3 per 1 m^3 of ore	Ensures cleaning of the ore mass and reduces secondary metal precipitation

Auxiliary reagents, including humic substances, oxidizing agents, and surfactants, do not govern the process independently; however, they can significantly affect its stability and completeness. Their use is advisable when it is necessary to reduce mineral-surface passivation, improve wetting of the ore mass, stabilize the biochemical environment, or enhance uniformity of reagent distribution within the leaching zone.

The results obtained in this study regarding the expediency of the combined use of sulfuric acid, iron(III) sulfate, and thionic bacteria are in good agreement with current understanding of the leaching mechanism of chalcopyrite and other sulfide copper minerals. In particular, Córdoba et al. [89] showed that, in an acidic medium, the ferric sulfate-sulfuric acid system is one of the fundamental systems for the oxidative dissolution of chalcopyrite, while the process rate is governed by oxidant concentration, pH, temperature, and surface passivation phenomena. A similar conclusion was reached by Li et al. [90], who noted that the ferric sulfate-sulfuric acid system, including in the presence of acidophilic microorganisms, represents one of the most promising routes for the hydrometallurgical recovery of copper from low-grade sulfide raw materials. In this context, the recommended application parameters established in the present study for H_2SO_4, $Fe_2(SO_4)_3$, and *Thiobacillus ferrooxidans* with the available literature, while providing a more specific basis for underground leaching and borehole hydromining conditions.

The conclusion regarding the decisive influence of ore granulometric composition and external mass transfer on leaching intensity is also consistent with the findings of other researchers. In the present study, a particle size of about 0.35 mm was shown to be rational, as it provides a compromise between increasing the specific reaction surface area and maintaining acceptable filtration characteristics of the medium. A similar pattern was reported by Kang et al. [91] in their study of copper ore leaching, where a reduction in particle size was shown to intensify copper recovery, whereas an increase in particle size slowed the process and reduced its efficiency. Although the results reported by those authors relate primarily to the heap leaching of oxidized ores, the observed trend is consistent with our findings and confirms that excessively coarse particles impair dissolution kinetics, while excessive size reduction in real filtration systems may adversely affect the permeability of the ore mass.

Of practical importance is our finding that additional reagents, such as oxidizing agents and surfactants, play an auxiliary yet significant role in reducing mineral-surface passivation and improving reagent penetration into the ore mass. This conclusion is supported by the study of Zhang et al. [92], which showed that the addition of the nonionic surfactant Triton X-100 increased copper recovery during chalcopyrite bioleaching by improving surface hydrophilicity, enhancing bacterial adhesion, and mitigating the effect of passivating sulfur-bearing species. Consistent with this, Nicol [93] demonstrated that hydrogen peroxide can serve as an effective oxidant in acidic sulfate solutions, although its performance depends strongly on system composition and process conditions. In addition, Sokić et al. [94] noted that increasing the concentrations of H_2SO_4 and H_2O_2 accelerate chalcopyrite leaching, although the effect is governed by kinetic limitations and the mineralogical characteristics of the raw material. Thus, our results confirm that auxiliary reagents should not be regarded as an alternative to the core acid-Fe(III)-bacteria system; rather, they represent an effective means of controlling passivation, wettability, and process stability under the complex conditions of underground leaching.

Overall, the results of this study confirm the expediency of combining chemical, physicomechanical, and biotechnological approaches in underground sulfide copper leaching. Such an approach creates the prerequisites for improving metal recovery efficiency, achieving more complete utilization of mineral resources, and reducing the environmental risks associated with the development of complex sulfide ores.

4. Conclusions

Approaches to the selection of acidic reagents for underground sulfide copper leaching were substantiated with due regard to the physicochemical characteristics of the system and the role of biological agents in the oxidation of sulfide minerals. It was shown that the use of acidic reagents in combination with thionic bacteria is advisable for intensifying the leaching process and increasing the degree of copper recovery.

It was established that process efficiency depends substantially on ore granulometric composition, the hydrodynamic parameters of hydraulic erosion, and the conditions governing mass-transfer processes. The rational parameters for process implementation were found to be a disintegrated ore particle size of about 0.35 mm, a hydraulic erosion pressure of 1.6 MPa, a nozzle diameter of 25 mm, an H_2SO_4 concentration in the range of 5-10%, a medium pH of 1.5-1.8, and a temperature of 25-32°C.

Mathematical models of moisture transfer were developed that take into account volumetric moisture content, permeability, porosity, and suction pressure, and make it possible to describe reagent transport in a porous ore medium. Model verification showed high agreement with experimental data, confirming the possibility of its application for predicting filtration conditions and substantiating the parameters of underground leaching.

It was shown that mineral dissolution kinetics are governed by mineralogical nature: for oxidized copper forms, a diffusion-controlled mass-transfer regime predominates, whereas sulfide minerals are characterized by a more complex multistage mechanism associated with the combined action of chemical, diffusive, and biochemical processes.

The obtained results can be used to substantiate and design technological schemes for borehole hydromining and underground leaching of sulfide copper. The integration of chemical, hydrodynamic, and biotechnological approaches creates the prerequisites for improving the completeness of metal recovery, promoting the rational use of the mineral resource base, and reducing the technogenic burden on the environment.

Author contributions

Conceptualization: ZM, VK; Data curation: VM, YM, OV; Formal analysis: VS, VZ; Investigation: ZM, VK, VZ; Methodology: ZM, VK; Project administration: VM; Resources: YM, OV; Supervision: ZM; Validation: VK, VS; Visualization: VZ, OV; Writing – original draft: ZM, VM, YM, VK; Writing – review & editing: VK, VS, VZ, OV. All authors have read and agreed to the published version of the manuscript.

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Conflicts of interest

The authors declare no conflict of interest.

Data availability statement

The original contributions presented in the study are included in the article, further inquiries can be directed to the corresponding author.

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Моделювання процесу вологопереносу кислотних реагентів при гідровидобутку сульфідної міді

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Мета. Обґрунтувати вибір кислотних реагентів і розробити математичний опис процесів вологопереносу при гідровидобутку сульфідної міді з урахуванням фізико-хімічних властивостей реагентів, параметрів пористого рудного середовища та біологічних факторів окиснення сульфідних мінералів.

Методика. В роботі використано поєднання теоретичного аналізу, математичного моделювання та узагальнення експериментальних даних. Вибір реагентів, зокрема сірчаної кислоти та солей заліза, обґрунтовано із застосуванням функції кислотності Гаммета (H_0) та принципу жорстких та м'яких кислот і основ. Для опису процесів масо- та вологопереносу використано модифіковані рівняння дифузії і фільтрації, що враховують об'ємний вологовміст, проникність та всмоктуючий тиск. Гідродинамічні умови руху гідросуміші оцінювали за критеріями Фруда та Рейнольдса.

Результати. Встановлено, що ефективність підземного вилуговування сульфідної міді визначається сукупним впливом гранулометричного складу руди, гідродинамічних параметрів, концентрації реагенту та біохімічних факторів окиснення. Раціональними параметрами процесу є крупність руди близько 0.35 мм, тиск гідравлічного розмиву 1.6 МПа, концентрація H_2SO_4 у межах 5-10%, pH середовища 1.5-1.8 та температура 25-32°C. Розроблені математичні моделі дають змогу описувати динаміку вологопереносу в пористому середовищі та прогнозувати характер переміщення кислотного реагенту в рудному масиві.

Наукова новизна. Розроблено комплексний підхід до моделювання процесу вологопереносу кислотних реагентів при гідровидобутку сульфідної міді, який поєднує фізико-хімічне обґрунтування вибору реагентів, опис гідродинамічних умов руху гідросуміші та врахування біотехнологічних факторів окиснення сульфідів. На відміну від наявних підходів, запропонована модель у єдиній постановці враховує об'ємний вологовміст, проникність і всмоктуючий тиск пористого середовища.

Практична значимість. Отримані результати можуть бути використані при обґрунтуванні та проєктуванні технологічних схем свердловинного гідровидобутку і підземного вилуговування сульфідної міді з бідних та забалансових родовищ. Поєднання хімічних, гідродинамічних і біотехнологічних підходів створює передумови для підвищення повноти вилучення міді, раціонального використання мінерально-сировинної бази та зниження техногенного навантаження на довкілля.

Ключові слова: *підземне вилуговування; сульфідна мідь; свердловинний гідровидобуток; кислотний реагент; вологоперенос; масоперенос*

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