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Review Article

Biooxidation of refractory silver minerals as pretreatment for silver recovery

Biooxidación de minerales sulfurados argentíferos como pretratamiento para la recuperación de plata

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ABSTRACT

The worldwide demand for silver is increasing due to the technological development in different areas, especially for the requirements in the production of renewable energies; therefore, for the treatment of refractory minerals by conventional processes, it is necessary to study mechanisms to identify the viability of their processing and the feasibility of their recovery, ensuring profitability. Gold and silver present in the matrix of sulfide-based ores are not affordably recovered by the conventional method involving leaching in sodium cyanide solution. Therefore, it must be subjected to oxidation pretreatment or conversion of the sulfides to simple compounds that allow recovery of the precious metals. The methods of oxidation of minerals considered refractory are carried out by hydro and pyrometallurgical methods, seeking the dissolution of sulfur, iron, arsenic and other elements present. It is important to identify and consider the mineralogical structure, microbiological variety, physicochemical characteristics, types of reactors, bacterial interactions, among other factors. The choice of biooxidation as the preferred process in projects for the recovery of gold, silver, copper or other metals, in most cases is based on the economic advantage, compared to the conventional processes of roasting, chemical leaching and pressurized leaching. The biooxidation of refractory argentiferous ores has been extensively studied in recent years, concluding that its application is feasible, mainly because it is environmentally friendly.

Keywords: Biooxidation, silver, refractory minerals, biohydrometallurgy, sulfide minerals.

RESUMEN

La demanda mundial de plata está aumentando debido al desarrollo tecnológico en diferentes áreas, especialmente por los requerimientos en la producción de energías renovables; por lo tanto, para el tratamiento de minerales refractarios por procesos convencionales, es necesario estudiar mecanismos para identificar la viabilidad de su procesamiento y la factibilidad de su recuperación, asegurando la rentabilidad. El oro y la plata presentes en la matriz de los minerales de sulfuros de metales base no se recuperan de forma adecuada por el método convencional de lixiviación en solución de cianuro de sodio. Por lo tanto, deben someterse a un pretratamiento de oxidación o conversión de los sulfuros en compuestos simples que permitan la recuperación de los metales preciosos. Los métodos de oxidación de los minerales considerados refractarios se realizan por métodos hidro y pirometalúrgicos, buscando la disolución del azufre, hierro, arsénico y otros elementos presentes. Es importante identificar y considerar la estructura mineralógica, variedad microbiológica, características fisicoquímicas, tipos de reactores, interacciones bacterianas, entre otros factores. La elección de la biooxidación como proceso preferido en proyectos de recuperación de oro, plata, cobre u otros metales, en la mayoría de los casos se basa en la ventaja económica, en comparación con los procesos convencionales de tostación, lixiviación química y lixiviación presurizada. La biooxidación de minerales argentíferos refractarios ha sido ampliamente estudiada en los últimos años, concluyéndose que su aplicación es factible, principalmente por ser amigable con el medio ambiente.

Palabras claves: Biooxidación, plata, minerales refractarios, biohidrometalurgia, minerales sulfurados.

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I. INTRODUCTION

Gold and/or silver refractory ores, are those that subjected to direct cyanidation have low recoveries; originating the need to implement a previous processing or pretreatment with the ability to contribute to the prevention, detection and remediation of environmental pollution and waste degradation, it is a sustainable way to develop clean processes and products (Singh R., 2017). Refractory sulfide minerals (RSM) of gold and silver have generally been subjected to processes such as roasting, flotation and leaching, with unsatisfactory results. Over time, the growing consumption of metals and alloys by mankind has driven the search for and development of technologies for the beneficiation of difficult to treat or complex mineral deposits (Sánchez-Yañez, 2006). Currently, four industrial processes are used to recover precious metals from sulfide ores: pressure oxidation, roasting, biooxidation and cyanidation.

Bioleaching is an economical, efficient technique and an environmentally friendly alternative, used for the extraction of valuable minerals from low-grade ores (Gahan et al., 2012; Erüst et al., 2013), which involves a biphasic suspension of solid particles in a liquid medium (Srichandan et al, 2019), where chemolithotrophic bacteria are indirectly involved in the oxidation of ferrous ion (Fe2+) to ferric ion (Fe3+). An example of this group are bacteria of the genus Acidithiobacillus that are present in ore deposits (Fowler et al, 1999).

The treatment in continuous and discontinuous modalities, the variables that are generally subjected to consideration and study are the pulp density, Fe2+ concentration, pH, bacterial population, sulfides, arsenides, among others. Bacterial leaching or bioleaching by means of chemolithotrophic bacteria is an alternative due to its capacity to oxidize sulfur and iron contained in the mineral sulfides, facilitating the dissolution of metals and/or the liberation of others. Consequently, recovery rates are increased (Asamoah et al, 2018).

The application of traditional technologies has relative efficiency, high cost and unfortunately environmental damage; therefore, the use of biotechnological alternatives such as bacterial leaching to oxidize sulfur and iron from sulfide ores facilitate the extraction of commercially valuable metals, at low cost, in an environmentally friendly manner and without polluting the environment (Maluckov, 2017). Biomining methods are affordable, non-toxic, effective and also environmentally friendly (Mahajan et al, 2017).

1.1 Is there interest in biohydrometallurgical processes?

There are numerous biohydrometallurgical methods that have allowed the recovery of copper and the liberation of gold; mainly through the action of acidithiobacillus chemolithotrophic bacteria and other sulfide reducing bacteria to extract metals from ores, concentrates, tailings and residues. Biohydrometallurgy has contributed to alleviate the problems related to the depletion of high grade mineral resources, transforming secondary or marginal mineral resources into reserves. Processing techniques range from leaching in reactors, ponds, heaps and dumps. The current focus is on the treatment of high grade ores or concentrates.

To oxidize refractory gold concentrate with high sulfur and arsenic contents, a chemical-biological process is planned. High temperature chemical oxidation includes roasting and pressure leaching, and biological oxidation or microbiological oxidation.

Capital costs in the application of mining bioprocesses are reduced due to the simplicity of biooxidation plants which consist of fewer unit processes compared to conventional smelting plants, which generally require a series of sequential operations. By using a combination of biooxidation and biocyanidation processes, total capital costs could potentially be greatly reduced provided the optimal system design and potential microorganisms are in place (Karthikeyan et al, 2015).

The impediments or difficulties in the extraction of gold and silver from their sources, which determines refractoriness are mainly: low dissolution kinetics due to the presence of sulfides and sulfosalts associated with iron, leachate adsorption due to the presence of carbonaceous material, high reagent consumption due to the existence of cyanide compounds and the abundance of base metals such as sulfides of copper, zinc, nickel and others.

In the case of the presence of sulfides it is common to carry out oxidation pretreatments to totally or partially eliminate and/or transform them. Processes considered as strategies aimed at improving dissolution through cyanidation (Larrabure & Rodríguez, 2021). When the ore contains carbonaceous material, during cyanidation gold retention occurs due to the affinity of AuCN2- on the surface of the material. Looking for carbon removal, the material may undergo combustion and the consequent formation of CO2, SO2 and As2O3, causing environmental pollution. Therefore, biohydrometallurgy is a rapidly evolving biotechnology that has already provided entrenched solutions to old problems associated with metal recovery by conventional pyrometallurgy or chemical metallurgy (Gahan et al., 2012; Erüst et al., 2013). Its application to low-grade gold-bearing sulfide ores allows for a reduction in cyanide consumption (Rodrigues et al, 2021).

1.2. Biological oxidation as pretreatment.

The biological oxidation or BIOX process is the use of a mixed bacterial population composed of: Acidithiobacillus ferooxidans, Acidithiobacillus thiooxidans and Leptospirillum ferooxidans. These bacteria are capable of oxidizing auriferous sulfide ores and concentrates under controlled conditions. Therefore, they offer an alternative to conventional roasting or pressure oxidation techniques to recover Au and Ag from sulfide ores and concentrates (Groza et al, 2008).

Biooxidation as a pretreatment applied to refractory sulfidic gold ores allows improving recovery and reducing cyanide consumption during leaching. Therefore, it is important to consider the concentration of the bacterial population, which is directly proportional to the increase in pyrite oxidation (Cheng et al, 2021). The presence of a mixed community of bacteria and acidophilic archaea, iron and sulfur oxidizers, facilitates the recovery of precious metals. Changes in the community during the oxidation period can help create conditions that allow the formation of a bacterial consortium to achieve biooxidation and optimize the recovery of metal values (Smart et al, 2017).

Critical aspects for the selection of the BIOX process for the treatment of a refractory ore are: susceptibility to biooxidation, process design, economic evaluation and environmental implication (Van Niekerk, 2015). The effectiveness of pure and mixed cultures of three moderately thermophilic and extremely acidophilic bacterial strains (Acidimicrobium ferrooxidans, Sulfobacillus sibiricus, Acidithiobacillus caldus), was evaluated by Tanaka et al (2015), in the biooxidation of highly refractory ore concentrates, with very satisfactory results in Au and Ag recovery. Oxidative dissolution with the presence of microorganisms is a necessity in the mining industry, associated with the treatment of refractory sulfide ores containing precious metals (Lv et al, 2020), and in other metals of commercial interest.

1.3. What is the catalytic role of Acidithibacillus Ferrooxidans in the recovery of precious metals?

The bacterium Acidithiobacillus ferrooxidans exists naturally in mines, it participates in geochemical processes by oxidizing sulfide minerals and causing the formation of acid mine water. Its use in the pretreatment of refractory minerals dates back to the 1980's (Gilbert et al. 1988). The bacterial community that constitutes the group of sulfur and iron oxidizing bacteria of sulfide ores, is formed by the genera Acidithiobacillus, Acidiferrobacter, Acidiphilum, Leptospirillum and Ferroplasma and within them, Acidithiobacillus ferrooxidans, Acidithiobacillus thiooxidans, Acidiphilium multivorum, Leptospirillum ferriphilum, Sulfobacillus thermotolerans, Ferroplasma acidarmanus, and Ferroplasma acidiphilum strains (Bulaev et al, 2017). The ability to oxidize minerals is due to their status as chemolithotrophs; that is, they obtain their energy source from the oxidation of inorganic substances (Nordstrom and Southam, 1997).

The addition of certain cations such as Ag+ and Fe2+ and amino acids, as well as cysteine and other precursor molecules of bacterial metabolism, therefore, contribute to the oxidation of the metallic sulfides present in the mining-metallurgical resources, as well as the control of temperature, pH, O2 , nutrients and other physicochemical and biological factors. Using a mixed culture of Acidithiobacillus ferroxidans, Acidithiobacillus thiooxidans and Leptospirillum ferrooxidans, as a pretreatment to cyanidation, it was possible to improve gold and silver extractions with respect to direct cyanidation (McNeice et al, 2021).

The main catalytic mechanism of the bacteria consists of the oxidation of Fe2+ to Fe3+ to maintain an adequate Fe3+/Fe2+ ratio and to allow the oxidation of sulfides to sulfites, sulfates, thionates and tetrathionates. The sulfuric acid generated lowers the pH and the ferric ion is converted to ferrous ion to maintain the acidity cycle, in conjunction with the oxidation-reduction cycle to generate ATP molecules within the bacterial metabolism.

1.4 BACKGROUND

The development of biotechnology has allowed the incursion into mining activities; thus, approximately since the late 80's biooxidation has become an alternative for hydrometallurgical treatment of ores and refractory concentrates of precious metals (Torma et al, 1992). From a refractory gold concentrate with high sulfur and arsenic content, Wang (2018), analyzes chemical oxidation and then biological oxidation, by the latter he manages to improve the level of gold recovery.

The physiology of bacterial genera and species tolerant to extreme acidity has been investigated, as well as the sub-lethal concentrations of Ag, as in the case of Leptospirillum and Thiobacillus genera, which maintain the oxidation capacity in extreme acidity, which is key in biohydrometallurgy for the efficient recovery of precious metals such as: Au and Ag (Sánchez-Yáñez, 2006; Sand et al, 2001). Likewise, the rate of arrest and production of extracellular polymeric substances of At. ferrooxidans, At. thiooxidans and Leptospirillum ferrooxidans strains has been studied, opening new perspectives for the mining industry, seeking to improve the bioleaching process (Ruiz et al, 2008).

Through the BIOX process, a 50 kg/day pilot plant was set up to treat an arsenopyrite concentrate containing gold, using a native bacterial culture isolated from acid mine drainage (Ly & Van Nekierk, 2006). Also, in flotation concentrates, to reduce the refractory behavior of the material, favor the recovery of metallic values and contribute to preserve the environment (Marchevsky et al, 2017). In pilot scale studies, with concentrates with high pyrite and arsenopyrite contents, in continuous and batch process, sulfide degradation and bacterial population dynamics were evaluated (Hu et al, 2017).

In stepwise oxidation studies. Treatment of mine tailings with copper, zinc and gold values in three stages: (1) acid sulfuric leaching to remove soluble forms of copper and zinc, (2) biogenic ferric iron leaching for oxidation of copper, zinc and auriferous pyrite sulfides, and (3) bacterial oxidation of remaining sulfides. Biooxidation for a more complete oxidation of pyrite, with the consequent liberation of the associated gold. Achieving very satisfactory recoveries (Muravyov & Fomchenko, 2018). In the work by Wang et al (2018), the two-stage pretreatment: chemical oxidation and biological oxidation, allowed increasing the solubility of Fe, As and S; consequently, gold recovery was improved.

In a commercial process, using the bacterium Leptospirillum ferriphilum, the ability to adhere to sulfide minerals and perform the oxidation action was identified. Being important to consider the temperature, pH and characteristics of the passive layer of the mineral to achieve contact leaching of the mineral (Liu et al, 2017). The study of the microstructural and rheological properties of slurries during a bioleaching process reveals mineral attrition (particle size), presence of excreted bacterial exopolysaccharide, pH change and changes in the viscosity of the medium; resulting in flow phenomena such as thixotropy, elastic limit and rheopexy (Núñez et al, 2018).

II. METHODOLOGY

The present research has a qualitative approach of applied type and non-experimental design of documented review. The articles reviewed refer to practical work carried out in relation to the research topic "Biooxidation of argentiferous sulfide ores as pretreatment for silver recovery" and are evidence of its application.

For the collection of information, several types of documents were used, such as articles and reviews of journals indexed in the Scopus and Web of Science databases from 1988 and in English and Spanish language.

Once the search results were obtained, a classification was made by reading titles and abstracts. The following inclusion and exclusion criteria were used:

- Inclusion:
  - Treatments and pretreatments of ores and concentrates with application to gold and silver.
  - Use of various types of microorganisms.
- Exclusion:
  - Heap and column bioleaching treatments and pre-treatments.
  - Copper and uranium ores and concentrates.

III. STATE OF THE ART

Studies carried out in the last 5 years demonstrate the benefits of bacterial oxidation, with several studies having been conducted to achieve sulfide oxidation. Currently, roasting and pressure oxidation have microorganism treatment as an alternative (Ahn J., 2019). Gold and silver particles trapped in pyrite, chalcopyrite and arsenopyrite matrices are released by the action of iron oxidizing bacteria, determining that a higher concentration of bacterial inoculum led to an increase in the amount of bioleached metals (Brinza et al, 2021).

During the treatment of two refractory gold ores. Subjected to flotation, concentrates containing mainly apatite and dolomite were obtained, the biooxidation products generated jarosite and graphitic carbon, respectively. The cyanidation results were dulled, the former by the encapsulating action of jarosite and the latter by the preg-robing effect of carbon, in both cases gold extraction was reduced (Asamoah et al, 2021). Similarly, the presence of gypsum, jarosite and graphitic carbon negatively affect cyanidation (Ofori-Sarpong et al., 2020).

The application of multi-stage bioleaching of pyrite by arresting the growth of the bacterial population makes it possible to determine the rate of pyrite dissolution, surface roughness and corrosion intensity (Yin et al., 2020; Arias-Arce et al., 2023). In the treatment of ores with high pyrite content, it is important to identify the source and dosage of carbon; as well as, the temperature of the processing medium. The studies carried out by Bulaev et al. (2021), determine that at 50°C greater benefit is obtained.

In doubly refractory gold ores, due to the gold content at ppt levels and the considerable amount of carbonaceous material, the sequential treatment first seeking the destruction of iron sulfides and then the decomposition of carbonaceous matter by enzymes produced by the fungus phanerochaete chrysosporium with the capacity to degrade lignite, improved gold recovery (Sasaki & konadu, 2021). On the other hand, fungal chemoorganotrophic microorganisms capable of dissolving manganese and silver in mine tailings have been identified (Huerta-Rosas et al., 2020).

The processes to which refractory flotation concentrates are subjected are considered to have mechanochemical activation by physical and chemical changes, from size reduction with surface increase, increased particle tension, reagent consumption and gold extraction efficiency (Asamoah et al., 2018a). Mechanochemical activation by the action of biooxidation helped to overcome the refractoriness of the sulfide ore, achieving significant gold recovery (Asamoah, R.K., 2021). Also, the electrochemical behavior of arsenopyrite in iron-free 9 K culture medium in the presence of Ag+ significantly enhanced the dissolution of arsenopyrite. The formation of Ag2S on the passive surface preventing the dissolution of sulfides. The electrochemical-catalytic mechanism provides insight into the role of Ag+ as a catalyst and inhibitor of the process (Zhang et al, 2021a).

However, there are also cases where biooxidation is not considered for the processing of refractory ores. They, subjected to roasting and cyanidation, allow obtaining high gold recoveries in less time and with minimal cyanide consumption (Lin et al, 2022; Yoğurtcuoğlu & Ibrahim, 2023). For pyrite concentrates with gold and silver, by roasting between 500 and 600 ºC, it allows to significantly improve gold and silver recovery (Zhang et al, 2021). Also, there is the technology of roasting with improved chlorination by adding pyrite to promote the oxidation of calcium chloride and produce chlorine gas, and the process achieved the recovery of gold and silver in an efficient and environmentally friendly manner (Qin et al, 2020).

4.1. Alternative Processes to Cyanidation

An alternative to the cyanidation process is thiosulfation, this process requires the presence of copper sulfate and ammonia, as leaching agent and stabilizer of copper ions, respectively; and provides an ecological, non-toxic and relatively low cost model for the leaching of gold and silver from refractory ores or concentrates (Chen et al., 2022). Also, by mixing sulfate-tartrate-copper thiosulfate, Chen et al. (2023), achieves the dissolution of silver sulfide. A study by Zhao et al (2020), biooxidation of gold concentrate, followed by leaching with thiosulfate, obtained good results. The application of ultrasound to refractory silver ore, followed by cyanidation, as well as the control of other variables and by means of a statistical experimental design, allowed a significant increase in silver recovery (Cilek et al., 2020).

Studies on bacteria-mineral interaction identify the formation of biofilms, which largely depend on the chemical characteristics of the mineral surface. The adhesion of Leptospirillum sp. to surface-modified and weathered pyrite using a non-invasive electrochemical technique such as electrochemical impedance spectroscopy revealed significant changes at low frequencies, achieving different degrees of bacterial adhesion, evaluated by cell counting with Neubauer chamber (Saavedra et al, 2021).

Also, in research to extract gold from ores using iodide oxidizing, cyanogenic and amino acid excreting bacteria, flow charts have been developed for different scenarios to achieve gold dissolution from refractory and non-refractory ores (Jorjani. 2021).

IV. RESULTS

In the work “Bacterial leaching of an argentiferous refractory sulphide ore”, Sulfide Minerals and Refractory Silver Concentrates (SMRS) are treated by various methods such as: roasting, leaching with chemical chelating agents, pressure oxidation and chemical oxidation. These methods have relative efficiency, high investment and operating costs and, unfortunately, in some cases environmental damage; Therefore, alternatives such as bioleaching (BL) or bacterial leaching, which is a natural process in which chemolithotrophic bacteria oxidize the sulfur and/or iron in the minerals, thus facilitating the extraction of metals of commercial value at low cost and without contaminating the environment, although it has the limitation that the silver in the leaching solution inhibits the growth of the microorganisms that participate in the process, so an attempt was made to increase the resistance of these bacteria to silver. The objectives of this work were: 1) To isolate and select bacteria that leach SMRS, 2) To adapt these microorganisms to high silver concentration. For this purpose, erlenmeyers were prepared with modified 9K culture medium mixed with SMRS as an energy source for the microorganisms existing in the ore. The capacity was measured by the concentration of SO4-2 (sulfate) generated by the oxidation of sulfur from the SMRS, by the chemolithotrophic bacteria, the isolate with the maximum leaching activity of the ore, adapted to increasing concentrations of silver, which again was measured by the production of SO4-2 in the presence of an elevated concentration of silver nitrate. The results indicate that SMRSs contain chemolithotrophic bacteria that leach that ore to extract silver. One of these bacterial isolates was adapted to high silver concentration. Based on the type of leaching activity and physiological characteristics of the bacterium, it was classified as a species of the genus Thiobacillus sp. It is concluded that leaching is an economically viable alternative for the extraction of precious metals from SMRSs and does not cause environmental damage (Sánchez-Yáñez et al, 2006).

In the work “Bioleaching process for silver recovery: Structural and rheological studies”, it characterize the microstructural and rheological properties of silver manganese mineral pulps during a bioleaching process in a continuous stirred-tank reactor (CSTR). Analysis of the dissolution kinetics of Manganese in the pulp during the bioleaching process reveals a dissolution level of 20–23% during 36–48h. This percentage allows the extraction of large percentages of silver (Ag) during the cyanidation process, thus obtaining 64wt% of Ag. The maximum value of viscosity attained in the medium (especially between 48 and 72h) is an important parameter, since it may cause processing setbacks such as in-homogeneous agitation and increase in transport energy. Several factors contribute to the continuous change of viscosity in the media such as mineral wearing, the presence of the excreted bacterial exopolysaccharide (EPS), pH changes, particle size modifications due to mineral wear (plus corrosion) and changes in density of the medium. For this reason, it is of great importance to monitor the rheological behavior of the mineral pulp during the bioleaching and cyanidation processes. The mineral pulp behaves as a weak gel as reported in linear viscoelastic measurements (Núñez-Rámirez et al, 2018).

V. DISCUSSION

The removal rates of iron, sulfur, arsenic, among others, can be expressed by means of a mathematical equation, as well as the kinetics of the chemical reaction or biological reaction. The favorable kinetics of sulfur and iron removal in the biooxidation process studied by Huang & Yang (2021), corresponded to the gradual increase of the bacterial population, consequently the recovery of gold by cyanidation was favored.

High grade sulfide ores or sulfide concentrates can be processed by roasting or pressure oxidation processes. For low grade ores, it is possible to identify feasibility and viability by biohydrometallurgical processes due to requiring low capital and production costs (Ahn et al, 2019). It is also mentioned by Purmomo et al. (2019), by sulfur and iron oxidizing bacteria and, in media containing ferrous sulfate.

High temperature oxidation could be profitable in some cases where the necessary infrastructure for the treatment of the gases generated and due respect for the environment is available. There is the case of chloride roasting to produce soluble chlorides in subsequent processing stages. The chlorinating capacity is enhanced with pyrite, due to its ability to reduce calcium chloride and produce chlorine gas (Qin et al, 2020).

The evaluation of the microbiological diversity present in the processing system and the characterization of the mineralogical and physicochemical structures of the ore can significantly influence the identification of biooxidation in the different bioreactors (bioleaching heaps, stirred tanks and/or tempered bioreactors). Data processing using artificial intelligence will allow new and improved dissolution rates of compounds present in mineral resources to be obtained (Darvanjooghi et al., 2022).

VI. CONCLUSIONS

If the submicroscopic particles of gold and silver present in the matrix of sulfide ores are not recovered in an affordable way by the conventional method that involves leaching in sodium cyanide solution, the ore must be subjected to an oxidation pretreatment or conversion of the sulfides to simple compounds that allow or do not hinder the recovery of the precious metals.

The oxidation methods are the main fundamentals in the kinetic determination of the process and the relationship of the dissolution of sulfur, iron, arsenic and other elements present in the minerals considered refractory. In addition, they can be represented by various linear and nonlinear mathematical models, according to the handling of the variables and the conditions of the medium.

The selection of biooxidation as the preferred process in projects for the recovery of gold, silver, copper or other metals, in most cases is based on the economic advantage, compared to roasting, chemical leaching and pressurized leaching processes.

The variety of mineralogical, microbiological and physicochemical structures of sulfide ores, reactor types, bacterial interactions, among other factors, significantly influence biooxidation reactions. As well as microbial characterization (Kaksonen et al, 2018).

Elemental, mineralogical and textural characteristics must be carefully understood to identify potential solutions, such as those described in the present review, but these potential solutions should be evaluated through research and development (R&D) projects. Identification of the types of gold and silver mineralization in sulfide deposits is necessary to develop and improve rational extraction schemes (Palyanova, 2020).

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Conflict of interests

The authors declares no conflicts of interest.