Biohydrometallurgy of Sulphide Minerals

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Mineral processing industries today face enormous challenging tasks of meeting the ever-incresing demand of metals in view of rapidly depleting high-grade ore resources. several factors such as occurances of low grade complex ores, high energy and labour costs and stringent environment proting processes as these processes (i) can handle low grade complex ores or wastes from previous operations efficiently, (ii) can be incorporated in existing mineral processing circuits with very little modifications and (iii) have minimal environmental impact. From among two dozen or more non-ferrous metals extracted by hydrometallurgical routes, biotechnological route can be used for Al, Au, Cu, Ni, U and Zn as shown in Table 1. At present, commercial biotechnological processes (also referred to as bioleaching or biooxidation) for extraction of Au, Cu, and U from their ores has been practiced in countries like Australia, Canada, Chile, South Africa, Russia, United States, to name a few. In case of refractory gold ores, the native gold locked in pyrite-arsenopyrite matrix is exposed by bioleaching of sulphide matrix, and therefore, process is sometimes referred to as bioliberation.

Association of microorganisms in the formation and solubilisation of mineral deposits is known since geological times [Ehrlich, 1990]. Mining operations have long benefited from the activities of such naturally occurring microbes, especially from the ability of some bacteria to leach the metals from insoluble ores. In spite of the fact that bioleaching has been occurring in nature for a long time, the contribution of bacteria in metal leaching was recognized only in 1947 when Colmer and Hinkle [1947] identified a bacterium, *Thiobacillus ferrooxidans* from the acid drainage of bituminous coal mines. But the presence of bacteria in the leach waters of Rio Tinto was first confirmed in 1963 [Razzell and Trussell 1963]. Although leaching of metal ions from the ores is the only commercially practiced process, several different type of processes based on the interactions between microorganisms and ores are of interest to mineral processing industries as illustrated in Table 2. The bioleaching process in principle is operationally similar to the chemical leaching process by mineral acids or other lixivants. However, the main difference is the use of microorganisms and their metabolites in solubilizing metals from their sulphide matrix. Some of salient features of chemical and bioleaching are compared in Table 3.

The focus of this review article is bioleaching of sulphide minerals and ores. Several aspects of the bioleaching process, namely, microorganisms involved, mechanisms of leaching, factors affecting the kinetics of leaching process and operating strategies are reviewed. The application of biotechnology in processing of refractory gold ores is illustrated.

Microbiological Aspects

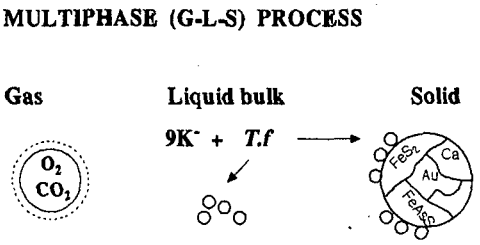
A variety of acidophilic bacteria that can participate in the oxidation of mineral sulphides with consequent solubilization of the metal are listed in Table 4. It is clear from Table 4 that *Thiobacillus ferrooxidans* is the most versatile microorganism which can solubilize sulphide minerals both by direct oxidation of sulphide mineral and indirect oxidation through lixiviants ($Fe_2(SO_4)_3$ and H_2SO_4) generated by the oxidation of ferrous iron in the liquid. Consequently, *Thiobacillus ferrooxidans* is the most widely implicated microorganism in bioleaching applications [Torma, 1977].

Thiobacillus ferrooxidans is a motile, mono-flagellated, non-spore forming and gram- negative acidophilic bacterium. Cells are short, straight rods, approximately 1.0 μm long and 0.5 μm in diameter, occur singly or occasionally in pairs and divide by binary fission. Different strains may possess flagella and/or pili. The energy source for *Thiobacillus ferrooxidans* is either oxidation of ferrous iron or reuced valence inorganic sulfur compounds. The details of the complex mechanisms involved in sulfur and iron oxidation are beyond the scope of this article.

Several physical and chemical factors that influence the growth and bioleaching ability of *Thiobacillus ferrooxidans* are summarized below with their optimal levels indicated in the brackets: (i) pH (1.5-2.5), (ii) Temperature (28-35°C), (iii) Redox potential (750-850 mV), (iv) CO_2 (1-2 vol%), (v) O_2 (3-5 ppm) and (vi) Growth medium (9K Medium, Silverman and Lundgren, 1959).

Mechanisms of Bioleaching

Mechanisms involved in bioleaching of ores are very complex. The complexity arises from different types of chemical, biochemical and electrochemical interactions between microorganisms, metabolites produced from them and minerals. Two main bioleaching mechanisms identified are (i) direct, and (ii) indirect leaching. It must be cautioned that there is no general agreement in the reported literature regarding the dominance or lack of it of any one particular mechanism. In *direct mechanism*, *Thiobacillus ferrooxidans* attaches to the sulphide mineral and oxidizes the mineral which provides energy for bacterial growth. For example, pyrite is oxidized and iron is released in the leach liquor as ferrous (Fe^{2+}). In *indirect mechanism* ferrous iron is oxidized to ferric (Fe^{3+}) by the bacteria (energy source) in the liquid phase. Fe^{3+} in turn can chemically leach sulphide mineral. The sulphide ore is oxidized by *Thiobacillus ferrooxidans* and metal solubilizes in the leach liquor in the sulphate form. Various processes involved in bioleaching of sulphidic minerals are shown schematically in Figure 1.



Pyrite

(1 - S) $4FeS_2 + 14O_2 + 4H_2O \longrightarrow 4FeSO_4 + 4H_2SO_4$

(2 - L) $4FeSO_4 + O_2 + 2H_2SO_4 \longrightarrow 2Fe_2(SO_4)_3 + 2H_2O$

(3 - SL) $FeS_2 + Fe_2(SO_4)_3 \longrightarrow 3FeSO_4 + 2S^0$

(4 - L) $2S^0 + 2H_2O + 3O_2 \longrightarrow 2H_2SO_4$

Fig. 1 Bacterial Oxidation Mechanism

Technological Aspects of Sulphide Bioleaching

Apart from the physical and chemical parameters that influence the growth of *Thiobacillus ferrooxidans*, there are several technological parameters which influence the efficiency of bioleaching of sulphide ores. It is clear from above discussions that the bioleaching process is a highly complex multiphase process involving interactions of four different phases: gas (O_2 and CO_2), liquid (growth medium, metabolites), solid (sulphide ore and minerals) and microorganism. In the frame work of multiphase reaction engineering, the overall bioleaching process can be thought of consisting of following subprocess involving mass transfer and chemical/biochemical reactions:

(A) Mass tranfer of O_2/CO_2 from gas phase to liquid phase
(B) Mass tranfer of O_2/CO_2 and chemical lixiviants from liquid phase to solid surface
(C) Diffusion of O_2/CO_2 and chemical lixiviants in porous solid to sulphide sites
(D) Attachment of microorganisms to sulphide mineral
(E) Microorganism mediated oxidation of sulphide mineral/elemental sulphur (direct mechanism)
(F) Microorganism mediated oxidation of ferrous iron in liquid phase
(E) Chemical leaching of sulphide mineral by lixiviants (indirect mechanism).

Since all these processes are occurring simultaneously, the rate of overall process is determined by the rate of the slowest step which is the rate-determining step. Apart from the factors affecting the growth and activity of microorganism additional parameters such as particle size, pulp density, agitation and aeration rates also influence the kinetics of these processes. The effect of these and other parameters on the mass transfer and biochemical reactions is summarized below.

Mass transfer : The transfer of O_2/CO_2 from gas phase (air) to the reaction site (sulphide mineral) is absolutely essential for efficient bioleaching. This rate depends on the gas-liquid/liquid-solid mass transfer rate (K_L), gas-liquid/liquid-solid interfacial area (a) and the solubility of oxygen in liquid. The mass transfer rate constant (K_{la}) increases as the agitation speed and superficial gas velocity are increased [Liu etal., 1988]. The decrease in gas bubble/solid

paraticledius also increases the mass transfer rate constant. However, in the presence of solid particles, the gas-liquid mass transfer rate constant is adversely affected [Liu *et al.*, 1988]. This results in overall reduction in bioleaching rates as the sulphide mineralpulp density is increased beyond 20-30% [Torma *et al.*, 1972]. Similarly, the solid-liquid mass transfer rate is also lowered in the presence of gas. The solubility of O_2/CO_2 in aqueous media is also lower in the presence of salts of nutrient media which lowers the mass transfer rate further.

Bioleaching kinetics : In a bioleaching process, *Thiobacillus ferrooxidans* is continually exposed to different energy sources, namely, ferrous iron and sulphide/sulphur. As a result, the activity of bacteria in the presence and the absence of the sulphide mineral are different and this would have impact on the overall kinetics of the process. Bioleaching is a heterogeneous process and irrespective whether it occurs through direct or indirect mechanisms, surface area is an important parameter which influence the bioleaching kinetics. It can be expected that bioleaching rates also increase with decrease in particle size or increase in pulp density since both factors increase the surface area. Indeed, higher leaching rates with lower particle sizes have indeed been reported for pyrite and chalcopyrite [Asai *et al.*, 1992]. This increase is also related to increase in number density of bacteria attached as the surface area is increased. However, it should be pointed out that the particle size in these studies was 70-80 μm or lower. At higher particle sizes(70 μm - 5 μm), the leaching rates were found to increase with the particle size, reach maxima and then decrease [Shrihari et al., 1995] in case of pyrite leaching. Furthermore, only direct mode of operation was found to be responsible for bioleaching of sulphide. Similar results were also reported for chalcopyrite [Shrihari, 1993] and refractory gold ores [Natarajan *et al.*, 1995].

Since dissolved metals accumulate in the leach liquor, a slowdown in bioleaching rates is observed as a result of toxic effect of some of the metal ions on the growth. As pointed out earlier, the activity of the bacteria can be regained through prolonged exposure to the toxic metal. The bacteria thus adapted loose their metal tolerance if they are grown in the absence of the metal. These observations have important practical application in the use of such tolerant strains in bioleaching. In a typical bioleaching process initiated with unadapted or wild strains, the strain will be exposed to increasing concentrations of dissolved metals as leaching progresses. Thus, the cell population present at the end of the operation will be different from those at the start. It has been reported [van Aswegen et al., 1991] that during pilot plant studies on bioliberation of gold, considerable reduction in residence time and increase in pulp densities could be achieved over a period of 3-4 years. One of the factors that could have contributed to such improvement is the continual adaptation of the bacteria to the dissolved metal.

A summary of various parameters and their influence on technological and economic performance of bioleaching process is given in Table 5.

Reactors for Bioleaching : Reactors used in biohydrometallurgical leaching of sulphide minerals can be broadly classified in two categories: the fixed bed reactor and reactors working with suspension (slurries) of finely ground mineral particles and microbes in aqueous medium [Rossi, 1990]. The commercial heaps or dumps and laboratory airlift percolators and leach columns are examples of fixed bed reactors. The slurry reactors can be classified in two categories depending on the mode of mixing slurry, either by agitation be external pump, mechanically agitated reactor or by rising bubble column of injected air, air-lift reactors. In hydrometallurgical operations such as cyanidation of gold ores, airlift reactors or pachuca tank are most commonly used. Figure 2 shows schematics of these reactors.

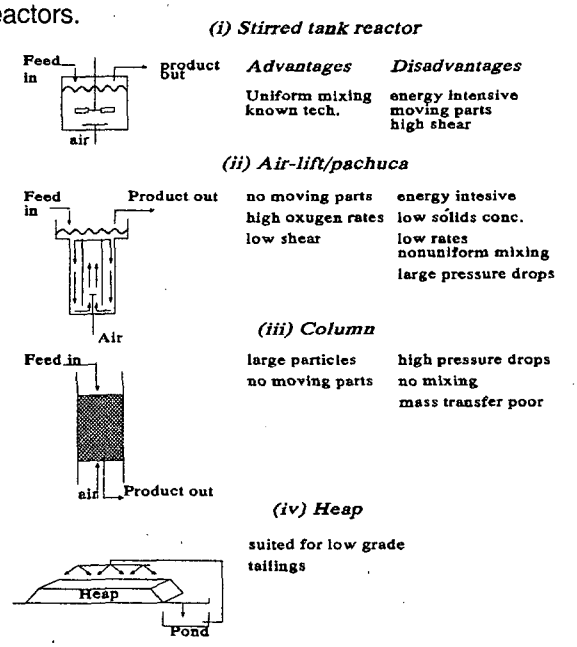


Fig. 2 Reactors for Bioleaching

The fixed bed reactors are ideally suited for treatment of high volume low-grade sulphide ores or

wastes from previous workings. The slurry reactor is economical only for low volume operation which essentially involve bioleaching of flotation concentrates of sulphide ore. The suspension of particles in the slurry reactor requires the use of fine particles while coarser particles are usually handled in fixed bed reactors. Due to mass transfer and other limitations, the kinetics of bioleaching processes is very low in fixed-bed reactors, requiring several months to years for the desired treatment. The kinetics is fairly rapid in slurry reactors as compared to fixed-bed bioreactors. The commercial processes for copper mainly utilize dump and heap leaching techniques while refractory gold ore processing has been reported with slurry reactors [Natarajan, 1992].

Biotechnology for Proccessing of Refractory Gold Ores : The Hutti Experience

The Hutti Gold Mines Company Limited (HGML), Hutti Karnataka is a premier gold producing company in India. During the past 53 years, the Company has produced more than 45 tonnes of gold and is currently producing average of 1.5 tons per annum. Gold, a precious noble metal, occurs in nature in its native form and usually associated with quartz. Such ores, referred to as free milling ores, are treated in a conventional process with cyanide to solubilize gold-cyanide complex. The gold is then recovered from the solution by using either zinc (old technology) or more recent, carbon-in-pulp technology. Recent history of mining activity at Hutti suggests that both the quantity and quality (gold content) of free milling ores are rapidly decreasing. The excavations within Hutti zone as well as in areas such as G.R. Halli, show that the gold ores refractory in nature. G. R. Halli is a very promising newly opened mine containing both gold and silver in mixed sulphide matrix incorporating copper, lead, zinc, cobalt etc. The typical composition of G.R. Halli flotation concentrate is 47% pyrite and 23% arsenopyrite with gold and silver content of 22-25 and 1000 g/MT, respectively. The gold in the sulphidic ores is finely disseminated within the pyritic and arsenopyritic minerals and cannot be liberated even after finer grinding. Only gold particles located on the surface of the ore are leached by cyanide solution thereby reducing the extraction efficiency. Sulphide minerals and organics associated with refractory ores consume cyanide and the organics may retain gold by robbing, which further reduces the cyanidation efficiency. Various process options are available for the treatment of refractory gold ores prior to cyanidation. These options include pressure oxidation, roasting and bacterial oxidation. Bacterial oxidation offers the advantage of significantly reduced capital costs while producing environmentally acceptable effluents.

Bacterial oxidation as a process technology for refractory gold was first implemented at the Fairview plant in South Africa in 1986. This operation treated a refractory arsenopyrite-pyrite concentrate by roasting before adopting bacterial oxidation as its replacement. Despite the novelty of the new technology, bacterial oxidation was able to displace roasting which at the time was the accepted method of treating refractory gold concentrates containing significant levels of arsenic. The technology has so far been applied in North and South America, Australia, South Africa and West Africa and there are about 11 commercial plants worldwide. As confidence in commercial bioprocessing grows and experience extends the application's knowledge base, innovations and new commercial practices will emerge.

In early nineties, HGML faced with dwindling grade free milling ores and excavations of sulphidic zones, recognized the need for exploring new technology for gold processing. The multidisciplinary research team consisting of Metallurgist, Chemical Engineers and Microbiologists at Indian Institute of Science (IISc) took up this challenge, resulting in collaborative research project sponsored by HGML. The project were essentially a laboratory scale study of investigating microbial ecology of Hutti Gold Mines and establishing a process for biotreatment of sulphidic ores. Having successfully demonstrated the technology at laboratory level, a need was felt to scale up this process and demonstrate it on a larger scale. The research team at IISc conceived a unique concept of combining the strengths of a research organization, Indian Institute of Science, Bangalore, an engineering consultancy firm, Engineers India Limited, New Delhi and the industry, The Hutti Gold Mines Company Limited. A collaborative project "Setting of Demonstration Bioreactor Plant for Biotreatment of Refractory Gold Ores and Concentrates at Hutti Gold Mines" took shape with financial assistance from Department of Biotechnology, Government of India. The main objectives of the project are to set-up and operate a demonstration bioreactor plant for treatment of refractory gold ores and concentrates.

Based on the laboratory scale experimental data provided by Indian Institute of Science, Engineers India Limited developed a process design package for the demonstration bioreactor plant at Hutti. The nominal capacity of the plant is to treat 100 kg/day of flotation concentrating but can be enhanced up to 200 kg/day. The plant consists of three sections: mixing, bioleaching and product separation.

Mixing Section: Refractory gold concentrates and process water are added to a Mixing vessel to prepare slurry of 10% wt/vol. The preparation of slurry is done in a batch mode. Continuous agitation is required to main uniform pulp density. The transfer of mixed slurry from mixing vessel to reactors I and II is done in continuous mode using multi-channel feed. Slurry samples would be collected from mixing vessel at regular intervals to measure pH and pulp density.

Bioleaching section: The bioleaching section consists of three reactors, each 2 m^3 in size. Reactors I and II are operated in parallel followed by Reactor III in series (Figure 3). The bioleaching process starts from Reactor I and II. Each reactor is equipped with agitator, air sparging, CO_2 sparging and jacket facility. It also has inoculum and nutrient addition facility as and when required. An agitator is provided to maintain uniform pulp density, gas distribution as well as low shear to prevent bacterial death. Air and CO_2 sparging are required to maintain bacterial growth and biooxidation of sulphides. The pH of the slurry is to be maintained between 1.5-2.0 by addition of sulfuric acid as and when required. Slurry sample has to be collected from reactors at regular intervals to measure pH, redox potential, ferrous and ferric iron concentrations and bacterial activity. Air sparging rate, pH and agitation speed are to be maintained manually. Outer jacket water flow rate is maintained automatically by a flow control valve to maintain the temperature in the reactors constant. After a residence period of 80 hours in reactor I and II, bioleached slurry flows into reactor III by multichannel slurry pump. Operating conditions and their control are the same as in reactors I and II except the reactor temperature is maintained manually. After a residence period of 40 hrs in this reactor, the slurry flows to a settling tank through a multichannel slurry pump.

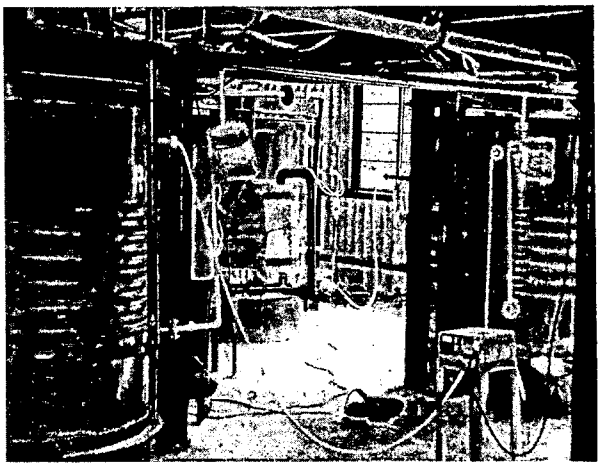


Fig. 3 Bioleaching Section

Product separation section: Settling tank would operate in a batch mode. The capacity of this tank is to hold bioleached slurry for 5 days. Bioleached residue will be collected from the bottom of the tank and would be sent for cyanidation.

The plant has been in operation for past six months and initial results are quite encouraging (Figure 4).

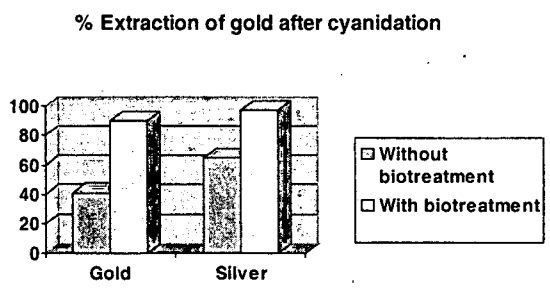


Fig. 4 % Extraction of Gold after Cyanidation

Relevance to other mining sectors in India.

Biomineral technology holds great promise and practical significance in India. Although extensive research efforts on various aspects of bioleaching of different types of Indian ferrous and nonferrous ores have been made, no fruitful commercial adaptation of biotechnology has so far been made in India. The following aspects of biomineral technology have national relevance: (i) biomineralization of Indian deposits, (ii) biobeneficiation of ferrous and non-ferrous minerals and (iii) bioleaching of low grade ores, mine wastes, tailings as well as flotation concentrates. Apart from gold ores, bioleaching of copper, uranium and zinc

has tremendous potential. Further, sea nodules which are rich sources of nonferrous metals such as cobalt, nickel, copper and manganese could also be bioprocessed. India has great stake in the commercial utilization of this ocean resource and biotechnology holds great promise in this context. The demonstration bioreactor plant at Hutti is a first and only effort in this direction in India. Although the plant is designed for processing gold ores, it can be suitably modified to test several other ores. The facility created at Hutti Gold mines can serve as national center for testing biotechnological solutions to problems faced by mining industries all over the country. Our efforts at Hutti mark only a beginning and lots of ground remains to be covered. What is needed is a mission oriented, dedicated and time-bound national policy devoted to adaptation of biotechnology to Indian Mining. We hope that our initiative at Hutti inspires other research laboratories and mining industries to adapt this environmentally benign, energy-efficient and cost effective biotechnological approach.

Summary

Biohydrometallurgy holds tremendous promise in industrial situations with respect to recovery of base and noble metals from lean grade ores and flotation concentrates. The machenisms in biooxidation are complex which involve microbiological, biochemical, Physicochemical and electrochemical factors. Reactor design and engneering holds the key for improving bioleaching kinetics, with reference to treatment of Sulphide minerals and concentrates.

Table 1 World production of some nonferrous metals

Metal	Important ores	World Production (million tons)	Production	Process
			Hydrometallurgy	Biotechnological
Al	bauxite, kaolin, nephelin, Aluminite	19	practically all	potential exists
Au	Native gold. gold associated with sulphides	2000 tons	all primary gold	commercial nearly 1/15th
Cu	chalcopyrite, chalcocite, covellite, bornite, azurite, cuprite, malachite, chyrscocolla	> 10	15-20%	commercial nearly 35% of hydrometallurgicall y produced
Ni	pentlandite, laterite, garnierite	0.8	60%	potential exists
U	uraninite, pitchbende, carnotite, autunite, torbernite	not known	100%	commercial nearly 1/8th
Zn	sphalerite, marmatite, zincite, smithsonite	< 8	85%	potential exists

Table 2 Processes based of interactions between microorganisms and ores

Process	Description
Biomineralization	formation of mineral and ore bodies mediated by microorganisms and/or their metabolies. example: biomineralization of bauxite [Ehrlich, 1990]
Bioleaching	dissolution of metal of interest from the ore, example: copper from chalcopyrite and liberation of native gold from pyrite-arsenopyrite matrix by Thiobacillus ferrooxidans [Tuovinen, 1990].
Biobeneficiation	dissolution of impurity from the ore therby enriching the ore with metal of interest example: calcium and iron from bauxite by Bacillus polymyxa [Anand et al, 1996]
Bioflotation	mineral separation by selective flotation or depression of minerals, example: silica from haematite by Bacillus polymyxa[Deo and Natarajan, 1997]
Bioflocculation	mineral separation based on floculation of minerals by cell-cell, cell-mineral adhesion, example: pyrite from coal by Microbacterium phlei [Misra et al., 1993]
Biosorption	removal of metal ions from aqueous effluents, example Uranium from waste waters by Rhizopos arrhizus [Tsezos and Volesky, 1982]
Biocorrosion	deterioation of metal surfaces, example: aluminium corrosion by pseudomonas spp. [Iverson, 1987]

Table 3

Comparison of chemical leaching and bioleaching process

	Chemical leaching	Bioleaching
Raw materials	costly synthetic chemicals such as mineral acids, ferric sulphate etc.	microorganisms: many time indigenous, capable of growth and therefore renewable, characterisitics of microorganisms different depending on the source, ameanable to change through adaptations, mutations and genetic engineering, few simple nutrients required, chemical lixivants such as sulphuric acid, ferric sulphate generated by microbial action
Ores	preferably high grade ores	low grade ores and wastes can be treated, selective dissolution from complex sulphide ores possible.
Operating conditions	high temperature and pressure may be required	mainly ambient processes narrow range of temperature and pH for microbial growth.
Kinetics	rapid	slow due to microorganisms
Environmental impact	highly polluting	minimal as processes are natural, no air pollution, liquid byproducts more ameanble to containment and treatment.
Scale-up	large data base due to years of operation	very few commercial venture, limited database, site specific process

Table 4
Microorganisms for bioleaching of sulphide minerals and ores

Microrganisms	*Thiobacillus ferrooxidans*	*Thiobacillus thiooxidans*	*Leptrospirillum ferrooxidans*	*Sulfolobus spp*	*Sulfolobus brierleyi*
	IOB + SOB	SOB	IOB	IOB	IOB + SOB
Characteristics	Autotrophic	Autotrophic	Autotrophic	Autotrophic	Autotrophic
	Aerobic	Aerobic	Aerobic	Aerobic	Anaerobic Aerobic
Temperature	25-30°C	30°C	30-40°C	50-60°C	50-90°C
	1.5 - 3.0	1.5 - 3.0	1.5 - 3.0	1.5 - 3.0	1.5 - 3.0
Sulphides dissolved	CoS, $CuFeS_2$ FeS_2, PbS, ZnS NiS	ZnS, CdS	FeS_2	MoS_2, Cu_2S $CuFeS_2$	MoS_2, Cu_2S, $CuFeS_2$
Mode of action	direct oxidation indirect oxidation	indirect oxidation[a]	direct oxidation indirect oxidation[a]	direct oxidation	direct oxidation indirect oxidation[a]

IOB - Iron Oxidising bacteria SOB - Sulphur Oxidising bacteria
a : Mainly in mixed culture with *Thiobacillus ferrooxidans*

Table 5

Summary of important parameters and their influence on bioleaching process

Parameter	Influence on the performance of bioleaching process	Probable reasons
Particle size	bioleaching rate increases as particle size decreases for finer particles (< 100 μm)	higher surface area resulting in better mass transfer and indirect oxidation rate , increase in direct oxidation rate due to larger number of attached cells
	larger particles (500 μm - 5mm), rate first increases, reaches maxima and then decreases as particle size increases	only direct mechanism operating, attachment of cells showing a maxima
	higher grinding costs for finer particles	
Pulp density	bioleaching rates and metal dissolution efficiency decreases as pulp density is increased	mass transfer rates lower, abrasion effects high, loss in ferrous iron oxidation ability by bacteria
	low throughputs for smaller pulp densities	liquid volumes very large
	slurry handling difficult at higher pulp densities	slurry density and viscocity very high
	energy costs high at high pulp densities	liquid/air pumping rates, agitation speed high for slurry suspensions
Agitation	bioleaching rates increase as impellar speeds are increased upto critical agitation speeds	uniform mixing and better suspension of slurries, better mass transfer rates
	bioleaching rates decrease if agitation rates increased beyond critical speeds	bacterial damage due to high shear and particle attrition
	power consumptions high	
Aeration	bioleaching rates increase as aeration rates are increased	availability of O_2 & CO_2 higher
	bioleaching rates increase as size of gas bubbles is decreased	better mass transfer rates
	power consumption high for higher aeration rates and finer bubble sizes	
Residence time	bioleaching rates high at lower residence times, low productivity at low and high dilution rates	slow kinetics, bacterial wash-out at high dilution rates

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