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Chapter

Rare Earth Elements Biorecovery from Mineral Ores and Industrial Wastes

Laura Castro, M. Luisa Blázquez, Felisa González and Jesús A. Muñoz

Abstract

Rare earth elements (REEs) are critical raw materials and are attracting interest because of their applications in novel technologies and green economy. Biohydrometallurgy has been used to extract other base metals; however, bioleaching studies of REE mineral extraction from mineral ores and wastes are yet in their infancy. Mineral ores have been treated with a variety of microorganisms. Phosphate-solubilizing microorganisms are particularly relevant in the bioleaching of monazite because transform insoluble phosphate into more soluble form which directly and/or indirectly contributes to their metabolism. The increase of wastes containing REEs turns them into an important alternative source. The application of bioleaching techniques to the treatment of solid wastes might contribute to the conversion towards a more sustainable and environmental friendly economy minimizing the amount of tailings or residues that exert a harmful impact on the environment.

Keywords: bioleaching, rare earth elements, recycling, wastes, minerals

1. Introduction

Rare earth elements (REEs) are strategic materials in order to facilitate the transition from current economy based on petroleum to an efficient circular economy based on clean energy. Although often needed in small quantities, these metals are essential to produce a huge number of technologically sophisticated products for electronic, optical and magnetic applications. Among other applications, these elements play a crucial role in the development of clean emerging low-carbon energy technologies [1].

In spite of the archaic term, most of rare earths are common elements and some of them are even more abundant than other metals, such as copper or lead. Though moderately abundant in the Earth’s crust, rare earth elements are scarcely concentrated in mineral deposits and this fact complicates its Extractive Metallurgy which is complex and requires economic solutions. The world production of REEs has undergone an exponential growth since its discovery in the 18th century, with a notably increase overtime from 1,000 t in 1930 to 133,600 t in 2010 [2]. The rising REEs production has been the consequence of an escalating demand for REEs as well.
Based on their strong affinity for oxygen, REEs resources are mostly present in oxidic form, mainly as rare earth oxides, phosphates, carbonates and silicates. According to recent estimates, 100 Mt of rare earth oxides are accessible in more than thirty countries all around the world. More than 200 REEs ores have been identified as rock-forming minerals, however, only three are considered mineral ores for economic extraction: bastnasite \( ((\text{Ce},\text{La})(\text{CO}_3)_2)\text{F}) \), monazite \( ((\text{Ce},\text{La},\text{Nd},\text{Th})\text{PO}_4) \) and xenotime \( (\text{YPO}_4) \) [3]. Thus, the primary sources of REEs are mineral deposits spread out worldwide, but confined mainly in China, Australia and USA.

Furthermore, REEs are also found in industrial wastes in vast amounts and they have been investigated as potential resources for rare earth metals [4–6]. Among REEs-bearing industrial residues, phosphogypsum, generated during the wet phosphoric acid process from fertilizer production, and red mud residues from the digestion of bauxites in the Bayer are rich in valuable rare earth metals as to be economically treated.

In addition, end-of-life materials can be recycled due to their significant quantities of REE, among them: magnets (38%), lamp phosphors (32%) and metal alloys (13%), retain more than 80% of REE market. Modern fluorescent lamps typically contain more than 20% (w/w) REE (Ce, Eu, La, Tb and Y) [7].

After ore and/or industrial waste concentration processing, rare earth metals are dissolved selectively from raw materials. Actinides, such as uranium and thorium, with similar chemical properties to REEs, are often co-dissolved during hydrometallurgical processes and this could pose a problem. REE primary ores are leached using acid \( (\text{H}_2\text{SO}_4, \text{HCl, HNO}_3, \text{H}_3\text{PO}_4) \) or alkaline \( (\text{Na}_2\text{CO}_3, \text{NaHCO}_3) \) reagents and \( \text{NaCl} \) or \( (\text{NH}_4)\text{SO}_4 \) for REE-ion adsorbed clays [8–10]. Nonetheless, the hydrometallurgical treatment is ore-dependent and has been well established in the case of some REE ores, especially monazite, but is less evident for other key minerals with a very complex mineralogy.

Biohydrometallurgy and more specifically its application to the extraction of metals through bioleaching processes have been successfully practiced at industrial level for the recovery of uranium, copper and gold [11, 12]. Biohydrometallurgical technologies could play a fundamental role for the treatment of REE-bearing materials since they offer an alternative to physico-chemically based methods. Bioleaching is connected to the development of more cost-effective, less energy demanding and less polluting metal extraction processes than pyro- and hydrometallurgical processes and is able to treat low-grade mineral ores and a great variety of residues. These biotechnological processes involve interactions between microorganisms and metal-bearing ores that render valuable metals in solution. REE mobilization from solid matrices has been done with a wide range of microorganisms, both autotrophic and heterotrophic, and using both pure and mixed microbial cultures [13–15].

This chapter provide an insight into the global situation of REEs and the potential application of microorganisms in the extraction of REEs from both REEs-bearing minerals and industrial residues.

2. Global situation of REEs: market and technology

The demand of REEs have increased in the past decade because of their extensive use in several fields related to electronics, in renewable energy capture technologies, biomedical devices, and other industrial products. In 2018, a list of 35 critical minerals, including rare earth elements, was published by the U.S. Department of the Interior and other executive branch agencies [16]. Likewise, the European
Commission developed a critical assessment on non-energy and non-agricultural raw materials in 2017 including heavy rare earth elements, light rare earth elements and platinum group metals [17].

Global mine production was estimated to be 210,000 tons of rare earth oxide (REO) equivalent, which means an 11% increase in comparison with 2018 (Figure 1a). China dominates the global production of rare earth minerals, separated compounds and metals. China exports REEs to United States (31%), Japan (27%), the Republic of Korea (11%), the Netherlands (9%) and Germany (6%).

Other countries are making efforts to increase their domestic production of mineral concentrates. For example, United States enhanced the production, all of which was exported, a 44% in 2019 compared with 2018 [18].

Rare earths are relatively abundant in the Earth's crust; however, REEs resources with minable concentrations are less common. Nowadays, about 85% REE deposits have been identified worldwide, which are mainly located in China, Vietnam, Brazil, Russia, India and Australia (Figure 1b) [18, 19].

Prices for most rare earth products are increasing compared with those in 2016 reversing the falling trend that began after prices spikes in 2011. Gadolinium, praseodymium and neodymium experienced the greatest increase in the price, while the yttrium and dysprosium prices decreased. The estimated unit value of rare-earth compounds was $11.60 per kg in 2017 based on information from the U.S. Census Bureau on imports [20].

The estimation of global consumption of rare earth varies significantly due to the limited data transparency and it generally ranges about 140,000 and 170,000 tons of REO equivalent. Furthermore, the global consumption of scandium was estimated in 10–20 tons per year [21].

The amount of specific REEs used strongly depends on the market sector and application. Lanthanum and cerium, and lower amounts of neodymium, are consumed in the catalysts sector. There are different types of permanent magnets but neodymium-iron-boron magnets are fabricated with neodymium and praseodymium, and samarium-cobalt magnets uses samarium and gadolinium. Batteries contain mainly lanthanum, and ceramics, yttrium. Europium, yttrium and terbium are commonly associated with the phosphors sector.

The global growth rate of REEs consumption is expected to be 5–7% per year through 2022. The magnet materials sector would grow more than other sector such as catalysts, ceramic or phosphors. The increasing global demand of REEs as well as the enforcement of environmental and production legislation beyond 2022 lead to higher prices and, consequently, this situation may drive the mining and processing development outside China.

Figure 1. World mine production (a) and reserves (b).
3. Bioleaching of REEs mineral ores

The application of biohydrometallurgy for extracting REEs offer a green alternative to the conventional methods, which are complex and energy intensive. The main economic REEs-bearing minerals are bastnaesite, loparite, monazite, xenotime, and the lateric ion-adsorption clays. Both autotrophic and heterotrophic microorganisms are capable of solubilize REEs and the selection of these microbes for bioleaching processes depends on the type of mineral. Autotrophic bacteria have been employed for the extraction of scandium from ore minerals containing metal sulfides, whereas heterotrophic bacteria are mostly employed for REEs carbonates and phosphates [22].

Bastnaesite is a rare earth fluoro-carbonate ore (REE(CO$_3$)$_2$F) containing commonly cerium, lanthanum or yttrium. REE fluorides from bastnaesite are removed by sulfuric acid roasting (400–500°C) emitting CO$_2$ and HF gases or alkaline pre-treatment (96°C). An alternative method consist on a thermal activation step at 400°C followed by HCl leaching that reduce the release of fluorine but not carbonate [23]. Until now only one attempt has been made to leach biologically bastnaesite minerals. Four actinobacterial strains were able to leach REEs from a bastnaesite-bearing rock and bastnaesite reaching. a total REEs concentration that ranged from 56 to 342 μg/l when grew in a nutrient-rich medium. Only the strain Streptomyces sp. grew in an oligotrophic medium in the presence of the bastnaesite-bearing rock and bastnaeasite, leaching 548 μg/l of REEs. The leaching efficiency of total REEs was very low (0.008–0.08%); however, this amount should be due to the REEs precipitation and/or sorption onto residual rock and cell surfaces. Several molecules, such as organic acids, complexing ligands, and siderophores secreted by the actinobacteria, are involved in the mechanism of REEs bioleaching from bastnaesite-bearing rock [13].

Xenotime is a phosphate ore and its content of yttrium is high and the presence of other light rare earths is low. Furthermore, the composition of xenotime is very complicated containing large amount of tungsten and other impurities such as iron and silicon. Generally, xenotime is more difficult to decompose than monazite and the mineral has been treated using concentrated sulfuric acid, alkaline solution at elevated pressure, fusing with molten caustic soda, and mixing with sodium carbonate and roasting [24]. These technological difficulties are probably linked to the fact that there are no biological approaches for REEs leaching from xenotime yet.

Monazite is also a phosphate mineral and is the major commercial source of cerium. Alkali treatment is also used for monazite dissolution and the phosphate is recovered as a marketable by-product, trisodium phosphate, by using caustic soda at high temperature and high pressure.

The bioleaching of REEs has been developed especially on monazite using microorganisms able to dissolve phosphorous from inorganic rocks, named phosphate solubilizing microorganisms (Figure 2). Numerous organisms including bacteria, fungi, actinomycetes and algae mobilize insoluble phosphorous. Bacterial species belonging to the genera Bacillus, Pseudomonas, Burkholderia, Enterobacter or Azotobacter, and fungi species of the genus Aspergillus, Penicillium, Trichoderma or Rhizoctonia are reported as phosphate solubilizers [25]. Phosphate solubilizing microorganisms have been previously used as biofertilizers in agriculture to promote crop production [26]; nevertheless, there are few works related to the recovery of valuable metals from phosphate minerals [27, 28].

Bacteria such as Enterobacter aerogenes, Pantoea agglomerans and Pseudomonas putida are able to grow in presence of natural rare earth phosphate minerals dissolving phosphorous, iron, thorium and REEs generating numerous organic acids [3].
Species such as *Azospirillum brasilense*, *Azospirillum lipoferum*, *Pseudomonas rhizosphaerae* and *Mesorhizobium cicer*, but especially *Acetobacter aceti*, solubilize cerium and lanthanum from monazite, although the efficiency of the process was low [28].

Several fungal strains able to solubilize phosphate minerals have been also used to leach monazite releasing rare earth elements to the aqueous phase, such as *Aspergillus niger* ATCC 1015, *Aspergillus terreus* strain ML3-1 and a *Paecilomyces* spp. strain WE3-F [14]. *Aspergillus niger* has been extensively studied because it generates organic acids such as gluconic, citric and oxalic acids [29, 30]. Nevertheless, the formation of REE oxalates limits the long-term dissolution of these elements [31].

The fungus *Penicillium* sp. reached a total concentration of 12.32 mg l$^{-1}$ REEs in aqueous phase after 8 days. Although monazite also contains radioactive thorium, bioleaching by these fungi preferentially solubilized rare earth elements over thorium that remained in the solid residual [3]. Fungus from the genus *Paecilomyces* produces several metabolites potentially associated with bioleaching, and citric and citramalic acids present a significant contribution to REEs dissolution, 6.4 and 15.0 mg l$^{-1}$ total REEs solubilized respectively. Furthermore, citramalic acid exhibits more interesting properties to those of citric acid due to the lower radioactive thorium release [32].

Not only heterotrophic but also autotrophic bacteria has been used for the treatment of monazite. The bioleaching of REEs using *Enterobacter aerogenes* and *Acidithiobacillus ferrooxidans* from various grades of monazite and the effect of glycine have been examined. Nevertheless, a decreased REEs bioleaching was observed in comparison to abiotic leaching or bioleaching in the absence of glycine [33].

Previous works have determined remarkable alterations in the natural microbial populations during bioleaching of monazite ores. The existence of native *Firmicutes* on the monazite seems to have significantly contributed to the increase REEs leaching observed when using non-sterilized monazite. A syntrophic effect between the native consortia and introduced microorganisms leads to a greater amount of leached REEs reaching 23.7 mg l$^{-1}$ [34, 35].

There are a number of mechanisms explaining the inorganic phosphate solubilization. The main mechanism is the production of biological dissolving compounds.
such as organic acids, siderophores, extracellular polymeric substances, protons, hydroxyl ions and CO₂.

The production of organic acids leads to the chelation of cations with their carboxyl and hydroxyl groups and/or to the reduction of the pH releasing phosphorous. The decrease of the pH results in the release of phosphorous by substitution of H⁺ for cations in the rocks [36]. Nevertheless, there is no correlation between pH and the concentration of solubilized phosphate and the cation assimilation is also required in the process [37] or the action of the H⁺ traslocation ATPase [38].

Gluconic acid is the most common organic acid involved in the mineral phosphate solubilization. This acid is generated by bacteria by direct oxidation of glucose and chelates the cations bound to phosphates [3]. Microorganisms not only produce organic acids but also inorganic acids and siderophores; however, these mechanisms are less effective in the release of phosphorous.

Extracellular polymeric substances have an indirect impact on phosphate solubilization due to their ability to bind metals influencing solubility of metal phosphates in soil. However, further investigation is needed to elucidate the role of high-molecular-weight polysaccharides in phosphate mobilization [39, 40].

Microbial phosphate solubilization can also take place through the liberation of enzymes like phosphatase or phosphohydrolase, phytase, phosphonatase, and C–P lyase. For example, phosphatase enzymes transform high-molecular-weight organic phosphate into low-molecular-weight products by the hydrolysis of ester phosphate bonds, releasing phosphate ions [41].

Despite of the recent work performed in this field, the selection of novel strains and a deeper study of bioleaching mechanisms is required to optimize the extraction of REEs. Biotechnology advances could have a key role in the development of cleaner strategies for the recovery of REEs.

4. Biorecovery of REEs from industrial and electronic wastes

Base metals as well as precious metals recycling have achieved high rates; however, the recycling rates of REEs are still very low (<1%). The low recycling of REEs can be explained by different factors, such as technological difficulties, low toxicities of the REEs, and, until few years ago, low prices and lack of incentives. The technological issues of the recycling of rare earths are due to the low concentrations of these elements in consumer goods.

The growing generation of industrial and electronic wastes and its significant content in critical metals has become these materials in an alternative economic source for the recovery of REE. Recently, secondary sources of REE including industrial wastes, mine wastes, and electronic wastes are being treated using bioprocess technology for the metal recovery. Nevertheless, bioleaching studies of REEs extraction from wastes are yet in their infancy.

Among REE-bearing industrial residues, phosphogypsum is worthy to be mentioned. REEs are often associated with phosphate deposits and phosphogypsum wastes are generated during the wet phosphoric acid process from fertilizer production in large amounts (100–280 Mt per year) with an estimate of 21 Mt of REEs locked into the total of phosphogypsum wastes accumulated to date [2]. A biolixiviant produced by the growth of the bacterium *Gluconobacteroxydans* on glucose containing organic acids was used for the leaching of synthetic phosphogypsum doped with six rare earth elements (yttrium, cerium, neodymium, samarium, europium and ytterbium). The pH of the lixiviant generated by the bacteria was 2.1 and the main organic acid present in the bioreagent was gluconic acid (220 mM). REEs leaching yield from phosphogypsum using the biolixiviant was compared
with the REEs leaching results using sulfuric acid, phosphoric acid, and commercial gluconic acid. The lixiviant produced by *Gluconobacter oxydans* was more efficient at REEs dissolution than gluconic acid and phosphoric acid but less efficient than sulfuric acid [42].

Other important waste material as REE resource is the red mud from the digestion of bauxites in the Bayer process. According to estimates, about 2700 Mt of red mud residues have been accumulated in Bayer plants all over the world and its generation increases at a rate of 120 Mt per year [43]. These residues are harmful due to its alkalinity, but also are rich in valuable rare earth metals as to be economically treated. Scandium represents about 95% of the economic value of the REEs present in red mud containing between 130 and 390 ppm [44].

*Penicillium tricolor* RM-10, a filamentous acid-producing fungi isolated from red mud, was used in bioleaching experiments. The optimum leaching ratios of the REEs were obtained under one-step bioleaching process at 2% pulp density due to the production of citric and oxalic acids. Nevertheless, the highest extraction yields were achieved under two-step process at 10% (w/v) pulp density [45]. The chemoheterotrophic bacterium *Acetobacter* sp. leached 53% of Lu, 61% of Y, and 52% of Sc under one-step process at 2% pulp density. In addition, this bacterium was suitable for red mud bioleaching at high pulp densities, because the production of organic acids excreted by the bacteria increased with the waste concentration [46].

Spent cracking catalysts are solid wastes generated in large amounts in oil refining and biocombustible industries. It is estimated that 700,000–900,000 tons of spent fluid catalytic cracking catalyst per year are generated worldwide and the management of this waste and the recycling of rare earth metals have become a challenge [47]. Cerium and especially lanthanum are the main REEs present in cracking catalyst. Cell-free culture supernatants of *Gluconobacter oxydans* containing gluconic acid were used for REEs leaching from spent cracking catalysts and dissolved 49% of the total REE with preferential recovery of lanthanum over cerium [4]. Furthermore, several studies have optimized this biolixiviant reaching a yield up to 56% and a continuous bioreactor system was developed achieving leaching efficiencies of 51% [48]. In addition, techno-economic analysis indicated that the use of agricultural wastes as substrate for bacterial growth instead of glucose provides a cost-effective process REEs recovery [49].

Bioleaching of spent cracking catalysts using the fungus *Aspergillus niger* at 1, 3 and 5% pulp densities achieved 63%, 52%, and 33% of lanthanum recovery, respectively; while the cell-free supernatant at 1% pulp density resulted in 30.8% leaching recovery. Furthermore, chemical leaching by strong inorganic (hydrochloric, sulfuric and nitric) and organic (citric, gluconic and oxalic) acids at 100 mM was evaluated. In chemical leaching, sulfuric, nitric and oxalic acids showed lower efficiency than bioleaching. Hydrochloric, citric and gluconic acids dissolved 68%, 65%, and 64% of REEs, respectively, which is nearly the same results than bioleaching with the fungus [50].

The recovery of REEs from fluid catalytic cracking spent catalyst by biochemical processes using *Yarrowia lipolytica* while also examining a route for the valorization of biodiesel-derived glycerin, which is the main carbon source, has been investigated. Remarkable bioleaching yields were obtained, 53% of La, and 99% of Ce and Nd, using *Y. lipolytica* IM-UFRJ 50678 at 50°C [51].

Compact fluorescent lamps contain on average glass (88 wt.%), metals (5 wt.%), plastic (4 wt.%), lamp phosphor powder (3 wt.%) and mercury (0.005 wt.%). The lamp phosphor fraction contains about 10% of rare-earth phosphors bound in the triband dyes [52]. Consequently, owing to the presence of REEs along with mercury, the disposal of lamp phosphors not only would lead to loss of resources but also to environmental hazards. Some countries collect great amounts of fluorescent
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phosphors as a distinct fraction from the recycling of fluorescent lamps (175 tons per year in Germany) and thus it is suitable as a secondary resource of REEs.

The most common rare-earth phosphors in these lamps are: $\text{Y}_2\text{O}_3: \text{Eu}^{3+}$, $\text{LaPO}_4: \text{Ce}^{3+}$, $(\text{Gd,Mg})_2\text{O}_2\text{O}_4: \text{Ce}^{3+}, \text{Tb}^{3+}$, $(\text{Ce,Tb})\text{MgAl}_2\text{O}_4$ and $\text{BaMgAl}_2\text{O}_4: \text{Eu}^{3+}$ [52] and it is possible to dissolve these REE-compounds through microbial processes (Figure 3).

A symbiotic mixed culture from tea Kombucha, consisting of yeasts and acetic acid bacteria, was used to leach REE from fluorescent powder. The highest leaching yields were achieved using the entire Kombucha-consortium or its supernatant as leaching agent compared to experiments using the isolates $\text{Zygosaccharomyces lentus}$ and $\text{Komagataeibacter hansenii}$. The pH decreased during the microbial growth as a result of organic acid production, acetic and gluconic acid [53].

Other study tested a broad spectrum of different microorganisms to evaluate their potential to dissolve REE from the lamps residues. Larger amounts of REE were leached with the strains $\text{Komagataeibacter xylinus}$, $\text{Lactobacillus casei}$, and $\text{Yarrowia lipolytica}$ comprising a total release of REEs of 12.6%, 10.6%, and 6.1%, respectively. Yttrium and europium were dissolved selectively during the experiments [54]. The supernatant generated by the bacterium $\text{Gluconobacter oxydans}$ was also tested for the recovery of REEs from phosphor powder; however, only about 2% of the total REEs was leached [4].

The phosphor powder solubilization is probably linked to the carboxyl-functionality or a proton excess. Among the different REE components preferably the red dye $\text{Y}_2\text{O}_3: \text{Eu}^{3+}$ was shown to be preferentially solubilized in accordance with the higher solubility of REE-oxides compared to REE-phosphates and –aluminates.

Electronic wastes are discarded devices that are at the end of their economic use and cannot be utilized by consumers anymore. The total global e-waste generation in 2021 is expected to achieve 52.2 Mt. The biggest economic interest is focused on gold with 50% of the possible revenue, but e-wastes contain other metals in significant amounts and still worth to be recovered. Investigations on bioleaching associated to printed circuit boards (PCB) recycling has mostly centered on copper and gold recovery. Ferric iron generated by iron-oxidizing bacteria is involved in

![Figure 3. SEM image of the fungus Aspergillus niger grown on fluorescent lamp powder.](image-url)
copper extraction. Cyanogenic bacteria or fungi have been tested to recover gold from e-wastes [55]. Nevertheless, these processes need to be upscaled and optimized. Recently, a two-step reactor has been developed to separate the production of biogenic ferric iron from the valuable metals leaching reaction achieving a 96% recovery of Cu [56]. Many e-wastes contain magnets with an important amount of REEs, 20–30%. The content of Nd, Dy, and Pr in NdFeB magnets is 259.5, 42.1 and 3.4 ppm, respectively [57]. Acidithiobacillus ferrooxidans and Leptospirillum ferrooxidans showed high leaching efficiencies when grew in presence of magnets but also in abiotic controls. Consequently, leaching mainly took place by chemical processes due to the presence of H₂SO₄ [58]. Furthermore, biodismantling is a new application of bioleaching in the recycling process of electronic waste to enhance the concentration of critical and precious materials imbedded in the electronic components. Considering sufficient sorting of the components after separation, some rare earth elements may become economically available by reaching a grade similar to commercial ores. A concentration of 9000 μg/g of dysprosium has been detected in one of these separated fractions [59].

The growing demand and applications of REEs in many fields lead to an increasing generation of industrial and electronic wastes. Thus, these materials become an alternative economic for REE recovery due to mineral scarcity and the environmental degradation, and the developments in bioprocess technology have a key role in sustainable mining for the green economy.

5. Conclusions

Rare earth elements possess unique properties that make them useful in a wide variety of applications, such as catalysts, magnets, batteries, phosphors and polishing compounds. Consequently, the demand of these elements is growing and the REEs extraction become an important issue. Biohydrometallurgy is a potential technological solution to conventional chemical processes that employ corrosive reagents with harmful effects on the environment. Bioleaching for the extraction of rare earth metals from mineral ores and industrial wastes can be performed by autotrophic and heterotrophic microorganisms, although the phosphate solubilizing microorganisms have been especially investigated. Several mechanisms are involved in the mobilization of REEs: organic acids, enzymes, bacterial attachment, phosphate regulation, siderophores... Moreover, the application of biotechnological strategies to the treatment of solid wastes might contribute to maximize the amount of resources minimizing the amount of tailings or residues that exert a harmful impact on the environment. Bioleaching of REEs is in its infancy, but the development of global market and the environmental policies as well as the appearance of new drivers such as synthetic biology and digital revolution could influence the evolution of biohydrometallurgy.

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Conflict of interest

The authors declare no conflict of interest.
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