

Recycling of spent electric vehicle (EV) batteries through the biohydrometallurgy process

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Abstract. Lithium-ion batteries constitute a primary component of electric vehicles (EV). The proliferation of EV on a global scale is expected to result in a rise in the quantity of spent EV batteries. The spent EV batteries comprise various heavy metals that possess a higher content than naturally available ores. These metals are valuable and have the potential to adversely affect the environment and human health if not managed appropriately. Conventional recycling techniques, such as pyrometallurgical and hydrometallurgical processes, have proven to be effective in the recovery of precious metals from used EV batteries. These techniques are used to recycle wasted EV batteries. Nonetheless, it should be noted that these processes are associated with a considerable cost, require high levels of energy consumption, present challenges in terms of regulation, and produce byproducts that can be classified as secondary pollutants. Biohydrometallurgy is a component of the discipline of hydrometallurgy that is widely recognized or thought of as an ecologically friendly and cost-effective extraction metallurgical technique as an alternative of extracting and recovering valuable metals from spent EV batteries. This approach involves the utilization of microorganisms. The present study employs a consortium of microorganisms comprising fungi, chemolithotrophic bacteria, mixotrophic bacteria, and acidophilic bacteria. These microorganisms have demonstrated their proficiency in metal recovery by generating acids and biosurfactants and utilizing ferrous ions and sulfur as energy sources. This article presents a review of biohydrometallurgical techniques as potential strategies for cost-effective and environmentally friendly technologies for the recycling of spent EV batteries. These techniques encompass the fundamental principles of biohydrometallurgy, in addition to the roles that microbes play in biohydrometallurgy.

Keywords: Biohydrometallurgy; Electronic vehicles batteries; Recycling technology; Valuable metals

1 Introduction

Lithium-ion batteries represent one of the most significant technological advancements in recent decades. Their convenience as energy storage devices is integral to daily life, owing to their high power and energy densities, as well as their extended lifespan. These attributes make lithium-ion batteries essential for powering a wide range of portable electronics, including laptops, cell phones, and power tools. Additionally, they are extensively utilized in various medical devices and equipment, such as portable medical monitors, defibrillators, and infusion pumps [1, 2]. In current research, the focus is on understanding the mechanisms of battery ageing specifically for automotive use.

Additionally, efforts are being made to enhance the safety features of lithium-ion batteries. These steps are crucial for the development of EV battery designs that prioritize both durability and performance [3–5].

In recent years, the swift progression of EVs technology has guided the automotive industry towards a more environmentally friendly and sustainable direction. EVs confer several advantages, including a reduction in greenhouse gas emissions, diminished operational costs, and less reliance on fossil fuels. A critical component of these vehicles is the lithium-ion batteries, which furnish the essential energy storage for effective and prolonged travel distances. However, the escalating production of EVs has led to concerns regarding the environmental impact associated with the lifecycle of these batteries,

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specifically the challenge of appropriately disposing of them once they have reached the end of their functional lifespan. [6] highlight the adverse effects of the EV recycling process on carbon neutrality in China, emphasizing the growing issues of waste accumulation and resource consumption. The build-up of waste from depleted lithium-ion batteries can lead to harmful consequences for both the environment and human health [7]. Conventional methods of battery disposal, often involving the direct placement of used batteries in landfills or incineration, not only exacerbate environmental pollution but also waste valuable metals contained within these batteries [8]. The metals in spent lithium-ion batteries (LIBs) typically targeted for recycling and recovery include lithium, cobalt, copper, aluminium, nickel, and manganese. The concentrations of these materials in spent LIBs are significantly higher than those found in natural ore sources, rendering depleted LIBs a vital resource for metal recovery and recycling [9, 10].

The extraction and recovery of valuable materials from spent LIBs present several advantages. This process aids in conserving essential natural resources by reducing the need for new mining sites, thereby mitigating the environmental impact associated with mining activities [11]. Moreover, recycling typically consumes less energy than extracting raw materials from ores [12]. This approach also fosters economic growth by creating new job opportunities and promoting the development of a circular economy [13]. Wang (2022) provides a multifaceted analysis of EV battery recycling and utilization, identifying potential challenges and limitations within the existing system [14]. These challenges encompass issues related to lithium metal anodes, dendrite growth, battery performance, and the overall recycling infrastructure. There is a clear need for a more robust and efficient recycling system for LIBs from EVs to address the potential waste and resource consumption that may arise with the escalating production of EVs.

Bioleaching is a hydrometallurgical technique that utilises metabolites produced by microorganisms to leach ores and waste materials, including EV batteries [11, 15]. This method involves the utilization of specific bacteria to catalyze the oxidation of metal sulfides, thereby releasing metal ions into a solution where they can be subsequently recovered and processed for recycling [15, 16]. Compared to conventional pyrometallurgical and hydrometallurgical methods, bioleaching offers several advantages in the context of spent LIBs. These include reduced energy consumption, minimized environmental impact, and the capability to selectively recover particular metals [15, 17]. Numerous studies have been conducted to investigate the bioleaching of metals from electronic waste, including spent LIBs, employing various bacterial strains [15, 16, 18]. For instance, the bacterium *Chromobacterium violaceum* has been demonstrated to effectively leach metals such as gold from electronic waste [15, 17]. Additionally, some research has explored the synergistic effects of using

mixed bacterial cultures, such as a combination of *C. violaceum* and *Pseudomonas aeruginosa*, to enhance metal mobilization [15]. These results highlight the significant potential of bioleaching as an eco-friendly and efficient method for metal extraction from discarded EV batteries.

The bioleaching process for spent LIBs is typically conducted in a series of stages, including the pre-treatment of the batteries to eliminate organic materials and enhance metal leaching, the leaching of metals utilizing bacterial cultures or cell-free media, and the subsequent recovery of metals from the leachate [15, 18, 19]. Several factors can influence the efficiency of bioleaching, such as the battery's composition, the conditions under which leaching occurs (including temperature, pH, and duration), and the selection of bacterial strains. The specific types and concentrations of metals within the battery can indeed affect the bioleaching process, as different metals may exhibit varying solubilities and susceptibilities to microbial leaching, thereby influencing the overall extraction efficiency [20]. Leaching conditions are also pivotal in determining bioleaching efficiency. Factors such as temperature, pH, and leaching duration must be optimized according to the particular bacterial strains used and the metals targeted for extraction [20]. Since some bacterial strains may demonstrate enhanced activity and metal solubilization within specific pH or temperature ranges, fine-tuning these parameters is vital for maximizing metal extraction efficiency. Furthermore, the selection of bacterial strains for bioleaching can have a significant impact on the process's effectiveness. Different strains may vary in their ability to solubilize metals and produce metabolites that aid in leaching [20]. Therefore, the choice of strains with high metal solubilization potential and compatibility with the battery materials is essential for achieving optimal bioleaching results.

The utilization of microorganisms for recycling spent LIBs represents a promising, environmentally friendly, and sustainable approach for the extraction of valuable metals. However, further research is required to optimize the bioleaching process, encompassing the identification of appropriate bacterial strains, the fine-tuning of leaching conditions, and the creation of effective methods for metal recovery. Additionally, a comprehensive evaluation of the economic feasibility and scalability of bioleaching for industrial applications remains necessary. The conclusion can be drawn that bioleaching is viable on a small scale for recovering specific metals from depleted electric vehicle batteries, meeting current standards. However, its applicability on an industrial scale remains to be comprehensively investigated [21]. A detailed assessment of the cost-effectiveness of the bioleaching process, including factors such as the cultivation and maintenance of microbial cultures, the optimization of leaching parameters, and the subsequent processing of extracted metals, must be conducted [21–23]. The advancement of economically efficient bioleaching technologies is vital

to render the recycling of spent LIBs a financially viable endeavour.

The challenge of scaling bioleaching processes for industrial applications is substantial. Transitioning from laboratory-scale experiments to large-scale industrial implementation necessitates meticulous evaluation of various factors, including process efficiency, throughput, and environmental ramifications [22]. The complexity of amplifying bioleaching processes, while preserving high metal recovery rates and minimizing both resource utilization and waste production, calls for additional research and technological advancement [22]. Furthermore, the heterogeneity in the composition and condition of spent EV batteries introduces an obstacle to the scalability of bioleaching processes. Variations in battery chemistry, electrode materials, and states of degradation can influence the efficacy of metal extraction, as well as the overall economic feasibility of the recycling procedure. The formulation of resilient and versatile bioleaching methodologies, capable of accommodating the wide array of spent battery materials, is a critical requirement for the realization of industrial-scale applications.

2 Pre-treatment of spent LIBs

The pre-treatment of spent EV batteries involves five key stages: sorting, discharge or stabilisation, disassembly, grinding, and separation [24, 25]. These steps are essential due to the intricate chemical composition and complex structure of spent batteries, which preclude direct treatment. The objective of pre-treatment is to simplify later stages and enhance the overall efficacy of the bioleaching process [26]. The first stage, termed 'battery sorting', aims to classify batteries based on various physical and chemical attributes such as shape, size, density, magnetic properties, and cathode material. Before initiating the disassembly phase, it is imperative to completely discharge the battery to prevent risks such as accidental short circuits, which can lead to heat generation or even ignition of flammable internal components. Typically, the discharge process involves immersing the LIBs in a sodium chloride (NaCl) solution with a concentration between 5% and 20%. Alternatively, submerging the batteries in liquid nitrogen can also reduce lithium metal reactivity. Disassembly can be manual or automated. Manual methods are generally suitable for small-scale settings like laboratories, while automated disassembly is designed for large-scale recycling efforts [27].

The composition of spent LIBs primarily includes the external metal casing, plastic components, separator, cathode, anode, and other materials such as adhesives, binders, electrolytes, and wires. In the disassembly phase, these components are segregated and processed as required [27]. Following dismantling, the LIBs are subjected to a crushing and grinding process. This step is crucial in the pre-treatment stage, aiming to mechanically

reduce the size of LIBs, thereby minimizing their volume and concentrating valuable components to enhance the efficiency of the subsequent bioleaching process. During the separation stages, the crushed EV battery components, now reduced to small flakes, undergo a differentiation process. This process sorts the materials based on their distinct physicochemical properties, including size, density, hydrophobicity, and ferromagnetism. The complexity of this differentiation lies in the fact that certain components, despite being different in size, may possess similar densities. This complexity necessitates the use of a multifaceted approach, combining various separation techniques to ensure the maximum efficiency of the process [25]. The primary goal of this pre-treatment is to extract a material commonly known as "black mass", or the active cathode material, from the expended batteries. This black mass, once sieved to specific sizes, is then utilized in the bioleaching process. Implementing this comprehensive pre-treatment procedure optimizes the recovery of target metals in the ensuing bioleaching stage.

3 Microorganisms employed in bioleaching of spent LIBs

Bioleaching is a technique that employs microorganisms or their metabolic byproducts to liberate valuable metals from ores, minerals, or waste materials, such as LIBs. The primary goal of bioleaching in the context of spent LIBs is to efficiently extract target metals through microbial degradation of battery components, leading to the release of these metals [20]. During this process, selected microorganisms oxidize sulfur and iron compounds, thereby generating acid and facilitating the dissolution of metal sulfides. Concurrently, the organic acids synthesized by the microorganisms function as chelating agents, forming complexes with metal ions, which further aids in their solubilization into the leaching solution. The efficacy of the bioleaching process is contingent on several factors, encompassing the selection of appropriate microorganisms, optimization of leaching conditions, and the specific composition of the spent LIBs.

Utilizing bioleaching for the recycling of spent LIBs presents several distinct advantages. Primarily, bioleaching is recognized as an environmentally benign and sustainable alternative to conventional chemical leaching techniques. This method diminishes the reliance on aggressive chemicals and curtails the production of toxic wastes [28]. Additionally, bioleaching has demonstrated superior efficacy in the removal of heavy metals relative to traditional chemical leaching approaches [28]. The inorganic acids and other metabolic byproducts generated by microorganisms can effectively solubilize metal compounds within battery materials, thereby facilitating the extraction of valuable metals. Furthermore, bioleaching may represent a cost-efficient strategy for metal recovery from spent LIBs. This is attributed to its potential for reduced operational expenses and energy consumption in comparison to other

recycling methodologies [29]. The employment of microorganisms as catalysts in the bioleaching process may also mitigate the requirement for costly reagents and specialized equipment [29].

Horeh et al. [20] conducted a study exploring the use of the fungus *Aspergillus niger* for bioleaching target metals from spent LIBs in mobile phones. The study evaluated the efficacy of *Aspergillus niger* in one-step, two-step, and spent medium bioleaching processes. The findings have demonstrated that both *Aspergillus niger* and *Penicillium* spp. are effective microorganisms for the bioleaching of spent LIBs. The bioleaching mechanism involves the synthesis of acids such as citric, oxalic, and malic acids by the fungi, which facilitate the dissolution of metal ions from the battery components. Notably, *Aspergillus niger* and *Penicillium* spp. exhibit superior bioleaching abilities, achieving higher heavy metal removal efficiencies compared to traditional chemical leaching methods [20, 30].

Additionally, a study by Liu et al. [31] examined the oxidative stress caused by metal ions during the bioleaching of LiCoO₂ using an acidophilic microbial consortium (AMC). This research focused on the effects of metal-ion stress on the bio-oxidative capabilities of the consortium during the bioleaching process of spent LIBs.

The results indicated that the bio-oxidative activity of the AMC was significantly influenced by the level of metal-ion stress encountered during the process [31]. This insight highlights the complex interplay between microbial activity and metal ion concentrations in the bioleaching environment.

In addition to *Aspergillus niger*, a diverse array of microorganisms has been utilized in the bioleaching processes for the extraction of valuable metals from LIBs and other forms of electronic waste. A microorganism exhibiting significant potential in this domain is *Acidithiobacillus ferrooxidans* [32]. A recent study by [33] demonstrated the use of acidophilic *A. ferrooxidans*, isolated from acid mine drainage, for bioleaching aged LIBs. The research indicated that cobalt content in LIBs could be recycled through a bioleaching process employing acidophilic *A. ferrooxidans*, a method characterized by high efficiency, cost-effectiveness, and environmental compatibility. The utilization of various microorganisms in the bioleaching of spent LIBs is summarized in **Table 1**.

Table 1. Microorganisms employed in bioleaching process of spent LIBs.

Organism employed	Battery powder	Bioleaching Medium	Bioleaching Condition				Leaching Efficiency (%)		References
			Temp (°C)	pH	Slurry Density (g/L)	Inoculum (v/v)			
<i>Acidithiobacillus ferrooxidans</i> (PTCC 1647)	<75µm (CR2032)	9K	30	10.6	40	10%	Li 100 Mn 20	Co 88	[34]
<i>A. ferrooxidans</i> (ATCC 19859)	<150µm	9K + 1 g/L elemental sulfur and 3g/L Fe(II) ion	30	2.5	5	10%	Li 9	Co 65	[35]
<i>A. ferrooxidans</i> (DSMZ, 1927)	<100 µm (Nanyang Technological University)	Modified 9K + 45 g/L FeSO ₄ ·7H ₂ O	30	2.0	100	10%	Li 60	Co 94	[36]
<i>A. ferrooxidans</i> (DSMZ 1927)	<100 µm (Nanyang Technological University)	Modified 9K + 150 g/L FeSO ₄ ·7H ₂ O	30	2.0	100	10%	Li 89 Mn 92	Co 82 Ni 90	[37]
<i>A. ferrooxidans</i> (PTCC 1647)	<75 µm	Modified 9K + 44.22 g/L FeSO ₄ ·7H ₂ O	30	2.0	10	5%	Li 57 Mn 42	Co 13 Ni 25	[38]
<i>Acidithiobacillus thiooxidans</i> (PTCC 1717)	<75 µm (Iran Computer Center)	Sulfur (5 g L ⁻¹), (NH ₄) ₂ SO ₄ (2 g L ⁻¹), K ₂ HPO ₄ ·3H ₂ O (0.25 g L ⁻¹), MgSO ₄ ·7H ₂ O (0.25 g L ⁻¹), KCl (0.1 g L ⁻¹)	30	2.0	30	10%	Li 99 Mn 20	Co 60	[39]
<i>A. thiooxidans</i> (80191)	<300 µm	Basal 317	30	2.4	2.5	10%	Li 60	Co 23	[40]
<i>A. thiooxidans</i>	<150 µm (CITIC GUOAN MGL)	Basic medium + 16 g/ L elemental S	30	1.0	10	5%	Li 98		[41]

<i>Leptospirillum ferriphilum</i>	<150 µm (CITIC GUOAN MGL)	Basic medium + 16 g/ L pyrite	30	2.0	10	5%	Li 49	[41]
<i>A. thiooxidans</i> and <i>L. ferriphilum</i>	<150 µm (CITIC GUOAN MGL)	Basic medium + 8 g/ L elemental S + 8 g/L pyrite	30	1.5	10	5%	Li 80 Co 90	[41]
<i>A. ferrooxidans</i> (PTCC 1647) and <i>A. thiooxidans</i> (PTCC 1717)	<75 µm (Moulinex, AR1044)	Silverman and Lundgren 1959 (<i>A. f</i>) and 8 g/L S, 2 g/L (NH ₄) ₂ SO ₄ , 0.1 g/L KCl, 0.25 g/L K ₂ HPO ₄ , 0.25 g/L MgSO ₄ .7H ₂ O (<i>A. t</i>)	30	2.0	10	10%	Co 99.5 (direct), Ni 99.5 (direct) Co 42.5 (indirect), Ni 71.5 (Indirect)	[42]
<i>A. ferrooxidans</i> = <i>A. f</i> (PTCC 1647) and <i>A. thiooxidans</i> = <i>A. t</i> (PTCC 1717)	<75 µm	Modified 9K medium + 5 g/L elemental S + 36.7 g/L FeSO ₄ .7H ₂ O	32	1.5	40	10% (<i>A. f</i> : <i>A. t</i> = 3 : 2)	Li 99.2 Co 50.4 Ni 89.4	[43]
<i>Aspergillus niger</i> (PTCC 5210)	<75 µm	Sucrose medium	30	6.0	10	1%	Li 95 Co 45 Mn 70 Ni 38 Cu 100	[20]
<i>A. niger</i> (PTCC 5010)	<75 µm	Sucrose medium	30	6.0	10	1%	Li 100 Co 38 Mn 72 Ni 45 Cu 94 Al 62	[44]
<i>A. niger</i> and <i>Aspergillus tubingenensis</i>	<250 µm	Sabouraud Dextrose Agar (SDA) medium	30	6.1	10	1%	Li ~95 Co ~60 Mn ~98 Ni ~80 Al ~82	[45]

In the context of spent LIB bioleaching, various methodologies can be employed, encompassing one-step, two-step, and spent medium bioleaching techniques.

- One-step bioleaching (direct bioleaching): This traditional approach involves a singular process where microorganisms are directly introduced to the battery materials, and the leaching occurs in one continuous stage [40, 46]. While this method is relatively uncomplicated, it may present constraints concerning efficiency and the extent of metal recovery.
- Two-step bioleaching (semi-direct bioleaching): This method consists of a two-phase process. Initially, the battery materials undergo pretreatment to augment the accessibility of metals to the microorganisms. Such pretreatment may encompass physical procedures like dismantling, crushing, screening, and magnetic separation [40]. Subsequently, the conditioned materials are subjected to bioleaching with microorganisms. This bifurcated approach facilitates enhanced control and fine-tuning of the bioleaching process, potentially leading to superior metal recovery [40, 46].
- Spent medium bioleaching (indirect bioleaching): This variant of bioleaching utilizes a medium containing organic acids generated by the microorganisms themselves [46]. The spent medium, rich in organic acids, is employed to leach metals from the battery constituents [47]. By circumventing the need for direct interaction between the microorganisms and battery materials, this method may offer a more efficient and scalable avenue for metal recovery [47].

These diverse bioleaching strategies have been the subject of extensive research. For instance, [20] investigated fungal bioleaching of spent mobile phone batteries using *Aspergillus niger* through one-step, two-step, and spent medium bioleaching, discovering that the mycelium of *A. niger* exhibited greater tolerance to battery powder toxicity compared to spores. In a separate investigation, Bahaloo-Horeh and Mousavi [44] conducted bioleaching of spent LIB using organic acids synthesized by *A. niger* for the extraction of metals. The selection of a particular bioleaching approach is contingent upon factors such as the specific microorganisms employed, the targeted metal recovery efficiency, and the scalability of the process. Each approach comes with its own inherent advantages and limitations, emphasizing the significance of further research to enhance and optimize bioleaching technologies for the efficient extraction of valuable metals from spent EV batteries.

4 Biohydrometallurgy mechanisms

The bioleaching process for spent LIBs generally involves three primary mechanisms: redoxolysis, acidolysis, and complexolysis. These mechanisms synergistically function to solubilize metals from the solid matrix of the spent LIBs, thereby enhancing their solubility. The specific mechanisms may differ based on the microorganisms utilized, the battery materials' composition, and the prevailing environmental

conditions. A comprehensive understanding of these processes requires further investigation to optimize bioleaching and maximize the recovery efficiency of valuable metals from spent lithium-ion batteries.

- **Redoxolysis:** This microbial-mediated process involves the oxidation of metal ions by microorganisms, leading to the dissolution of targeted metals from the battery materials' solid matrix [48–51]. The microorganisms engaged in redoxolysis can exploit various electron donors and acceptors, thereby catalyzing the redox reactions essential for metal dissolution [48–51].
- **Acidolysis:** This pathway involves microbial synthesis of organic acids that possess the ability to dissolve metals embedded within battery components [48–51]. Organic acids such as citric acid, oxalic acid, malic acid, and gluconic acid function as leaching agents, facilitating the solubilization of metals from the solid matrix [50–52].
- **Complexolysis:** This mechanism entails the microbial production of complexing agents, such as siderophores, which form complexes with metal ions, thereby augmenting their solubility and facilitating extraction from the battery materials [50, 52–54]. These complexing agents have the ability to chelate metal ions, rendering them more soluble and amenable to extraction.

In summary, the bioleaching of spent LIBs is a multifaceted process that leverages microbial activity to achieve the extraction of valuable metals. The interplay of redoxolysis, acidolysis, and complexolysis pathways, tailored to the specific characteristics of the battery materials and the microorganisms employed, constitutes the core of this environmentally sustainable approach to metal recovery. Continued research is essential to refine these processes and realize their full potential in the context of spent LIBs recycling.

4.1 Direct leaching (Contact leaching)

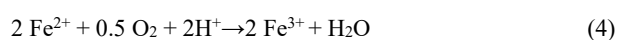
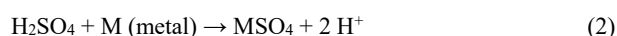
Direct leaching or contact leaching in bioleaching is a technique in which microorganisms engage directly with the solid matrix containing metals, such as those found in spent LIBs, bypassing the need for an intermediate leach solution. This method consists of several stages designed to enhance metal extraction.

- **Pre-processing of solid matrix:** Initially, the solid matrix, comprising crushed and sieved LIBs, is prepared to augment the surface area available for microbial interactions. This increased surface area facilitates more effective engagement between the microorganisms and the metal-containing matrix.
- **Introduction of microorganisms:** Subsequently, acidophilic or metal-mobilizing microorganisms are introduced to the pre-processed solid matrix. Acidophilic microorganisms generate an acidic milieu by producing sulfuric acid (H_2SO_4) through the oxidation of sulfur and ferrous ions. Conversely, metal-mobilizing microorganisms can directly oxidize

metal sulfides within the solid matrix, liberating metal ions and producing electrons (stoichiometric reactions 1 – 4) [23].

- **Oxidizing agents and redox reactions:** Both sulfuric acid (H_2SO_4) and ferrous ions (Fe^{2+}) act as oxidizing agents, aiding in the leaching of metal ions from the spent LIBs' solid matrices. The electrons generated in this process partake in redox reactions, resulting in the dissolution of metal ions from the solid matrix.
- **Microbial interaction and complex formation:** Microorganisms facilitate metal extraction through direct interaction with the material [55]. The metal ions released from the solid matrix may form metal-bearing complexes in the presence of organic acids synthesized by the microorganisms, thereby enhancing metal solubilisation [55].
- **Biofilm formation:** Microorganisms may adhere to the surface of the solid matrix and potentially form a biofilm. This biofilm creates a microenvironment that fosters microbial activity and promotes metal extraction.
- **Control of environmental factors:** The regulation of pH and nutrient availability is crucial to sustain optimal microbial activity and metal solubilisation [55, 56].

In summary, direct leaching or contact leaching in bioleaching is a multifaceted process that leverages microbial activity to extract metals from spent LIBs. By employing a series of carefully coordinated steps, this method offers a direct and efficient approach to metal recovery from complex solid matrices. Further optimization and understanding of this process could enhance its applicability in the recycling of spent LIBs.



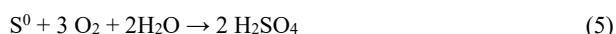
4.2 Indirect Leaching (Non-contact leaching)

In the indirect leaching or non-contact leaching process, bacteria function as catalysts without making direct contact with the surface of the LIBs. In this context, these microorganisms serve as catalysts, expediting key reactions essential for metal extraction, particularly the re-oxidation of ferrous ions (Fe^{2+}). This microbial acceleration occurs at a rate significantly faster than chemical oxidation at pH levels of 2 to 3. The enhancement in reaction rates observed is typically between 10^5 to 10^6 times faster [57–59]. The process can be described as follows:

- **Oxidation of elemental sulfur:** Bacteria are involved in the oxidation of elemental sulfur to sulfate ions. In the presence of protons in the surrounding environment, this oxidation leads to the production of biogenic sulfuric acid.

- **Creation of acidic conditions:** The biogenic sulfuric acid generated plays a crucial role in establishing the acidic conditions necessary for metal leaching. This acidic environment is conducive to the dissolution of metals from the LIBs.
- **Oxidation of ferrous ions:** The microorganisms assist in converting ferrous ions (Fe^{2+}) to ferric ions (Fe^{3+}), a change essential for the redox reactions within the system (Reactions 5-7).
- **Metal dissolution:** The redox reactions facilitated by the bacteria result in the dissolution of metals from the LIBs, enabling their extraction and recovery.

In summary, indirect leaching or non-contact leaching leverages bacterial activity to catalyze specific reactions that enable metal extraction from LIBs. By accelerating critical oxidation processes and creating favourable acidic conditions, this method offers an efficient pathway for the recovery of valuable metals from spent batteries.



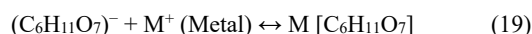
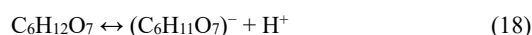
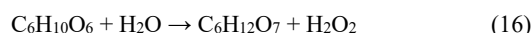
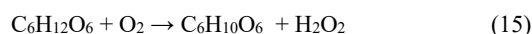
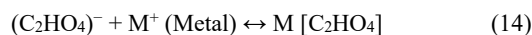
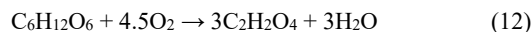
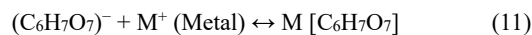
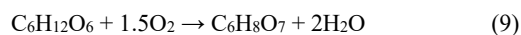
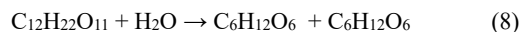
4.3 Fungal bioleaching mechanism

The fungal bioleaching of spent LIBs commonly employs microorganisms like *Aspergillus niger*. These microorganisms can produce organic acids that serve as catalysts, facilitating the effective extraction of metals from battery waste [20, 53, 60, 61]. Key organic acids, including citric acid, gluconic acid, oxalic acid, and malic acid, are instrumental in this process [20, 30, 48]. These organic acids function through several distinct mechanisms:

- **Acidolysis:** This mechanism involves the direct dissolution of metals through the action of organic acids, leading to the release of metal ions from the solid matrix [48, 52].
- **Complexolysis:** In this process, metal-organic acid complexes are formed, enhancing the solubility of metals. The complexation of metal ions with organic acids facilitates their extraction from the battery materials (Reactions 8 – 19) [48, 52].
- **Redoxolysis:** This mechanism encompasses reduction and oxidation reactions that enable metal leaching. The redox reactions mediated by organic acids contribute to the mobilization of metal ions [48, 52, 53].
- **Bioaccumulation:** This refers to the adsorption or binding of metals by fungal mycelia. The fungal cells can accumulate metal ions on their surface, aiding in their removal from the battery waste [48, 52, 53].

It can be summarized that the fungal bioleaching process of spent LIBs is a holistic procedure that harnesses the distinctive characteristics of organic acids synthesized by fungi. These acids act through various mechanisms to dissolve, complex, reduce, oxidize, and

accumulate metals, thereby enabling their extraction from the battery wastes.



5 Future prospects and challenges

The potential for utilising bioleaching techniques to extract valuable metals from spent LIBs is substantial. This method offers several advantages over traditional chemical leaching, including enhanced efficiency in extracting heavy metals and a smaller environmental impact. Recent studies have highlighted the notable efficacy of specific microorganisms like *Acidithiobacillus* spp. and *Aspergillus niger* in both detoxifying and recovering precious metals from spent LIBs [20].

Future investigations may focus on optimizing bioleaching parameters, such as pH, temperature, pulp density, and nutrient content, to enhance the extraction efficiency of valuable metals. A systematic examination of these factors and their influence on metal recovery efficiency could lead to improvements in the bioleaching process [20]. Furthermore, the selection and engineering of microorganisms to augment their bioleaching capabilities could be explored more extensively. Genetic modification or the employment of microbial consortia may enhance metal leaching efficiency. Given the presence of hazardous components in spent LIBs, such as heavy metals and organic electrolytes/solvents, the application of synthetic biology techniques, including genetic engineering, to alter metal tolerance genes within the microbial genome may be warranted. Such modifications could potentially increase the microorganisms' resilience to the toxic elements found in spent LIBs. The deployment of genetically modified microorganisms in the bioleaching process may also contribute to cost reduction in recycling by obviating certain pre-treatment procedures [47, 62]. In summary,

current research and refinement of bioleaching techniques present a promising path for the efficient and sustainable recovery of valuable metals from spent EV batteries.

The challenges and impediments that must be addressed throughout the recycling process of spent LIBs encompass the following aspects:

- **Collection and sorting:** The initial challenge lies in the effective collection and categorization of spent LIBs. The establishment of robust collection systems is essential to ensure that batteries are appropriately segregated and directed towards recycling processes [11].
- **Battery design and evolution:** LIB designs are in a continuous state of evolution to enhance energy density and performance. Consequently, recycling methodologies must adapt to these changes and be compatible with various battery configurations [11].
- **Efficiency of metal recovery:** The refinement of bioleaching procedures is vital for achieving elevated rates of metal recovery from spent LIBs. This necessitates the careful selection of suitable microorganisms, optimization of process parameters, and a comprehensive understanding of the bioleaching mechanisms [20, 63].
- **Scaling up and industrial implementation:** While bioleaching has demonstrated promise in laboratory-scale experiments, challenges arise in its transition to large-scale industrial applications. The widespread adoption of bioleaching in the recycling industry requires the development and refinement of cost-efficient and effective bioleaching technologies that can be seamlessly scaled up [63].
- **Economic viability:** Assessing the economic feasibility of recycling spent LIBs is a critical factor. To foster sufficient incentives for the recycling industry, the associated costs must be competitive with, or even lower than, those related to extracting metals from natural ore sources [64].

In summary, the recycling of spent LIBs presents a multifaceted set of challenges that require coordinated efforts across various domains, including technological innovation, process optimization, economic analysis, and industrial scalability.

6 Conclusions

The escalating concerns regarding the environmental impact of disposing of LIBs at the end of their lifecycle have stimulated research into innovative and ecologically sustainable methods for recycling and metal extraction. Bioleaching, an emerging hydrometallurgical technique, has been identified as a potential solution to the challenges associated with the growing volume of electronic wastes, including spent LIBs. The use of specific microorganisms, such as *Aspergillus niger* and *Acidithiobacillus ferrooxidans*, to aid in the solubilization of valuable metals from battery components is especially promising. This approach is

avored for its environmentally benign characteristics and the potential to achieve high metal recovery efficiencies.

The efficacy of bioleaching processes is contingent on several pivotal factors, encompassing the selection of appropriate microorganisms, the fine-tuning of leaching conditions, and the specific composition of the battery materials. Studies have elucidated that various bioleaching strategies, such as one-step (direct), two-step (semi-direct), and spent medium (indirect) bioleaching, can be customized to facilitate efficient metal recovery from spent LIBs. Despite the manifest advantages of bioleaching, obstacles persist in areas such as scalability, economic feasibility, and the heterogeneity of spent battery compositions. Continued investigation is requisite to enhance bioleaching techniques, formulate industrially cost-effective procedures, and navigate the complexities associated with diverse battery chemistries and constituent configurations.

The full potential of bioleaching in the recycling of spent LIBs necessitates interdisciplinary collaboration. Accelerating the development of efficient and scalable bioleaching solutions requires concerted efforts among microbiologists, metallurgists, materials scientists, chemical engineers, and industrial stakeholders. Advances in genetic engineering may lead to the creation of microorganisms with enhanced metal tolerance, thereby augmenting the overall efficacy of the bioleaching process.

As the demand for electronic products and EV increases, identifying sustainable methods for recycling and recovering metals from spent LIBs becomes essential. Bioleaching presents a potential pathway towards a circular economy for battery materials, mitigating adverse environmental impacts, conserving resources, and promoting a more sustainable future. By addressing the challenges associated with bioleaching and enhancing its application through continued research and technological innovation, there is potential to establish a more ecologically sustainable method for managing electronic waste and efficiently recovering precious metal resources.

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