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Advances in bioleaching as a sustainable method for metal recovery from e-waste: A review

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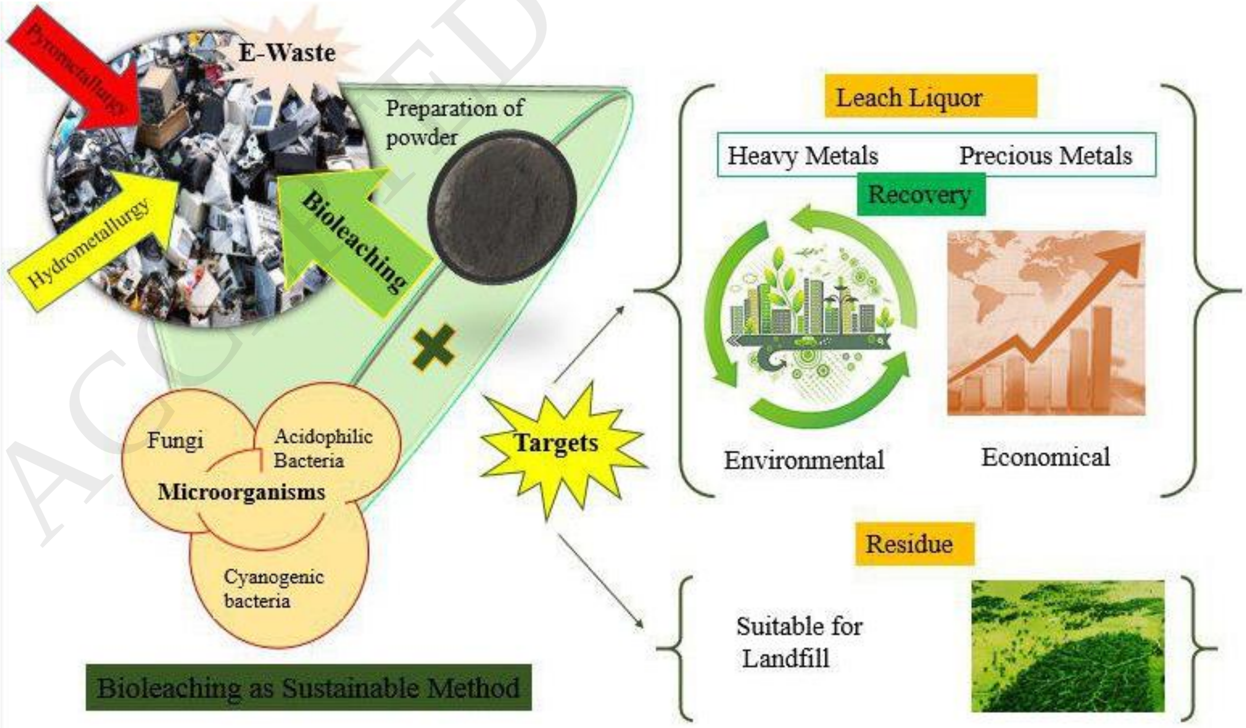
Graphical Abstract



Highlights

- In this review available methods for bioleaching of E-waste are summarized.
- Metal recovery from E-waste such as PCBs and batteries using bioleaching are reviewed.
- Available kinetic models for describing the bioleaching process are explained.
- State of the art, limitations and future work to be carried out in this field are reviewed.

Abstract

Electronic waste (e-waste) accumulation on earth is a serious environmental challenge. The need for heavy metal recovery, together with the profitability of precious and base metals, are strong incentives for researchers to find a sustainable method for metal recovery from e-waste. The scientific community is trying to improve the efficiency of metal recovery from e-wastes using bioleaching, a more sustainable method in comparison to traditional methods. In this review, available methods and the kinetic models that describe the bioleaching processes, but also their limitations, are reviewed. In addition, the application of new approaches to understand how the contribution of microorganisms and their genetic modification can affect the processes, are reviewed.

Key words: Bioleaching; E-waste; Heavy metals; Waste PCBs; Spent batteries

1. Introduction

Waste of electric and electronic equipment (WEEE) is defined as amalgams of various metals including copper, nickel, iron, aluminum, and zinc, to name only a few, trapped in plastic and ceramic matrices through mixing, coating or binding mechanism. High value waste contains precious metals like silver, gold and platinum group metals (PGMs), which are used for their high conductivity and high chemical stability [1]. The need for metal recovery from e-wastes is highlighted by the toxicity of these metals (especially heavy metals) for human health and environment, as well as by the economic benefits of WEEE as a secondary source of rare and precious metals [2]. According to the statistics, 40 million tons of e-waste are produced annually, which comprise 5% of the total solid wastes worldwide [3].

The content of urban mines has attracted the interest of both academic and industrial sectors due to the high amount of precious and rare metals, which are becoming depleted in natural resources but can be recovered from e-wastes with much less energy consumption [4].

The conventional methods available for metal removal from WEEE are pyrometallurgical and hydrometallurgical routes, and are considered to be environmentally unsustainable due to the difficulty of control, the production of secondary waste together with a high cost and risk of the process. In pyrometallurgical methods, which has high energy demand solid wastes are mixed with metal concentrates and smelted for metal recovery [5]. Energy crisis and concerns about emission of greenhouse gases led industry to look for alternatives. Emission of atmospheric pollutants such as dioxins and furans due to the presence of some flame retardants in electronic equipment is also a growing concern. Disadvantages of this method include high capital and operational costs, low selectivity, negative environmental impact, low social acceptance, and harsh thermal treatment conditions. However, this method has been proved to be feasible and is a developed technology on commercial scale. Hydrometallurgy is the process of solubilizing metals by using large quantities of chemical agents (acids). This method also needs to pretreatment of waste. However, this process is a slow, and hazardous process with negative environmental impact and high operating cost [6]. As a result, hydrometallurgy process includes the need for treatment and management of large amount of acidic wastewater effluent [7][8]. Biological metal recovery from solid waste is based on the same mechanism of hydrometallurgical process with employing microorganisms for production of reagents to extract metals [6]. Specificity, cost-effectiveness, and environmental acceptability are key advantages for metal recovery through microbial processes [9]. Bioleaching as a bio-hydrometallurgical process is simple and has advantages such as higher efficiency and safety, lower operating costs and energy consumption, easier management, implementation of operating conditions at atmospheric pressure and room temperature, eco-friendly, no need for skilled workers and use of few industrial requirements [10]. As an economic point of view, conventional processes require larger investments than bioleaching process [7].

Bioleaching is performed by different microorganisms (fungus and bacteria) with the ability to secret inorganic or organic acids or cyanide which enhance enzymatic oxidation-reduction, proton-promoted mechanisms and/or ligand and complex formation [7] [10].

Microorganisms with the bioleaching ability can be categorized in three major groups [11]:

a) Chemolithoautotrophic bacteria by acidolysis and redoxolysis mechanism

b) Organic acid producing fungi by acidolysis and complexolysis mechanism

c) Cyanogenic bacteria by complexolysis

Astonishingly diverse groups of microbes such as bacteria, fungi and yeast are used for leaching activity, although acidophilic sulfur-oxidizing bacteria and iron-oxidizing bacteria are the most widely used in this process [12]. The most well-known chemolithotrophs are *Acidithiobacillus ferrooxidans* and *Acidithiobacillus thiooxidans* which catalyze the oxidation of ferrous to ferric iron ions [13]. In addition, Bacillus species as the most effective bacteria as well as some fungi such as *Aspergillus niger* and *Penicillium simplicissimum* genera can be used in bioleaching process. *Chromobacterium violaceum* and *Bacillus megaterium* proved to be the most promising cyanogenic bacteria [10,14].

Generally three mechanisms of acidolysis, redoxolysis and complexolysis are involved for bioleaching of e-waste. In acidolysis, the protonation of oxygen atoms that covers the metallic compound surface occurs. Protons of organic acids produced by heterotrophs (malic, oxalic, gluconic, acetic, citric, succinic, pyruvic and formic acids) and also bacterial inorganic acids (e.g. H_2SO_4) have the ability for acidolysis [15,16]. Redoxolysis is the metal solubilisation mechanism through oxidation-reduction reactions. By redoxolysis, energy transfer necessary for microbial growth occurs through electron transfer. In the redoxolysis reactions of acidophiles, ferric ions are reduced enzymatically under anaerobic conditions, where hydrogen or sulfur acts as the electron donor [15,17]. Complexolysis is another mechanism by fungi and also is significant for precious metal recovery by cyanogenic bacteria. Cyanide is produced by decarboxylation of glycine in the late stationary phase of microorganism growth [18]. Many cyanide-producing bacteria have the ability to detoxify the cyanide to β-cyanoalanine by β-cyanoalanine synthase. This makes bio-cyanidation process an attractive process in the case of availability of less hazardous cyanide in the wastewater streams [9].

As shown in Table 1, microorganisms used for bioleaching of electronic waste were from all three groups mentioned earlier, considering chemolithoautotrophs, fungi and cyanogenic bacteria. It can be seen that the most common e-waste for bioleaching tests were PCBs and different types of batteries. Both mesophilic and thermophilic chemolithoautrophs have been used for bioleaching. When precious metals are targeted, cyanogenic bacteria are commonly used.

Different mixtures of chemolithoautotrophs bacteria have been used by researchers for the bioleaching of e-wastes including PCBs [5,36–38], LIBs [39,40], ZMBs [41–43] and cathode ray tubes [44] for better metal recovery due to the synergic effect of bacteria.

Target metals available in the electronic waste are mostly critical metals (including precious metals and some base metals). These are metals with large and essential consumption in industrial and technical applications, that have led to the depletion of their natural sources [45]. Among the e-wastes, printed circuit boards (PCBs) are the most heterogeneous, since they contain many elements including base metals, heavy metals, alkali metals, precious metals, lanthanides, actinides and some non-metal elements. Precious or noble metals are resistant to oxidation, corrosion and acids. PCBs contain the highest amount of precious metals, which are used due to their desirable conductivity and stability; as such, they can be considered as “artificial ores” as they contain significant amount of Pt, Au and Ag [7,46].

E-waste contains a wide range of discarded devices from small to voluminous electronic equipment. However, bioleaching studies mostly focus on the devices that contain metals of interest which are heavy or precious metals and rare earth elements in most of the cases. In previous research available in the literature which will be reviewed in this work, bioleaching has been most widely studied for PCBs and batteries (lithium ion batteries (LIBs), Ni-Cd batteries and Zn-Mn batteries (ZMBs)) (sections 2.1 and 2.2). Other form of e-waste such as panels of liquid crystal displays (LCD), light emitting diodes (LED) can be also noteworthy for research (section 2.3).

This work is a review of the current state of the art in the field of bioleaching of e-wastes. Previous reviews on metal recovery from e-waste focused mostly on pyrometallurgical and hydrometallurgical methods, and often only mentioned bioleaching briefly [47]. Lee and Pandey (2012) instead focused on bioprocesses available for metal recovery of different industrial and urban waste including some e-wastes [48]. The aim of this review is to describe the fundamentals of bioleaching and the obtained results of the up-to-date published research in the field of bioleaching of e-wastes. For this purpose, the authors describe the principles of the bioleaching including mechanism, microorganism and bioleaching methods and process. Previous studies on bioleaching of e-waste are reviewed and classified according to waste type, microorganism and target metal. The bioleaching stages include waste preparation, microorganism adaptation, as well as one-step, two-step and spent-medium bioleaching methods are described in this review. The limitations of the bioleaching process are also described. The kinetic study of the process is also discussed and the catalysts that were used in previous studies are reviewed. Finally, future prospects and open issues for the bioleaching of metals from e-wastes are discussed in the last part of this review.

2. Bioleaching of e-waste

2.1. Discarded printed circuit boards (PCBs)

PCBs are the most common target of the recycling process due to the presence of precious metals and hazardous materials content as a step forward for resource preservation and circular economy [49].

The heterogeneity of electronic scrap (ES) makes the recycling process very complicated [1]. PCBs are important component of electronic scraps although they comprise only 6% of e-wastes [19]. PCBs composition can be varied with the year of production and the manufacturers [50]. It has been reported that annually 17 million computers are discarded, which corresponds to the disposal of 50 million kg of CPUs annually [32]. It has been reported that 0.5 million tons waste PCBs are disposed annually [51].

The most important and studied PCBs are computer and mobile phone PCBs, because of their high precious metal content, high disposal amount and short lifespan estimated to 4 years, considering also reuse [19].

The composition of PCBs varies between sources. However, polymers, ceramics and metals are only fractions of this waste. Around 28% of PCBs is the metal fraction including Cu (10-20%), Pb (1-5%) and Ni (1-3%), plastic materials compose 19%, glass and ceramics 49%, Br 4% and very little percentage (0.3-0.4%) precious metals like Pt (10-200 ppm), Au (20-500 ppm) and Ag (200-3000 ppm) [19] which are used as a contact material due to their suitable conducting properties and chemical stability [1]. Ceramics available in PCBs are composed of silica, alkaline earth oxides, alumina, mica and barium titanate [52].

Regarding the metal recovery process, PCBs are categorized into three classes of high, medium and low grade scrap on the basis of their precious metal quantity [52]. High and medium grade materials are those having high concentrations of Pd and Au. It must be considered that almost 90% of PCBs value is for these two metals. In high and medium grade PCBs, the metals are easily separated, and are available in integrated circuits, pin boards, and thermally coupled modules of mainframe. The second category is medium grade waste materials and includes wastes with high amount of precious metals which are in pin and edge connectors together with aluminum capacitors. The low grade PCBs are those available in televisions, and power supply units with transformers of heavy ferrite and aluminum heat sink assemblies. The metal recovery from this category is not cost effective [53]. Two fractions of PCBs are high grade or metal-rich (RPCBs) and low grade or metal-poor (PPCBs) fractions are separated through mechanical methods by shaking table separator after grinding and sieving. The high grade fractions are suitable for pyrometallurgy and the low grade fractions are suitable for bioleaching [36].

PCBs are traditionally disposed by incineration and landfills. Informal disposal of waste is done in backyard workshop by techniques such as acid washing and incineration. The risks for environment and human health on one hand, and the economic motivations coming from metal recovery on the other hand, have motivated the scientific community for finding alternative recycling methods [7].

Bioleaching has been reported to be a commercially feasible and as a low cost solution for recovery of metals from low grade ores. However its application to the PCBs and other electronic wastes is still in the research phase [8]. One of the limitations of bioleaching of PCBs is the toxicity of the waste to the microorganism, although it is not clear whether this toxicity is the result of the metal or organic content [53].

The most common microorganisms used for the bioleaching of PCBs are chemolithoautotroph (mesophilic) bacteria or cyanogenic bacteria like *C. violaceum* depending on the target metal [49]. Among cyanogenic organisms, *C. violaceum* can be more important due to its six HCN synthase enzymes which could produce more cyanide. Compared to other microorganisms, it has better versatility of energy metabolism [9].

The first category (chemolithoautotrophs) are suitable for base metal recovery while the cyanogenic bacteria are the most promising microorganism for gold recovery. For Au recovery from PCBs, which contain also high amount of copper, the Cu^{2+} ions may interfere with Au recovery, due to the formation of Cu–cyanide complexes that are formed faster and at higher concentration than Au–cyanide complexes [20,30]. In the other words, the Cu dissolution occurrence is dominant to Au dissolution due to two following reasons [54]:

- ✓ Base metals such as Cu consume free cyanide and make it unavailable for complexation of Au

- ✓ The standard electrode potential (E^0) of gold (Au^0/Au^+) equals to -1.83 V which makes it less reactive than base metals such as Ni (E^0=-0.67 V), Cu (E^0=-0.34 V) and Fe with E^0=-0.44 V)

Au complexes with cyanide by the Elsner's reaction [55]:

$$2\mathrm{Au} + 4\mathrm{CN}^- + \mathrm{H_2O} + 1/2\mathrm{O_2} \rightarrow 2\mathrm{Au(CN)}_2^- + 2\mathrm{OH}^- \quad (1)$$

Therefore, separation of base metals in the first step and precious metals in the second step should be envisaged for the bioleaching of PCBs. This process named pre-treatment, has been performed by chemical method (acid leaching) or biological methods (using chemolithoautotrophs). Işıldar et al., used a novel method to separate the base metals in the first step by chemolithoautotrophic acidophilic bacteria and the precious metals in the second step by cyanogenic bacteria [54].

Results using bioleaching of PCBs with chemolithoautotrophs for base metal recovery is summarized in Table 2, while the work focused on gold recovery which is using cyanogenic bacteria was mentioned earlier in Table 1.

Xia et al. have performed PCB bioleaching in stirred tank reactor. They firstly separated PCBs into two fractions which were metal rich and metal poor using a shaking table separator, and performed the bioleaching on the metal poor fraction. They have proposed a semi industrial process based on this method and did the economic analysis to prove the feasibility of the process [36]. The most comprehensive techno-economic assessment has been done by Isildar (2018) [59]. The process used for metal recovery from PCBs in that work had two steps including a pretreatment method for copper recovery and then gold recovery in the next step. Three different technologies has been assessed for that process, which were biological, chemical and hybrid technologies. In the biological method, both steps were bioleaching and in the chemical method both steps were chemical leaching, while the hybrid method was the application of acidophiles for copper recovery and using commercial cyanide in the second step for gold recovery. The comparison of the operational and capital cost of the three technology has been carried out and can be seen in the Table 3.

It was concluded that the most economical process for metal recovery was the biological method. However, considering the revenue and return on the investment, chemical method was found to be the most competitive. The environmental aspect of each process, was also evaluated by calculating the contribution of each method to the climate change, which showed that the most environmental friendly process is the biological method [59].

Jagannath et al., have used a pulsed plate reactor in order to perform the large scale bioleaching. They have studied the effect of parameters including inoculum size, waste load, frequency and amplitude of the pulsation [2].

In most of the cases Cu and Au were the target metals for PCB bioleaching. The use of catalyst to improve the bioleaching of PCBs was also done in several works [51][57]. In the work of Wang et al. three different pathways, carbon-mediated (C-mediated) , sulfur mediated and iron-mediated processes, were tested for determining the role of biochar in promoting the metal recovery in bioleaching pathways [57] which can facilitate the electron transfer in the process. It was observed that in C-mediated process where glucose was used as electron donor against the metabolite production, and the addition of biochar did not change the function of electron transfer chain. It was suggested that inefficiency of the C-mediated pathway was due to the lack of functional bacteria, organic acid and cyanide. In S-mediated pathway, where the sulfur reduced as substrate, the biochar addition blocked the process. It was due to the coverage of sulfur powder with biochar, which limited the sulfur consumption by microorganism. This led to less biomass production and lack of H^+ release. In other word, the electron transfer chain (from S^0 to bacteria) was blocked in the presence of biochar, and also the dominant microbial species was changed. However, in the Fe-mediated pathway, the electron transfer for oxidation of Fe^{2+} was enhanced due to the transformation between oxidation and reduction states of quinone and aromatic structures. Redox-active biochar in Fe-mediated pathway enhance the transfer of electrons and the rate of redox reaction of Fe^{2+} to Fe^{3+} and efficiency of Cu leaching from PCBs [57].

A low-cost oxidant such as ferric iron that is frequently used in hydrometallurgy and can particularly leach various metals from PCBs. Hubau et al. investigated generation of biogenic ferric iron by using BRGM-KCC acidophilic consortium in a bubble column in the presence of activated charcoal. The effects of concentration of ferrous iron in the range of 1 to 9 g L^{-1} on the bio-oxidation rate was studied in the presence of a solid support. The impacts of solid support type and quantity, hydraulic residence time (HRT), and culture medium composition on bio-oxidation yield were investigated. The study showed that colonization by the microorganisms was affected by solid support clogging with jarosite precipitates. The structure and the abundance of the microbial community was also influenced by operating conditions, ferrous iron concentration and nutrient composition of medium [17].

Priya and Hait suggested a hybrid bioleaching and hydrometallurgy approach, in which citric acid as a chelating agent improved the base metal recovery from PCBs. Citric acid improves production of expolymeric substance by *A. ferroxidans* and decrease the jarosite formation [58].

The most recent findings in the application of chemolithoautotrophs for base metal recovery are summarized in Table 2, while the findings of the recent research in the field of cyanogenic bacteria application for precious metal recovery from PCBs can be summarized as follows:

- ✓ The optimum pH for cyanogenic bacteria growth is in the range of 7 to 8, while the chemically the cyanide might be lost at this pH due to the volatilization (pKa= 9.3), therefore there is a need to compromise between the chemical and biological effect of pH [34]

- ✓ Higher amount of glycine might improve the yield of cyanide and gold recovery, but glycine can have an inhibitory effect after a certain period [54]
- ✓ Better recoveries can be obtained at lower particle size and lower pulp densities [32]
- ✓ The amount of dissolved oxygen has a determining effect on the metal recovery by cyanogenic bacteria as oxygen apart from being needed for bacterial respiration is needed as electron acceptor [60]
- ✓ Mixed culture of autotrophs and heterotrophs can be more in metal removal efficiency effective compared to pure cultures of single-strain and also synergistic effects occurs between microorganisms of bioleaching [61].The mixed culture of different bacteria mostly from two groups of iron and sulfur oxidizing have shown to be beneficial for the bacterial growth and activity and as a consequence metal recovery. For example *A. thiooxidans* can provide the favorable condition for the growth of iron oxidizing bacteria [62]. The superior rate of extraction with mixed culture was also observed for bioleaching of ores [63].
- ✓ When iron-oxidizing and sulfur-oxidizing chemolitoautotrophs coexist in the medium, more copper mobilization was observed from PCBs. In this case, both redoxolysis and acidolysis mechanisms are available [54].
- ✓ In addition, mixture of *Thermoplasma acidophilum* and chemolithoautotroph bacteria (*S. thermosulfidooxidans*) for bioleaching of PCBs have also been studied by Ilyas et al in years 2010 and 2013 [64][1]. The authors explored the potential of adapted consortia of moderately thermophilic bacteria (autotrophs and heterotrophs) for metal recovery from

scrap on a small scale such as column reactors [1]. In the next paper, they focused on applying consortia of moderately thermophilic bacteria (iron and sulfur oxidizing) in metal recovery from e-waste [64].

2.2. Disposal of spent batteries

Two types of batteries are available which are primary non-rechargeable batteries and secondary rechargeable ones [28]. Primary batteries include ZMBs while the secondary batteries are mostly LIBs, Ni-Cd batteries and nickel metal hydrid batteries [41].

Accumulation of spent batteries in general, causes serious problems to the ecosystem due to the presence of hazardous materials. They are also a rich resource of the metals to be recovered [28].

EU Battery Directive 2006/66/EC imposes that at least 50% (weight) of waste batteries must be recycled to be used for new batteries or other purposes but not energy recovery [26]. Therefore, there is a need to find technical solutions for the treatment and recycling of these wastes [65]. It must be noted that the batteries built from recovered material has 51% less life cycle impact in comparison to the ones that was produced by primary materials [66].

2.2.1. Spent lithium ion batteries (LIBs)

Technology, industry and communication development has led to a sharp increase in the consumption of portable electronic device such as mobile phones, laptops, and cameras [26]. Therefore, there is a huge demand for electrochemical power sources such as rechargeable batteries and as a result a large number of used batteries is discarded in the environment [67].

LIBs are preferred to aqueous electrolytes rechargeable batteries such as Ni-Cd [65], representing 60% of total portable battery sales in the year 2011 [67]. In 2011, the global consumption of LIBs was reported to be 4.49×10^9 unit with annual growth rate of 14.5% from 2006 to 2011 [68].

The significant difference between primary lithium batteries (LBs) and secondary rechargeable LIBs should be noted. Primary LIBs are prone to catch fire after corrosion since they have metallic Li as cathode material and no toxic metals; on the other hand, secondary rechargeable LIBs contain material like $LiXMA_2$ ($LiCoO_2$, $LiNiO_2$, $LiNiMnCo_2$) instead of metallic Li in their cathode in order to overcome the fire hazard [65].

Currently there are strict regulations on the fate of hazardous wastes such as spent portable batteries containing heavy metals [65]. This waste should be managed not only for their potential hazard to ecosystem and human health, but also for the presence of valuable metals like Co (5-20%) and Li (5-7%) which are experiencing a shortage in the natural resources [69][70]. Co as a quite critical and high cost metal and Li have application in several industries. The concentration levels of some metals in LIBs are even higher than natural ores or their processing concentrate [65]. Cathode materials are usually the most expensive part of the battery. However, the different compositions of the cathode material are available and are being developed to produce LIBs with low cost and high energy density. Most of the current recycling efforts have focused on Co recovery from $LiCoO_2$ cathode material. However, other materials have been substituted to Cobalt in the LIBs cathode material to lower use of Co and Ni for their high price [67].

The hazards of this waste is due to their flammable and toxic ingredients, which during landfill may leach to the soil, groundwater and rivers. The valuable metals available in LIBs are Co, Li, Ni, Al, Cu, and Mn [26].

The effective LIB waste management is usually through recycling which leads to valuable metal recovery, and environmental impacts reduction. The regulations must be observed during all production, use, collection, recycling and disposal steps [68]. Before recycling, the spent LIBs should be manually dismantled into plastic shell, cathode, anode, aluminum foil, etc. Then the cathode and anode rich in Co, and Li are mixed, grounded, and sieved to the specific size [40].

For the bioleaching of spent LIBs, usually acidophilic bacteria like *A. ferrooxidans* are used. Bioleaching of Co from LIBs was studied using copper ions as catalyst to solve the problem of low metal recovery [65].

In the work of Niu et al. the thermodynamic of bioleaching was compared with chemical leaching. The negative value obtained for ΔG with bioleaching process indicated that bioleaching of LIBs is more probable to occur spontaneously in comparison to chemical leaching. However, the speed of the reaction is very low [40].

In some studies the catalytic effect of metals (Cu, Ag) was studied [65,71]. Xin et al. have studied the metal recovery from electronic vehicle LIBs by bioleaching [39]. They have determined the dominant mechanism for bioleaching of different metals (Li, Co, Ni, Mn) from LIBs. The addition of Fe^{2+} did not affect the Li release in the presence of H_2SO_4. This proved that the dominant mechanism for Li mobilization was acid dissolution. However, Ni, Co and Mn recoveries were enhanced by increase of initial Fe^{2+} concentration. This results were further approved in the work of Heydarian et al. for bioleaching of spent laptop LIBs with mixed *A. ferroxidans* and *A. thiooxidanse* by statistical analysis of the influence of different parameters which were initial sulfur and initial Fe^{2+} concentration [72].

Table 4 summarizes the findings in the previous works in the field of LIBs bioleaching.

Li, Ni and Co available in LIBs are mostly in the form of metal oxides such as $LiCoO_2$ and $LiNiO_2$. Eqs. (2)-(4) were suggested by different researchers for metal recovery from LIBs [73][40][74]:

$$4LiCoO_{2(s)} + 6\ H_2SO_{4(aq)} \rightarrow 2Li_2SO_{4(aq)} + 4CoSO_{4(aq)} + 6H_2O_{(aq)} + O_{2(g)} \quad (2)$$

$$4LiNiO_2 + 6H_2SO_4 \rightarrow 2Li_2SO_4 + 4NiSO_4 + O_2 + 6H_2O \quad (3)$$

$$2FeSO_4 + 2LiCoO_2 + 4H_2SO_4 \rightarrow Fe_2(SO_4)_3 + 2CoSO_4 + Li_2SO_4 + 4H_2O \quad (4)$$

2.2.2. Spent nickel cadmium batteries

Ni-Cd and nickel metal hydride (NMHs) belong to the group of rechargeable secondary batteries with several industrial applications specially in portable electronic devices such as wireless communication and computing equipment [28].

High content (51-67%) of heavy metals is available in Ni-Cd batteries. The electrode material contains 22-46% Cd, which is much higher than earth crust Cd content (only 0.1-0.5 ppm). Different from the metal ores which contain mostly metal sulfides, metals in Ni-Cd batteries are mostly in the form of elemental metals, metal oxides and hydroxides [23].

Around 3.9% of discarded batteries are Ni-Cd batteries, which are classified as hazardous waste due to the presence of heavy metals and carcinogenic effect [75]. Therefore, different EU directives have been established for their collection. Several treatments are available based on pyrometallurgical and hydrometallurgical or combined techniques for Ni-Cd batteries recovery [23].

Despite of environmental concerns, Cd has several industrial applications such as stabilizers and alloys, coating, pigments, and battery production. Therefore reuse of metals available in battery could be considered as secondary resources [23].

Several researchers have used a bioreactor system in which, the microbial growth or biogenic acid production was conducted in a separate reactor from bioleaching reactor in which the batteries were placed [75][76][77].

Table 5 summarizes the recent research in the field of Ni-Cd bioleaching.

2.2.3. Spent zinc manganese batteries (ZMBs)

The most frequently used primary batteries are ZMBs [28]. It was estimated that the annual consumption of ZMBs is up to 40 billion unites in the world [42] and they comprise 90% of the total portable batteries annual sales [43]. This indicates the large accumulation of these batteries in the nature with serious threat to environment and human health [42]. The presence of Hg, Zn, Mn and heavy metals, makes spent ZMBs among the toxic and hazardous waste [43].

ZMBs are non-rechargeable batteries and therefore their consumption is far more than secondary ones (LIBs and Ni-Cd batteries). Zinc-Carbon and alkaline batteries are other rechargeable batteries and are used in several household devices including radios, calculators, cameras, remote controls and toys [42].

ZMBs contain high percentage of Zn (12-28%) and Mn (26-45%) and can be considered as secondary resources of these metals [42]. The rest of the composition include graphite, K and Fe which are quite abundant and nontoxic [43].

Large amount of batteries are currently landfilled or incinerated. However, several countries established regulations to ban landfill and incineration in favor of collection and recycling. In addition, Mn and Zn are among strategic metals due to their high demand and limited natural resources and must be recovered and reused [43].

Recently bioleaching of this waste for recovery of metals has been investigated in several studies. The high content of alkaline and toxic matters in the form of oxides and hydroxides present in the ZMBs, urges to metal recovery from the waste before landfill [42].

The catalytic effect of metallic ions Ni^{2+}, Co^{2+}, Cu^{2+} and Ag^{+} was analyzed for bioleaching of ZMBs. Among them Cu^{2+} proved to be an effective catalyst of the bioleaching process, via formation of $CuMn_2O_4$ [42].

Kim et al. have used the spent-medium obtained from fungi to recover the metals from ZMBs [28]. Sun et al. performed LCA for bio-hydrometallurgical metal recovery from ZMBs. They found out that the main environmental impact categories were human toxicity and marine eco-toxicity comes from metal emission in mechanical cutting and crushing step. Therefore, there is a need for well-designed, and closed battery cutting and crushing equipment to meet environmental requirement. The authors suggested that the process must be optimized considering large bacteria consortia, high pulp density and reuse of solid residue [78]. Table 6 summarizes the findings in the previous works in the field of bioleaching of ZMBs.

In ZMBs the metals (Zn and Mn) are in the form of hetaerolite ($ZnMN_2O_4$), manganese oxides (MnO_2), simonkilleite ($Zn_5(OH)_8Cl_2H_2O$) and zinc ammine chloride ($Zn(NH_3)_2Cl_2$). Zn^{2+} mobilization from zinc ammine chloride compounds is straightforward. Metal extraction from simonkolleite as Zn^{2+} can be obtained by biogenic acid dissolution as alkaline matter [42].

Mn recovery from manganese oxide is by combined reaction of acid dissolution and reduction by ferrous ions generated by biogenic and chemical reactions (Eq. 5) [42]:

$$\mathrm{MnO_2 + 2Fe^{2+} + 4H^+ \rightarrow Mn^{2+} + 2Fe^{3+} + 2H_2O} \tag{5}$$

However, hetaerolite remains even after bioleaching [42].

For the bioleaching of ZMBs, the highest pulp density used was 10% in the work of Niu et al. They used the catalyst to improve the pulp density and Zn and Mn recovery from ZMBs by boosting the bioleaching of hetaerolite. However, jarositie formation led to further blockage between cell and energy matter and stopped the Fe^{3+}/Fe^{2+} cycle. Bioleaching of hetaerolite could not be continued thereafter. The kinetic of ZMBs bioleaching was best described by chemical reaction controlled model [42].

In the study of the parameters influencing metal extraction from ZMBs, it was observed that metal recovery was not affected by type of energy source and bacterial species (sulfur oxidizing or iron oxidizing bacteria), while it influenced strongly by initial pH. The metal recovery decreased almost linearly with the increase of pH. Mn recovery from ZMBs occurred during contact mechanism between microbial cells and the waste material. The XRD analysis was performed on the ZMB powder before bioleaching which showed the presence of Mn in the form of Mn_2O_3 and Mn_3O_4 in the waste. When the bioleaching was performed in non-contact mechanism (through separation of waste from microbial cell by dialysis bag) the existence of Mn was still observed but in the form of MnO_2. However, Zn was completely absent in the bioleached sample even in non-contact approach [79].

2.3. Miscellaneous e-waste

Several studies have shown the bioleaching of miscellaneous e-waste including fluorescent powder obtained from cathode ray tube glass recycling process [44], obsolete SIM cards [31], TV circuit boards [80] and electrolyte manganese slag [79]. However, there are still several other e-wastes for bioleaching which should be investigated for recovery of hazardous heavy metals and also reuse of some precious and economically viable metals. These include panels of liquid crystal displays, light emitting diodes in lamps and TV to name only a few.

In the work by Beolchini et al. the acidophilic mixture of iron and sulfur oxidizing bacteria were used for the bioleaching of zinc and yttrium from fluorescent powder coming from cathode ray tube glass recycling process, studying the effect of pulp density and substrate. This waste comes from recycling process of cathode ray tubes by diamond cut technology. Around 30% of metal recovery was obtained in this study [44].

Obsolete SIM cards are another group of e-wastes that is increasing in the environment due to the expansion of cellular networking. This waste contains high quantity of Cu (75.84%), and small amount of precious metals (0.042% Au and 0.01% Ag). In the work that studied the bioleaching of SIM cards [31], *C. violaceum* was used as microorganism. However, the obtained yield were not satisfactory (13.79% for Cu, 0.44% Au and 2.55% Ag). Therefore, they have suggested a hybrid chemo-biohydrometallurgy technology, in which Cu recovery was achieved in an acid pretreatment step and later when there is no hindrance of Cu, Au and Ag recovery was performed by bioleaching.

The bioleaching of TV circuit boards which is low grade scrap, which has low content of Fe was studied by Bas et al. using a mixed culture of acidophilic bacteria (*A. ferrooxidans, L. ferrooxidans, A. thiooxidans*) [80]. By addition of Fe (II) ions, the Cu recovery improved significantly up to 54%. However, the acid consumption increased. In another approach, in which pyrite was used as a source of iron and sulfur, Cu recovery improved from 24% to 84% with significant decrease in acid consumption.

In another study, Xin et al. evaluated the effectiveness of bioleaching in metal recovery from electrolytic manganese slag, which is produced from electrolytic manganese residue [79]. In this work, the series of bioleaching was conducted by sulfur-oxidizing bacteria followed by pyrite leaching bacteria to promote Mn recovery (98.1%).

3. Process of e-waste bioleaching

3.1. E-waste preparation

In most of the e-waste bioleaching experiments, the waste should be prepared and/or pre-treated prior to metal recovery. Usually, the electronic components must be disassembled manually, and the desirable compartments be separated. In the case of batteries, usually anodes and cathodes are used for metal recovery in bioleaching process. The desirable compartments are powdered and grinded by blenders and ball mills to obtain a homogenous mixture. The powder is then sieved to obtain a specific particle size. Drying is applied in some cases prior to bioleaching. Several analytical tests can be performed on the obtained powder for characterization of sample before bioleaching. The metal content quality and quantity can be measured by Inductively Coupled Plasma-Optical Emission Spectrometry (ICP-OES). X-ray diffraction (XRD) analysis for detecting component phases of the powder is helpful as well.

3.2. Adaptation of microorganism to the e-waste

The alkaline nature of e-wastes causes a sharp increase in initial pH of the medium after addition of the solid waste [47]. This is mostly due to the presence of heavy metals in the e-waste. In bioleaching, the toxicity of e-waste to microorganism is not only affected by heavy metal concentration, but is also dependent on the cumulative contact time [81]. Therefore, their application to the medium can have toxic effect which can hinder the microorganism growth and activity.

One of the challenges in this bioprocess is to obtain a stable microbial population with sufficient functionality, since some inhibitory factors, in particular the bacteriostatic and bactericidal effects of the heavy metal may cause toxicity to the microorganism [82]. The response of microorganism to heavy metals is in biphasic mode. Low concentration of heavy metals can stimulate the microorganism growth, but higher concentration inhibits the microorganism growth and metabolism [82].

The microorganism must be adapted to the e-waste before the start of bioleaching process to become resistant to the toxic nature of e-waste and be able to continue their growth and activity;

in addition , it must also sustain its functionality in acidic condition, and in the presence of heavy metals and a wide range of inorganic ions and organic plastic content of e-waste [62][83].

For a better understanding of the adaptation process, the bacterial growth process must be understood. In the dynamic of bacterial growth, five distinct growth phases have been distinguished. The first phase is the lag phase which refers to the delay period before the start of the exponential growth. Lag phase is the less understood stage of the bacterial growth. Next steps is exponential growth phase which is the period in which the cell division proceeds at a constant rate followed by stationary phase, when the bacterial replication stops due to the unfavorable conditions. After these steps, death phase starts which is the period in which the cells lose their viability [84]. Apart from toxic nature of waste to microorganisms, at higher pulp densities, collision and friction of solid particles may lead to the decrease of oxidation activity of bacterial cells [85].

The transition to the stationary phase in Gram-negative and Gram-positive bacteria is ruled by sigma regulatory proteins. It is believed that the adaptability of the bacteria is due to the role of sigma factors. These factors are a subunit of the holoenzyme RNA polymerase that is responsible for recognizing the specific regulons of the bound sigma factor and can regulate the gene activity at the transcriptional level [84].

When the microorganism is exposed to high amount (higher than its adaptation capacity) of toxic material such as heavy metals, the integrity of cellular membrane is disrupted and its population in the leaching medium is affected. Some fungal strains have the ability to efflux heavy metal ions out of their cell to reduce the toxic effects [82]. A tool for mapping the adaptive evolution of bacterium is measuring the growth tolerance index (TI) with time which is defined by Eq. 6 [82].

$$\mathrm{TI} = \frac{\text{Cell count in presence of heavy metals (cell mL}^{-1}\text{)}}{\text{Cell count in the absence of heavy metals (cell mL}^{-1}\text{)}} \qquad (6)$$

High concentration of heavy metals inhibit the metabolism and growth of the microorganism. The toxic effect could be considered bactericidal or bacteriostatic. The bactericidal is lethal effect in which the microorganisms are unable to continue their growth even after toxin removal. This is in contrast to bacteriostatic method where the toxic effect is reversible [82].

The measurement of bacteriostatic effect by TI method indicates the tolerance development of a microorganism over an extended period of time after sequential exposure to the toxic material. This is a better method than measuring through minimum inhibitory concentration (MIC) approach that has shorter duration of exposure to toxin. This short period is not sufficient for adaptive evolution of bacterial strains [82].

The bacteriostatic effect of heavy metals on the bacterium can be limited by low pH, since high acidity increases the positively charged groups on the cell surface and consequently reduces the metal biosorption capacity of the bacteria. In general, less pH and chaperone activity gives the acidophilic bacteria the ability to detoxify and proliferate even in the presence of heavy metals [82].

The most common method used for the adaptation of microorganisms to the e-waste is by gradual increase of the e-waste pulp density in medium and sub-culturing of microorganism. In this method, the microorganism uses its natural intra and extra detoxification strategies to attain the tolerance to toxic environment [62]. Repeated sub-cultivation of microorganism and gradual increase of heavy metals available in the medium, may leads to isolation of metal tolerant mutants and activation of some biochemical pathways that allow cell growth [81].

More pulp density (the weight of solid waste per volume of bioleaching media) is a crucial parameter in the size of bioreactor, medium consumption and operation cost per weight of treated solid material and therefore a significant improvement toward large scale application of bioleaching. High pulp density can minimize the capital cost of the bioleaching process as well [86].

In comparison to e-waste, mineral ores have less toxic nature, since they are mainly reduced sulfides and contain less alkaline and toxic matter. The metals in e-waste (in the form of oxides and hydroxides) severely harm the growth and activity of the microorganism. As an example the highest pulp density achieved for spent batteries was less than 10% [42].

Xin et al. have studied the decline in the rate of bioleaching with pulp density increase. They have observed that there is a linear relation between the reduction of bacterial growth and activity with pulp density increase and could achieve an acceptable metal recovery (100% Zn and 89% Mn) at 4% pulp density [41].

For the adaptation of microorganisms to the mineral ores, different protocols have been used. However for the e-waste bioleaching only gradual sub-culturing of microorganism has been reported. Different criteria have been considered for considering the microorganism as adapted. In the case of bioleaching of e-waste, the microorganisms are usually considered adapted when the cell concentration reach 10^7 cell mL^{-1}. However, other criteria such as appearance of significant amount of soluble Fe and Cu and Fe(II)/Fe(III) lower than one, ferrous iron oxidation rate similar to those obtained in the waste free medium and equal metal dissolution rate in two consecutive sub-cultures have been used for considering the microorganisms as adapted when working with mineral ores [87]. Easier adaptation was reported for bioleaching of solid waste containing heavy metals when larger particle size of the waste were used [88].

During the adaptation of microorganism by repeated sub-culturing and gradual increase of waste concentration, the bacteria gain resistance by physiological changes. Therefore, in future due to the further adaptation and function of gene transformation working at higher pulp densities will be possible [36].

3.3. Bioleaching methods

There are three different bioleaching methods suggested by researchers based on the type of biomass exposure to the waste, including one-step, two-step and spent-medium.

In the one-step method the e-waste is added immediately to the culture medium, while in the two-step leaching substrate is added in the second step after the microorganism reaches its logarithmic growth phase. In two-step method, the medium is inoculated in the first step and pre-cultured without e-waste [53],[34],[89]. Since in the one-step process the bacterial growth occur in the presence of e-waste, the growth and metabolite production are inhibited [30].

The two-step method is reported to be a more efficient metal mobilization process, in which biomass growth is firstly a separate step. Advantages reported for two-step process are better bacterial growth and appropriate Fe^{2+} oxidation rate [7].

It was reported by Bryan et al. that if a decline in the culture viability is observed after the addition of e-waste, fresh media and inoculum will be required and might be impossible to sub-culture from one run to another [53]. It was observed in the work of Zhu et al. that two-step process can shorten the leaching time significantly with obtaining high recovery [89].

Işıldar et al. used multiple-step method, in which, the Cu was removed from PCBs with proteobacterium in one-step, and then Au was separated in the following step by cyanide produced by cyanogenic from Cu-leached waste. This is an attractive method in order to remove the metals, based on their different chemical properties and leaching mechanism. The idea comes from the fact that presence of high concentrations of base metals (such as Cu) will interfere with Au cyanidation, and their removal in the prior step is important [54]. In the work of Natarajan and

Ting, by increase of pulp density, the efficiency of bioleaching for Au and Cu decreased in two-step process, as a result of toxicity of e-waste for cyanide production [34].

As mentioned before in the two-step process, the e-waste is added to the pure culture and the metabolite concentration (acids or cyanides) declines during metal bioleaching. The decrease is sharper, in the presence of higher pulp densities [30].

The third method to perform bioleaching process is the spent-medium approach, in which cells are separated with filtering and centrifuging from the culture after the microorganism growth and maximum cell density and production of metabolite [28][34]. In this approach only the metabolite free of cells is used for bioleaching. The advantages of that are: the non-direct contact of biomass with metal-containing waste during growth period which enables the biomass recycling, optimization of metabolite production in the absence of waste and possibility of applying higher waste concentrations and no concern about contamination of waste with microbial biomass during growth [47].

Natarajan and Ting, observed for cyanogenic bacteria that with a spent-medium, increasing the pH will shift the equilibrium in favor of cyanide ions production which will lead to improved Au solubilization and higher metal recovery from electronic scrap (from 18% to 30%). The better performance of spent-medium in comparison to the two-step method for Au recovery by cyanogenic bacteria could be due to the several facts. In the spent-medium, the oxygen is available to form Au-cyanide complex, while in the two-step process the oxygen is consumed by bacteria. The cyanide is also consumed by bacteria in the two-step process during the mid and late stationary phase, while in the spent-medium the biogenic cyanide is available to be totally utilized for the Au leaching. The biosorption and bioaccumulation of metals on the inactivated bacteria after bioleaching may cause the reduction of metal concentration in the bioleached solution. Finally, working at higher pH and pulp densities in the spent-medium could be applied, since there is no limitation due to the toxicity of the waste [34].

In the work of Kim et al., different *Aspergillus* species were used for recovery of metals from spent Zn–Mn or Ni–Cd batteries in the spent-medium containing organic acids produced by fungi [28]. In the work of Bahaloo-Horeh and Mousavi,*A. niger* was used for the recovery of Cu, Li, Mn, Al, Co and Ni from spent phone mobile LIBs using one-step, two-step and spent-medium for comparison. The maximum recovery was obtained by the spent-medium at pulp density of 1%. The organic acids produced by *A. niger* were citric, gluconic, oxalic and malic acids, among them citric acid had principle role. Spent-medium also provides shorter processing time and ease of the operation. According to this study the spent-medium is the most effective among the three available methods in the leaching of heavy metals [68].

3.4. Kinetic studies

Kinetic study is an important step toward the understanding of the nature and mechanism of leaching process. The aim of kinetic studies is to develop the best kinetic model that describes the process and deduces kinetic parameters to obtain insight and appropriate results to be used for plant design, optimization of operating conditions of a working plant, real time optimization for automatic control and maximizing the metal recovery. The common belief according to the research is that the rate of bioleaching is controlled by the diffusion controlled model described as shrinking core model. In this model, the irreversible desorption is followed by diffusion in the porous solid through the pores. The assumption of this model is that the solute inside the particles is located inside a core that shrinks by the extraction of solute [90]. The three major steps of the reaction according to this model are diffusion of biogenic acids and metabolites through the solution and reaching the solid surface, diffusion of acids and metabolites inside the solid particles and ultimately chemical reaction. In a suitably shaking medium the first step is not limiting step, therefore the process is controlled either by diffusion through the solid or chemical reaction.

The method is fitting the changes in metal recovery with time according available for the process controlled by diffusion (Eq. 7) or chemical reaction (Eq. 8)

$$\mathrm{kt} = 1 - \frac{2}{3}\mathrm{X} - (1 - \mathrm{X})^{\frac{2}{3}} \qquad (7)$$

$$kt = 1 - (1 - X)^{1/3} \qquad (8)$$

The shrinking core model can be described by Eq. 7 and Eq. 8 only if the concentration of the leaching agent is constant during the process, which is far from the reality in most of the cases. The shrinking core model can be modified to consider the change of the leaching by using Eqs. 9 and 10 instead for the cases when the diffusion or chemical reaction are dominant respectively [91]:

$$\mathrm{E(X)} = 1 - {}^{2}/_{3}\,\mathrm{X(t)} - (1 - \mathrm{X(t)})^{\frac{2}{3}} = \frac{2\mathrm{bD_e}}{\rho_{\mathrm{B}}\mathrm{R}^2(1-\varepsilon)}\int_0^{\mathrm{t}} \mathrm{C_A}\mathrm{dt} \qquad (9)$$

$$\mathrm{G(X)} = 1 - (1 - \mathrm{X(t)})^{\frac{1}{3}} = \frac{\mathrm{bk}^*}{\rho_{\mathrm{B}}\mathrm{R}(1-\varepsilon)}\int_0^{\mathrm{t}} \mathrm{C_A}\mathrm{dt} \qquad (10)$$

In the above equations, ρ_{B} is molar density of metallic compounds in solid waste, C_A is proton concentration in the solution, D_e is the diffusion coefficient of proton, R is the particle radius, ε is porosity of the solid waste and $\int_0^{\mathrm{t}} \mathrm{C_A}\mathrm{dt}$ is the function of time that can be calculated by the area underneath of the graph of proton concentration which can be estimated by theoretical calculations of acid concentration and time zero is the moment that the solid waste was added. By plotting G(X) and E(X) versus $\int_0^{\mathrm{t}} \mathrm{C_A}\mathrm{dt}$, a linear must be fit. The equation that better fits the experimental data shows which model (diffusion and chemical reaction control) is better for describing the kinetics of the process.

The kinetic study of bioleaching of waste of PCBs was performed in the work of Yang et al. In this process precipitation due to jarosite formation influences the kinetic of process. Due to the formation of a product layer of jarosite, shrinking core model would be a suitable model for describing metal recovery from waste PCBs by *A. ferrooxidans.* In this process the iron cycle relates the metal recovery and H^+ consumption and therefore second-order may apply for kinetic of metal recovery [90].

At high pulp densities, the mass transfer of oxygen and carbon dioxide may be determining and as a result the mechanism is combination of both diffusion and reaction. However, the mentioned conventional methods are not enough for the cases that more than one mechanism is determining in the overall rate of equation. They will not be useful also in the cases where the correlation coefficients of the equations are too similar that distinguishing of the controlling mechanism is not possible.

3.5. Catalytic bioleaching of e-waste

The effectiveness of the bioleaching is restricted by several reasons mostly coming from slow dissolution kinetics and low metal leaching yield which hinders the use of bioleaching in commercial scale [92].

It was reported that, for obtaining acceptable metal recovery, long period (300 days in laboratory experiment and as long as 900 days in industrial scale) is required to obtain 60% recovery [92].

For solving this problem, different materials were suggested by different researchers as catalysts. Catalyst, a substance that can lower the activation energy and increase the rate of reaction, can be used for modifying the slow kinetic of bioleaching from e-waste [92]. Using metal ions, surfactant and carbonaceous materials were proved to improve the bioleaching yield from e-wastes [51].

Table 7 summarizers the studies that examined the catalyst application for improving the efficiency of metal recovery from e-waste by bioleaching.

Metallic ions such as Ag^+, Cu^{2+}, Bi^{3+} and Hg^{2+} have been used as catalyst for bioleaching of metals from ore. However, only Cu^{2+} and Ag^+ were used for bioleaching of e-waste (LIBs [65],[71] and ZMBs [42]). Therefore, the mechanisms of these two metallic ions are explained in the following. The role of Cu^{2+} ions in metal recovery from mineral ores has been studied several times. The metals of Li and Co in LIBs (an e-waste for which catalytic bioleaching have been studied) are mostly in the form of $LiCoO_2$ [65]. Zeng et al. suggested that $LiCoO_2$ during bioleaching in the presence of Cu^{2+} undergo a cationic interchange reaction with Cu^{2+} formed $CuCo_2O_4$ on the surface of the sample, which could be dissolved easily by Fe^{3+}. The role of metal ions as the catalyst is formation of this intermediate. $CuCo_2O_4$ can then oxidizes to Cu^{2+} in the presence of Fe(III). The Fe(III) ions are then produced from ferrous ions. Eqs. (11)-(13) show the catalytic reactions of LIB bioleaching [65]:

$$\mathrm{Cu^{2+} + 2LiCoO_2 \rightarrow CuCo_2O_4 + 2Li^+} \quad (11)$$

$$\mathrm{CuCo_2O_4 + 6Fe^{3+} \rightarrow 6Fe^{2+} + Cu^{2+} + 2O_2 + 2Co^{2+}} \quad (12)$$

$$\mathrm{4Fe^{2+} + O_2 + 4H^+ \rightarrow 4Fe^{3+} + 2H_2O} \quad (13)$$

Zeng et al. proposed a mechanism for metal removal from LIBs, which is mostly in the form of $LiCoO_2$ by using Ag metal ions [71]. Compounds such as silver sulfate, silver nitrate and silver chloride have been used previously assources of Ag^+ ions and has proved to be efficient for improving the dissolution kinetics and metal recovery leaching yields from different solid wastes [92]. The role of Ag^+ is formation of an intermediate compound $AgCoO_2$ which is further oxidized by ferric ions and release Ag^+ ions again. The ferrous ions are oxidized back in to ferric ions by bacterial activity. The proposed reactions are as Eqs. 14 and 15 [71]:

$$\mathrm{Ag^{+} + LiCoO_2 \rightarrow AgCoO_2 + Li^{+}} \tag{14}$$

$$\mathrm{AgCoO_2 + 3Fe^{3+} \rightarrow 3Fe^{2+} + Ag^{+} + O_2 + Co^{2+}} \tag{15}$$

In the work of Niu et al. four different metal ions were tested as the catalyst for the metal recovery from ZMBs. The metal ions were Co^{2+}, Ni^{2+}, Cu^{2+} and Ag^{+}; among them only Cu^{2+} proved to be an effective catalyst [42]. This work inspired from the work of Zeng et al. [65] and considered the catalytic mechanism due to the formation of intermediate similar to $CuCoO_2$ which was $CuMn_2O_4$ in the case of ZMBs. This intermediate will undergo the same mechanism as the Eqs. 16 and 17 [42]:

$$\mathrm{Cu^{2+} + ZnMn_2O_4 \rightarrow CuMn_2O_4 + Zn^{2+}} \tag{16}$$

$$\mathrm{CuMn_2O_4 + 2Fe^{3+} + 4H^{+} \rightarrow 2Fe^{2+} + 2Mn^{2+} + O_2 + Cu^{2+} + 2H_2O} \tag{17}$$

In this mechanism, Fe^{2+}/Fe^{3+} have great importance to metal release. However, even with the metal ion as catalyst the obtained recovery did not exceed 62.5% for Zn and 62.4% for Mn. This was reported to be due to the high dose of alkaline substance and jarosite formation [42].

On the other hand, the high cost of the metals ions as catalyst must be considered. The combined application of metal ion catalysts and non-metal catalysts are worth studying as well. As an example, simultaneous application of metal ions (such as Ag^{+}) and activated carbon as non-metal catalyst may have synergic effects that promotes greater catalysis [92].

Surfactants such as Tween 20 and Tween 80 are believed to be effective in the improvement of efficiency of bioleaching from e-waste since they were proved to be an effective catalyst for the bioleaching of mineral ores. By changing the surface condition of the minerals or wastes they can accelerate the oxidation rate of ferrous ions and the bio-oxidation of elemental sulfur. This is due to the improved attachment of bacteria to the elemental sulfur, increased rate of sulfur oxidation and sulfuric acid production, and finally boosting the reaction kinetics. However, at a very high concentration of surfactants bacterial growth may be inhibited [93].

The combination of surfactants and activated carbon and their synergistic effect gave even a better result for bioleaching behavior of mineral ores than that of either individual application. In their combined application, the surfactant not only reduces the surface energy of the surface, but also the surface energy of activated carbon which enhances the particles dispersion and decreases the agglomeration of solid particles (ores or waste) and activated carbon [93].

In the work of Karwowska et al. biological surface-active compounds, were produced by two surfactant producing strains *Bacillus subtilis* PCM 2021 and *Bacillus cereus* PCM 2019 for bioleaching of PCBs [52].

Different forms of carbon based catalyst have been used for enhancing bioleaching including activated carbon, graphene, biochar and nitrogen doped carbon nanotubes (NCNT).

Activated carbon is electrically conductive catalyst that can form a galvanic couple. This material have been used for bioleaching of minerals and reported to be thermodynamically more stable than minerals [93].

Biochar is the by-product of the biomass pyrolysis process, rich in carbon and inorganic materials [94], with several applications due to its high carbon content and high porosity which provide the suitable matrix for the attachment of microorganism. Biochar can be used as the catalyst also in bioleaching process by facilitating the electron transfer and consequently promoting the redox reactions. Fe mediated bioleaching was promoted significantly using biochar in the bioleaching of PCBs using *Alicyclobacillus spp*. and *Sulfobacillus spp.* Three different basic processes were examined for determining the role of biochar. Carbon-mediated, Iron-mediated and sulfur-mediated. The biochar can facilitate the redox action between Fe(II) and Fe(III), and then effective leaching of Cu [57].

Graphene is a nano material with a high surface area, desirable stability and favorable conductivity. Therefore, graphene has a common use as a catalyst in different fields. It was tested as a catalyst in bioleaching of PCBs and resulted in improved Cu recovery. The enhanced bioleaching process in the presence of graphene, could be due to the adsorption of microorganism on the catalyst surface, which leads to the extended reaction time between the microorganism and waste. Another hypothesis is that the leached metals are adsorbed on the graphene surface, therefore the concentration of metal ions decrease in the solution, so more metals move to the solution from solid waste. However, the amount of graphene must not exceed a certain limit that may impede the normal growth of bacteria. The graphene particles are surrounded with ferrite precipitations at the end of bioleaching [51].

Nitrogen doped carbon nanotube are materials with large amount of surface area, desirable selectivity and stability and promising conductivity. Therefore, this material is used largely as catalyst [50]. The structure of NCNTs is a one-dimensional tubular form, which is made of single layer or multilayer graphene [56]. Characteristics such as high specific surface area, favorable adsorption, and good electric and heat transfer properties, nominated NCNTs as catalyst in different application.

In the work of Gu et al., NCNTs was used for the bioleaching of PCBs by *A. ferrooxidans*. The optimal amount of NCNT catalysts and recycling effects of NCNT modified electrodes were studied in that work [50].

In the work of Bai et al., NCNTs catalytic role was studied for bioleaching of WPCBs by *A. ferrooxidans*. Better results obtained for bioleaching with NCNTs as catalyst was due to the increased contact area between cell and the waste and extended reaction time in large surface area of the catalyst. The NCNTs with the gathered bacteria in their surface are adsorbed on the surface of the waste and the residence time of bacteria in the waste is increased. However, addition of excess NCNTs prevents CO_2 from entering to the culture medium and causes agglomeration and limits the bacterial normal growth [56].

3.6. Large scale bioleaching of e-waste

Few studies have investigated the bioleaching process of e-waste in different bioreactor configurations. However, the knowledge for large scale bioleaching of e-waste is still limited. The only commercialized e-waste treatment is through pyrometallurgical methods, which as mentioned earlier are energy intensive, with the possibility of hazardous gas emission [5].

The first large scale application of bioleaching for metal recovery from e-waste was done in the year 2003 by Zhu et al. [76]. In this work, the metal recovery from Ni-Cd batteries was performed inside two successive reactors. The first was a bioreactor inoculated with indigenous *Acidithiobacilli*, in which the sulfuric acid was produced. The overflow of the reactor was transferred to settling tank where the sewage sludge was thickened. The effluent was then conducted to leaching reactor that contained the powder obtained from Ni-Cd batteries. The whole process took 50 days, during which 100% Cd and 66.1% Ni were recovered. The effect of retention time in both bioreactor and leaching reactor on recovery was also studied.

The same method was applied in the work of Zhao et al. and obtained the total recovery of Co, Ni, Cd. They made an important conclusion that iron oxidizing system is more efficient in plants with high loading rate due to its shorter bioleaching time, while sulfur oxidizing systems are more suitable for long leaching due to more acid generation [77].

Ilyas et al. studied the bioleaching in a column reactor loaded with electronic scrap by mixed adapted consortium. On the bottom of the column there was support plate with holes for air injection. The temperature was maintained with a jacket at 45 °C. The obtained recovery was 80% for Zn, 64% for Al, 86% for Cu, and 74% for Ni [1].

In the work of Chen et al., the washed e-waste samples were placed in a column while the inoculated medium was put in an incubator and fed continuously to the column. The effluent of the reactor was entered the medium in incubator. The whole system including column and incubator were located in a thermotank [21].

Mäkinen et al. first separated the PCBs into metal rich and metal poor fractions by flotation. The metal rich fraction was treated by pyrometallurgical methods and the metal poor fraction was transfered to the bioleaching reactor. The scaled-up experiments were conducted in three steps with a three continuous CSTR [5].

A rotating drum reactor was used by Rodrigues et al. for bioleaching of PCBs. The reactor was a perforated drum rotating with a motor. This drum was located in a cylindrical tank filled with liquid medium. The PCBs were in the form of sheets with 20 mm long immersed in the medium containing inoculum [25].

Jagannath et al. applied perturbations to overcome the problems of fixed bed reactors for bioleaching through provision of proper mixing and distribution of substrate, renewal of interfering surface area and improving mass transfer. Bioleaching was performed in pulsed plate column with solid e-waste located in the space between plates as packed bed. Different frequency and amplitude were studied as variables. The entire working volume of the reactor was 1.5 L [2].

In the work of Xia et al. the bioleaching of e-waste was performed in 3 L stirred reactor placed in thermostatic bath at 45 °C [36]. The catalytic bioleaching of e-waste was also conducted in bioreactor [56].

3.7. Restriction of e-waste bioleaching

Several phenomena can hinder the proper bioleaching of e-wastes. Among them toxicity and precipitation due to the jarosite formation and complexation are the major ones.

As mentioned earlier, the heavy metals available in the e-wastes may hinder the microorganism growth and activity, which urge the adaptation of microorganism to the e-waste prior to bioleaching usually by gradual sub-culturing.

However, the toxicity of the e-waste to microorganism is also due to organic compounds available in the e-waste specially epoxy plastics and brominated flame retardant. The organic matter of the plastic content of the e-waste cannot be consumed by microorganism and at certain concentration could be toxic for bacterial growth [62]. The sensitivity of the microorganism restricts the amount of waste that can be treated in each batch, therefore limits the large scale and high pulp densities application of bioleaching process for e-waste treatment.

For bioleaching of PCBs toxic effects of organic materials such as aromatics and brominated flame retardants (BFRs), caused the decrease of iron oxidation rate and inhibition of microbial enzymes involved in carbon dioxide fixation [54].

Precipitation of target metal due to jarosite formation can hinder bioleaching and decrease the rate of metal recovery as well. This is promoted by saturation of metals available in solution and presence of precipitating agents [62]. Jarosite can be represented by $M_n(Fe^{3+})_6(SO_4)_4(OH)_{12}$ where M could be K, NH_4, Na, Ag or Pb. For monovalent and divalent cations, n equals 1 and 2 respectively. The heavy metals can be trapped in the lattice of jarosie crystal if they replace the M (K, NH_4, Na, Ag or Pb) in the structure [95].

As a consequence of ferric ions generation from ferrous by bacterial activity, the pH will increase due to H^+ consumption. When pH is more than 2.0 the formation of Fe oxyhydroxides through ferric hydrolysis is promoted.

The generated H^+, will reduce the pH again and at that condition, the ferric hydroxide reacts with monovalent cations (K^+, NH_4^+, …) [62].

Velgosova et al. have described the jarosite formation by Eq. 18 for K^+ [23].

$$K^+ + 5OH^- + 3Fe^{3+} + 2SO_4^{2-} + H_2O \rightarrow KFe_3(SO_4)_2\,(OH)_6 + H^+ \tag{18}$$

Jarosite precipitation can adversely affect the metal bioleaching. Several parameters including high concentration of ferrous iron, pH and temperature can affect the jarosite formation [95]. The precipitates can settle down on the reactive surfaces of the waste and make a barrier toward the diffusion which slows down the reactant and product fluxes [96]. As an example in the work of Mishra et al. for bioleaching of LIBs, ferric ion precipitation with ions available in leaching medium and formation of metal complexes were reported [69].

In general, jarosite is the undesirable by-product of the bioleaching process, which destroys the effectiveness of the metal removal process due to passivation of leaching reactions by precipitation on solid waste surface and consumption of Fe^{3+} in solution which lowers the oxidation–reduction potential value. Rashidi et al. have proposed the following correlation for the rate of Fe(III) precipitation**,** in which k_1, k_2, q and p are the constants [97].

$$r_{Fe^{3+}precepitated} = -k_1[Fe^{3+}]^q(k_2 + \frac{[Fe^{3+}]}{[Fe^{2+}]})^p \tag{19}$$

Therefore, the ferric balance in the system can be described by the Eq. 20:

$$\frac{d[Fe^{3+}]}{dt} = \frac{1}{Y_{cells/Fe^{2+}}}(r_{cell}) - 2\frac{M_{Fe}}{M_{t.m}}(-r_{t.m}) - (-r_{Fe^{3+}precepitated}) \tag{20}$$

In which $\mathrm{M_{Fe}}$ is the molecular mass of iron, $\mathrm{M_{t.m}}$ is the molecular mass of target metal and $\mathrm{r_{t.m}}$ is the target metal dissolution rate, $\mathrm{Y_{cells/Fe^{2+}}}$ is the yield coefficient in the oxidation of iron [97].

During fungal leaching, the metal oxalate can precipitate on the non-leached powder and cause lower metal recovery. Most of metal oxalate complexes (such as nickel oxalate and cobalt oxalate) are insoluble, while other metal oxalate such as lithium and aluminum oxalate are more soluble. Therefore metal oxalate formation can decrease the yield of nickel and cobalt recovery. For fungal bioleaching of LIBs, Eqs. 21 and 22 were proposed to describe the undesirable precipitation of cobalt oxalate [68]:

$$\mathrm{2LiCoO_2 + 7C_2H_2O_4 \rightarrow 2LiC_2HO_4 + 2\,Co(C_2HO_4)_2 + 4H_2O + 2CO_2} \quad (21)$$

$$\mathrm{2LiCoO_2 + 4C_2H_2O_4 \rightarrow Li_2C_2O_4 + 2CoC_2O_4 + 4H_2O + 2CO_2} \quad (22)$$

In the bioleaching of Ni-Cd batteries with fungi, it was observed that the larger quantities of oxalic acids inhibit metal recovery for Ni, Cd, Co and Zn, since these metals precipitate with oxalic acid [28].

4. Future of bioleaching perspective in OMICS and bioinformatics

The term OMICS refers to innovative technologies platforms such as genetics, genomics, metabolomics and proteomics. OMICS approach along with bioinformatics is a new approach to be applied for bioleaching for clarifying the questions related to the complex contribution of microorganisms. This approach will help to the discovery of new aspects and characteristics of microorganisms regarding to interaction of genes, proteins, macromolecules, and the environment. Prediction of metabolic models can be also achieved with the aid of genomics (study of the structure, function and evolution of genomes), metabolomics (study the chemical fingerprint of a cellular process), proteomics (study of structure, function and interaction of proteins produced by genome) and other OMICS approach in addition to bioinformatics applications which helps on advancing the scientific knowledge on the physiology of the microorganisms involved in bioleaching including metabolism of the extremophiles, and synergetic effect of microorganism in bioleaching system [98].

In the work of Tay et al. the metabolically-engineered pBAD and pTAC *C. violaceum* strains were used for recovery of Au from electronic scrap and significant increase was observed for gold recovery by using both strains. Considering the cyanide lixiviant production, under optimal conditions, pBAD strain produced 68% and pTAC 51% more cyanide lixiviant in comparison to non-engineered cyanogenic bacteria and the consequence was the improved Au recovery from e-waste [99].

Tay et al., have studied the modulations of the lixiviant metabolic network of the pBAD by proteomics study and comparison of the engineered strains with natural ones which can be seen in Fig.1. By decreasing the availability of the substrate glycine, cyanide production through HCN synthase can be decreased. As a consequence glycine flux can be also achieved by enzymatic actions such as serine hydroxymethyltransferase (glyA), useful in the biosynthesis of serine from glycine, glycine decarboxylase (gcvP), the enzyme for glycine cleavage system, serine dehydratase (sdaA1) the enzyme for serine catabolism, threonine aldolase and glycine C-acetyltransferase (CV_4309 and kbl) for biosynthesis of threonine. The performed study in the mentioned work, revealed that by increase of cyanide production in the engineered strain, no significant direct or indirect change in the protein levels of the mentioned enzymes was associated with the cyanolytic pathways [99]. This concept is depicted in Fig.1.

The cyanide production is increased significantly by dissociating the cyanogenesis from quorum control which consequently lead to the improved Au recovery. In the engineered strain in this work (pBAD), the cyanide production increased above wild-type levels at mid-log phase using Larabinose as the exogenous inducer. In engineered pTAC strain, an analogous doseresponsive profile was obtained with IPTG as the exogenous inducer, where the addition of 1 mM IPTG resulted in maximal cyanide production [99].

Genomics, metagenomics and proteomics are used to study the mechanism that microorganisms use for adaptation to their changing environment [100]. The tools needed for the identification of the steps that must be done for manipulation of strains genetically and/or chemically for enhancing the heavy metal storage capacity and efficiency are provided by system-biology experimental approaches (metabolic control analysis and kinetic modeling) application to the cellular processes involved in the chelation and intracellular accumulation of heavy metals. The cellular responses to the stress caused by heavy metals relies majorly on the activation of the Cys, GSH and PCs biosynthesis. The most logical strategy for enhancing the heavy metal accumulation capacity of a microorganism is by handling of the basic controlling steps of these pathways which is done through genetic engineering and selection by growth under stressful conditions [101]. As an example, *Acinetobacter calcoaceticus* by excreting extracellular substances which are metabolic products (such as glycoproteins, polysaccharides, proteins, lipids, cellulose and glycolipids) provide the sites for binding cation. This provides them protection capacities against heavy metals. Another mechanism which reduces the ionic force of metals is neutralizing heavy metal toxicity on the cells through nanoparticle formation. In this mechanism, the metallic nano particles are enclosed in vesicles that can be released later. This mechanism gives the microorganisms the ability to survive in contaminated environments due to transition between metal ions and non-toxic form [102].

OMICS data analysis method (neural networks and genetic algorithm) has been used by Abdollahi et al. (2019) to study the effect of parameters on metal dissolution from mineral ores using bioleaching. This method can be applied also on bioleaching of E-waste [103].

Another attempt to apply genetic modification to bioleaching of PCBs was performed by Natarajan and Ting. The authors examined the mutation of alkali-tolerant bacteria (*C. violaceum*) with targeting the growth of bacteria under higher alkaline conditions which is more favorable for chemical stability of the produced cyanide. In that work *C. violaceum* was exposed to the mutagen, NNitroso-N-ethyl urea at pHs 9, 9.5 and 10 to make the bacteria more resistant to higher pH ranges. The mutations were random and genome wide. Bioleaching by mutated *C. violaceum* resulted in higher recovery of gold at pH 9.5 compared to the mutated strains at pHs 9 and 10 [30].

5. Conclusion

Bioleaching is a sustainable method for metal recovery from e-waste and development of urban mining, which can help on saving the non-renewable energy consumed in mining industry, and provide mitigation of greenhouse gas emission. In this paper, previous published results obtained in the field of bioleaching of e-wastes are reviewed and presented based on the type of microorganism, the type of e-waste and bioleaching method. The microorganisms used for bioleaching of E-waste are divided into two groups of chemolithoautotrophs and heterotrophs. The biogenic acid produced by microorganisms such as sulfuric acid in the case of chemolithoautotrophs can solubilize the metals from the solid matrix via acidolysis and redoxolysis. For fungal strains that produce organic acids, the complexolysis becomes important as well due to the chelating capabilities of the organic acids. The cyanogenic bacteria are the other class of the heterotrophs that produce cyanide and promote the metal dissolution via complexolysis. For investigating the kinetics of the bioleaching, the common believe is that the

process is controlled by diffusion controlled model described as shrinking core model. However, the slow rate of the process is still a limitation toward large scale application of the bioleaching for metal recovery from e-wastes. Therefore, the application of catalyst has been used by several researchers to improve the kinetic of the process. The application of different catalysts (metal ions, surfactants and carbon based catalyst) for bioleaching of e-waste was reviewed here. The challenges and limitations toward the process including precipitation, toxicity of solid waste to microorganisms and necessity of adaptation were explained. The future of research in the field of bioleaching of e-wastes should focus on the methods for metal recovery from the solution and application of OMICS approaches; this will provide a better understanding of the microorganism contribution in the process and the manipulation of their genetic makeup toward the generation of more efficient microorganisms with enhanced bioleaching capabilities.

Table of content

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Fig.1. Comparative proteome analyses after increasing cyanogenesis in *C. violaceum* [99]

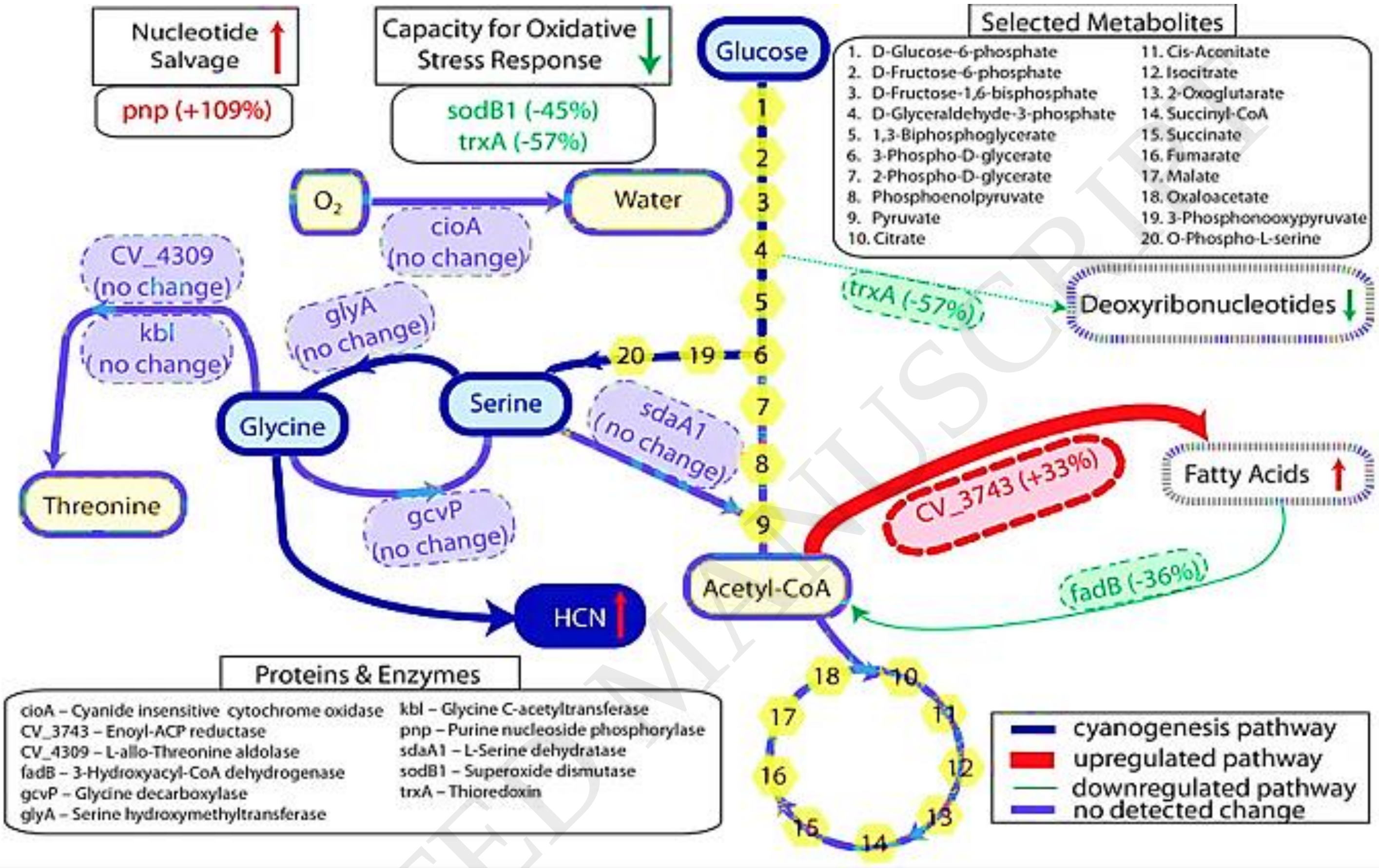

Table 1. Microorganisms used for bioleaching of e-waste with autotrophs

Microorganism	**E-waste**	**Target metal**	**References**
Chemolithoautotrophs Mesophiles *A. ferrooxidans*	Mobile and computer PCBs	Ni, Cd	[19,20]
	PCBs	Cu	[21]
	PCBs	Al	[22]
	Ni-Cd batteries	Cd	[23]
A. thiooxidans	PCBs	Al, Cu, Mg, Zn	[24]
Thermophiles *S. thermosulfidooxidans*	PCBs	Cu	[25]
Fungi *Aspergillus niger*	LIBs	Ni, Co, Mn, Li, Cu, Al	[26,27]
Mixture of six *Aspergillus* species: *A. fumigatus, A. flavipes, A. japonicus , A. tubingensis* *A. versicolor, A. niger*	ZMBs and Ni-Cd Battery	Ni, Cd, Co, Zn	[28]
Cyanogenic bacteria *C. violaceum*	Mobile PCBs Electronic scrap Sim card	Au, Cu Au Au, Ag, Cu	[29] [30] [31]
B. megaterium	Computer PCBs Mobile PCBs	Au, Cu Au, Cu	[32]
Pseudomonas Chlororaphis	PCBs	Au, Ag, Cu	[33]
Mixture of: *C. violaceu/ Pseudomonas aeruginosa* *P. fluorescens*	PCBs	Au	[34]
Mixture of: *C. violaceum* *P. fluorescens* *B. megaterium*	Electronic scrap	Au, Ni, Cu	[35]

Table 2. Recent studies in bioleaching of PCBs

Microorganism	**Bioleaching type**	**Recovered metals**	**Findings**	**Reference**
Acinetobacter sp. Cr B2	Pulsed plate rector	Cu	✓Studied large scale application by the function of extracellular enzymes and metabolites ✓Used ambient conditions, less energy intensive, controllable conditions	[2]
L. ferriphilum, *A. caldus*	Stirred Tank Reactor	Zn, Cu, Al, Pb, Sn	✓ Thermophilic bacteria have better performance than mesophilic ✓Proposed the semi industrial process	[36]
A. ferrooxidans	One-step	Cu	✓Used nitrogen doped carbon nanotubes modified electrode as catalyst ✓Optimized the recovery based on the effects of pH, ORP, Fe^{2+} and Cu^{2+} concentration ✓Analyzed the mechanism of catalysis ✓The modified electrode was recyclable	[50]
A. ferrooxidans	One-step	Cu	✓Graphene is a promising catalyst for bioleaching of Cu from PCBs	[51]
A. ferrooxidans	One-step-Bioreactor	Cu	✓Studied catalytic effect of carbon nanotubes	[56]
Alicyclobacillus sp. and *Sulfobacillus sp.*	One-step	Cu	✓Biocahr used to facilitate the redox action between Fe^{2+} and Fe^{3+} ✓Catalyst promoted the leaching recovery by Fe mediated pathway	[57]
Step 1 *A. ferrivorans/ A.thiooxidans* **Step2** *P.fluorescens and*	2 steps	Cu, Au	✓Bioleaching was done for base and precious metals separately ✓There is a need to enhance bacterial cyanide production	[54]

Pseudomonas putida				
A. ferrooxidans	One-step-Column	Cu	✓The kinetic of the bioleaching process was studied in which the variation in Fe^{3+} concentration considered in the conventional kinetic ✓The rate of Cu dissolution is controlled by external diffusion rather than internal one since Fe hydrolyze and form jarosite. ✓Jarosite formation can be prevented by adding dilute sulfuric acid.	[39]
L. ferriphilum	Two-step	Cu, Cr, Ni, Sn, Zn	✓Using pyrite as a source of lixiviant ✓High content of elemental iron caused a lag period in copper solubilisation due to displacement reactions.	[53]
A. ferroxidanse	One-step	Cu, Zn, Pb, Ni	✓Addition of natural citric acid (lemon juice) improved the metal recovery due to the role of acid as chelating agent	[58]

Table 3. Techno-economic and environmental assessment of biological and chemical leaching for metal recovery from PCBs [59]

Technology	Operational cost	Investment cost	Total cost	Net revenue	Return of interest	Climate change contribution
	(Euro/kg PCB)				year	Kg CO_2/Kg PCB
Biological	0.159	0.457	0.616	4.41	5.1	8.26
Chemical	0.224	0.446	0.670	8.97	2.4	14.6
Hybrid	0.232	0.777	1.008	8.25	4.3	11.6

Table 4. Previous studies on LIBs bioleaching

Microorganism	**Bioleaching type**	**Recovered metals**	**Findings**	**Reference**
A. niger	One-step, Two-step, Spent-medium	Li, Co, Al, Cu, Ni, Mn	✓Studied the growth characteristics of fungi (pH variation, biomass and organic acid concentration) ✓Maximum recovery obtained by spent medium (100% Cu, 95% Li, 70% Mn, 65% Al, 45% Co and 38% Ni) ✓The major organic acids produced was citric acid	[26]
A. niger	Spent-medium	Li, Co, Al, Cu, Ni, Mn	✓Optimized the bioleaching parameters (the variables of sucrose concentration, initial pH, and inoculum size) for organic acid production and then metal recovery.	[68]
A. ferrooxidans	One-step	Li, Co	✓Possibility of the recovery of cathode material of LIBs with acidophilic bacteria by producing sulfuric acid ✓Li dissolved slower than Co. ✓Higher density of solid prevents the bacterial activity due to its toxicity.	[69]
A. ferrooxidans	One-step	Co	✓Cu metal ions could enhance not only the oxidation rate of $LiCoO_2$, but also the leaching amount of Co from spent LIBs.	[65]
A. ferrooxidans	One-step	Co	✓Silver ions can also be used as a catalyst for the metal recovery from LIBs.	[71]
Alicyclobacillus sp. and *Sulfobacillus sp.*	One-step	Li, Co	✓Increase of pulp density reduces sharply the metal recovery by bioleaching. ✓Product layer diffusion model described best the bioleaching behavior of Co and Li from the spent LIBs.	[40]
A. thiooxidans *L. ferriphilum*	One-step	Li, Co	✓Metal dissolution is exclusively dependent on the non-contact mechanism not on the contact mechanism.	[12]

			✓Acid dissolution was the sole mechanism for Co release in the sulfur containing environment.	
A.thiooxidans *L. ferriphilum*	One-step	Li, Co, Ni, Mn	✓Effective bioleaching of metals from LIBs of electronic vehicles was obtained. ✓Li extraction was due to acid dissolution by biogenic H_2SO_4. ✓Co, Ni and Mn mobilization were due to the combined actions of Fe^{2+} reduction and acid dissolution. ✓Non-contact mechanism observed for Li extraction, while a contact mechanism was observed for efficient mobilization of Co, Ni and Mn.	[39]
A. thiooxidans *A. ferroxidans*	Two-step	Li, Co, Ni	✓ Energy sources concentration and pH were optimized in spent LIBs detoxification. ✓The results confirmed acid dissolution mechanism for Li recovery and combination of acid dissolution and oxidation reduction for Ni and Co recovery.	[72]

Table 5. Previous studies on bioleaching of Ni-Cd batteries

Microorganism	**Bioleaching type**	**Recovered metals**	**Findings**	**Reference**
A. niger *A. tubingensis*	Spent-medium	Ni, Cd, Co, Zn	✓ More than 95% of metals were extracted.	[28]
A. ferrooxidanse	One-step	Cd	✓ H_2SO_4 and $Fe_2(SO_4)_3$ solutions were used as media and different Fe (III) concentration were tested. ✓ Ferric iron plays an important role in bioleaching from Ni-Cd batteries, due to its high oxidative function and ability to maintain the low pH.	[23]
Indigenous *Thiobacilli* sludge	Two-step	Ni-Cd	✓ Bioleaching was conducted in a system consisting of a bioreactor, settling tank and leaching reactor. ✓ Total Cd recovered, and partial Ni recovery was achieved.	[76]
Indigenous *Thiobacilli* sludge	Two-step	Ni, Cd, Co	✓ System includes an acidifying reactor and a leaching reactor. ✓ Iron oxidizing system is more efficient in short time, and is suitable for high loading rate plants. ✓ Sulfur oxidizing system with its strong acid generation capacity is more suitable for a long time leaching. ✓ Cadmium hydroxide, elemental Cd and Co were more easily leached than nickel hydroxide and metallic Ni.	[77]
A. ferrooxidanse	Two-step	Cd, Ni	✓ Production of biogenic acid was done in the first reactor and bioleaching of batteries in the second reactor. ✓ More than 90% of metal recovery was obtained.	[75]

Table 6. Previous studies on ZMBs bioleaching

Microorganism	**Bioleaching type**	**Recovered metals**	**Findings**	**Reference**
A. thiooxidans *L. ferriphilum*	One-step	Zn, Mn	✓ Bioleaching was performed at high pulp densities (10%) in comparison to previous works on ZMBs. ✓ Metal ion (Cu^{2+}) catalyst improved the results through formation of $CuMn_2O_4$. ✓ Poor microorganism growth and jarosite formation destroyed the Fe^{3+}/Fe^{2+} cycle and impede the bioleaching process.	[42]
Six *Aspergillus* species	Spent-medium	Zn, Mn	✓ Two different nutrients, malt extract and sucrose, were used to produce different types of organic acids. The acid were then used for the metal recovery. ✓ Oxalic acid was the dominant acid in malt extract media and citric acid in sucrose media. ✓ *A.tunbingensis* resulted to be a useful fungus for bioleaching.	[28]
Alicyclobacillus sp. *Sulfobacillus sp.*	One- step	Zn, Mn	✓ Zn recovery occurs effectively regardless of the bacterial type and energy source. ✓ Zn recovery was done in non-contact mechanism and Mn recovery in both contact and non-contact mechanism.	[43]
Alicyclobacillus sp. *Sulfobacillus sp*	One-step	Zn-Mn	✓ Analyzed the reason of ZMBs bioleaching failure at high pulp densities due to cell number reduction and activity decrease.	[41]

Table 7. Previous studies on catalytic bioleaching of e-waste

Microorganism	**Catalyst**	**Waste**	**Recovery**	**Reference**
A. ferrooxidans	Graphene	WPCBs	✓ Cu recovery 75% without catalyst, 84% with catalyst	[51]
Aerobic activated sludge	Biochar	PCBs	✓ Enhancement of Cu recovery by iron mediated pathway	[57]
A. ferrooxidans	NCNT	WPCBs	✓ Cu recovery 99% without catalyst, 79% with catalyst	[50]
A. ferrooxidans	Cu^{2+}	LIBs	✓ Co recovery achieved 99% with catalyst, while it was 43.1% without catalyst	[65]
A. ferrooxidans	Ag^{+}	LIBs	✓ Co recovery achieved 98.4% with catalyst, while it was 43.1% without the use of catalyst	[71]
A. ferrooxidans.	Co^{2+} Ni^{2+} Cu^{2+} Ag^{+}	ZMBs	✓ Zn recovery achieved 62.5% with catalyst, while it was 47.7% without catalyst ✓ Mn recovery 62.4% with catalyst, while it was 30.9% without catalyst	[42]
Acidithiobacillus sp.	Surfactant producing bacteria	PCBs	✓ Pb recovery reached 70% from 30% in the absence of surfactant producing bacteria ✓ For Zn, Cu and Cd quite the same recovery achieved with and without catalyst	[52]
A. ferrooxidans	NCNTs	PCBs	✓ Catalyst promoted Cu recovery by 10%	[50]