



Gunarathne, V., Rajapaksha, A. U., Vithanage, M., Alessi, D. S., Selvasembian, R., Naushad, M., You, S., Oleszczuk, P. and Ok, Y. S. (2021) Hydrometallurgical processes for heavy metals recovery from industrial sludges. *Critical Reviews in Environmental Science and Technology* (Early Online Publication)

(doi: [10.1080/10643389.2020.1847949](https://doi.org/10.1080/10643389.2020.1847949))

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Deposited on: 3 November 2020

Hydrometallurgical processes for heavy metals recovery from industrial sludges

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Hydrometallurgical processes for heavy metal recovery from industrial sludges

Hydrometallurgical approaches have been successfully employed for metal separation and recovery from various types of waste materials. Therefore, hydrometallurgy is a promising technology for metal recovery and the removal of potentially toxic heavy metals found in industrial sludge. However, a comprehensive review that focuses on the heavy metal recovery from industrial sludge using hydrometallurgical approaches has not been conducted in the recent past. The present review discusses the capacity of hydrometallurgical techniques in recovering heavy metals sourced from different types of industrial sludges, highlighting recent scientific findings. Hydrometallurgical approaches primarily consist of three process stages: metal dissolution, concentration and purification, and metal recovery. The chemical characteristics of industrial sludge, including the type, concentration and speciation of heavy metals, directly impact selection of the best recovery method. Solvent extraction, ion-exchange, and adsorption are the major techniques employed in concentration and purification, whereas electrodeposition and precipitation are the main methods used in metals recovery. Future research should focus on the development of more efficient and environmentally-friendly methods for metal dissolution from industrial sludges contaminated with multiple metals, while increasing selectivity and energy use efficiency in the concentration and purification, and recovery steps.

Keywords: Industrial waste; Resource recovery; Precious metals; Acid leaching; Solvent extraction

Introduction

Heavy metals are raw materials employed in numerous industrial processes; their demand in the modern world is increasing exponentially owing to extensive usage in electronics and other manufactured products. For instance, the production of crude steel has approximately doubled from 2000 to 2013 (i.e., 850 to 1630 million tons), and the production of Cu has increased from 12.9 million tons in 2000 to 19.7 million tons in 2017 (Kaksonen et al., 2018; Mikoda et al., 2019). Metals are typically extracted from metal ores using industrial metallurgical processes. However, the increasing global demand for metals has caused a rapid depletion of high-grade metal ores. Metal recovery from readily available and continuously generated secondary resources and waste has become more appealing as compared to treatment and landfill disposal (Shemi et al., 2015; Sethurajan et al., 2018). Industrial metallurgical processes involve the production of metals from naturally occurring ores result in substantial amounts of metal-rich solid wastes. More than 50% of wastes generated in Europe originate from the quarrying and mining industries (Lottermoser, 2010). Therefore, the development of economically viable and environmentally friendly approaches for recovering and recycling metals from secondary waste sources is an important and timely concern as it helps in achieving sustainable resource management and mitigating possible environmental threats. Several scientific studies have been conducted for the recovery of metals from wastes including sludges, slags, fly ashes, shales, spent catalysts, and spent liquors; industrial-scale treatments units have been developed recently (Sethurajan et al., 2018; Sethurajan et al., 2017). These studies and developments are the result of growing global concern about industrial wastes, metal recovery, and hydrometallurgy over the past two decades (Figure 1).

Industrial sludge is recognized as an important source of heavy metal-bearing secondary waste materials that is generated in high quantities every year. For example, the worldwide annual generation of Al-rich paper industrial sludge exceeded 1.6 metric tons (Cusidó et al., 2015). Moreover, 100,000 tons of various precious metals in the form of electroplating sludge are wasted annually in China (Li et al., 2010). Land disposals of these toxic metal-rich sludges have a high potential to cause numerous environmental and health-related impacts. A study conducted by Ozdemir & Piskin (2012) showed that rainwater has a high capacity to dissolve heavy metals from galvanic sludge, which has been classified as one of the highly polluting industrial wastes in the natural environment (Bocanegra et al., 2019). To ensure comprehensive environmental protection and resource recycling, it is essential to determine effective technologies for extracting hazardous heavy metals from industrial sludges. In comparison to various methods of metal recovery from metal-enriched waste materials, hydrometallurgical processes have gained considerable attention as they show effectiveness in the extraction of metals. Hydrometallurgical approaches are primarily based on acid leaching with the use of inorganic acid, such as sulfuric, nitric, and hydrochloric acids, in the metal extraction process (Huyen et al., 2016). However, most recent studies focus on the bioleaching approach with various types of microbial strains to convert heavy metals from industrial sludges into the solubilized state (Zhang et al., 2020; Wu et al., 2020; Zhou et al., 2019b). Some recent studies have attempted to compare the leaching rates of heavy metals from industrial sludge with several inorganic and organic acids, and reveal the mechanisms of metal dissolution that resulted from different acids (Gunarathne et al., 2019b). Consequently, it is essential to provide insights into the development of hydrometallurgical technologies for metal recovery from industrial sludges as it serves as a viable method for utilizing sustainable waste in an environment-friendly manner.

In this review, we emphasize the necessity for the extraction of heavy metals from industrial waste sludges by highlighting their hazardous properties, associated environmental consequences, and provide extensive evidence of sustainable and effective recovery of metals through hydrometallurgical processes with the support of recently published scientific literature.

[Figure 1 near here]

Chemical characteristics of industrial sludge

The properties of industrial sludge depend on the raw material, unit processes, and unit operations involved in the industries that generate it (Silva et al., 2005). However, properties such as pH, organic carbon, nitrate, phosphate, and metal contents of industrial sludge are influenced not only by the industrial processes but also by the performance of industrial wastewater treatment. Table 1 indicates the changes in chemical properties of different types of industrial sludge based on the type of industry that generates them. It is important to understand the general chemistry of sludges because their disposal in the environment may alter the chemical properties of soil and water environments. For example, pH directly influences heavy metal solubility and sludges having low pH may contain elevated concentrations of soluble metals. In addition, chemical parameters such as pH and nutrient content (e.g., N and P) readily interfere with the ideal reaction conditions that are maintained during most treatment and resource recovery processes for sludge.

[Table 1 near here]

Heavy metals content

Various industrial processes generate metal-bearing wastes in large volumes. Typically,

industrial sludges contain elevated concentrations of heavy metals, such as Cr, Ni, Zn, Cu, and Pb (Cui et al., 2010). Table 2 details these heavy metals and their concentrations in waste sludge derived from different industrial sources.

Electroplating sludge is derived from spent electrolytes or rinse water sources of electroplating industries (Li et al., 2011; Peng & Tian, 2010). This sludge contains characteristically high concentrations of heavy metals: Ni, Cu, Zn, Cr, Mn, and Fe (Yue et al., 2019b; Zhang et al., 2020; Özbaş et al., 2013). Typical heavy metal contents of Cu, Ni, Zn, and Cr in electroplating sludge range between mass ratios of 1–8%, 1–10%, 0.5–5%, and 1.5–15%, respectively (Yan et al., 2019). Arsenic sulfide sludge, which is generated from acidic wastewater of sulfide precipitation technique, contains elevated concentrations of As and has long been identified as a problematic sludge. Xu et al. (2020) reported that As accounts for 46.9% of the total elemental composition of arsenic sulfide sludge. Arsenic-iron sludge generated from electrochemical arsenic remediation plants also contains elevated As. In a recent study, Roy et al. (2019) reported an As range of 250–1219 mg kg⁻¹ in arsenic-iron sludge collected from two different contaminated groundwater remediation plants in India.

The granitoid quarry industry also produces large volumes of heavy metal (e.g., As, Mo)-contaminated sludge through sawing of rock masses (Lieberman et al., 2019). Sludge generated from the paper industry contains high amounts of Al. The battery industrial sludge contains elevated amounts of Fe, Zn, Ni, and Cd, while agro-industrial wastewater sludge contains a considerable amount of Fe (Table 2). Similarly, the mixed sludge derived from wastewater sources of electroplating, pickling-rolling, and textiles industries possesses elevated concentrations of Fe, Ni, Cu, Mn, Cr, and Pb. Moreover, the sludge derived from petrochemical,

brew, and paper industries primarily contain Zn and Cu. Thus, the type of heavy metals present and their concentrations depend on the origin of the industrial sludge.

Characterization of industrial sludge for metals content is vital for decision-making and to adopt the most suitable management practice such as stabilization, metal recycling, or use as a raw material in other industrial processes (Ozdemir & Piskin, 2012). The total metal content of sludge can be evaluated either through microwave digestion and detection of heavy metals, or through more sophisticated direct techniques such as X-ray fluorescence spectroscopy (XRF) or energy-dispersive X-ray spectroscopy (EDS). In addition, DTPA (diethylenetriaminepentaacetic acid) and CaCl_2 extraction procedures provide viable approaches for evaluating bioavailable heavy metal concentrations (Kashem et al., 2007; Peijnenburg et al., 2007).

[Table 2 near here]

It is also important to understand the fractionation of heavy metals in different phases of sludge to select an appropriate treatment procedure or metal recovery approach. While various procedures for determining the heavy metals associated with different phases in soil and sediments have been developed, the methodology developed by Tessier et al. (1979) is a widely used analytical method to determine the speciation of heavy metals in soil and sediment samples. This procedure consists of five consecutive extractions to separate the heavy metals that are bound to exchangeable, carbonate, Fe-Mn oxide, organic matter, and residual fractions. Gunarathne et al. (2019b) successfully employed the aforementioned method to evaluate heavy metals such as Cu, Pb, and Ni that are associated with different fractions of sludge generated from the electrical appliance manufacturing industry. They observed that the majority of Cu was bound to the carbonate and Fe-Mn oxide fractions (i.e., 65.0% and 25.8%, respectively), whereas Pb was primarily associated with residual and organic matter-bound fractions (i.e., 63.7% and

32.3%, respectively). Furthermore, they observed that Ni was associated with Fe-Mn oxide and carbonate fractions (i.e., 46.1% and 41.8%, respectively). A similar study conducted by Gunarathne et al. (2019a) indicated the presence of Pb in two major fractions of sludge generated from the battery manufacturing industry—5413.6 mg kg⁻¹ in the Fe-Mn oxide fraction and 2962.4 mg kg⁻¹ in the carbonate fraction. Figure 2 illustrates the association of metals (M) in different fractions of sludge particles based on the classification of Tessier et al. (1979).

[Figure 2 near here]

The European Community Bureau of Reference (BCR) sequential leaching test is another frequently used analytical method of determining the speciation of heavy metals in industrial sludge (Mossop & Davidson, 2003). The modified BCR sequential test separates metals associated with five different fractions: water soluble, acid soluble and carbonate, Fe-Mn oxides, organic compounds or sulfides, and residual fractions. Su et al. (2019) applied this method to evaluate the phase association of metals in stainless-steel pickling sludge before and after sodium sulfide hydrate stabilization. The study revealed that Cr and Cu in stainless-steel pickling sludge are primarily associated with the Fe-Mn oxides fraction and Ni and Zn are mainly associated with the acid soluble and carbonate fractions. Forty-five days of sodium sulfide hydrate stabilization treatment was involved to transform a high percentage of the Cr into the residual fraction and Cu into the organic compounds or the sulfides fractions, while most of the Ni and Zn remained with the acid soluble and carbonate fractions.

Environmental consequences and health risks related to heavy metals

The need for metal recovery from heavy metal-enriched sludges is two directional, as these sludges supply a viable source for economically important metals and poor sludge management

practices lead to environmental issues. Therefore, the heavy metal content in industrial waste sludge is one of the most important parameters used to evaluate the toxicity and possible hazards of sludge. Metals can be released from the solid wastes through natural weathering processes and microbial activities (Sethurajan et al., 2018). The release of heavy metals into the natural environment through the direct disposal of non-treated or partially treated industrial waste may contribute to human health risks and ecosystem damage (Tou et al., 2017; Pecht et al., 2018). Therefore, with regard to environmental contamination, threshold limits have been set for heavy metals content in sludge by many nations across the world (Wang et al., 2005). Moreover, the chemical form or speciation, and concentrations of heavy metals are indicative of the extent of hazardous effects. For example, studies have demonstrated that the available fraction of heavy metals in sludge can impact the bioavailability, mobility, and phytotoxicity of heavy metals in soil, rather than the total concentration of heavy metals in the sludge (Rauret, 1998).

Landfill disposal is the most widely used management practice for the disposal of hazardous sludge (Cusidó et al., 2015). However, landfill failures can introduce significant amounts of toxic heavy metals into the surrounding land areas and to the groundwater table. The storage of waste sludges inside storage dams is one of the methods widely used by industry. Specifically, waste materials including sludges that are released from metallurgical industries are stored inside storage dams. However, the breakdown or leakage of dams leads to severe environmental consequences (Sethurajan et al., 2018). A few cases of storage dam failures and release of heavy metals into the environment have been reported in Hungary and Brazil (Ruyters et al., 2011; Segura et al., 2016). One of the significant risks that has been associated with storage dam failures is the contamination of water resources, including rivers, lakes, drainage systems, and pits, by metals released from industrial waste materials (Johnson & Bangor, 2009). Heavy

metals, such as Pb, Cd, and Zn, are toxic toward plants, and they have adverse effects on crop production. In addition, heavy metals can be absorbed by plant roots, bioaccumulated and transferred from one organism to another via food chains (Nagajyoti et al., 2010). Ultimately, the bioaccumulation of heavy metals can reach humans through contaminated vegetables and meat products. For example, significant concentrations of heavy metals were reported in the bloodstreams of humans residing in the Rudnaya River valley, Russia, as a consequence of the bioaccumulation of toxic metals in crop plants (Kachur et al., 2003). Furthermore, heavy metals can enter the human body via consumption of contaminated water sources, and inhalation or dermal contact during the handling of metals-contaminated industrial sludge. Therefore, the possible ecological and human health-related risks associated with heavy metals in industrial waste must be assessed carefully. Figure 3 provides a concise picture of health risks and environmental threats caused by the improper disposal of heavy metals-rich industrial sludges.

[Figure 3 near here]

Hydrometallurgy

Metal recovery from industrial sludge includes three main steps: collection, pre-processing, and end-processing. The pre-processing stage mainly involves the refining and recovering of metal fractions from waste sludges, and it comprises a number of hydrometallurgical, pyrometallurgical, electrometallurgical, or integrated processes (Khaliq et al., 2014; Meskers & Hagelüken, 2009). However, pyrometallurgy and hydrometallurgy are the most widely used techniques to recover metals from wastes in industrial-scale applications. The pyrometallurgical process uses heat to smelt the constituents of waste, whereas hydrometallurgical processes use an appropriate solvent to dissolve the components of waste.

Hydrometallurgy can be defined as a metal recovery method used to obtain metals from ores and waste materials using aqueous media by combining water, oxygen, and other chemical reagents with or without the use of a pressurized environment. Hydrometallurgical metal recovery is typically performed in three main stages: metal dissolution, concentration and purification, and metal recovery (Gupta, 2006) (Figure 4).

Advantages over other techniques

Pyrometallurgical, electrometallurgical, and hydrometallurgical techniques are the main approaches employed to recover metals from low-grade ores and industrial wastes. However, pyrometallurgical approaches are the most intense methods that are applied in the recovery of metals from wastes (Ashiq et al., 2019). Pyrometallurgical processes consist of incineration and high-temperature roasting using selective gases or arc furnaces (Kaya, 2016; Kaya & Sözeri, 2009). These stages of pyrometallurgical processing are associated with disadvantages and limitations owing to either high energy consumption or poor end-product quality (Miškufova et al., 2006). Additionally, they generate a significant amount of slag with residual metals that cannot be recovered using other approaches, thereby rendering them unsuitable for resource recovery in a sustainable manner (Cui & Zhang, 2008; Kaya, 2016). Electrometallurgical techniques also often have high energy-consumption rates. Therefore, investigations into hydrometallurgical approaches are of increasing research interest.

During the past decade, hydrometallurgical recovery of metals has been considered the most suitable alternative to pyrometallurgy as the latter requires more energy and incurs higher costs. Hydrometallurgy is the most efficient, economically viable method for recovering metals from wastes as it regulates the levels of impurities at different stages and consumes less energy

(Safarzadeh et al., 2007; Kaya et al., 2020; Hao et al., 2020). Hydrometallurgical approaches such as bioleaching are considered more environment friendly as they consume minimal amounts of chemical compounds and energy.

Process stages

Hydrometallurgy involves the extraction of valuable metals into acidic or alkaline solutions by employing leaching, which is the first step that must be investigated before understanding the subsequent metal separation techniques. This method was conventionally developed for the separation of valuable metals from ores. However, hydrometallurgical processes have been adopted for recovering the precious metals from industrial wastes, and previous studies have shown promising results (Ashiq et al., 2019). Metal dissolution, which involves chemical and bioleaching approaches, is the first step in hydrometallurgy after the sorting and grading of waste materials (Wu et al., 2020).

The leach liquor resulting from the leaching step typically undergoes concentration and purification processes to concentrate metal ions that are to be recovered and for removing impurities that interfere with the metal recovery process. Solvent extraction, ion exchange, adsorption, precipitation, gaseous reduction, and electrowinning are the main procedures used in the concentration and purification step. However, some industrial procedures skip this step if the impurities do not interfere with the metal recovery step or if the leach solution contains a sufficient concentration of the target metal ions.

The third process stage is the separation of targeted metals, and it is typically carried out using either chemical reactions or voltage (Kaya, 2016). The most widely used methods for recovering metals are precipitation, electrodeposition in electrolytic cells, and gaseous reduction. Note that

methods such as precipitation and gaseous reduction can be employed in both purification and metal recovery stages.

[Figure 4 near here]

Metal dissolution

Bioleaching

The bioleaching technique uses microorganisms to solubilize insoluble forms of metals (Mishra et al., 2005). This process utilizes a wide range of microorganisms that belong to different taxonomic groups (Figure 5). Still, the microorganisms utilized in the bioleaching of metals from certain waste materials may belong to the same or different species. Initially, this process was mostly utilized for the extraction of base metals from ores (Sethurajan et al., 2018). However, many researchers reported that this technique is promising for recovering metals from metal-enriched industrial materials (Srichandan et al., 2019; Mikoda et al., 2019; Wang et al., 2018b). In addition, this technique has been successfully utilized for the solubilization of a number of valuable metals including U, Ni, Co, Cu, and Zn (Rohwerder et al., 2003). Table 3 indicates the different bioleaching approaches employed for recovering metals from different types of waste sludge.

[Figure 5 near here]

[Table 3 near here]

Bioleaching has several benefits over conventional chemical leaching approaches. Those are cost-effectiveness, facilitation of in-situ leaching, no energy consumption, no requirement for toxic chemical compounds, and less secondary waste generation (Johnson, 2013; Watling, 2006).

Therefore, bioleaching is considered one of the most environment-friendly and cost-effective approaches that can be applied for recovering metals from the industrial sludge. However, bioleaching is also associated with some drawbacks: specific microorganism strains are required for specific metals; conditions must be controlled for proper microbial activities; the recovery process can be time-consuming. Considering its applicability to a wide range of industrial wastes and associated heavy metals, and the process duration that is required to obtain an acceptable amount of metals, chemical leaching is often considered a better technique for recovering metals from the industrial sludge, as compared to bioleaching (Mikoda et al., 2019).

Chemical leaching

Chemical leaching is a method involving the reaction of a solid when exposed to a liquid. Most valuable metals exist as complexes with other metals; this poses a challenge for separation and extraction. However, chemical leaching has proven to be advantageous for separation of metallic components from sludges when compared with pyrometallurgical processes as it does not emit harmful gaseous compounds, does not generate particulate matters, and consumes a significantly lower amount of energy because it only uses solvents and acids. In addition, it features considerably higher recovery rates and can be easily performed. Therefore, chemical leaching is considered both economically and environmentally viable to recover metals from the sludge, and it is well-established among hydrometallurgical processes (Karwowska et al., 2014; Wu et al., 2020). Leaching is a process of extracting targeted valuable metals using protons and ligands resulting from acids such as hydrochloric and sulfuric acids, and several leaching agents such as thiosulfate, alkali, and cyanide (Guo et al., 2015; Awasthi & Li, 2017; Rocchetti et al., 2013; Bas et al., 2014). Based on the leaching solution used, hydrometallurgical approaches can be further

divided into different groups such as acid, alkaline, thiosulfate, thiourea, halide, and cyanide leaching. Among these, acid leaching is the most frequently used method for recovering metals from industrial sludge as it is a cost effective and an environmentally-friendly approach, whereas cyanide, thiosulfate, thiourea, and halide leaching can be employed primarily for the recovery of precious metals from sludge.

Acid leaching

The use of an acidic medium to dissolve valuable metals from industrial wastes is the basis for most hydrometallurgical processes. This process typically occurs between pH 1.0–3.0, which facilitates the synchronous dissolution of targeted metals into the leaching solution (Zhang et al., 2020; Gaber et al., 2011). Metals embedded in solid waste particles are dissolved and released into solution after being transformed into their ionic states by H^+ ions introduced by the acidic solvent. However, the solubility of metals from waste is predominantly attributed to the pH of the leaching medium (Wu et al., 2020). Therefore, acid leaching frequently utilizes different types of inorganic acids including sulfuric, hydrochloric, nitric, phosphoric, and mixtures of acids such as aqua regia (Huyen et al., 2016; Ajiboye et al., 2019; Chen et al., 2017). A few organic acids, such as citric, ascorbic, malic, succinic, and acetic acids, have also been studied for the removal of some of the light metals from waste materials (Meng et al., 2020; Sun et al., 2018; Li et al., 2015). However, most studies reveal that a higher yield of metals is obtained by leaching with inorganic acids, including sulfuric, nitric, and hydrochloric acids, rather than by leaching with organic acids such as oxalic and citric acids (Xie et al., 2018; Özbaş et al., 2013; Gaber et al., 2011).

Apart from the type of acid and acid concentration used, the solid/liquid (S/L) ratio, stirring speed, temperature, and leaching period also control the concentration of heavy metals released into the leach liquor by acid leaching. However, intrinsic factors such as the fractionation of heavy metals in different phases of the sludge directly influence the leaching efficiencies of a particular acid (Wang et al., 2015). The carbonate-bound fraction of metal is highly vulnerable to dissolution, even with a weak acid; however, by increasing the acid strength, the rate of leaching can be increased (Gunarathne et al., 2019b). In contrast, it is difficult to separate metals associated with the residual fraction through acid leaching. The metals encapsulated in crystalline assemblages of silica in the residual fraction are less susceptible to leaching with acid as Si—O bonds are highly stable (Terry, 1983). Gunarathne et al. (2019b) suggested that high acid leaching rates observed when recovering Cu from sludge generated from the electrical appliance manufacturing industry occur because much of the Cu is associated with the carbonate-bound fraction in these wastes.

Sulfuric acid (H₂SO₄) leaching is the most prominent method utilized for separating heavy metals from electroplating sludge (Yan et al., 2019). A recent study conducted by Wu et al. (2019) examined optimal conditions for the extraction of the heavy metals Cu, Zn, Cr, and Ni, from electroplating sludge using H₂SO₄ leaching. Their study revealed an improvement in the extraction of selected heavy metals by an initial addition of Fe³⁺ at a rate of 1 g L⁻¹. The addition of Fe³⁺ significantly improved the extraction of metals that were bound to the residual and organic matter/sulfide fractions. Further, the extraction of metals was improved using a higher redox potential, greater than 500 mV. Moreover, the same study observed a remarkable influence of pH on the extraction efficiency of metals, with lower solvent pH values (i.e., ≤2.2) increasing the dissolution of metals that exist in the acid-soluble and Fe-Mn oxide bound fractions.

However, it was difficult to dissolve the metals in the residual and organic matter/sulfide-bound fractions without the addition of Fe^{3+} .

Heavy metals in sludge particles are typically precipitated or adsorbed into a solid matrix (Özbaş et al., 2013). Acid leaching dissolves or desorbs the heavy metals from the matrix of waste materials by a two-way process including proton promoted and ligand promoted mechanisms. Ion exchange is the most prominent proton promoted mechanism involved in the release of metals from sludge particles by acids. Strong acids can supply elevated concentrations of protons into the leaching solution; these protons are involved in the dissolution of the matrix, resulting in the consequent release of heavy metals (Gunarathne et al., 2019b). Inorganic acids facilitate metal dissolution via proton promoted mechanisms because of the lack of chelating ligands in their molecular structures. In contrast, organic acids, which are typically associated with ligands, can promote the release of metals from sludge particles by the formation of soluble metal-ligand complexes. Organic acids with low molecular weights, such as citric, malic, tartaric, oxalic, and acetic acids, have been successfully used for the mobilization of heavy metals from solid matrices (Qin et al., 2004; Jing et al., 2007). Gunarathne et al. (2019b) evaluated heavy metal dissolution rates from electrical appliances sludge using six different inorganic and organic acids. Their study observed that the high ligand activities of organic acids resulted in higher metal dissolution rates, compared to those resulting from inorganic acids at the same pH values. Figure 6 shows the mechanisms of metals dissolution by inorganic and organic acids from sludge particles.

[Figure 6 near here]

Acid leaching applications have also been successfully studied as a treatment method as well as a resource recovery technique for a wide range of waste materials; however, the usability of acid

leaching for resource recovery from industrial sludge has not been investigated sufficiently. Table 4 shows the efficiencies of metals leaching from industrial sludge and sediments using different types of acid solvents.

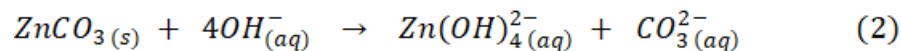
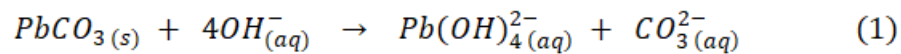
[Table 4 near here]

Alkaline leaching

Alkaline leaching is considered a more selective method to leach targeted heavy metals than acid leaching (Ghasemi & Azizi, 2018). Further, the alkaline leaching technique is recognized as an economically feasible and easy-to-use technique for some heavy metals such as Zn obtained from low-grade oxidized ores or waste with impurities including Cu, Cd, Ni, and Co. These impurities are not easily leached with alkaline media; thus, employing electrowinning in the metal recovery step consumes less energy than that in acidic media (Youcai & Stanforth, 2001; Zhao & Stanforth, 2000).

A recent study conducted by Klein et al. (2019) applied alkaline leaching to electrolytic tinning sludge to recover Sn, and a recovery of 90% was achieved under optimal conditions using 3M NaOH at 30 °C. Another study conducted by Gargul et al. (2016) investigated the applicability of alkaline leaching for the separation of Zn from iron-bearing sludge derived from basic oxygen furnaces. This sludge is characterized by a significantly lower concentration of Zn than that found in sludge and dusts derived from electric arc furnaces. Leaching using either 5 M NaOH or KOH at optimal conditions of 95 °C for 60 min reduced the Zn content of sludge by 66%. In contrast, at ambient temperature (25 °C), the reaction required 100 h to reduce the Zn content of the sludge by up to 60%.

Although recent studies that used alkaline leaching for industrial sludge are rare, this method has been sufficiently studied for the separation of heavy metals from low-grade ores and metal-rich dusts resulting from industrial activities. Ghasemi & Azizi (2018) studied the alkaline leaching of Pb and Zn from low-grade mine tailings using NaOH. They reported recoveries of 72.15 and 85.52% for Pb and Zn, respectively, using 4 M NaOH at a temperature of 80 °C and a liquid-to-solid ratio of 20 ml g⁻¹. The percentage recoveries of Pb and Zn increased with increased NaOH concentration. Specifically, the leaching of Pb increased from 23.55% to 64.01% with an increase in NaOH concentration from 0.5 M to 4 M, and the leaching of Zn increased from 24.12% to 80.92% with the same increase in NaOH concentration. The release of Pb and Zn from mine tailing using NaOH can be explained by following reactions:



Alkaline leaching has the potential to be applied in the selective separation and recovery of specific metals from multi-metal contaminated industrial sludges. However, further studies must be conducted to uncover the possible applications of alkaline leaching for industrial sludge.

Thiosulfate leaching

Thiosulfate solutions can be used as an alternative to cyanide-based leaching solutions that are highly toxic and are known to lead to potentially severe environmental consequences.

Thiosulfate leaching is highly effective for recovering Au and Ag, which are found in waste

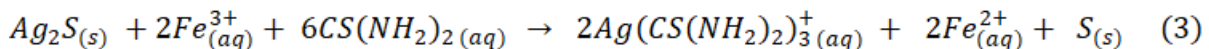
materials resulting from the cell phone industry in considerable amounts (Abbruzzese et al., 1994). Therefore, this leaching technique is suitable for the treatment of wastewater sludge resulting from the mobile phone manufacturing industry. The primary benefit of using thiosulfate for the recovery of valuable metals is that it solubilizes the targeted metals by making recoverable complexes while resulting in less interference with other cationic species (Abbruzzese et al., 1995). Solvents such as ammonium thiosulfate or sodium thiosulfate solutions solubilize Au, Ag, Pt, and other metals in the form of stable anionic complexes over a wide range of pH. The ammonia in the “pregnant solution” (i.e., the acidic metal-laden solution that results from the leaching process) stabilizes the complex by inhibiting the formation of further oxides of metals (i.e., Au to Au(I)) (Abbruzzese et al., 1995; Ubaldini et al., 2000). Two forms of thiosulfates, sodium and ammonium thiosulfate, have been effectively employed to recover Au from waste materials. These solutions are nontoxic, non corrosive, and moderately stable in alkaline conditions. However, the requirement of the solvent is considerably high as compared with other leaching solvents.

An oxidizing agent, such as Cu(II) or Fe(III), is also required along with the primary leaching solvent and co-leaching agents such as ammonia or thiourea, to achieve reaction rates sufficient to recover economic concentrations of precious metals. The reaction of Au with ammoniacal thiosulfate results in a complex having high stability and solubility; thus, the resulting environmental consequences are minimal when compared to those of cyanide-based solvents. This method consumes lower amounts of oxidizing agents such as Cu ions to facilitate the dissolution, oxidation, and complexation of metal ions. However, additives such as ethylenediaminetetraacetic acid (EDTA) and monoethanolamine acid (MEA) are used to

stabilize the oxidizing agents in ammoniacal solutions to promote the formation of metal complexes (Puente-Siller et al., 2017; Cho et al., 2014; Puente-Siller et al., 2013).

Thiourea leaching

Thiourea leaching was developed as an alternative to the well-known cyanide leaching as the latter may be detrimental to the environment (Birich et al., 2019); however, thiourea is 25% more expensive than cyanide (Boboev et al., 2019). Thiourea ($SC(NH_2)_2$) is a sulfur-based complexing agent that has the ability to form soluble, cationic complexes with targeted metals, in contrast to other anionic complexes. The leaching rate of this complex extends up to 99%, making it readily soluble in the solvent medium. Thus, thiourea is one of the most widely used leaching techniques for recovering precious metals from industrial waste sources. The following equation, which utilizes thiourea as the leaching agent and Fe(III) ions as the oxidant, represents the complexation of Ag from Ag_2S using thiourea (Bernaola-Flores et al., 2019; Bruckard et al., 1993):

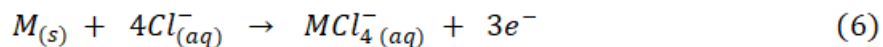
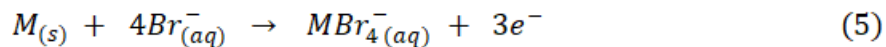
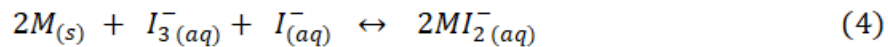


The reaction requires an acidic medium because thiourea is not stable and decomposes easily in alkaline conditions (Ashiq et al., 2019). The use of thiourea is an environmentally-friendly and low-cost method, while simultaneously offering rapid leaching and little co-ion interference issues (Birich et al., 2019). However, due to sulfur precipitation and the oxidation of solution into disulfide formamidine, which result in surface passivation, this extraction method consumes large quantities of reagents as compared to other leaching methods (Syed, 2012). The threshold mass of thiourea concentration, reaction temperature, contact time, initial feed concentrations,

and solid interface characteristics (e.g. surface area) influence the leaching rates of the target metal ions from solid wastes (Xu & Li, 2011; Veit & Bernardes, 2015a).

Halide leaching

Studies have uncovered that the leaching rate of metals is high in halogenic environments, and so chloride, bromide, and iodide have been employed as leaching agents. For example, tri-iodide ions serve as an oxidizing agent for a number of metal ions and produce metal-iodide complexes by reacting with metals in waste materials such as industrial sludge. Iodine has been recognized as the most suitable halogen for use as a leaching solvent owing to its less hazardous nature and low volatility. However, the industrial-scale use of iodine is rare because of the high market price of industrial-grade iodine. In contrast, chlorine and bromine are volatile and hazardous because of their highly corrosive nature (Birich et al., 2019). The following reactions demonstrate the leaching of a divalent metal (i.e., M) with different halide solutions.

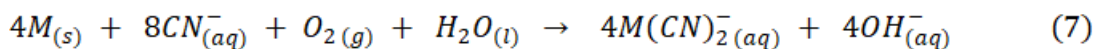


Halides have been utilized, in most of the cases, as a replacement for cyanide-based leaching solvents to minimize the environmental consequences of cyanide compounds (Seisko et al.,

2018). High solubility, improved rates of release, and high redox potentials have been demonstrated for the removal of Au in a halide medium (Liddicoat & Dreisinger, 2007). Halogenic leaching agents are economically viable, selective toward target metals, and do not affect downstream processes; thus, they meet the criteria for being ideal leaching agents for metal recovery.

Cyanide leaching

Regardless of toxicity issues, the use of cyanide-based solvents is a conventional technique that has been extensively used for recovering precious metals such as Au and Ag from primary ores and industrial wastes. Cyanide ions act as the leaching agent, whereas oxygen plays the role of an oxidant (Bernaola-Flores et al., 2019). Alkaline solutions or alkaline-based metal-cyanide solutions delivered promising results in recovering Au and Ag by dissolution under aerated conditions (Bisceglie et al., 2017; Marsden & House, 2006). Cyanide dissolves the metals by forming readily soluble dicyano-metal complexes. However, the availability of oxygen in the reaction medium is essential for metal dissolution via cyanide leaching. To prevent the formation of volatile hydrogen cyanide, the reaction medium must be maintained in a basic state (pH~10) (Birich et al., 2019). The following equation represents the complexation of Au and Ag by the reaction of cyanide, where M represents the metal (i.e., Au or Ag) ions (Jiang et al., 2015).



Copper is readily soluble in cyanide-based solvents. Previous studies have shown that the use of a simple acid sulfate is the most successful pathway for recovering Cu in the first stage, followed

by cyanide treatment for forming complexes in the “pregnant solution” in the next phase (Kamberović et al., 2011; Oh et al., 2003; Quinet et al., 2005). Therefore, cyanide leaching is considered a promising method of recovering Cu from Cu-rich sludge generated by industries such as the electroplating and electrical product manufacturing industry.

Potassium cyanide, sodium cyanide, and 3-nitrobenzene sulfonic acid sodium salts are the most widely used cyanide-based solvents for leaching metals from waste materials, as they are readily soluble in water (Birich et al., 2019). Large-scale treatment plants use a mixture of potassium cyanide and 3-nitrobenzene sulfonic acid sodium salts to upsurge the dissolution rates of metals by forming complexes at stoichiometric levels (Bisceglie et al., 2017). The essential factors that must be considered during metal leaching with cyanide-based solutions are the presence of air/oxygen, pH of the solvent, temperature, cyanide concentration, stirring rate, area of the contact surface, and the presence of anions/cations in the liquid-solid interface.

Concentration and purification

Concentration and purification steps are typically carried out using solvent extraction, ion-exchange or adsorption techniques according to the type and concentrations of targeted metals and impurities present in the leach liquor solution (Safari et al., 2020). In some cases, chemical precipitation is also utilized during concentration and purification steps. However, this step can be skipped and directly employ the metal recovery if the concentration of targeted metals is sufficient or the concentration of impurities is sufficiently low in the leach liquor solution.

Solvent extraction

Solvent extraction or liquid–liquid extraction is the process of separating target elements using a

liquid solution with an insoluble liquid such as an organic solvent. The extraction mechanism involves the transfer of a solute from the “pregnant solution,” which is obtained from leaching and known as the “raffinate,” to the organic solvent (Theodore & Ricci, 2010). The main principle used in extraction is the solubility of both the solution and the solute. All the components are soluble in each other at some level; however, the separation becomes feasible if their solubilities are sufficiently different, thereby forming a two-phase system (Veit & Bernardes, 2015b).

Different types of extractants, such as anionic, cationic, or solvating, must be utilized depending on the metal ions present in the leaching solution. The anionic type solvents primarily consist of amines or amides and are used for the extraction of valuable metals including, gold, vanadium, iridium, rhodium, and tungsten (Jha et al., 2012). For both cationic and anionic types, the pH, composition of the solution, concentration and type of the extractor, and aqueous–organic volume ratio also determine the efficiency of the solvent extraction process. The main advantages of using solvent extraction are the selectivity and high purity of targeted metals and the ability to process large quantities at one time (Chagnes & Pospiech, 2013).

Solvent extraction was utilized by Guimarães & Mansur (2017) with di-2-ethylhexylphosphoric acid (D2EHPA) and Cyanex 272 extractants for the selective removal of Ca and Mg to purify Ni in a concentrated sulfate solution. During the first extraction comprising two stages, 98.6% of Ca was removed with the use of D2EHPA extractant under the conditions of pH 3.0 and 50 °C. In the second extraction consisting of a single stage, Cyanex 272 extractant was utilized and 99% of Mg was removed (pH 5.7 and 50 °C). The solution resulting from the purification process was suitable for recovering Ni through electrowinning. Additionally, the simultaneous removal of Ca

and Mg from a Ni sulfuric solution was demonstrated by the same authors with the use of three extractants: Cyanex 272, D2EHPA, and naphthenic acid (Guimarães & Mansur, 2018).

In the study conducted by Innocenzi & Veglio (2012), two stages of extraction using D2EHPA in n-dodecane was found to be sufficient for the effective separation of Zn and Mn from a leaching solution containing Ni at the optimal conditions of $\text{pH} \leq 2.5$ and $\text{O/A} = 1/1$, with a 30-min leaching period at ambient temperature ($\text{O/A} = \text{organic phase volume/aqueous phase volume}$).

Wang et al. (2018a) had studied the application of solvent extraction for the removal of trace V from an ammonium tungstate solution. The organic solvent, comprising 1% (v/v) Aliquat336 and 5% (v/v) 2-octanol, could reduce V(V) concentration from 0.42 g L^{-1} to 0.004 g L^{-1} after a six-stage extraction with $\text{pH} 8.74$, $\text{A/O} = 1/1.25$, and a 10-min of separation period at $25 \text{ }^\circ\text{C}$.

Ion-exchange

Resins that contain chelating functional groups to form complexes with metal ions can be used for selective separation of targeted metal ions. Impregnated resins are developed by adsorbing solvent extraction reagents onto polymer beads (Tavlarides et al., 1987). The first hydrometallurgical application of ion exchange resins was for uranium recovery. However, their usage became widespread with the development of chelating and impregnated ion-exchange resins. The effectiveness of an ion-exchange resin is typically expressed as the equilibrium loading capacity or exchange capacity. Other characteristics of resins are functional groups, their selectivity ratio, cross-linking, porosity, and matrix geometry. Ion exchange can successfully be used for selective separation and recovery of metal ions by changing the properties of resins, specifically their functional groups. The ease of operation, no reagent losses, no disengagement of phases, economic feasibility for use in low concentrations of metal ions, and environmental

safety can be considered as the advantages of using ion-exchange resins for metal recovery (Nikoloski & Ang, 2014; Tavlarides et al., 1987).

Ion-exchange resins have been used to separate several metals from aqueous solutions. Nguyen & Lee (2013) utilized two cationic and anionic resins (Diphonix and AGI-X8 resins, respectively) to separate Mo and V from hydrochloric and sulfuric acid solutions. Owing to the similar chemical properties, the separation of Mo(VI) and V(V) has been long considered a difficult task. The maximum separation of Mo from the sulfuric acid solution was observed with the AGI-X8 resin at pH 1.2. In those conditions, 176 mg g⁻¹ of Mo was loaded into the AGI-x8 resin, and no V was loaded. The efficient recovery of Mo that was loaded into the resin was demonstrated using the NaOH solution.

In a recent study conducted by Zhou et al. (2019a), the ion-exchange resin D201 was utilized for the removal of Zn(II) from manganese-zinc chloride mixed liquor solution. Manganese-zinc chloride waste liquor generated during the Co smelting process is an excellent source for the synthesis of high-purity manganese sulfate; however, the presence of Zn(II) as an impurity interferes in the synthesis process. The conventional technique does not facilitate adequate separation rates for Zn(II) from mixed liquor solutions owing to the analogous physicochemical characteristics of Zn and Mn. A maximum of 158.98 mg g⁻¹ of Zn(II) was removed using D201 resin at ambient temperature. The removal of Zn(II) was significantly controlled by reaction conditions such as the contact time, dosage of the resin, temperature, and chloride concentration.

Adsorption

One of the most utilized hydrometallurgical techniques for the removal of metal ions present as impurities in leach liquor is adsorption. The removal of Tungsten (W), which is present in leach

liquor as an impurity, to facilitate efficient the recovery of Mo is one of the most critical applications of adsorption. The separation of these two metal ions is difficult because of their almost equal ionic radii due to the phenomenon of lanthanide contraction. The study conducted by (Srivastava et al., 2012) showed the separation of W from a mixed liquor solution that was obtained from the leaching of the hydro-desulphurization catalyst using manganese and ferric compounds. The results indicated approximately 90% adsorption of W and 100% adsorption of V and Co on freshly prepared adsorption material of hydrous ferric oxyhydroxide with optimal conditions of 0.5 M W/Fe ratio, 50 °C temperature, and a reaction period of 2 h. However, the loss of Mo was noted as 10%. A similar study conducted by Zhang et al. (2014) to separate W from mixed Mo solution using microporous resin D301 (analogous to Amberlite IRA-94) showed better selectivity for W at near-neutral pH. The W adsorption rate was enhanced with the increase in temperature and decrease in the particle sizes of resin. The adsorption kinetics of W was explained by the pseudo-second order adsorption model, and diffusion was recognized as the mechanism that regulated the adsorption rates for W and Mo.

Moreover, the adsorption of metal ions by activated carbon for the selective separation of metals from different sources has been extensively studied. Dai et al. (2010) studied the equilibrium adsorption of copper cyanide species by activated carbon as they interfere in the process of Au recovery from mixed liquor solutions. Some studies show that the adsorption of copper cyanide complexes onto activated carbon is considerably increased with the increase in ionic strength caused by cations such as Ca^{2+} (Tauetsile et al., 2018). The Ca^{2+} ions can interact with acid groups of activated carbon to generate excess positive charge on the activated carbon surface, thereby enhancing the adsorption of negatively charged copper cyanide complexes, regardless of the net surface charge (Souza et al., 2018).

Liu et al. (2013) investigated the mechanism and adsorption properties of Fe(III) onto 2-ethylhexyl phosphonic acid mono(2-ethylhexyl) ester (HEHEHP) resins. The selective removal of Fe(III) from leach liquor solutions is important as these ions lead to common problems in metal recovery. The study showed the maximum adsorption of Fe(III) onto HEHEHP resins with the optimal pH value of 1.5. The adsorption capacities of different HEHEHP resins that were examined ranged from 15.9 mg g⁻¹ to 25.6 mg g⁻¹ at 25 °C. Several bio-sorbents, including bacteria, fungi, algae, and chitosan derivatives, have also been used for the separation of various types of metals (Ogata & Nakano, 2005; Wang et al., 2017; Yurtsever & Şengil, 2009).

Metal recovery

Metal recovery is typically carried out employing either electrodeposition or precipitation techniques. In this step, solid metals are recovered from leach liquor.

Electrodeposition

Electrodeposition is the most widely used method for recovering metals from metal-containing industrial wastes. It facilitates high recovery efficiencies for metals with a small amount of leaching medium; therefore, it is considered an economically feasible and environmentally-friendly method (Fogarasi et al., 2015).

Heavy metals, such as lead, copper, and tin, can be easily recovered through leaching, followed by electrodeposition. However, conventional electrodeposition has a lower capacity to recover copper and lead. Therefore, rotating cylinder electrode reactors, which deposit copper at the cathode and lead dioxide at the anode, are utilized for the simultaneous recovery of copper and lead to better recovery efficiencies (Mecucci & Scott, 2002). Further research should be

conducted to determine the optimal conditions for electrodeposition, as it is a solitary end process for recovering copper from waste materials. The study conducted by Fogarasi et al. (2014) involved the development of a direct and mediated electrochemical oxidation method for the recovery of metals. This process dissolves metals from waste materials and leaches them into the leaching solvent, thereby achieving simultaneous cathodic electrodeposition of copper. The utilization of the $\text{Fe}^{3+}/\text{Fe}^{2+}$ redox couple in the acid electrolytes enables high recovery of copper along with enrichment of gold in the residue (Fogarasi et al., 2014). Moreover, the deposition of copper at the cathode and current efficiency are functions of the flow rate of the system. This process yields 99.95% copper content at the cathode, along with gold and silver enrichment in the residue, after further modification with ceramic separators and establishing an optimal flow rate. Moreover, gallium, indium, and copper can be easily recovered through electrodeposition using a solvent made with a mineral acid (Gustafsson et al., 2014).

Precipitation

Precipitation involves the selective separation of heavy metals from leach liquor solution using chemical compounds. This method has wide applicability in the separation of Cr and Fe from mixed liquor solutions resulting from acid leaching. Liu et al. (2014) effectively separated Cr from a leach solution by removing 98.50% of the iron impurities by leveraging the low solubility of ferrous oxalate ($k_{sp} = 2.1 \times 10^{-7}$) at pH 4.0 and 25 °C.

A recent study conducted by Yue et al. (2019a) investigated Cr(III) and Fe(II) recovery from a multi-metallic acid leach solution of electroplating sludge via Cr(III)-Fe(III) co-precipitation on the surface of maghemite ($\gamma\text{-Fe}_2\text{O}_3$). In this study, $\text{Fe}_2(\text{SO}_4)_3$ was used as a source for Fe to facilitate the co-precipitation of Cr(III)-Fe(III) on the surface of maghemite. A two-stage co-

precipitation process successfully recovered 96.17% of Cr and 99.39% of Fe from the leach solution with minimal impurities of Ni, Cu, and Zn (0.41, 0.38, and 0.22%, respectively). The results indicate that the high Fe(III):Cr(III) ratio in the solution facilitated the hydrolysis and corresponding precipitation of Cr(III). The fine particles of maghemite behaved similarly to crystalline nuclei to induce the co-precipitation of “core-shell structured” Cr(III)-Fe(III) on its surface. This process enhances the sedimentation of co-precipitates inside the magnetic field. The resulting co-precipitates can be directly used as a raw material for Cr smelting after employing heat treatment. This study showed the economic and practical use of precipitation for recovering Cr from leach solutions. Furthermore, the Cr(III)-Fe(III) co-precipitation method could effectively separate Fe and Cr from a mixed liquor solution, preventing their detrimental interference in the recovery of Ni, Cu, and Zn.

Yan et al. (2019) conducted a study to investigate the feasibility of using phosphate precipitation for the selective recovery of Cr(III) from a sulfuric acid leaching solution, which was obtained from a mixed electroplating sludge. The study indicated the effective separation of Cr and Fe using this Cr(III) – Fe(III) – PO_4^{3-} – H_2O system. The optimum conditions for the reactions were reported as a 1:1 ratio of PO_4^{3-} to Cr^{3+} , pH 2.0, and temperature of 80 °C, whereas maximum precipitation efficiencies of 97.04% for Cr and 14.75% for Fe were observed. In addition, the introduction of sulfite to the phosphate precipitation solution significantly reduced Fe precipitation. A purer CrPO_4 precipitate was obtained through the removal of impurities using dilute sulfuric acid.

Future perspectives

Industrial wastes are accumulating in the environment at an accelerated rate owing to industrial

development and extensive usage of electronic devices and other equipment in society.

Therefore, various technologies have been developed for the management of and resource recovery from industrial waste materials; however, research on metals recovery from industrial sludge is limited. In particular, more efficient, sustainable, and environment-friendly approaches are required for the management and resource recovery of industrial sludge.

When considering environmental aspects of metals recovery from sludge, bioleaching is a promising technology as it uses specialized microorganisms for the sustainable recovery of metals. However, it has the following limitations:

- existing microbial strains require specific conditions that must be consistently maintained in the system;
- particular microbial strains used for bioleaching are specialized, and they can leach only one or a limited number of metal types from wastes;
- microorganisms precipitate in the leaching solution, metals are often toxic toward microbes, and adaptation is required;
- a considerable amount of time is required for the adequate recovery of metals from waste materials.

Therefore,

- the identification/ development of new microbial strains with high metal tolerances and that target an extended range of metals, and
- the development of transgenic microbial strains with specific genes for effective metal dissolution from waste could be the best solution. Further research must be conducted to fill the gaps in current knowledge.

Chemical leaching has proven its effectiveness and efficiency in the recovery of a range of heavy metals from industrial wastes having widely ranging chemical characteristics. However, there are a few problems associated with that process.

- It involves the use of toxic chemicals (*i.e.*, Cyanide leaching), and
- secondary waste materials are produced.

Hence,

- the search for alternative, less toxic chemical solvents to be used as leaching agents must be considered in future research.

Finally, the removal of impurities and the recovery of metals from leach liquor solution is a challenging task as it requires high-energy inputs (e.g., electrodeposition) or expensive materials (e.g., ion exchange and adsorption). Thus,

- research on a low-cost and environmentally-friendly adsorbents or ion-exchange materials for efficient and more selective removal of impurities (e.g., biochar could be a promising alternative for a low cost and environment-friendly material for use as an adsorbent), and
- development of efficient methods for recovery of metals from leach liquor solution (e.g., more selective precipitation agents) are valuable.

Acknowledgement

The authors would like to acknowledge the Research Council, University of Sri Jayewardenepura, Sri Lanka, for providing financial support (Grant No. ASP/01/RE/SCI/2017/82) and the Ecosphere Resilience Research Centre, University of Sri Jayewardenepura, Sri Lanka, for providing the working environment.

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Table 1. General chemical characteristics of different types of industrial sludges.

Source of industrial sludge	Country	pH	Organic carbon (%)	N (%)	P (%)	K (%)	Ca (%)	Mg (%)	S (%)	Na (%)	CEC (Cmol kg ⁻¹)	EC (mS cm ⁻¹)	Reference
Paper industry	Spain	-	-	0.25	-	0.01	8.50	0.17	-	0.06	-	-	Cusidó et al. (2015)
	Bangladesh	7.39	58.70	2.04	3.14	0.98	-	-	-	-	52.40	-	Islam et al. (2017)
	India	-	39.52	0.50	-	-	-	-	0.11	-	-	-	Pradhan (2011)
Battery industry	China	11.50	-	-	-	-	13.05	0.93	-	-	-	-	Wu et al. (2015a)
Tannery industry	Bangladesh	7.85	64.10	2.11	3.62	1.11	-	-	-	-	79.60	-	Islam et al. (2017)
	Malaysia	7.40	20.00	-	0.09	0.42	7.70	0.12	-	0.10	-	9.00	Ahmed et al. (2007)
	China	7.83	43.60	1.35	0.69	-	-	-	-	-	-	-	Fang et al. (2007)
	China	7.32	38.26	2.21	-	-	-	-	5.63	-	-	-	Zeng et al. (2016)
		8.04	40.75	1.78	-	-	-	-	4.55	-	-	-	

		7.21	36.87	1.99	-	-	-	-	5.24	-	-	-	
Synthetic dye chemical industry	Banglade sh	5.37	55.60	3.01	2.24	1.98	-	-	-	-	66.10	-	Islam et al. (2017)
Textile industry	Banglade sh	8.91	49.10	1.92	2.05	1.47	-	-	-	-	63.30	-	Islam et al. (2017)
Jute mill industry	Banglade sh	6.52	44.10	1.79	3.71	0.87	-	-	-	-	38.90	-	Islam et al. (2017)
Metal processing industry	Banglade sh	6.93	36.90	2.61	2.96	1.66	-	-	-	-	37.10	-	Islam et al. (2017)
Battery manufacturing industry	Banglade sh	5.98	50.40	3.55	1.94	1.05	-	-	-	-	33.30	-	Islam et al. (2017)
TFT-LCD industry	Taiwan	6.98	-	-	2.50	2.50	41.09	-	-	0.13	-	-	Chen et al. (2015)
Agro- industrial waste water	Italy	10.2 0	68.10	-	1.54	-	4.32	0.84	-	-	-	2.00	Grigatti et al. (2017)

sludge													
Industrial	Pakistan	7.33	19.00	0.63	0.12	-	-	-	0.09	-	-	-	Kazi et al. (2005)
sewage sludge													
from													
industrial													
catchment													
areas													
Petrochemical	China	6.60	38.20	1.90	1.06	0.73	-	-	-	-	52.20	-	Wang et al. (2005)
, brew, and													
paper													
industries													
Electroplating	China	6.60	22.10	-	0.21	0.13	1190	-	-	0.17	-	-	Wu et al. (2020)
industry													

Note: The hyphen (-) indicates the absence of data for the particular parameter.

Table 2. Heavy metal contents of sludge derived from various industrial activities.

Source of industrial sludge	Country	Analytical method	Fe	Al	Zn	Pt	Ni	Ti	Cu	Mn	Cr	Hg	Pb	Cd	Mo	Co	In	As	Reference
mg kg ⁻¹ or %																			
Paper industry	Spain	ICP-AES	1615 ^T	14400 ^T 19500 ^E	231 ^E	1.90 ^T	6 ^E	33 ^E	33	34 ^E	17 ^E	0.20 ^E	37 ^E	-	-	-	-	-	Cusidó et al. (2015)
Textile industry	China	ICP-OES	5131.94	63958.33	154.51	-	233.80	-	40	430.32	434.498	-	19.38	0.15	-	-	-	-	Wang et al. (2019)
Battery industry	China	ICP-MS	56750	-	1700	-	22225	-	23	-	-	-	-	177300	-	-	-	-	Wu et al. (2015a)
Battery industry	China	AAS	-	-	1700	-	18135	-	23	-	31.507	-	0.00	112500	-	-	-	-	(Wu et al., 2015b)
TFT-LCD industry	Taiwan	ICP-AES	-	3180	-	-	-	-	-	-	-	-	-	-	61100	-	40330	-	Chen et al. (2015)
Agro-industrial waste	Italy	ICP-OES	26766	6444	-	-	-	-	-	-	-	-	-	-	-	-	-	-	Grigatti et al. (2017)

water																			
sludge																			
Granitoid	Spain	ICP-AES	-	-	57	-	64	-	78	590	92	<0.0	20	<0.01	25	10	-	7.10	Lieberman et al. (2019)
waste												01							
sludge																			
from																			
stations:																			
Station 1																			
Station 2			-	-	56	-	100	-	11	715	171	<0.0	23	<0.01	36	6.9	-	9.00	
									5			01				0			
Station 3			-	-	62	-	190	-	18	1143	320	<0.0	18	<0.01	59	17	-	10.0	
									8			01						0	
Station 4			-	-	58	-	17	-	43	337	9	<0.0	21	<0.01	1.48	84	-	4.00	
												01							
Industrial	India	-	414.5	-	152.7	-	52.30	-	92.	-	74.20	-	11.20	4.70	-	-	-	-	Sumalatha et al. (2019a)
wastes			0		0				30										
(KIADB																			
industrial																			
area,																			
India)																			
Tannery	Malaysi	AAS	1062	-	200	-	-	-	80	70	500	-	10	8	-	-	-	-	Ahmed et al. (2007)
industry	a																		

Metal plating industry	Turkey	AAS	45.46	-	132.7	-	25.94	-	83.	-	27.17	-	-	-	-	-	-	Balkan & Kocasoy (2004)
					4				62									
Steel industry	India	ICP-AES	3706	20200	490	-	300	-	60	1180	280	-	-	-	-	-	-	Singh & Garg (2020)
			00															
Stainless steel industry	China	ICP-OES	1034	5060	5060	-	2020	-	12	4360	19140	-	-	-	-	-	-	Su et al. (2019)
			00				0		48									
Electroplating industry	China	ICP-OES	4350	-	6660	-	8850	-	14	-	15400	-	-	-	-	-	-	Wu et al. (2020)
			0		0				80		0							
									0									
Electroplating, pickling-rolling and textiles industries	India	AAS	1197	-	410	-	1807	-	12	1985	16686	-	440	30.16	-	-	-	Bose & Bhattacharyya (2008)
			40						90	0								
Petrochemical, brew and paper industries (mean	China	ICP-MS	-	-	1754.	-	67.70	-	52	-	317.23	-	28.40	17.00	-	-	-	Wang et al. (2005)
					80				3.9		0							
									0									

values)																			
Food processing industry	India	AAS	642.7	-	2.07 ^D	-	1.60 ^D	-	0.9	27.30	4.27 ^D	-	5.25 ^D	0.31 ^D	-	6.0	-	-	Mahapatra et al. (2013)
			0 ^D		0.52		0.06		3 ^D	^D	0.34 ^W		0.59	0.04 ^W		9 ^D			
			19.10		w		w		0.1	2.54			w			0.2			
			w						5 ^W	w						1 ^W			
Arsenic sulfide sludge from a lead and zinc smelter	China	ICP-OES	0.50	-	0.86	-	-	-	0.0	-	0.05%	-	0.14	3.73%	-	-	-	46.9	Xu et al. (2020)
			%		%				1				%					0%	
									%										
Goethite sludge from zinc refining process	Belgium	XRF and ICP-OES (only for Mo)	24.1	0.9%	5.9%	-	-	-	0.4	0.4%	475	-	1.8%	-	82	-	-	-	Atia & Spooren (2020)
			%						%										
Electroplating industry	China	ICP-OES	16.12	-	21.03	-	0.80	-	3.1	-	1.79		-	-	-	-	-	-	Zhang et al. (2020)
			%		%		%		9		%								
									%										
Industrial filter	Finland	XRF	3.25	-	-	-	3.45	-	0.1	-	-	21.7	24.6	-	-	-	-	-	Piekkari et al. (2020)
			%				%		53			%	%						

sludge %
originated
by sulfuric
acid
production
from Ni
and Cu
smelter

^T – Total ^D – Dry sludge ^E – Extractable ^W – Wet sludge

Note: The hyphen (-) indicates the absence of data for the concentration of particular heavy metal.

The metal concentrations are indicated either with mg kg⁻¹ or %. The data represented with percentage (%) is indicated after the value and the unit for all other values is mg kg⁻¹.

Table 3. Different bioleaching approaches for recovering heavy metals from industrial sludges.

Sludge type	Metal content / (%) or (mg kg ⁻¹)	Bioleaching microorganisms	Condition applied					Leaching yield	Reference
			Pulp density	pH	Leaching period	Temperature	Other conditions		
Cu converter sludge	Zn: 1.7% Fe: 60.2%	<i>Acidithiobacillus spp.</i> and <i>Leptospirillum spp.</i>	1.0% w/v	1.5	79 days	-	-	Zn: 36%	Vestola et al. (2010)
Pyritic sludge	Zn: 0.13% Fe: 44.0%	<i>Acidithiobacillus ferrooxidans</i> , <i>Acidithiobacillus caldus</i> , and <i>Sulfolobus metallicus</i>	2.5% w/v	2.0	10 days	65 °C	-	Zn: 90%	Hita et al. (2008)
Electroplating sludge	Ni: 6.0%	<i>Acidithiobacillus thiooxidans</i>	10.0% w/v	1.0	9 days	30 °C	8.0 g L ⁻¹ of sulfur	Ni: 100%	Yang et al. (2016)

								content			
Electroplating sludge	Cr: 1.79% Cu: 3.19% Zn: 21.03% Ni: 0.80% Fe: 16.12%	Mixed culture of <i>Acidithiobacillus</i> <i>ferrooxidans</i> (78.0%), <i>Acidiphilium</i> sp. (15.5%), and small percentage of <i>At.</i> <i>ferrivorans</i> and <i>At.</i> <i>thiooxidans</i>	15% w/v	2.0	1 day	30 °C	9.0 g L ⁻¹	Cu: >95.6% Zn: >95.6% Fe ²⁺ Ni: >95.6% Cr: 90.3%	Zhang et al. (2020)		
Electroplating sludge	Cu: 1.48% Zn: 6.66% Cr: 15.50%	<i>Acidithiobacillus</i> <i>ferrooxidans</i>	8% w/v	2.2	9 h	30 °C	9.0 g L ⁻¹	Cu: 75.8% Zn: 84.4% Fe ²⁺ Cr: 80.9% Ni: 65.8%	Wu et al. (2020)		

	Ni: 0.85%						voltage		
	Fe: 4.35%						of 0.2		
							V		
Electroplating sludge	Zn: 4.030%	<i>Leptospirillum</i> <i>ferriphilum</i> CS13,	-	1.5	5 h	45 °C	bacteria 1 liquid ratio: 40%, liquid/s olid ratio: 4 L kg ⁻¹ , stirring speed: 120 rpm	Zn, Cu, and Ni: 95–100% Cr: up to 99%	Zhou et al. (2019b)
	Cu: 3.192%	<i>Acidithiobacillus caldus</i> <i>S2, and Sulfobacillus</i>							
	Ni: 0.941%	<i>acidophilus</i> CS5 mixed culture							
	Cr: 2.278%								
	Pb: 0.001%								
	As: 0.0014%								
	Cd: 0.0007%								

Hg:
0.0001%

Tannery sludge	Cr: 17190	A	sulfur-oxidizing	-	7.9	6 days	28 °C	5 g L ⁻¹	Cr: 98%	Fang et al.
	mg kg ⁻¹	mixed culture						of		(2007)
								element		
								al		
								sulfur		
								content		

Note: The hyphen (-) indicates the absence of data for the particular parameter.

Table 4. The efficiency of various acid solvents and extraction conditions for leaching metals from industrial sludges and sediments.

Waste type	Leaching	Extraction conditions					Metal	Amount recovered	Reference
	solvent	S/L ratio	Stirring speed	Leaching period	Temperature	Other conditions			
Printed circuit board industrial sludge	0.84 M H ₂ SO ₄	10 g L ⁻¹	200 rpm	80 min	60 °C	-	Cu	96.8%	Thawornchaisit et al. (2019)
Sludge generated from industrial area	0.1 N HCl	-	-	-	-	Column leaching with a flow of 0.2 x 10 ⁻⁵ cm s ⁻¹ and a pore volume of 56.55 cm ³	Cd Cu Zn Ni Pb Fe Cr	82.9% 82.5% 79.6% 59.3% 55.5% 49.3% 42.7%	Sumalatha et al. (2019b)

Galvanic sludge	1.0 M H ₂ SO ₄	-	-	30 min	-	10% w/v leaching solution	Cu	250000 mg kg ⁻¹ 1	Huyen et al. (2016)
Electroplating sludge	H ₂ SO ₄	-	500 rpm	2 h	85 °C	25 ml of concentrated H ₂ SO ₄ was slowly added to 2 kg L ⁻¹ feed solution at 500 rpm for 20 min	Cu Ni Fe Cr Zn	6.01 g L ⁻¹ 4.61 g L ⁻¹ 2.42 g L ⁻¹ 4.83 g L ⁻¹ 3.31 g L ⁻¹	Yan et al. (2019)
Tannery sludge ash	1.0 N H ₂ SO ₄	1:25	-	60 min	25 °C	-	Cr	93%	Kokkinos et al. (2019)
Low-grade	0.24 mL g ⁻¹	1:2	400 rpm	2 h	30 °C	-	Cu	97.47%	Chen et al.

copper sulfide tailings	¹ H ₂ SO ₄						Zn	26.56%	(2014)
							Mn	54.45%	
							Fe	15.47%	
Zinc slag generated from hot dip galvanizing plant	20 vol.% H ₂ SO ₄	150-200 g L ⁻¹	-	-	-	-	Zn	95–97%	Rudnik (2019)
Brass slag	30% H ₂ SO ₄	-	150 rpm	10 min	35 °C	-	Zn	95%	Ahmed et al.
							Cu	99%	(2016)

Note: The hyphen (-) indicates the absence of data for the particular parameter.

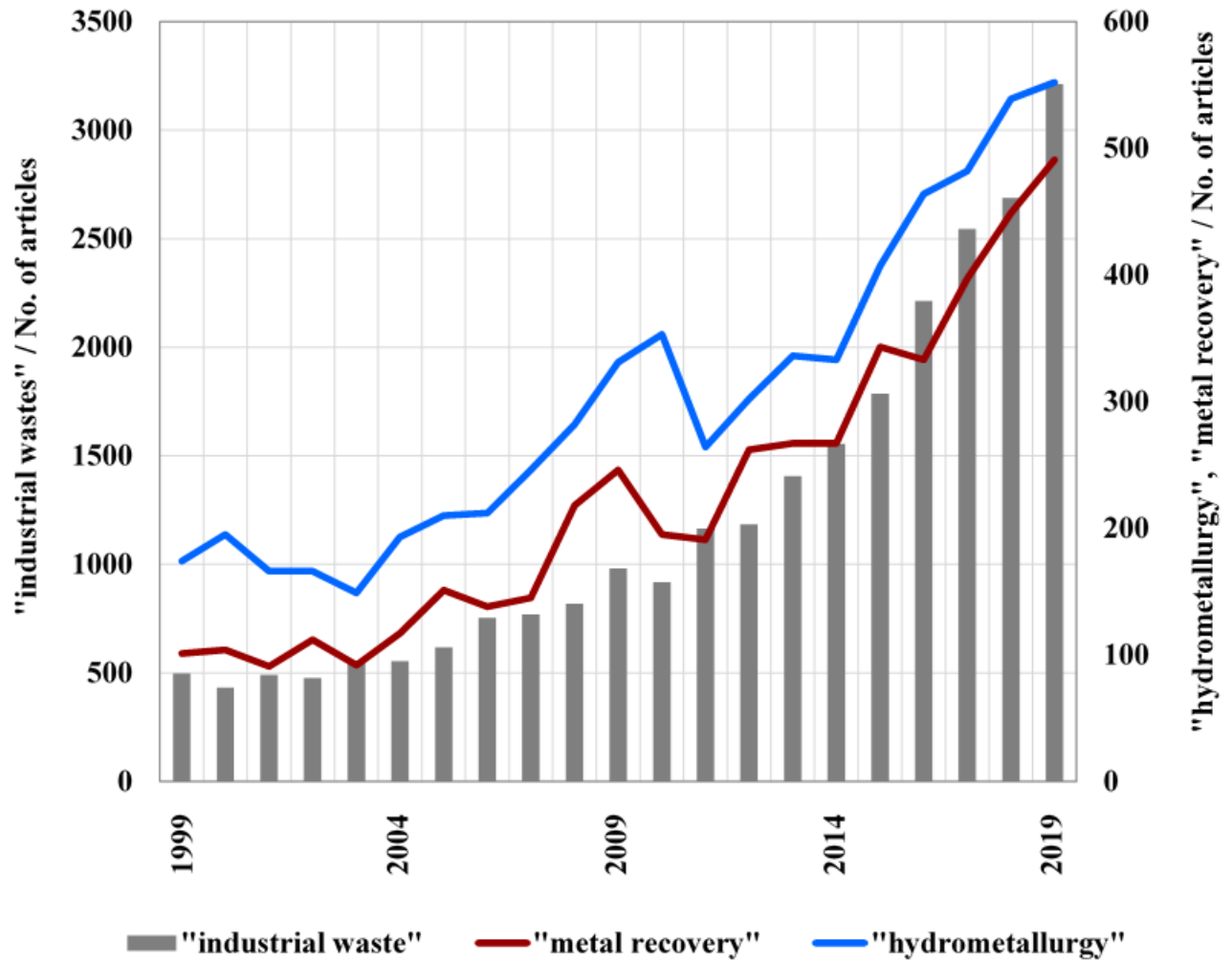


Figure 1. The number of research articles that appeared in Science Direct for keywords: “industrial wastes” and “metal recovery” (Date accessed on 28.02.2020).

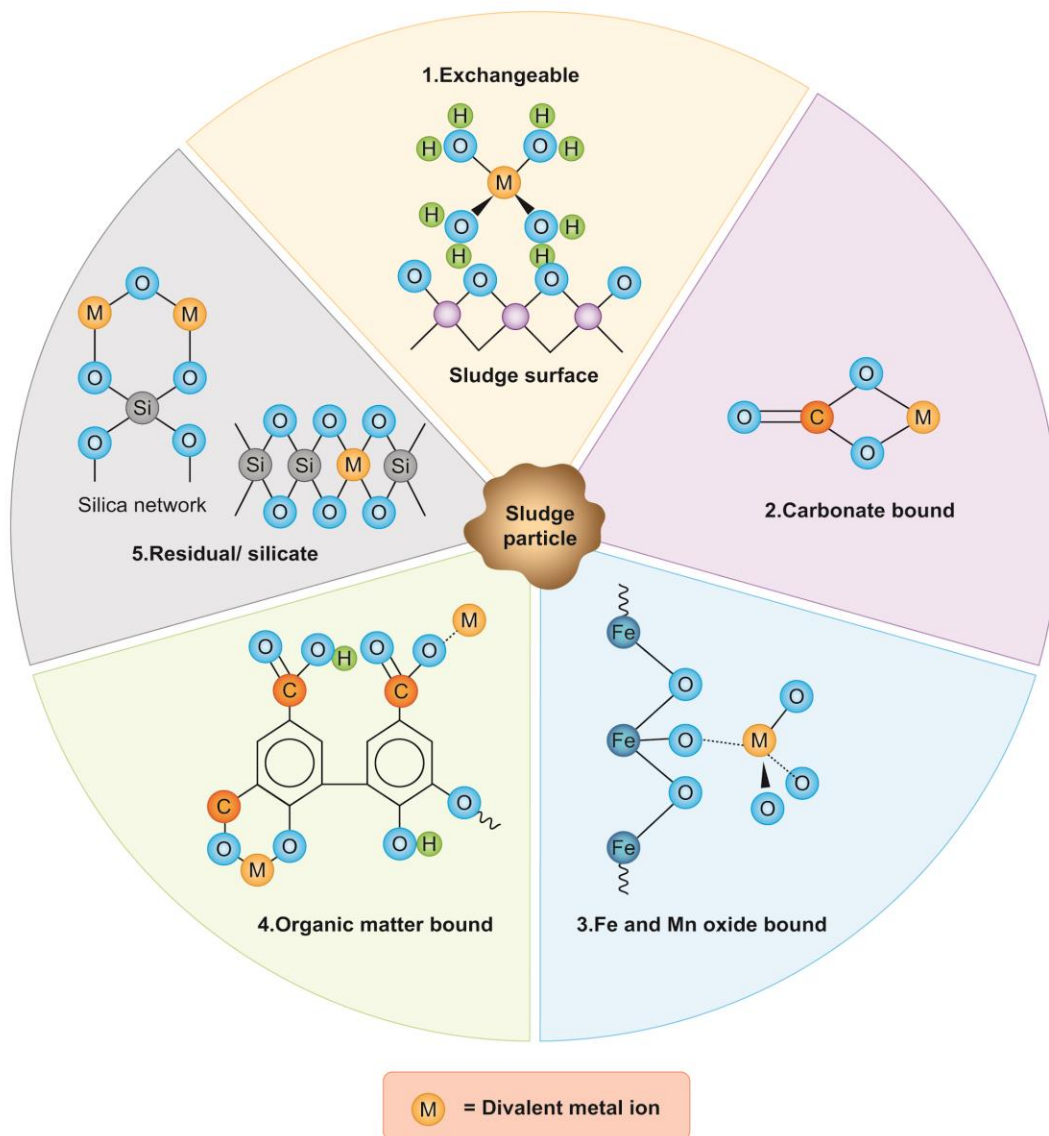


Figure 2. Speciation of metals in different phases of sludge particles (classification from Tessier et al. (1979)).

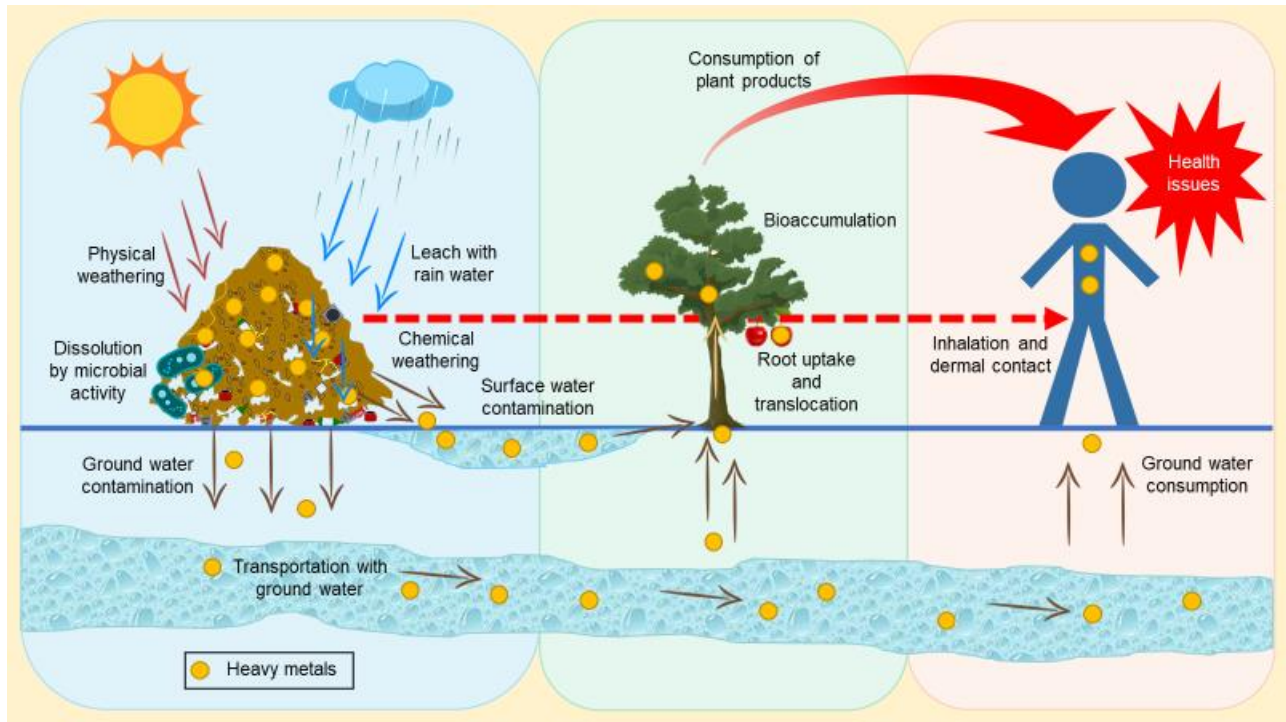


Figure 3. Release of heavy metals and the environmental threats that result from land disposal of industrial sludge.

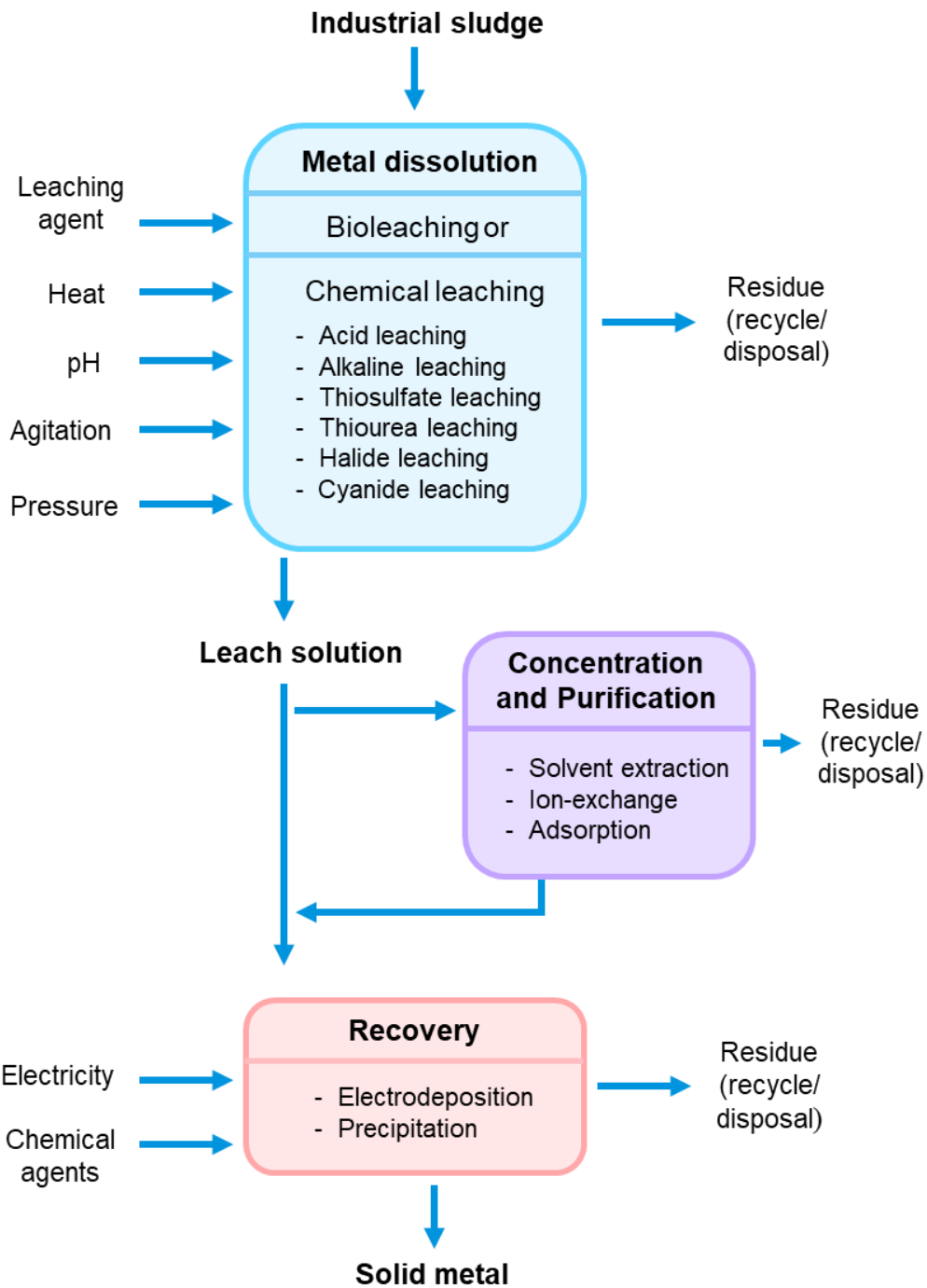


Figure 4. Basic process flow of hydrometallurgical metal recovery (adapted from Gupta (2006)).

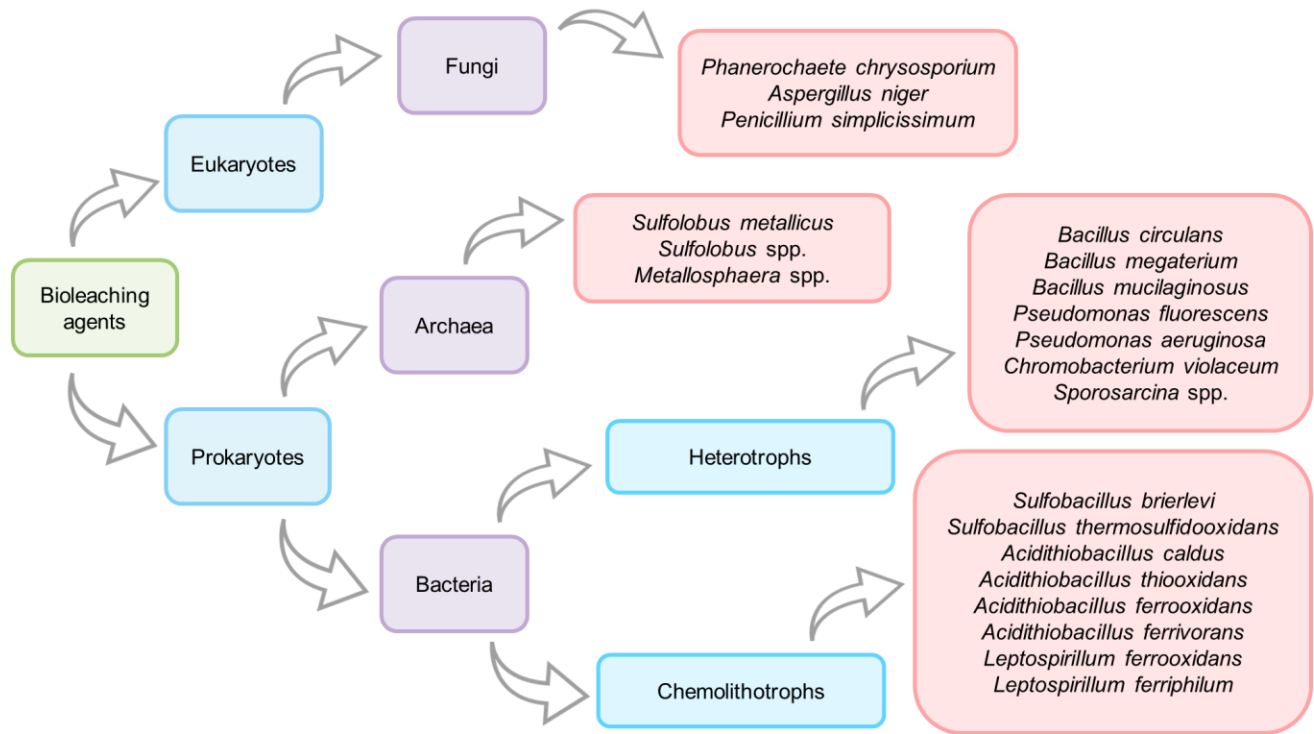


Figure 5. Types of microorganisms that are used in bioleaching for recovering metals from industrial wastes (adapted from Sethurajan et al. (2018)).

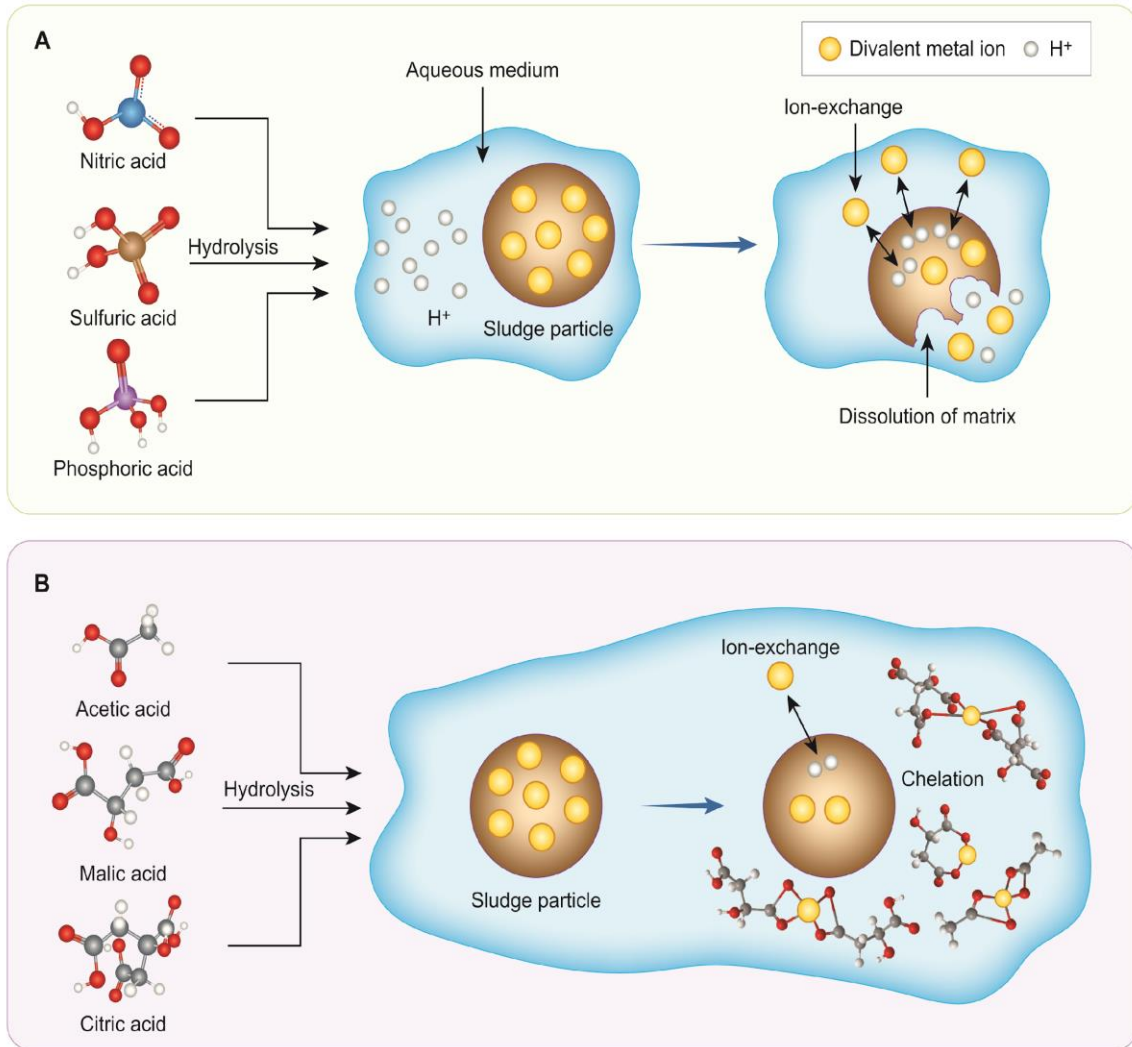


Figure 6. Release mechanisms of heavy metals with A) inorganic and B) organic acids.

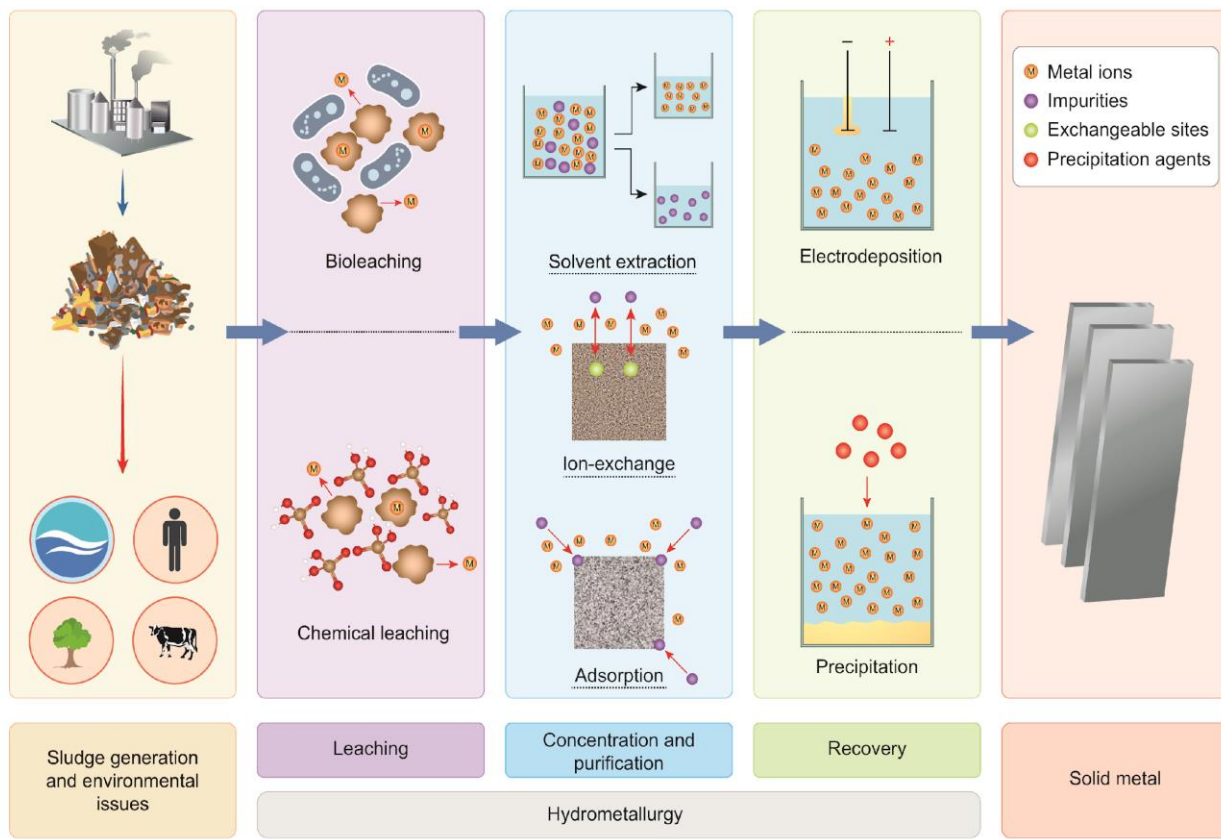


Figure: Graphical abstract