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Recovering metal(loids) and rare earth elements from closed landfill sites without excavation: leachate recirculation opportunities and challenges

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Abstract: Metal(loids) and Rare Earth Elements (REE) ('metals') are naturally scarce and economically high-value. They are used in a wide range of products, with demand continuing to grow. Metal-bearing wastes are a secondary source of raw material that can meet this demand by providing a previously unconsidered low impact supply source. Total annual leachate production is 1,056,716 m^3. Therefore, landfill leachate emerges as a significant potential resource as it contains high concentrations of metalloids, metal ions and REE. However, realising a profitable return on investment for leachate processing is a challenge due to relatively low recovery rates of approximately 0.02% of total heavy metals in a landfill being leached out in 30 years. Variation within the multi-element value and the effect of other chemicals (organic and inorganic) in these complex mixtures. There is a need to better understand the mechanisms and potential applicability of extraction methods for optimising metals recovery from leachate. This paper addresses this need by providing a systematic review of the critical factors and environmental conditions that influence the behaviour of metals within the landfilled waste. The paper provides a synthesis of how the factors and conditions may affect leachate recirculation efficiency for recovery in the context of a range of opportunities and challenges facing circular economy practitioners. To approach feasibility metal recovery economically from landfill leachate without energy-intensive and environmentally destructive, future research actions need to be initiated in lab-based and later on semi-pilot to pilot studies, which the review can help achieve the challenges.

Keywords: circular economy, non-intrusive investigation, organic compounds, mobility, metal recovery

Contents

1 Introduction

Metals, metalloids and Rare Earth Elements (REE), collectively termed ‘metals’, are finite natural resources with increasing demand. It is vital to find alternative sources to ensure supplies of these metals, especially for new technologies such as electric vehicles, renewable energy generation and battery storage (Jowitt et al., 2020). Several recent studies reported that a worthwhile amount of valuable secondary raw materials is available within closed landfill sites (Gutiérrez-Gutiérrez et al., 2015; Dino et al., 2017; Krook et al., 2018; Särkkä et al., 2018; Esguerra et al., 2019; Parrodi et al., 2019; Wagland et al., 2019). There are around 21,000 closed landfill sites across England and Wales (DEFRA, 2021; ENDS Report, 2021) and somewhere between 150,000 and 500,000 landfill sites within Europe, with an estimated 90% of them being non-sanitary (Jones et al., 2018).

Landfill technology is very different according to country. In many developing countries, open dumps still are in operation. In many developing countries, uncontrolled open dumpsites are more widely employed than controlled and engineered landfills as it requires minimum land and is an easy way of disposing of refuse. This means that no emission control occurs, and the waste is not compacted (Stegmann, 2005) thus posinga risk to human health and the environment. The risks posed by the leachate can be amplified by the lack of leachate containment systems, leading to high concentrations of organic and inorganic pollutants in the environment (Maiti et al., 2016; Vaccari et al., 2019). On the other hand, landfills are highly engineered facilities designed to minimise the adverse impact of waste on the surrounding environment. Even though are well-engineered many landfills still release methane and carbon dioxide from decomposing organic waste, affecting climate change. If leachate breaches the landfill lining, water contamination can occur.

Nevertheless, landfills are still the most common waste disposal method (Krook et al., 2018). A landfill site with good, engineered facilities should result in minimal negative environmental, social, and economic impact for waste compared to other waste management options. Also, waste from landfill sites can be used in several ways for energy generation and recycling resource. Closed landfills represent a significant opportunity across Europe and the UK to recover value from waste materials as they provide a previously unconsidered localised long-term storage deposit of secondary raw materials similar to traditional metal mineral resources. Enhanced Landfill Mining (ELFM) address the combined and integrated valorisation of distinct landfilled urban waste streams as both materials (Waste-to-Materials, WtM) and energy (Waste-to-Energy, WtE) while meeting the most stringent ecological and social criteria (Jones et al., 2013). ELFM provides an opportunity for combined resource recovery and reclamation of land while mitigating future environmental liabilities and remediation costs through excavation innovative transformation technologies (Jones et al., 2013; Esguerra et al., 2021; Vollprecht et al., 2021). However, existing technologies and good practices are unable to demonstrate the economic viability of such schemes. This is partly because mining processes for recovering material result in high capital costs. There are also important uncertainties about the actual abundance and concentration of suitable waste materials in landfill environments, which need to be known early on in the lifecycle of a metal's recovery project.

Two essential by-products of waste disposal by landfills are leachate and landfill gas. The former is formed when rainwater infiltrates and percolates through the degrading waste, the latter by microbial degradation of biodegradable waste materials under anaerobic conditions (Chu, 2008). Some are resistant to environmental degradation and have existed for a long time, referred to a persistent organic pollutants (POPs) (Alharbi et al., 2018). It is estimated that over 100,000 types of chemicals and their transformation products may present in the landfilled waste and are leached out by rainwater, forming landfill leachate (Öman and Junestedt, 2008). A wide range of POPs are present in landfill leachate (Paxeus, 2000). POPs associated with high production volume industrial chemicals directly or indirectly affect the environment and health even at low concentrations (Wojciechowska, 2013). Polluted environments increase the risk of exposure to contaminants, disease vectors, and other agents that may induce illnesses for humans (Adeola, 2004). Effective treatment methods are required as leachate contains trace chemicals, contaminating groundwater, surface water and soil, potentially polluting the environment and harming human health (Brennan et al., 2016). There are methods to remove organic pollutants from wastewater and water. For example, peroxymonosulphate can generate hydroxyl radicals and sulphate and the Z-scheme photocatalysts such as TiO_2 with high potential for degradation of organic pollutants (Ghanbari et al., 2020; Hassani et al., 2021). The electro-activated oxidant system also effectively removed organic pollutants (Ghanbari et al., 2021). However, ambitions for the landfill management should go beyond protecting human health and the environment, with conservation of energy and recovery of natural resources high on the agenda. Landfill leachate comprises recoverable metals, organics, phosphorus, ammonia, and water (Iskander et al., 2017; Kurinawan et al., 2021). The presence of recoverable metals means that landfill leachate can be of great importance as an alternative to conventional mineral exploration as the sediment of the leachate showed presence of REEs content was more than twice the content in landfilled waste (Gutiérrez-Gutiérrez et al., 2015), but also the presence of recoverable metals in landfill leachate can negate the need for full-scale landfill mining.

There are four objectives of this review paper: i) to give an overview of the properties and metals content in landfill leachate to gain insight into the opportunities for metal recovery from leachate; ii) to explore the knowledge on various factors affecting metals solubility; iii) to evaluate the efficiency of recirculation for increasing metal recovery rates and; iv) to discuss the opportunities for metal recovery from leachate, analyse the challenges associated with the recovery, and present the perspectives for future research and technology development to maximise the benefits of metals recovery from closed landfill leachate.

2 LANDFILL LEACHATE PROPERTIES AND METALS CONTENT

Leachate can be formed as a result of chemical and biochemical processes within the landfill. There is nonuniform and intermittent percolation of moisture through the solid waste in the landfill due to leachate generation (Hughes et al., 2013; Edokpayi et al., 2018). Several factors influence leachate composition, such as the age of landfill, depth of the waste in the landfill, location of the site, and weather condition of the landfill site; another critical factor affecting leachate composition is the composition of the waste deposited in the landfill (Jang and Townsend, 2003; Kalčíková et al., 2011; Moody et al., 2017). Generally, the waste composition is categorised as organic (food and garden waste), paper, plastic, glass, metals, etc. Table 1 shows the composition of global waste.

Table 1. Waste composition range

Component	Range (%)	References
Organic waste	15-58	Tapia, 2009; European Commission, 2016; Abdel-Shafy and Mansour, 2018; Kaza et al., 2018
Paper and cardboard	16-27	
Glass	2-4.5	
Plastic	9-20	
Wood	3-7	
Metal	3-9.2	
Textile	3-9	
Rest	3-18	

A wide variety of metal contents is collected into the leachate as it drains through the pile of waste in the landfill (Eggen et al., 2010; Edokpayi et al., 2018). Leachate is also rich in ammonia, which ranges from 50 to 11,000 mg/L and inorganic components such as iron, chlorine, sulphate, and metals (Öman and Junestedt, 2008; Luo et al., 2020; Zico et al., 2021). Previous studies concluded that ammonia is the principal pollutant in leachate, excluding organics (Kulikowska and Klimiuk, 2008). Ammonia nitrogen in the leachate accumulates due to there is no degradation pathway for ammonia. Therefore, a high concentration of ammonia-nitrogen in leachate occurs throughout the lifetime of a landfill (Berge et al., 2006; Couto et al., 2017), which means early leachate has a relatively low concentration of ammonia-nitrogen content. As landfill leachate ages, the concentration of ammonia increases, whilst the biodegradable fraction declines due to the stabilisation process (Abood et al., 2013). Ammonia nitrogen is rich in older landfill leachate due to hydrolysis and fermentation of the nitrogenous fractions of biodegradable substrates (Carley and

Mavinic, 1991). The ammonia nitrogen concentration remains for at least 50 years in the range of between 500 and 1500 mg/L (Kulikowska and Klimiuk, 2008). Nitrogen compounds are common in leachate. The total nitrogen content of leachate is in the range of 2.6-945 mg/L (Mukherjee et al., 2015). It affects environmental such as eutrophication, acidification of water, toxicity to aquatic animals, and increased algal blooms (Camargo and Alonso, 2006). Aside from ammonia-nitrogen, total phosphorus is considered for water quality as it is a major limiting nutrient in water ecosystems for algal growth, which are harmful to most aquatic organisms (Said et al., 2004). Total nitrogen and total phosphorus measures indicated that total nitrogen levels in the waste decreased over time, whereas phosphorus levels remained steady throughout (Zhao et al., 2007). Inorganic compounds may consist of potentially harmful elements such as Pb, Hg, and As in pure form or combined with other elements (Jan et al., 2015). As leachate contains a wide range of metals and nutrients, it can be considered a great potential for metal and nutrients recovery. However, there are also challenges associated with the method to meet the need for sustainable leachate management that maximises valuable metal recovery. The key challenge on metal recovery from leachate is the low concentration of metals which is often affected by landfill age and type (Table 2).

Table 2. The concentration range of chemical constituents of landfill leachate determined from available literature

Parameter	Concentration range (mg/L)	Parameter	Concentration range (mg/L)	References
Alkalinity (as $CaCO_3$)	0-20,850	Nitrogen (Ammonia)	0-1,250	Kjeldsen et al., 2002
Aluminium	0-2	Nitrogen (Nitrate)	0-9.8	Kjeldsen et al., 2002
Antimony	0-3.19	Nitrogen (Nitrite)	0-1.46	Christensen et al., 2001
Arsenic	0-0.04	Nitrogen (Organic)	0-1,000	Christensen et al., 2001
Barium	0-2	Nitrogen (Total Kjeldahl)	1-100	Christensen et al., 2001
Beryllium	0-0.36	Nickel	0-7.5	Christensen et al., 2001
BOD_5	0-4,000	Phenol	0.17-6.6	Akinbile et al., 2012
Boron	0.5-10	Phosphorus (Total)	0-234	Kjeldsen et al., 2002
Cadmium	0-0.01	Phosphate	1-10	Christensen et al., 2001
Calcium	100-1,000	pH	4.5-9	Adamcová et al., 2016
Chloride	20-2,500	Potassium	0.16-3,370	Akinbile et al., 2012
Chromium	0-0.05	Selenium	0-1.85	Adamcová et al., 2016
Cobalt	0-7.58	Silicon	0-12	Kjeldsen et al., 2002
COD	150-6,000	Silver	0-1.96	Christensen et al., 2001
Conductivity (µmho/cm)	480-72,500	Sodium	0-8,000	Kjeldsen et al., 2002
Copper	0-9.9	Thallium	0-0.32	Adamcová et al., 2016
Cyanide	0-6	Tin	0-0.16	Adamcová et al., 2016
Fluoride	0.1-1.3	TDS	0-42,300	Akinbile et al., 2012
Hardness (as $CaCO_3$)	400-2,000	Titanium	0-1.5	Christensen et al., 2001
Iron	0-5,500	TSS	140,900	Kjeldsen et al., 2002
Lead	0-5	TOC	335,000	Adamcová et al., 2016
Magnesium	16.5-15,600	TVA (as Acetic acid)	0-19,000	Akinbile et al., 2012
Manganese	0.05-1,400	Turbidity	40-500	Adamcová et al., 2016
Mercury	0-3	Sulphate	0-300	Adamcová et al., 2016
Organic halides	0.32-3.5	Zinc	0-1,000	Christensen et al., 2001
Benzene	0.1-0.6	Phenols	0-4	Christensen et al., 2001
Ethylbenzene	0-4.9	Toluene	0-3.2	Akinbile et al., 2012

Note: The grey shaded cell indicates metals and metalloids

The leachate produced in young landfills (< 5 years old) contains a substantial number of organic compounds derived from biodegradable organic water materials, which undergoes rapid anaerobic fermentation within confined landfills. As a result, volatile fatty acids (VFAs) are produced, e.g. acetic, propionic, iso-butyric, n-butyric, iso-valeric, and n-valeric acid. It is very well known that organic acids, such as VFAs, may play an essential role in the mobilisation of metals through either the formation of soluble ligand: metal complexes or a decrease of pH (Molaey et al., 2021). Thus, VFAs are considered valuable substrates for metal dissolution, increasing the release of metals in landfill environments. Young leachate is characteristic of its high content of biodegradable organic matter. BOD (Biochemical oxygen demand), COD (Chemical oxygen demand), and BOD/COD ratio act as indicators of microbial activities and organic pollution. BOD/COD describes the biodegradability level of materials by which organic matter containing leachate is readily broken down in the environment (Samudro and Mangkoedihardjo, 2010). Therefore, young leachate shows a high BOD/COD indicator.

As landfill age increases, the BOD/COD ratio in leachate decreases (Table 3). This is due to the decomposition of the majority of biodegradable compounds and small quantity changes of less degradable organic matter at the same time that acidic conditions begin neutralising (Talalaj, 2015). As a consequence, the higher pH condition results in decreasing metal release by complexation and precipitation (Zhang et al., 2018). Older leachate from the methanogenic phase is partially characterised by the lower concentration of VFAs. As the content of VFAs and other readily biodegradable organic compounds in the leachate decreases, the organic matter (OM) in the leachate becomes dominated by refractory compounds, such as humic acid (HA) and fulvic acid (FA), which are known to bind metals to their hydroxyl and carboxyl groups, and either mobilise metals or delay their release (Leung and Kimaro, 1997; Bozkurt et al., 2000; Kochany and Smith, 2001; Klavinsa et al., 2006; Gutiérrez-Gutiérrez et al., 2015). The humic substances (HS) give a dark colour with increasing pH due to the dissociation of protons (Stevenson, 1994). The decrease in VFAs increases pH; consequently, metals have a relatively low concentration in older landfill leachate as the solubility of metals is decreased with increasing pH.

Table 3. Selected characteristics of leachate according to landfill age

Parameter	Young	Intermediate	Old	References
Age (years)	<5	5-10	>10	Renou et al., 2008
pH	<6.5	6.5-7.5	>7.5	Bhalla et al., 2013
COD (mg/L)	>10,000	4,000-10,000	<4,000	Bhalla et al., 2013
TOC/COD	<0.3	0.3-0.5	<0.5	Abbas et al., 2009; Zhou et al., 2010
BOD_5/COD	>0.3	0.1-0.3	<0.1	Bhalla et al., 2013
Organic compounds	80% VFA	5-30% VFA + humic and fulvic acids	Humic and fulvic acids	Bhalla et al., 2013
Heavy metals (mg/l)	Low medium >2	Low <2	Very low <2	Renou et al., 2008
Molecular size distribution	Over a broad range-high fraction of low molecular weight organics	N/A	Over a narrow range-high fraction of high molecular weight organics	Abbas et al., 2009; Zhou et al., 2010
Biodegradability	Important	Medium	Low	Bhalla et al., 2013

3 PHYSICO-CHEMICAL FACTORS AFFECTING METALS SOLUBILITY

Several factors affect metals solubility within solid waste deposits (Fig 1). Important processes include abiotic redox processes, dissolution/precipitation of minerals, sorption, ion exchange, organic matter biodegradation, and complexation. The resulting matrix redox changes strongly influence both the inorganic and organic biogeochemistry of the landfill and therefore influencing the behaviour and fate of metals within landfills (Christensen et al., 2001). Gaining insights into the geochemistry of landfill is therefore needed to better understand the solubility of metals and predict metals recovery.

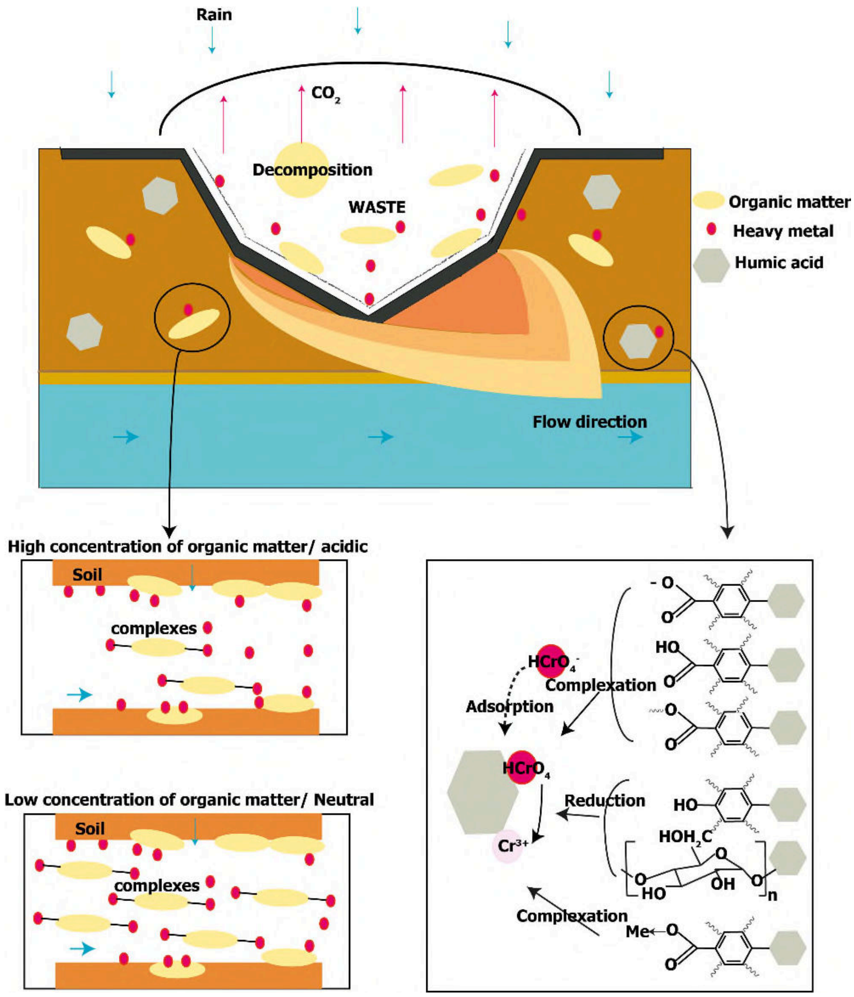

Figure 1. Processes occurring in landfills which affect metal solubility

3.1 Weathering and natural attenuation

The dissolution of metals can occur during natural events, such as weathering and natural attenuation. During weathering processes, a broad range of physical and chemical reactions such as hydrolysis, precipitation, pH neutralisation, oxidation/reduction of metals, sorption, and complexation will change the overall characteristics of metals (Chimenos et al., 2003; Polettini et al., 2004; Saffarzadeh et al., 2011; Takahashi and Shimaoka, 2012). Natural attenuation can be defined as a process by which the concentration of leachates is reduced to an acceptable level by natural processes. It can both mobilise and immobilise metals (Beaven et al., 2013). Based on the definition, *in-situ* natural attenuation mechanisms are identified as physical (diffusion, sorption, dispersion, dilution, and volatilisation), chemical (precipitation, adsorption, ion exchange, redox reaction) and biological (biodegradation) processes. For this reason, it is desirable to be able to predict how the metals in the landfill environment will behave over time when exposed to the weathering effects of infiltrating rainwater and the atmosphere. The effect of weathering on metals solubility is likely to be significant as pH is a dominant parameter in metals solubility and complexation (Stumm and Morgan, 1981). The redissolution of their respective hydroxide mainly causes the release of metals as pH is controlled by the solubility of $Ca(OH)_2$. Therefore, weathering reactions leads to a decrease in pH (Chimenos et al., 2003).

3.1.1 Oxidation

Several studies have been shown the formation of Fe/Al-(hydrate) oxides and calcite by weathering. It indicates that metals release may be controlled by sorption processes caused by weathering (Zevenbergen and Comans, 1994; Meima et al., 1997a; Meima and Comans, 1999; Saffarzadeh et al., 2011; Takahashi and Shimaoka, 2012). Saffarzadeh e*t al.* (2011) proposed the following order based on their direct metal uptake capacity: Fe-hydrate > Al-hydrate > calcite. Calcite is not adequate for direct metals sorption; however, they play a crucial role in buffering the system, pH neutralisation; consequently, it minimises metal leaching. Thus, weathering is expected to result in a reduced metal solubility in the long term (Meima and Comans, 1999).

3.1.2 Sorption and precipitation

Temporal studies of metal mobility in soils show that mobility decreases over time, suggesting that a high proportion of metals within Municipal solid waste (MSW) which consists of everyday items we use and then throw away, are insoluble (Peters and Shem, 1993; Aucott, 2006). The reasons for the reduced mobility of metals in soil include sorption on soil particles and particularly to HS, precipitation under anaerobic conditions, adsorption, and chelation with inorganic and organic ligands in landfills (Bozkurt et al., 2000). Christensen *et al.* (2001) reported that metals in landfills do not constitute a significant pollution problem due to strong attenuation by sorption and precipitation (Fig 2).

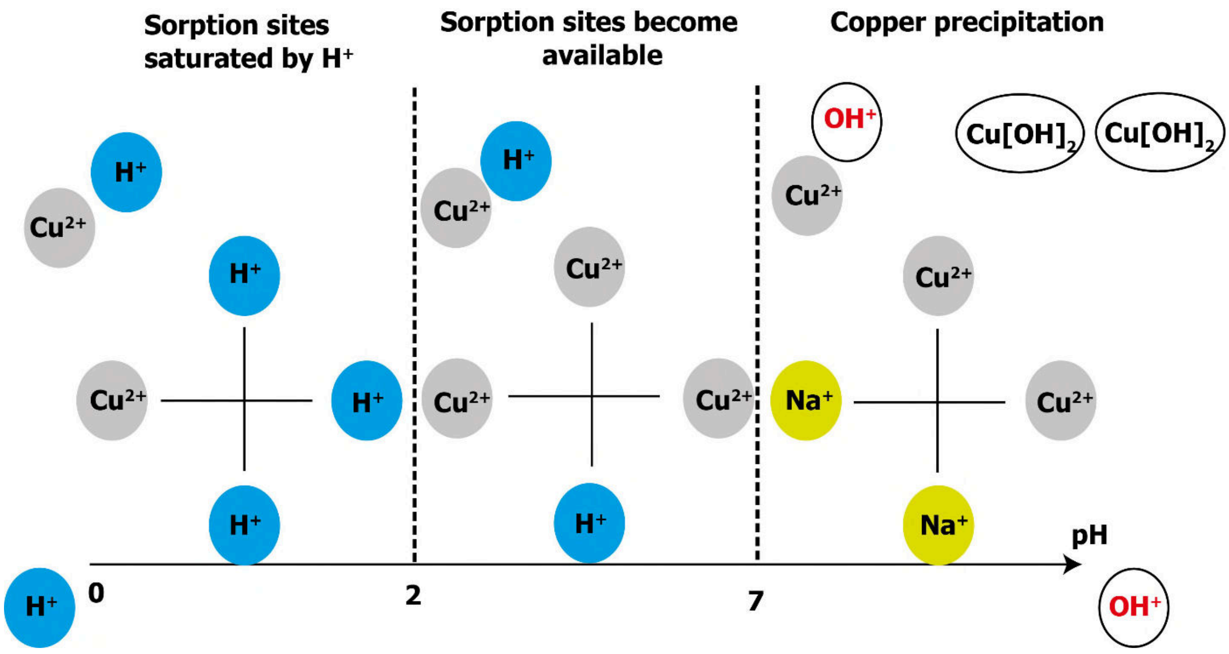

Figure 2. Copper sorption mechanisms (Adapted from Abbar et al., 2017)

In general, inorganic content of leachate ranges between 1 and 2000 mg/L. By raising the pH value, metallic hydroxide compounds become insoluble and precipitate from the solution. Alkaline conditions promote metal precipitation and adsorption, depending on the metal speciation (Lukman et al., 2013). Fig. 3 shows the solubility curves of selected metal ions and their respective solubility versus pH. Cu and Ni have a similar curve, albeit that the minimum solubility of Ni occurs at approximately pH 10.5 and the minimum solubility of Cu occurs at pH 9. Zn is amphoteric, being soluble in both acid and alkaline conditions. Cu and Zn readily form metallic complexes with ammonia. These metal complexes remain highly soluble at the higher pH values, prohibiting respective metal hydroxide precipitation. Cu sulphide is insoluble, and the presence of sulphide precipitates Cu as it dissociates from the ammoniacal complex. Precipitate in landfill environments strongly relates to organic decomposition and the formation of microorganisms during the process of methanogenesis (Li et al., 2015). According to Fig 3, precipitation is unlikely to occur in strongly acidic conditions except for Fe, Al, Pb and Zn.

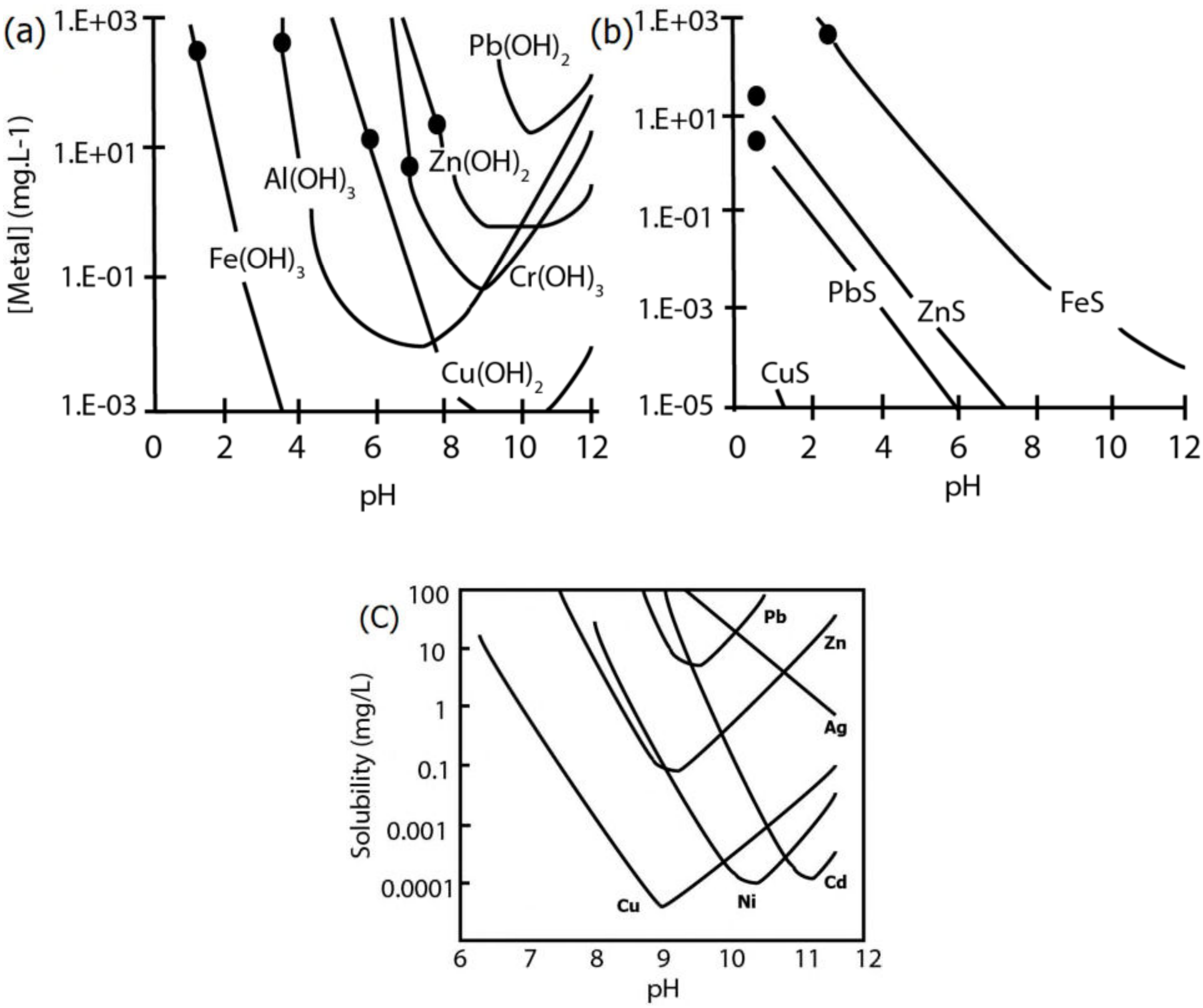

Figure 3. a) Solubility of metal hydroxides, b) Solubility of metal sulphide, c) Solubilities of metal hydroxides as a function of pH (Marchioretto et al., 2005)

Marchioretto *et al.* (2005) reported that when Fe and Al are present in landfill leachate, adsorption and co-precipitation may occur between Cr, Pb, and Zn with $Fe(OH)_3$ and $Al(OH)_3$ as pH increases.

The leachability of metals is also influenced by the chemical and physical affinity of metal ions and various waste materials under landfill conditions (Ward et al., 2005). Sulphates in waste are reduced to sulphide that forms insoluble precipitates with most metals or containing amino acids during anaerobic (Christensen et al., 2001). Dissimilatory microbial sulphate reduction is when certain bacteria use sulphate as the electron acceptor in the oxidation of organic matter. However, Cr does not form an insoluble sulphide; it is only precipitated out in the form of hydroxide. Sulphides of the metals are more difficult to dissolve, both in oxidising and reducing environments (Hammack and Edenborn, 1992).

3.1.3 Carbonation and redox

Carbonates are also capable of forming precipitates with metals and are abundant in landfill leachate. Nevertheless, the solubility of metal carbonates is generally high (Christensen et al., 2000). Metal precipitates of carbonate will dissolve, where the carbonate release will buffer the pH value as the pH decreases, which is called the humic phase (Kjeldsen et al., 2010). As attenuation mechanisms affect metal concentration and stability, it should be considered the metal adsorption and precipitation-pH relationship for recovering metals with high concentration from an economic point of view. The redox potential influences precipitation and should be considered when considering metal solubility. Redox potential is a measure of the propensity of a chemical or biological species to either acquire or lose electrons through ionisation (Lu and Marshall, 2013). Various parameters in landfill leachate can reflect transformations in redox potential. For example, as sulphate is reduced, their concentrations decrease. An increase in redox potential effects on the oxidation of reduced sulphur compounds to SO_4^{2-}. Oxidation of metal sulphides takes place, leading to metals release. The redox conditions in landfill leachate affect metal-organic interactions through the organic ligands, as organics are sensitive to redox conditions (Merian and Clarkson, 1991). Abundant OM tends to have low redox potential values. The speciation of metals, which is related to their mobility, is dependent on pH, redox, and organic compounds (Baun and Christensen, 2004). Each of the oxidation states has different metal complexation constants, and organic compounds may mobilise it to an extent critically dependent upon the redox conditions (Herbert et al., 1993). For example, Tingzong *et al.* (1997) found that Pb was bound to iron and manganese hydroxide under oxidised conditions. As the landfilled waste shifted to reducing conditions, Pb was leached out. Chuan *et al.* (1996) also reported that the solubility of Pb, Cd and Zn in soils increased when redox potential decreased, and this was due to the dissolution of Fe-Mn ox-hydroxides under reducing conditions resulting in the release of metals. In contrast, Sims and Patrick (1978) found that soluble Zn decreased at low redox potential, which may be caused by different environmental conditions and soil types. Also, Kamon *et al.* (2002) found that low redox potential and alkaline conditions induced by anaerobic respiration in landfill sites tend to prompt immobilization of Zn but a mobilization of Iron. Overall, redox potential strongly affects the behaviour of metals in leachate even though there uncertainty remains regarding to what degree such as different environmental conditions.

3.2 Organic matter decomposition and metal leachability

As landfill age increases, the leachate passes through successive stages of organic substance decomposition, which influences metal leachability. Metal leachability is highest when hydrolysis, fermentation and acetogenesis dominate due to an accumulation of VFA and a pH decrease (Fig 4). The primary acids formed during fermentation are acetic acid (CH_3COOH), propionic acid (CH_3CH_2COOH), butyric acid ($CH_3CH_2CH_2COOH$) and ethanol (C_2H_5OH). Carboxylic acids act as chelating agents, and there may be an increase in carboxylic functional groups on humic compounds due to the waste being oxidised (Kjeldsen et al., 2010). Qu *et al.* (2008) demonstrated that metals in leachate are bound to organic substances such as fatty acids, FAs and HAs. The fatty acids, FAs, and HAs content in leachates decrease as landfill age increases (Fan et al., 2006; He et al., 2006; Qu et al., 2008). The fatty acids are accumulated during the acid phase of the waste stabilisation (Christensen and Kjeldsen, 1989). FA predominates in young unstable leachates, and its concentration decreases as landfill age increases. The HA-forming processes are dependent on microbial degradation of OM, and the HA increases with the age of leachate, eventually reducing due to the leachate becomes more stable and diluted (Artiola-Fortuny and Fuller, 1982). HA has more carboxylic groups than FA and contains bands of aromatic C=C (Gustafsson and Berggren, 2005; Shirshova et al., 2006). The binding capacities of HS to metals within leachate and solid waste may imply that the solubility of HS strongly influences the mobility of metals (Qu et al., 2008). To the best of our knowledge, no work has been reported on the effects of organic matter decomposition on metal release in landfill environments. It is expected that different metals have different impacts on the decomposition processes of organic matter. Further research is required on the role of organic matter degradation on the release of individual metals.

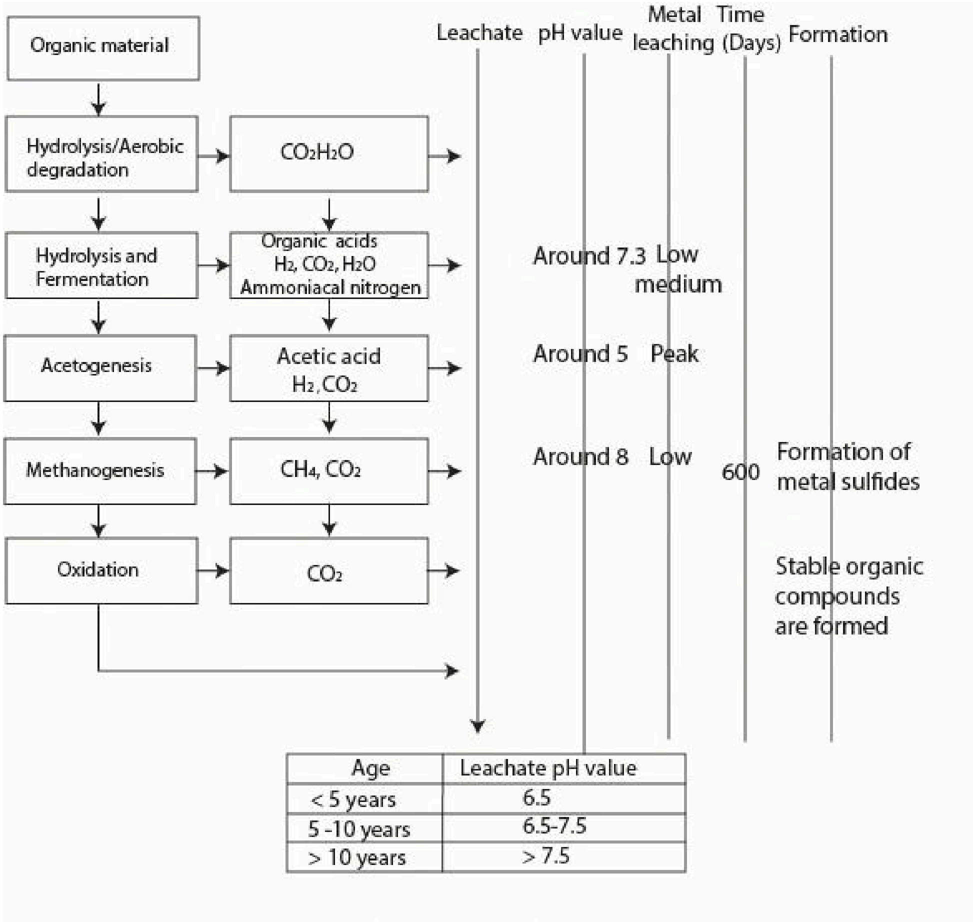

Figure 4. Metal leaching Process in a landfill and leachate pH value (Adapted from Zainol et al., 2012, Adhikari et al., 2014)

3.3 Chelation and complexation with organic substances

Most metal ions bind to neutral molecules in different oxidation states called a ligand, defined as an ion or molecule that binds to a central metal atom to form a complex (both organic; carboxylic acids, amino acids, HAs and inorganic) (Table 4). Ligands lead to the formation of metal complexes and metal chelates. Complexation with organic ligands is known to influence the mobility of metal by either increasing or decreasing its sorption on mineral surfaces. Many organic substances have been commonly identified in leachates worldwide (Details in supplementary data Table 1) (Paxéus, 2000; Staley et al., 2006; Zhang and Zhang, 2009).

Table 4. Mechanisms of adsorption for organic compounds in soils (adapted from Sposito, 1984)

Mechanisms	Principal organic functional group
Cation exchange protonation	amines, ring NH, heterocyclic N amines, ring NH, Heterocyclic N, carbonyl, carboxylate
Anion exchange water bridging	carboxylate amino, carboxylate, carbonyl alcoholic OH
Cation bridging	carboxylate, amines, carbonyl, alcoholic OH
Ligand exchange hydrogen bonding	carboxylate amines, carbonyl, carboxyl, phenylhydroxyl
Van der waals bonding	uncharged, nonpolar organic functional groups

Among the organic ligands, HS are the main organic compounds present in landfill leachate (Zhou et al., 2015). HS are the main component of soil OM or humus, most of which combine with the inorganic constituents in the soil (Pettit, 2004). HS have several functional chemical groups (carbonyl, hydroxyl carboxylic acid, phenolic ring, and quinine), which may combine with ions such as Fe^{3+}, Mg^{2+}, and Ca^{2+} and form chelate complexes; thus change the solubility of metals (Tipping et al., 2002). Generally, the potential for complex formation between metals and organics increases with pH alkalisation (Hummel et al., 2000). Farrah and Pickering (1997a) showed that the proportions of metal bound as hydroxyl complexes increase at pH 5 and above although the capacity for complexation shows no dependence on pH (Antelo et al., 2000). Instead the metal-organic interaction depends on the stability of complex formation and metal concentration. Esakku *et al.* (2003) reported higher stability constants for Cu complexes with OM and that these lead to higher Cu content in the organic fraction.

Phenolate, amino, and carboxylate groups enhance the formation of metal complexes at high pH, thus become increasingly stable at higher pH levels (Rieuwerts et al., 1998). Carboxylic and hydroxyl functional groups show acid-base behaviour.

At low pH, hydrogen ions compete with the metal ions for these sites, and as pH increases, less hydrogen ions are present and complex site availability for metal ions increases (Scott et al., 2005). There are challenges to understanding the complexation of different metal ions, e.g. i) organic compound functional groups influence the type of reaction it has with metals; and ii) the length of hydrocarbon chain length in carboxylic acid increases its metals adsorption capacity but decreases its stability as complex (Abollino et al., 2003).

Soil organic matter can influence the mobility and speciation of metal, where complexation reactions modify its accumulation potential (Kennou et al., 2015). For example, when organic materials, rich in soluble organic carbon and a large proportion of FAs are applied to soil, metal mobility increases due to the formation of soluble metal-organic complex (Pérez-Esteban et al., 2014). In contrast, when a chelating agent binds to a metal ion in more than one place simultaneously, chelated compounds become more stable (Pohlmeier, 2004). It has long been recognised that complexation may lead to increase metal solubility or decrease adsorption (Cavallaro and McBride, 1978; Bradl, 2004; Güngör and Bekbölet, 2010; Ahmed et al., 2019). Similar observations have been shown to occur within the landfilled waste. A variety of organic compounds can be expected in the leachates, which afford the potential for metal-organic interactions through the organic ligands. Previous studies established that dissolved organic matter (DOM) in MSW has a high affinity for metals, especially for Cu and Pb (Christensen et al., 1996; Christensen et al., 2000; Huo et al., 2008). Most insoluble metals are present in their refractory chemical form, i.e. $PbSO_4$. Over time the oxidation/ reduction of these metals to a soluble form through complexation appears likely (Takahashi et al., 2010). If insoluble metal-DOM complexes are formed, the mobility of the metals in question and the DOM to which they are complex decreases. Metal mobility is less clear when soluble complexes are created with DOM (Jansen et al., 2003). On the one hand, it may increase because, i) the mobility of DOM is affected by its functional groups and adsorption to soil particles (Kaiser et al., 1997); ii) binding to DOM prevents immobilisation by precipitation of inorganic metal complexes. On the other hand, the mobility of soluble metal-DOM could decrease complexes when they bind to soil particles through cation bridging (Guggenberger and Zech, 1993).

The leachability of metals could be enhanced through ligand complexation where organic acids such as carboxylic acids and phenols, formed during the decomposition of organic compounds, decrease pH. The pH determines the number of acidic functional groups on deprotonated DOM, which increases the availability of sorption sites for binding metals (Stevenson, 1994).

Jensen *et al*. (1999b) determined organic complexes of heavy metals in landfill leachate polluted groundwater in the Vejen landfill. They found that organic complexes made up a significant part of the total content of heavy metals: Cd 85%, Ni 27-62%, Zn 16-36%, Cu 59-95%, and Pb 71-91%. Kalis *et al.* (2006) found that the metal-humic acid complexes become the dominant complexed species when humic acid is present. Yu *et al.* (2018) reported that most of the complexes between Cd and the HS would be insoluble, and the complexation could contribute significantly to the reduction in the concentration of Cd in soil solution. Van Ginneken *et al*. (2001) discovered that the stability of chelated metals and noncyclic metal complexes depends on several factors, including pH, metal oxidation state, and ionic strength. Organic-metal complexes are increasingly stable at higher pH levels due to the ionization of functional groups (Jones and Jarvis, 1981; Rieuwerts et al., 1998). Conversely, organic acids present in the dissolved organic carbon (DOC) may act as chelating agents, enhancing the mobilisation of metals (Christensen et al., 1996).

Complexing behaviour significantly influences metal attenuation as it affects their mobility and saturation indices (Qu et al., 2019). As metal-organic complexation plays a critical role in the mobility of metals in landfill environments, lab and field experiments would be required to establish the relationship between complexing characteristics and observed metals leaching, performed under varying environmental conditions to optimise metals recovery.

4 LEACHATE RECIRCULATION STRATEGY FOR METAL RECOVERY

Leachate recirculation within landfills has been widely used for a range of purposes since the 1970s, including leachate management, enhanced landfill gas generation or recovery, and improved landfill sustainability (EPA, 2009). Leachate recirculation is a process where leachate is re-introduced into the landfill through an artificial recharge system (White et al., 2011). This technique aims to encourage saturation to stimulate the degradation processes, leading to more rapid stabilisation of the landfill (Scott et al., 2005). However, leachate recirculation can also increase the chloride content. Chloride contents may also be an important controlling factor for metal release. Chloride affects the behaviour of metals by binding the metals on humic acids and the adsorption of metals, such as the adsorption of Cd on iron hydroxides or their desorption mobility (Guevara-Riba et al., 2005; Begeal, 2008; Damikouka and Katsiri, 2020). The ionic forms of Cu and Cd can form metal compounds with the anions such as $CuCl_2$, $CdCl_2$ or $CuSO_4$, leading to chloro-complexation and the formation of dissolved metal-chloride compounds increase the mobility of metals (Kirkelund et al., 2010; Damikouka and Katsiri, 2020). This implies that leachate recirculation affects metal recovery. Leachate recirculation can significantly influence metal behaviour and fate within waste matrices (Ledakowicz and Kaczorek, 2004). For example, Yao *et al.* (2014) found that leachate recirculation contributed to faster stabilisation of the landfill and reduced leachability of Cu and Zn from the landfill. He *et al.* 2007 have also shown that recirculating leachate, which is by the sequential reactors, in landfills in the early stage allowed methanogenesis to be reached much earlier and that this was accompanied by a reduction of total metals released from landfills. In contrast, Qu *et al.* (2008) demonstrated that the initial stage of leachate recirculation had low leachate pH (5-6) and highly VFA levels (acetate 4500-700 mg/L, propionate 1450-2950 mg/L and butyrate 4500-7200 mg/L) due to the acidification stage, in resulting the concentration of the metals was high at this stage. Bilgili *et al*. (2007) showed that the release of metals can significantly increase at the beginning of leachate recirculation as in the early stages of the waste degradation, pH of the leachate is low contributing to higher solubility of metals and dissolution into leachate (Fig 5).

As stated previously, due to the low concentrations of metals, the way to make it economically viable in recovering metals is to maximise metal concentrations. Recirculating leachates will accentuate the potential for increased metal mobility within and from the landfill when oxidised conditions are introduced. Leachates have been shown to have an increased capability to enhance metal mobility when oxidised (Mårtensson et al., 1999). It implies that leachate recirculation provides higher extractable metals in the initial leaching phase.

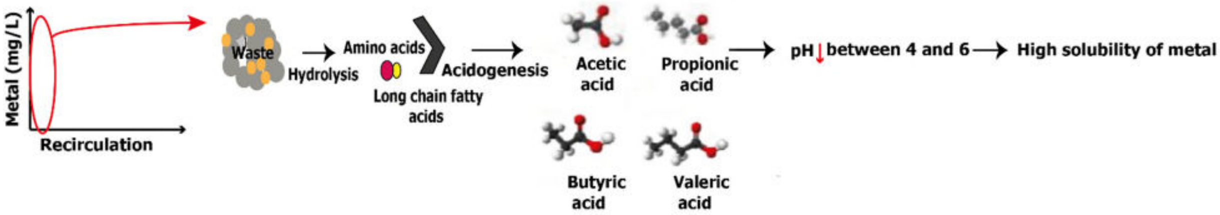

Figure 5. Leachate recirculation strategy for metal recovery

Therefore, regarding the critical challenges in metal recovery in leachate, this suggests that where leachate recirculation is applied with combining organic matter such as HA and pH could increase the economic feasibility.

5 Opportunities for metals recovery from landfill leachate

Technologies to extract metals are highly required to enhance and promote recovering metals as secondary raw materials. One of the modern economy models is a circular economy in which wastes should be considered a resource and used efficiently and sustainably (Wieczorek and Kwasniewska, 2018). Extracting metals from landfill leachate may solve the biggest challenges in landfill mining which is regarding the economy. Leachate contains a wide range of metal such as Cu, Zn, Cr, Cd in addition to REEs (Kjeldsen et al., 2002). Leachate in landfill sites has great potential for in-situ metal recovery processes.. Consequently, *in situ* metals recovery can help mitigate climate change and preserve biodiversity by reducing environmental exposure of landfilled wastes. There are several ways for recovering metals from waste, such as recycling, physico-chemical, thermo-chemical, pyro-metallurgical, hydro-metallurgical, bio-metallurgical, landfill-mining methods (Wang et al., 2017). Due to the lack of economically and operationally feasible primary resources for the production of metals, many countries are forced to depend on recycling metals from secondary sources such as industrial residues and end-of-life products. Table

5 shows various methods to recover metals from waste streams and leachate, suggesting the possibility of recovery metals in landfill leachate. Despite published research works on recovering metals from leachate, studies are scanty in an application towards the recovery of metals from real landfill leachate in the site. The main problem to recover metals from leachate is further compounded for metals that exist at low concentrations (Table 2), which can be affected by many factors such as landfill age, type, and chemical and physical mechanism in landfill environments. Also, the strong acidic condition can affect the environment; thus, it may later pose harmful risks to the environment if not managed well. The concentration of metals varies widely, and REEs has a very low concentration (1 or 2 μg/g waste), which limits the economic viability of recovery. Relatively, high concentrations of over 1% are required to ensure cost-effective recovery of metals (Umeda et al., 2011). There are many factors to be considered for the operational cost; leachate collection ($9.56 per m^3 for leachate treatment), labour cost for operating the plant ($30), electricity cost per hour ($0.1042) and recovery by chemical leaching ($1060 per m^3) (Priya and Hait, 2017; Leflay et al., 2020). Therefore, process optimisation is important to maximise the concentration of metals or co-extraction of other added-value materials such as nitrogen and phosphorus, improving the process's cost-effectiveness and efficiency. By understanding the processes, including organic-metal interaction in leachate, recovering metal can achieve higher average productivity.

Barriers remain in recovering valuable materials present due to the unknown concentrations and distributions of metals in landfills and not meet the reasonable financial level. Metal recovery from leachate has not been investigated before, but prior studies have demonstrated that metals can be recovered from wastewater and aqueous solutions. However, the methods remain limited; for example, physicochemical methods are energy and capital intensive due to the costs of chemicals, oxidants, and membranes; the biological treatment process is limited by treatment effectiveness and energy requirement (Ahn et al., 2002; Kargi and Pamukoglu, 2003).

Table 5. Metals recovery from wastewater and liquid solution

Source	Method	Characteristics	Effects	Reference
Leachate from landfill waste	Bioelectrochemical systems (BES)	·BES employs biological and electrochemical reactions to recovery resources from a wide range of substrates.	·The electrical conductivity of leachate makes it favorable for electricity generation, and it contains a high concentration of ammonium nitrogen, which may be recovered for agricultural application. ·Metal also may be recovered by the modified microbial electrolysis cells.	Iskander et al., 2016
Sulfate leachate of cathode scrap of Li-ion batteries	Liquid-liquid extraction	1) The cathode scraps undergo heat treatment to completely liberate the cathodic materials from aluminum foil. 2) Solubilise Co, Li, Fe, Mn, Ni, and Al by leaching the cathodic materials in sulphuric acid in the presence of H_2O_2. 3) Oxidative precipitation of Mn from liquor with $KMnO_4$ and extraction of Al and Fe using D2EHPA. 4) Treat to recover the metals.	·Focuses on selective recovery of Co, Ni, and Li from the sulphate leachate of cathode scarp generated during the manufacture of Li ion batteries. ·High-purity Co in a solution can be recovered by solvent extraction using the sodium salt of PC-88A. ·The metals extraction efficiency and separation factor depend upon the extractant concentration and the equilibrium pH of the aqueous phase.	Nguyen et al., 2014
Solution and leachate derived from electronic scrap	Biogas	·Precipitates metals from solution using the off-gas ·Recovery Au, Pd, and Ag from leachate derived from electronic scarp; safe microbiologically	·Au was recovered from electronic scrap leachate with selectivity against Cu using biogas as they could partially separate Au from Cu. ·In acidic conditions, Au and Cu are removed rapidly and separated from the liquor. ·The solid Pd and Ag will not easily be separated in water via biogas. ·Amines must be avoided for recovering metals using biogas.	Macaskie et al., 2007

End of life electronic wastes	Hydrometallurgical process	·Ferric sulphate concentration range (at 1:10 and 1:5 Cu to sulphide molar ratio 1) filled up by using N_2 gas for anaerobic conditions 2) add of 10 ml of $Na_2S{\cdot}9H_2O$ solution	·Effect of Fe^{3+} on leaching of Cu and selective recovery of Cu from the polymetallic leachate. ·Lixiviant concentration and pH were the important parameters in $CuSO_4$ precipitation. ·The precipitation mostly occurs in the acidic pH range (0.5 to 1.5). ·$CuSO_4$ can be further pyro/hydrometallurgical processed to produce Cu metal.	Sethurajan and Hullebusch, 2019
Aqueous solutions	Biosorption	·The phosphorylation yeast cells were used in Cu adsorption experiments with 0.1 M HCl, which is strongly influenced by the pH of the solution. ·Recovering metals from aqueous solutions. The biosorption of metals is a complex process affected by the adsorbent, the types, and the concentrations of metals in the solution.	98% of the Cu ions adsorbed to phosphor cells could be recovered by treating the cells with HCl.	Ojima et al., 2019
Sulfuric acid leaching liquor of spent Li-ion batteries	Hydrometallurgical process	·Needs to refine the residues into a purer form such as salts, hydroxides and metals. 1) selective precipitation method by adding dimethylglyoxime (DMG, $C_4H_8N_2O_2$) reagent 2) extraction using cobalt loaded phosphoric acid (D2EHPA) 3) 4Separation and recovery of metal (Ni, Mn, Co, and Li) from sulfuric acid leaching liquor	High purities of Co and Li were recovered as $CoC_2O_4{\cdot}2H_2O$ and Li_2CO_3.	Chen et al., 2015
Acid mine leachate	Sequential precipitation	1) Sequential precipitation; add a sodium hydroxide solution of 5 M 2) Selective dissolution; pre-concentrates of the valuable elements were re-dissolved into solution 3) Oxalic acid precipitation;	·95% of the Cu and Zn were recovered from the residual liquid using Na_2S at pH 2 and 3.	Zhang and Honaker, 2020

		dissolving 8 g oxalic acid dehydrate in 50 mL deionised water using an ultrasonic batch 4) Na_2S precipitation; 1 M Na_2S REEs, Cu, Zn Ni and Co recovery from an acid mining leachate. The sample was collected from a coal preparation plant.	·The optimise the oxalic precipitation for the REE recovery is using a solution pH of 1.2	
Sludge	Precipitation	·Add 3 mol/dm^3 NaOH, ·The recovery process of heavy metals from polluted sludge leachate with biosurfactant elution by batch and column experiments.	The recovery efficiency of heavy metals (Pb, Ni, and Cr) reached over 90% by the precipitation method with pH 10.9.	Gao et al., 2012
Wastewater	Cementation	·Add Fe, Al, and Zn metallic powders into 250 mL of wastewater ·Stir continuously with a magnetic stirrer	·Cu, Au, and Pd can be recovered by using Fe and Al powder. ·Precious metals can be effectively recovered by combining processes (cementation, neutralisation and reduction)	Umeda et al., 2011
Wastewater	Photoelectrochemical cell	·A stock solution was prepared by dissolving the metal salts into deionised water ·The photoanode and the Pt strip cathode were connected with a commercial Cu wire ·The photoanode was irradiated with a UV lamp	·Heavy metals were recovered by mechanical scratching of the cathode surface.	Wang et al., 2017
Wastewater	Electrochemical reactor	·Prepare solutions using deionised water ·Determining the quality characteristic to be optimised. ·Identification of the noise factors and test conditions. ·Identification of the control parameters.	·The highest efficiencies were obtained for Pb and Cu recovery from diluted solution: 75.8 % and 89.9 %	Kaminari et al., 2007

Bioelectrochemical systems (BES) is an environmental strategy that employs biological and electrochemical reactions to generate electricity and recovery resources from a wide range of substances. Organic compounds in BES tends to produce electricity and other value-added compounds by oxidising microorganisms. However, a high concentration of metals in landfill leachate can be recovered through BES, and the reduction in leachate volumes can be achieved using osmotic processes integrated with BES. Also, hydrometallurgical processes have gained considerable attention as they show effectiveness in the extraction of metals (Gunarathne et al., 2020). Hydrometallurgical metal recovery is typically performed in three main stages: metal dissolution, concentration and purification, and metal recovery (Gupta,2006). Thus, further research should be studied to take a circular approach, recovering metals from landfill leachate using BES after enhancing metal concentration through leachate recirculation or hydrometallurgical processes using less toxic chemical solvents to be used as leaching agents and assist of acids and pH value.

6 Conclusion

This critical review has appraised metal recovery opportunities in landfill leachate using factors influencing metal mobility in landfill environments. Landfill leachate is a significant potential resource in landfill as it contains a large variety of dissolved extractable metals. Metal's recovery opportunities may increase by several factors, influencing metal mobility in landfill environments as an excellent challenge for metal recovery from leachate is the low concentration of metals. Younger landfill leachate (<5 years old) has higher organic matter content due to the generation of dissolved and solubilised organic matter, consequently increasing metal release. Therefore, it implies that metal recovery may be effective in younger landfill leachate. Physio-chemical processes affect soluble metal concentration, which is critical to predicting metal recovery as they can govern the mobility of metals. More knowledge is required concerning the complexes in leachate in general and specifically on the importance of the organic matter in leachate, which led to an increase in the metal release rate. This article has also identified the gaps and has indicated that further efforts are required concerning leachate recirculation. Therefore, this review is paving a way forward that can inform lab-based and later on semi-pilot to pilot studies to be effective for the recovery of metals. It may ensure that maximising the concentration of metals from landfill leachate will be the economic feasible. Although mature technological advances provide opportunities for recovering metals from landfill leachate, significant challenges await us ahead as they can hardly be regarded as economical. It is clear that *in situ* recovery of metals from landfills is a novel technology area that links new sustainable remediation approaches for contaminated materials and land and is a great help to the development of a circular economy. Therefore, the chemistry mechanism of landfill environments should be well understood and fundamental and practical barriers of the recovering process in landfill leachate, which will lead us one step closer to resource recovery paradigm for a circular economy in closed landfill.

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