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In Situ Resource Recovery from Waste Repositories: Exploring the Potential for Mobilization and Capture of Metals from Anthropogenic Ores

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Abstract Wastes and the waste repositories in which they reside are becoming targets for resource recovery, both for legacy wastes and for future waste arisings as part of a desire to move toward a circular economy. There is an urgent requirement to explore concepts for practicable technologies that can be applied to these ends. This paper presents a synthesis of concepts concerning in situ technologies (developed from mining and contaminated land remediation industries) that have enormous potential for application to technospheric mining. Furthermore, potential target waste streams and their mineralogy and character are presented along with a discussion concerning lixiviant and metal capture systems that could be applied. Issues of preferential flow (critical to the success of in situ techniques) and how to control it with engineering measures are discussed in detail. It is clear that in situ recovery of metals from anthropogenic ores is a novel technology area that links new sustainable remediation approaches for contaminated materials and technospheric mining for closing material loops, and warrants the further research and development of technologies applicable to major waste streams.

Keywords Leaching · Mining · Industrial residues · Recycling · Technosphere · Remediation

Introduction

Waste repositories can be considered the ore deposits of the 'anthropocene.' Having historically disposed of vast quantities of industrial, municipal, metallurgical, and mining waste into or onto the ground, societies have put into geological storage an enormous quantity of resource in a range of materials of value such as metals and energy (in the form of biomass and polymers). Therefore, instead of considering these waste repositories to be a legacy waste issue and a long-term liability, a paradigm shift is required to view these installations as 'resource hubs' for future recovery. This has to some extent been recognized with commensurate but small-scale landfill mining occurring internationally [1], and the concept of mining materials from the 'technosphere' (rather than the lithosphere) is gaining ground. Following the definition of Johansson et al. [2], the 'technosphere' is defined as material stocks established by human agency, which originate from technological processes, in contrast to stocks in the lithosphere established by slow, primary geological processes. The technosphere can be distinguished from the lithosphere which is where historically humans have derived all of our metal resources. Of particular relevance, three major technospheric stocks have already been defined as 'controlled inactive stock' [2]: metals tailings, slag heaps (e.g., metallurgical wastes), and landfills, which are amenable to 'landfill' or 'secondary' mining.

The recovery of resources from waste repositories has not been widely adopted, beyond the limited extraction of landfill gas for energy generation. Attention is now turning toward recovering elements of value, for example, the so-called "E-tech" elements (Co, Te, Se, Nd, In, Ga, heavy rare earth elements) whose security of supply is an issue in addition to them being essential for current and future

green technologies. Furthermore, there are the 'EU-20' supply-threatened critical materials including platinum group metals (Pt, Ru, Rh, Os, and Ir) and precious and base metals: Au, Pd, Ag, Cu, Pb, Zn, Co, Ni, Sn, and Cr. Many of these elements are variously amenable to leaching (depending upon mineralogy) and found in significant concentrations in common wastes (e.g., tailings, metallurgical wastes, incineration ashes) but not in sufficiently high concentration to justify conventional ex situ processing. Additionally, there are predicted shortages in geological reserves for many metals, and exploitation of more complex deposits including anthropogenic deposits is a natural extension of this developing trend. There is also a clear synergy with the aims of a circular economy in returning as much resource as possible back into the production cycle. While recycling of metal goods is commonplace, metal-bearing industrial residues have received less attention despite constituting a considerable resource, although metal recovery processes do exist for certain residues such as FeCr slags and municipal solid waste (MSW) incineration ashes.

In order to successfully and sustainably mine technospheric stocks, new technologies and/or those adapted from existing mining or remediation technology are required. As such, this paper aims to provide an overview of in situ resource recovery within a conceptual framework that seeks to (i) explain why in situ resource recovery technologies are appropriate to waste repositories; (ii) identify existing technologies that can be transferred to this new area; (iii) highlight key wastes/waste repositories that could be targeted; (iv) explain how the waste mineralogy will be critical in devising lixiviant systems; (v) discuss the issue of preferential flow and how to control it with engineering measures; (vi) identify metal capture technologies; and (vii) explore current technology development level, legislation, and international applicability.

A Rationale for In Situ Recovery

Applicability of In Situ Recovery of Resources

In situ techniques in mining include in situ leaching (ISL), dump leaching, and stope leaching. Heap leaching is not sensu stricto 'in situ' but 'ex situ.' However, since it is similarly low intensity and the leaching mechanisms applied would be the same, i.e., a stationary solid phase is flushed by an extractant (lixiviant), or the biogeochemical environment around the solid phase is manipulated in situ, here it is grouped with 'in situ' technology. This family of in situ techniques has been applied for the recovery of metals such as uranium, gold, silver, copper, and nickel and

is applied to low-grade ores (for uranium ISL on grades as low as 0.05%).

The use of in situ techniques in mining is *prima facie* for economic reasons. The cost of extraction of metals from ores (and contaminants from contaminated mixtures) is demonstrably inversely proportional to the concentration, fundamentally this is related to the thermodynamics of separation of chemical mixtures [3–5]. An increase in the exergy cost for extraction is imposed as the target becomes more dilute within the mixture. Valero et al. [5] express a unit exergy cost which is the ratio of the energy (real) cost of processes to remove target metals from ores to the exergy cost. Such analyses demonstrate that while the exergy cost of removing metals from low-grade ores increases dramatically as ore grade decreases, the commensurate unit exergy cost increase can be offset by more energy-efficient mining processes. This is reflected in the economics of ore processing and is why in situ processes, which keep energy costs to a minimum by negating the large energy requirements of conventional mining and processing (e.g., rock-moving, comminution, pyrometallurgy), are favored for low-grade ores.

Wastes, as is implicit in the use of the term, usually contain sub-economic concentrations of valuable resources (at least at the time of their production), and it should be recognized that this can vary with time. They can be considered as very low- to ultra-low-grade materials, and the detriment that this causes is twofold: (i) the economic incentive to process these materials becomes less and less; and (ii) the exergy cost increases as grade decreases. This is a critical point that is often overlooked in research on valorization of wastes. Overcoming the decreased exergy of metals in wastes, the increased energy to extract them and the consequent economic disincentive to recover metals is a key challenge. The extraction of resource may have other drivers in addition to the direct economic recovery of resource (e.g., remediation of impacts to environment and human health), which needs to be assessed in careful cost–benefit analyses, but it is clear that minimal processing costs will be advantageous and thus low-intensity (energy-wise) processes to remove metals are required. In situ techniques established in the mining industry and contaminated land remediation offer a promising avenue to explore.

A Proposed Taxonomy for In Situ Techniques Applied for Resource Recovery

Leaching in mining and removal of contaminants from contaminated land can be considered as reciprocal approaches: in land remediation, the aim is the recovery of the material (soil/sediment) with leaching, resulting in purification/decontamination of target soil, rather than in

ore leaching where the target is the leached metal. However, these processes can be seen as being the same with simply a difference of perspective, where remediation recovers the soil material/value. The following taxonomy is proposed to clearly differentiate these two different aims when appraising the growing literature on in situ processes in resource recovery. Recovery can be subdivided into the following:

Direct recovery Recovery of resources from the waste repository while the waste remains in place, i.e., an efflux of the resource from the repository. Examples include the leaching and recovery of metals (as discussed herein) and methane capture and use from MSW landfills.

Indirect recovery This is the use of in situ decontamination or stabilization of the material in the waste repository to allow or facilitate the recovery of resources through subsequent exploitation, i.e., the focus is on the value recovered in the residue or the land rather than what is leached/removed. Thus, indirect recovery can be further subdivided into the following:

(i) *Indirect Material Recovery through decontamination* For example, Fedje et al. [6] discuss methods for leaching metals from MSW incineration fly ash so as to render the fly ash classifiable as non-hazardous waste—which facilitates reuse options.

(ii) *Indirect Material Recovery through changing the physicochemical nature of the waste* For example, the weathering/carbonation of blast furnace slags to remove problematic lime content ahead of its use as a product in aggregate applications and the accelerated carbonation of alkaline wastes, e.g., Van Gerven et al. [7].

(iii) *Indirect Land Recovery* The use of in situ decontamination or stabilization of the material in the waste repository to allow the recovery of land as a resource in itself. Examples include the range of in situ technologies in contaminated land remediation and waste stabilization.

Note that the process of recovering resource from waste repositories may involve a number of direct and indirect recovery technologies (e.g., Kim et al. [8]). In fact, the most desirable outcome for a waste repository might be a combination of direct and indirect recovery such that both the extracted resource and the residue are valorized. As such, in situ technologies both direct and indirect may contribute to the quest for zero-waste processes. The focus herein is on direct recovery of metals from wastes, while acknowledging that much of the presented information is also pertinent to indirect recovery.

In Situ Technologies

Mining Technologies

In situ leaching (ISL) involves accessing an ore deposit via a network of boreholes. The permeability may be enhanced through various stimulations, and then a lixiviant is pumped through the subsurface to solubilize the target metal which is then recovered at extraction bores. ISL has been used for the recovery of uranium, copper, nickel, zinc, cobalt, gold, and salt/trona. ISL has been most often applied to uranium leaching and accounts for a large proportion of uranium production. Acidic (sulfuric acid) or alkaline (ammonium or sodium bicarbonate/carbonate) lixiviant systems are typically used, although a much wider range of lixiviants are now being considered [9]. However, it is noteworthy that there have been many instances of poor environmental outcome from poorly/unmanaged uranium ISL operations [10].

Another important in situ technique applied at mine sites is dump leaching. This is where excavated materials are leached where they sit, contingent on the material sitting on a low-permeability base such that the leachate can be collected and metals removed. A variation of this is stope leaching, where the same process is conducted underground with piles of blasted low-grade ore. Furthermore, recirculation of water through the whole network of abandoned mine tunnels has also been practiced. As with ISL, clearly for in situ techniques the control and fate of the lixiviant is a vital issue for both process efficacy and environmental protection. It is feasible that a liner can be installed in situ through the use of grouts.

A more engineered version of dump and stope leaching is heap leaching. It involves the same principle but the material is crushed and stacked on an engineered liner system (either flat or in a valley-fill [11]) in order to capture all of the leachate which is applied with either sprinklers or emitters (drippers). Heaps can be 'multi-stack' where the material heap grows as progressive lifts are placed and leached, or 'dynamic' where materials are loaded and later unloaded from the pad. Dynamic pads might be useful where materials from a regional cluster of sites are to be processed. Where materials are fine-grained (as is common with many of the industrial and mining wastes considered here), then processing to agglomerate the materials prior to placing them on the heap leach pad is often undertaken to ensure sufficient permeability for leaching.

The metals are then extracted from the "pregnant" liquor which is collected from the base of the heap using approaches such as ion exchange/solvent extraction before

electrowinning from the concentrated solution. Heap leaching is typically employed where ore grades are not sufficiently high to justify more intensive hydrometallurgical/pyrometallurgical options. This illustrates the continuum of options between process intensity (cost) versus recovery and rate of recovery and highlights that in situ techniques, as has been recognized by other authors (e.g., Steen and Borg [12]), are applicable for materials with lower concentrations. A summary of the mining technologies and their potential application to in situ recovery from waste repositories is presented in Table 1.

Many commercial ISL, dump, and heap leaching operations involve bioleaching where sulfides are present. Metals bound as sulfides present an interesting case of a "self-extracting" system. Metals associated with sulfides, when exposed to the subaerial weathering environment, will be released as the sulfides spontaneously oxidize. The rate of weathering and release of metals from sulfides is kinetically controlled and influenced by microbes, including bacteria, archaea, and eukarya (see, for example, Schippers et al. [13]) with the ecosystem being driven by metabolic energy derived from the free energy available upon the oxidation of the ferrous and sulfur moiety of the sulfide minerals. This spontaneous breakdown of sulfide minerals is advantageous with regard to metal extraction (as is established with heap leaching of sulfide ores) but disadvantageous from an environmental perspective as it causes acid mine drainage and metal leaching from mines, which is one of the biggest causes of metal pollution worldwide. Therefore, wastes containing sulfides (e.g., mine tailings and mine wastes) will be amenable to self-extraction if engineered appropriately. Rather than trying to engineer mine waste repositories for isolation from oxygen/water ingress in perpetuity, allowing oxidation and leaching of metals from sulfides in an engineered and controlled fashion with metal capture and recovery might ultimately be a more sustainable approach to the long-term management of mining wastes.

Contaminated Land Remediation Technologies

A summary of the contaminated land technologies potentially applicable to in situ recovery from waste repositories is presented in Table 1. Both in situ and ex situ soil flushing have been investigated for the removal of metals from contaminated soils [14]. In situ recovery requires that the material to be flushed (as in the case of dump leaching in mining) overlies an impermeable stratum such that a pump-and-treat system can be applied to the soil capturing the extractant downstream in collection borehole(s) without polluting aquifers. A further variation of this approach is to excavate the material and place on a liner (as in heap leaching in mining), applying an extractant, collecting the

pregnant liquor, removing the metals, and then recycling the extractant. Again, while this is technically ex situ, the nature of the leaching is in situ. Chelates are typically used to enhance metal removal, but while metal recoveries are dramatically improved with chelating agents, the metals are also then conversely difficult to extract from the chelates without resorting to expensive oxidation methods. Costs for the various contaminated land technologies range from a reported value of 10–228 €/m^3 during 2001–2003 for in situ soil flushing to 22–222 €/m^3 for phytoremediation [15].

Phytoremediation is an established in situ technology (although still requires periodic harvesting) and is a very low-intensity 'land farming' technique for metal contamination remediation. Much recent research has been directed at chelate-enhanced phytoremediation (especially biodegradable chelates) to overcome the fact that many metals are not readily available for plants and/or have no natural tendency to hyperaccumulate in any plant species. Desirable plant species are those that are fast-growing, have a high biomass, and can be easily harvested [14]. Reported costs are scarce in the literature but a cost of 50–200 US $/m^3 is reported by Virkutyte et al. [16] for various methods applied in the 1990s. More recently, there has been interest in applying phytoremediation for Direct Recovery, this is called 'phytomining' (or agromining) and the estimated profitability of Ni phytomining using *Berkheya coddii* on serpentine soils rich in Ni (Australia) has been recently estimated at 11,500 AU$/ha/yield and the profitability of Au phytomining using *Brassica juncea* is about 26,000 AU$/ha/yield [17].

Electrokinetic (EK) techniques use the migration of suspended charged particles and chemical species under an applied electric field (known as electromigration) to remove metals from the subsurface. It is particularly applicable in fine-grained material, as it is not limited by low hydraulic conductivity. Indeed, the generation of electroosmotic water flow in fine-grained materials with an appreciable surface charge can aid in mobilizing contaminants. Typically, the process involves significant pH change within the treated material due to electrolysis of water at the electrodes, while the generation of a strongly acidic pH near the anode can mobilize metals and aid their recovery. The strongly alkaline environment near the cathode, to which most metals are attracted, can hinder recovery and so must be addressed. There is also an associated energy cost, as significant voltage gradients (typically 100 V m^{-1}) are often required, but this is comparable to other interventions. It is therefore likely to be a feasible option for flow generation and control in wastes. EK techniques have been widely explored in the laboratory, and to a degree in field applications for removal of contamination, delivery of chemicals (e.g., electron

Table 1 Existing technologies that have potential for application to in situ recovery of metals from waste repositories

Technology	Target/application	Process inputs	Advantages	Limitations/constraints	Technology status
In situ leaching (ISL)	Ore bodies, Cu, Au, Ni, Sc, Re, REES, Y, Se, Mo, and V [9]	Lixiviant addition required, e.g., sulphuric acid, sodium bicarbonate	Lower capital costs than conventional mining Environmental impacts (if process is well managed) are lower than conventional mining	Requires reasonable permeability of 1–5 m/d (natural or artificial enhanced) Targeted species may be occluded within rock inaccessible to lixiviant Not all material contacted by lixiviant leading to incomplete leaching/recovery Thicknesses and grades, distribution of mineralization, requires presence of aquicludes [9] Environmental concerns regarding fate of lixiviant in the subsurface	First used for U extraction in 1959. Has been used extensively. Future expansion in use predicted [9]
Dump leaching	Historically used to recover metals from surface dumps, underground dumps (stope leaching) and underground mine workings for Cu and U	Lixiviant addition, e.g., sulphuric acid, sodium bicarbonate	Lower capital costs than conventional mining Ease of implementation Can utilize mine workings as drainage conduits	Only permissable on prepared sites Leachability of target depends on liberation Not all material contacted by lixiviant leading to incomplete leaching/recovery Environmental concerns regarding fate of lixiviant	Discontinued on unprepared sites
Heap leaching	Cu, U, Au, Ag, Ni	Lixiviant addition required, e.g., sulphuric acid, sodium bicarbonate, cyanide	Controlled leaching on engineered liner, limits escapes to environment Economic extraction from low-grade ores possible	Leaching time can take several years Leachability of target depends on liberation Not all material contacted by lixiviant leading to incomplete leaching/recovery Capital costs of liner and material handling to load leach pad Fine-grained materials require agglomeration to ensure good contact with lixiviant	First gold heap leaching with cyanide 1900–1920s [11] Extensively used for uranium, gold and copper

Table 1 continued

Technology	Target/application	Process inputs	Advantages	Limitations/constraints	Technology status
Soil flushing	Any contaminated soils/sediment, though availability for transport is critical	Depending on situation—range of extractant solutions including chelators, surfactants, acids	Low cost compared to comparable methods such as in vessel soil washing	Leachability of target depends on solubility and/or degree of sorption. Highly sorbed materials will only slowly be removed, though this may be enhanced with chelators and other additives Not all material contacted by lixiviant leading to incomplete leaching Environmental concerns regarding fate of extractant in in situ soil flushing	Developing technology, not extensively applied
Phytoremediation/phytomining	Any—appropriate plant selection may tackle a wide range of specific metals, though availability of resource is critical For phytomining Au, Tl, Co, and Ni are considered most likely targets [17]	Planting (low cost), may need continued care and will require harvesting though this may provide benefits Often requires chelating agents, e.g., ethylene diamine tetraacetic acid (EDTA) to be used in conjunction with plant action	Non-invasive method Low cost relative to most other techniques	Low intensity—long period of time required Biomass requires non-straightforward processing for metals recovery Low efficiency. Recovery limited by root distribution and depth; phytotoxic metal build-up may adversely affect plant; plants may not tolerate more than one or two specific metals, limiting applicability in mixed wastes Dependence on the soil acidity, seasonal and climatic conditions	Phytoremediation has had some commercial application but is uncommon Phytomining expected to be commercial in near future in combination with production of biofuel [17]
Electrokinetics	Any, though limited to electrically charged materials (particularly ions) for rapid recovery. Availability of resource for transport is critical	Power supply (typically 100 V/m separation of electrodes) May need to be coupled with an extractant/lixiviant if target is not dissolved	Can be applied successfully to very fine-grained materials with low hydraulic conductivity	Energy intensive Electroosmotic flow limited to particulates with high surface charge Unlikely to work where large metal objects present (e.g., municipal waste)	Applied but not mainstream technique

acceptors to enhance bioremediation), and also dewatering of soils [18–21]. There is a good level of experience on the application of EK techniques in soils, but only limited consideration in wastes (e.g., Pedersen et al. [22]).

Practically, the technique has previously been implemented at relatively shallow depths for removal of contamination, but there do not appear to be significant barriers to operation at greater depth.

Candidate Wastes and Waste Repositories

Candidate Rationale

The candidate materials generally fall under the classification of "controlled inactive stocks" [2] and are usually the result of current waste management practices. The following are candidate wastes which may be amenable to in situ recovery of metals. This consideration is based on the following reasoning: (i) there are extremely large mass arisings of these wastes; (ii) there are significant concentrations of metals of interest in them or that the matrix (once decontaminated) is valuable; (iii) there is evidence of hydrometallurgical research work being done on recovering metals from these materials (see Table 2); and (iv) they have suitable physicochemical characteristics (e.g., mineralogy, particle size, etc.). It is not an exhaustive list but highlights some of the key arisings which may suit in situ leaching. Furthermore, in all cases the recovery of land may be an important driver in progressing with mining of the wastes. Another key criterion for in situ techniques is hydraulic conductivity conducive to the flow of leachates. Many of the wastes do not meet this criterion but agglomeration and the use of EK are potential existing technological solutions.

Ore Processing Residues

Pyrometallurgical and hydrometallurgical processing of ores leads to a variety of wastes. Notable candidates for leaching are red mud, zinc refinery solid waste, and phosphogypsum. Research and development (and commercial recovery) is already underway for some types of ore processing residue, such as fayalitic slags, originating from processing of Co and Cu ores. Red mud is a waste arising from the Bayer process, the process by which aluminum is produced from bauxite ore. For every tonne of alumina produced, between 1 and 2 tonnes (dry weight) of red mud residues are produced [23]. The global annual arisings are 120 million tonnes and 2.7 billion tonnes of this material has been stockpiled [24]. It is composed primarily of fine particles of silica and aluminum, iron, calcium, and titanium oxides and hydroxides. Due to its high sodium hydroxide content, elevated pH, and metal(loid) content (Al, As, Cr, Cu, Ni, Mo, V, and Zn) [25], red mud is viewed as hazardous waste. These elements might also be considered a resource, and red mud also contains rare earth elements (REEs) and scandium [26, 27].

Zinc refinery solid wastes are iron mineral-rich, often acidic, sludge residues from various processing routes of zinc ores. Ju et al. [28] estimate at least 1 million tonnes per annum of residue arising in China alone, containing Zn, Pb, Ag, Cd, Cu, Ni, and In in a matrix of jarosite with zinc ferrite and anglesite. Various secondary process routes have been developed for recovery of valuable components from such residues, e.g., Zn, Ge, and In recovery [29], Zn and Ga [30], and Pb and Ag [31].

Phosphogypsum is the main waste product of the production of phosphoric acid by sulphuric acid digestion of a concentrated slurry of pulverized phosphate ores [32]. Phosphate ores are often naturally enriched in REEs, which report significantly to the process residues and can be recovered with acidic leaching (e.g., Preston et al. [33]). Example figures for REE content are given by Binnemans et al. [34].

Coal Fly Ash and Bottom Ash

While the UK usage of coal for power generation is rapidly decreasing as old coal-fired power stations are decommissioned and not replaced, the global consumption of coal for power has, for various economic reasons, actually increased and as such coal will likely continue to play a role in the global energy mix for many years. Waste arisings from coal combustion, principally fly ash and furnace bottom ash, have been variously utilized in applications such as a cement component in concrete, grout, and road bases but supply outstrips demand with only 25% being utilized globally [35, 36] and the remainder is stockpiled. Example figures for the EU arisings of coal combustion for energy generation residues for 2010 were 100 Mt [37]. Arisings are circa 6 Mt per annum in the UK alone, of which approximately 50% are utilized and an estimated 53 Mt of fly ash lies in the UK repositories [38]. It is recognized that there is a pressing need for more applications [39] but most actual applications and research publications concern fly ash utilization in civil engineering applications. In terms of resource recovery, coal fly ash contains REEs [40] and there has been interest in coal ash as a resource of REEs (e.g., Franus et al. [41]), especially La, Ce, and Y. There has also been interest in the leaching of aluminum from coal ash [42]. There may also be potential for in situ recovery of potentially contaminating species such as Cr(VI), V, and B where they occur at high enough concentrations to hinder ex situ reuse (particularly in unbound forms).

Mine Wastes

With annual extraction of ores and coal running to billions of tonnes, there is a commensurate and large production of mining wastes. Typical mine wastes include piles of overburden, waste rock, marginal ore, and tailings and mine water treatment sludges. In addition to these stocks, mine wastes (overburden and sub-ore stockpiles) form

Table 2 Examples of studies concerning leaching of wastes and challenges in their potential application to in situ direct recovery from repositories

Waste/ repository	Mineralogy	Amount	Challenges for in situ applications	Example leaching systems		
				Reference	Leaching target	Lixiviant (optimum conditions)
Red mud	Hematite, goethite, boehmite, quartz, sodalite, anatase and gypsum [83] Minor presence of calcite, whewellite and gibbsite	2.7 billion tonnes globally	Average particle size <100 μm) is not conducive to flow, permeability would have to be enhanced by agglomeration pretreatment or electrokinetics	Agatzini-Leonardou et al. [84]	Ti	Sulphuric acid (6 N, temp 60 °C, 5% S:L)
				Ochsenkühn-Petropulu et al. [85]	Sc, Yt, REEs	Dilute nitric acid (0.5 mol/L, 25 °C, 24 h S:L of 1:50
				Qu and Lian [86]	REEs, radioactive elements	Bioleaching with organism isolated from red mud (*Penicillium tricolor*) two-step process at 10% (w/v) pulp density
			Lixiviant strength not conducive to environmental protection or cost Elevated temperature not practical Leaching tends to solubilize iron	Vachon et al. [87]	Al	Citric/oxalic/sulphuric acids. pH 1.5, 28 °C, 1:6 *Thiobacilli* cultures, 5% v/v red mud and 1% w/v sulfur
Zinc refinery residue	Jarosite-type: jarosite, zinc ferrite and anglesite Goethite-type: goethite (containing higher conc of In, Ge, Ga)	Unknown but at least tens of millions of tonnes from literature anecdotes	Particle size (<200 μm) in not conducive to flow, permeability would have to be enhanced by agglomeration pretreatment or electrokinetics	Li et al. [29]	Zn, In, Ge	Sulfuric acid/potassium permanganate stage for Zn, sodium chlorate for In, Ge
				Raghavan et al. [88]	Pb, Ag	Brine leach followed by ppt with sodium sulfide
				Farahmand et al. [89]	Zn, Pb	Sulfuric acid leach for Zn (pH 2.5, pulp density: 200 g/L, leaching time: 60 min and temperature: 80 °C) followed by brine leaching for Pb
Phosphogypsum	Gypsum or calcium sulfate hemihydrate, Minor silica, fluoride and unreacted phosphate rock	Unknown but of the order of billions of tonnes from literature anecdotes	Recovery can lead to dissolution of the phosphogypsum lattice [26]	Habashi [90]	REEs	Ambient temperature with a 0.1–0.5 M H2SO4 solution in a solid-to-liquid ratio of 1:10
				Preston et al. [33]	REEs	1.0 M nitric acid and 0.5 M calcium nitrate
Coal Fly ash and bottom ash	Mullite, quartz, anhydrite, lime, magnetite, feldspar, glass	750 Mt of coal ash per year globally [91]	Particle size ultrafine (<10 μm) is not conducive to flow, permeability would have to be enhanced by agglomeration pretreatment or electrokinetics.	Seidel and Zimmels [42]	Al	Sulfuric acid leaching for up to 12 days
				Moreno et al. [92]	Decontamination (As, B, Cd, Cr, Mn, Pb, Se, V)	Deionized water, upflow leaching
Mine wastes	Highly variable	See text	Danger of exacerbating pollution generation	Bulaev et al. [93]	Metals from flotation pyrite tailings (Zn, Cu, Au, Ag)	Percolation leaching 134 days leading to bio-oxidation
				Mulligan et al. [94]	Cu, Zn, Ni	Bioleaching at 20 °C with *A. niger*

Table 2 continued

Waste/ repository	Mineralogy	Amount	Challenges for in situ applications	Example leaching systems		
				Reference	Leaching target	Lixiviant (optimum conditions)
Steel-making dusts	Franklinite, iron, wustite, zincite, and haematite	0.02 t/t of steel	Fine particle size (<50 μm) not conducive to flow, permeability would have to be enhanced by agglomeration pretreatment or electrokinetics Recalcitrant mineralogy Leaching solubilizes Fe	Raza et al. [95]	Zn	0.816 M Sulfuric acid, (65 °C temperature, 44–63 μm particle size, 20 mL g^{-1} liquid to solid ratio
Dredged Sediments	Variable	See text for figures	Particle size fine (<50 μm) not conducive to flow, permeability would have to be enhanced by agglomeration pretreatment or electrokinetics	Zeng et al. [96]	Decontamination (Cd, Zn, Cu, Pb)	5 days bioleaching with *A. niger* followed by 2 g/L peroxide leach
Soils	Variable	Unknown	Lixiviant strength not conducive to environmental protection	Fedje and Strömvall [97]	Decontamination and recovery of Cu	Highly acidic process wastewater (pH around 0)

another group which although excavated by human agency have not been further processed so straddle the technosphere/lithosphere resource definitions.

For materials that are excavated, the classification of the materials as 'waste' is contingent on factors such as the value of the target metals at the time, their ease of extraction from the materials and the extraction process used for the particular site. Thus, what at certain times is considered a 'waste' is at other times an 'ore.' Tailings are the waste resulting from the beneficiation of mined materials to produce an ore concentrate for further processing. The tailings are thus concentrated in gangue (non-economic) minerals, but no beneficiation process is completely efficient and technologies are variously effective at separating ore from gangue minerals, so that a proportion of the target metals will report to the tailings. It is also the case that historic beneficiation processes were typically less efficient, and with changes in metal prices and technology the remining of old tailings or other mine wastes has occurred. For example, historic Pb–Zn mines in Wales (UK) were originally mined for Pb in the late 19th century and the Zn minerals were discarded; these wastes were later reworked for Zn when Zn prices rose in the early 20th century. There is therefore the possibility that metals that were not originally the target metals for mining, but have since become economically important (e.g., the EU critical metals), maybe be enriched in the waste and become the

valuable target (e.g., Dehain and Fillipov [43]). In terms of physical characterization, mine wastes tend to have a larger and wider particle size distribution than tailings which are typically fine (<60 μm), and thus hydraulic conductivity of the materials for in situ leaching is an important consideration.

Steel-Making Dusts

There are a range of wastes arising from the multitude of metallurgical processes and the potential exists to recover resources (e.g., Fe, Zn, Mo) from them with a range of hydro- and pyrometallurgical techniques. Wastes include slags, dusts, and sludges from metal finishing processes.

Steel-making dusts including blast furnace, basic oxygen steel (BOS), and electric arc furnace (EAF) dusts are considered here as they constitute a large metallurgical waste stream. The BOS process involves the introduction of oxygen into the crucible of molten pig iron in order to reduce its carbon content. This creates waste dust up to 0.02 t/t of steel. These dusts cannot be recycled into the steel-making process for the iron content because of the presence of Zn (circa 10%) [44] as a contaminant which has an adverse effect on the process performance and eventual quality of steel [45]. Zn is present because galvanized scrap is often used as an additional charge to the process and the Zn is volatilized into the furnace off-gas

where it cools and forms zinc oxides and zinc ferrites (e.g., franklinite) in the dusts. The Zn in the dusts is amenable to acidic hydrometallurgical extraction (e.g., Havlik et al. [46]) as well as pyrometallurgical extraction (e.g., Jaafar et al. [47]) but poor separation between Fe and Zn extraction means that further processing is often required. A major route for recycling scrap is the use of electric arc furnaces. EAF dusts from this process are also very rich in zinc (>20 %) with franklinite being a major phase in the dust. Manganese, cobalt, nickel, and chromium also occur isomorphously substituted for Zn in the franklinite. Fe is present (other than in franklinite) mostly as magnetite, Fe_3O_4 [48, 49].

Metallurgical dusts are a more likely target than metallurgical slags from a mobilization/dissolution perspective—although the dusts are composed of recalcitrant Fe and Zn oxides; they have a very small particle size, and thus limitations of solubility are somewhat mitigated. Again, with this comes the penalty of the reduced hydraulic conductivity for an in situ leaching process.

Dredged Sediments

Annually, approximately 40 million wet tonnes of sediments are disposed of in approximately 150 licensed disposal sites around the coast of England [50]. It is estimated that there are 100–200 million cubic meters dredged per year in Europe alone [51]. The main reasons for dredging include navigation, maintenance of operational shipping depth, flood control, construction and reclamation, and mining for infill materials. Reuse options for dredged sediments include both conditioning of agricultural land and fill applications. However, a considerable proportion of dredged materials are contaminated, which precludes reuse in many (especially unbound) applications. Of the approximately 300 million m³ of sediments dredged to deepen harbors and shipping lanes in the US, 3–12 million m³ are considered highly contaminated [52]. Metal removal from contaminated sediments would have the dual role of recovering metals and potentially allowing reuse of the decontaminated sediment. Several studies have examined soil flushing and/or heap leaching for decontamination of dredgings, [52–56]. In one study, 62% of the metals were leached in a 120-day percolation leaching test [55]. Akcil et al. [51] summarize the multifarious approaches that have been applied to metal recovery from aquatic contaminated sediments.

Landfill Soils

Metals have not historically been a target for recovery from MSW leachates as the leachates do not contain economically recoverable amounts of metals. Kjeldsen et al. [57]

report that leached metals may account for less than 0.02% of the total metals present. The majority of the metal mass resides within the landfill in largely insoluble metallic (i.e., zero-valent) components. While there is scope to increase this substantially, dissolution of these wastes to later upcycle back to the native metals might be excessively energy intensive compared to ex situ landfill mining and physical separation to recover the metals. Ex situ landfill mining is starting to become a serious proposition, and Krook et al. [1] point out that soils from landfills systems have been a major target of previous landfill mining efforts. Kaartinen et al. [58] suggest that the fine fraction of landfill may comprise soil materials used for, e.g., landfill intermediate covers and can account for up to 74% of the landfill mass. As such, the fate of the fine fraction may be crucial for the economics of a landfill mining project, having to re-landfill soils because they are contaminated would (at least in Europe where landfill taxes are common) render the landfill mining uneconomic. Landfill fines (fraction <75 μm) have been found to contain up to 30% metals, and high concentrations of Zn, Cu, Cr, and Pb in particular have been reported [59]. Soil flushing or EK (see above) could be a useful decontamination/metal recovery technique which may render this large amount of soil a higher value. Thus, in this instance, a leaching approach might be applicable to materials recovered post ex situ mining.

Hydrometallurgical Considerations

The success of in situ leaching as either a mining or remediation process depends on whether the component of interest can be (i) solubilized from the matrix in which it resides at a rate which gives a realistic timescale for the process and then (ii) transported out of the system to be recovered. The manifold considerations for the first step are well known within the field of hydrometallurgy and thus several universal hydrometallurgical concepts are important in any in situ leaching scenario.

The bulk solubility of wastes and the solubility of individual metals of interest in the lixiviant is dependent on the mineralogy, particle size, and crystallinity of the waste. The rate of recovery of metals will be contingent on the degree of liberation of metal-bearing minerals and their leaching kinetics. Wastes such as smelter slag, while metal-rich, may be vitreous and practically insoluble in all but the most aggressive lixiviants and thus not suitable for in situ leaching. Other metals may be soluble but physically occluded within the interior of large particles, which restricts mass transfer to the dissolved phase. Where metals within wastes are not sufficiently soluble in water alone, a lixiviant can be selected, which significantly enhances

solubility through pH change (e.g., sulphuric acid), redox manipulation, complexation/chelation, or by in situ conversion by reaction of the recalcitrant phase into a more water-soluble or environmentally reactive phase. This is where hydrometallurgical considerations for recovering metals from wastes come to the fore.

The choice of lixiviant will depend upon the deportment of metals within minerals comprising the waste and whether they can be solubilized by (bio)geochemical manipulation. Many existing heap leach processes use acid leaching (e.g., sulfuric acid in Cu and Ni recovery) or complexation (cyanide lixiviant for Au/Ag recovery or $NaHCO_3$ for uranium recovery). Chelation (multidentate complexation) has also been used extensively for treatment of contaminated soils. While ethylene diamine tetraacetic acid (EDTA) has been investigated (e.g., Sun et al. [60]), it is toxic and research into new biodegradable selective chelating agents is underway (e.g., Wu et al. [61]). There are also a range of microbially produced chelates that might be applied to enhance solubilization of target metals; for example, *Acidithiobacillus thiooxidans* and *Aspergillus niger* have been tested for application to the leaching of steel slag [62], while Cheng et al. [63] examined bioleaching from Pb/Zn smelter slags. A recent and thorough review on bioleaching of solid waste and secondary resources was conducted by Lee and Pandey [64]. Table 2 shows examples of (bio)hydrometallurgical systems from the literature which might be useful for in situ leaching of candidate waste repositories.

Control of Preferential Flow

Lixiviant contact with the metal-bearing particles and transport of the solubilized components from the wastes is clearly key to successful leaching of metals. Therefore, the control of preferential flow within the waste mass itself is likely to be a critical and challenging issue for the success of in situ leaching as it is within the mining industry. The efficiency of leaching techniques can be significantly affected by preferential flow, which occurs in porous media as a consequence of non-idealities at a range of scales. Resistance to hydraulic flow through such media is affected by pore structure and properties such as throat size, volume, particle angularity and orientation, etc. This causes fluid flow to be non-uniform both at the microscopic scale and at the macroscopic scale due to features such as stratigraphy, lenses, and other structural features in soil deposits. In rocks, preferential flow is governed primarily by distribution, size, and orientation of fractures rather than the porosity of the rock itself. Preferential flow has caused issues in remediation of contaminated soils, where technologies such as pump and treat require uniform flow to remove all contamination [65], and in agriculture where the loss of pesticides and fertilizers occurs due to non-uniform flow, with consequent environmental impacts [66, 67].

Wastes are, to a greater or lesser extent, heterogeneous materials and some (municipal wastes in particular) also change with time due to degradative effects. As with all varied particulate materials, they are susceptible to preferential flow, and this flow may in some cases be temporally variable. The likely target wastes described in "Candidate Wastes and Waste Repositories" section are all particulate and will all exhibit heterogeneity at a range of scales depending on both the nature of the grains (including size, shape, material variability, and durability) as well as the nature of their introduction into, and their distribution in, the repository (including packing and compaction, spatial distribution, saturation, and flow of pore fluids). Heterogeneity is an inherent property of all particle arrangements unless perfect 'crystalline' structures, with identical packing, and orientation of particles, can be obtained throughout the waste mass—this is practically unattainable. Particles of wastes such as ore processing wastes, steel-making dusts, and ashes, whose nature depends on the parent material and the process to which it is subjected, may be relatively uniform unless the process parameters change during production, but even with these heterogeneity will always arise in their arrangement within the repository (microscopically and macroscopically), thereby causing preferential flow.

In situ technologies for recovery of resource from waste bodies require fluid flow through the waste, and to maximize recovery from all parts of the waste body is likely to require some form of flow control. Control of preferential flow is not common practically. There are, however, examples of technologies in the scientific literature that describe potential methods of control and these are discussed below.

Hydraulic Flow Control and Non-idealities in Flow Processes

When using leaching to recover materials, flow is usually applied to the system. The manner in which flow is applied may help avoid preferential flow. In highly stratified deposits, for example, macroscopic preferential flow may possibly be overcome by applying flow perpendicular to the deposit orientation although at smaller scales it is likely to still occur. Cote et al. [68] describe how flow interruption can increase solute leaching efficiency by up to 20% compared to continuous leaching. Drainage of main flow paths was also practiced between leaching events. The effect was attributed to equalization of concentrations in any undrained zones during the periods of flow cessation. Note that the efficiency of leaching may not be in terms of

mass removal versus time, but in mass removal versus time of flow: efficiencies may therefore reduce overall operational costs but timescales may not be significantly affected.

Hydraulic flow controls are highly applicable and require little additional development. It is likely, however, that in practice it will prove more difficult to fully overcome preferential flow with such methods. It is not thought that the method has been applied in resource recovery, but has been considered in contaminated land remediation [68].

Where the particle sizes or ores are prohibitively small in terms of ensuring sufficient hydraulic conductivity for the lixiviant, then agglomeration techniques (using water and/or binders) have been applied to fines to improve lixiviant contact and flow in heap leaching, resulting in good recoveries even for fine-grained materials. However, agglomeration may not be feasible, particularly when using strictly in situ methods, and does not necessarily fully overcome the problem of preferential flow.

Cut-off Walls and Permeation Grouting

Many techniques for controlling groundwater borrowed from contaminated land control would be applicable for bulk control of groundwater/lixiviant flow during in situ applications. An excellent summary of these techniques is provided in LaGrega et al. [69], including circumferential and downgradient cut-off walls (keyed into an aquiclude), soil-bentonite and cement-bentonite slurry trench cut-off walls, plastic, concrete, diaphragm, and vibrating beam cut-off walls. Grout curtains as well as proposed horizontal barriers including grouted liners and lagoon sealing techniques are also considered. Furthermore, LaGrega et al. [69] describe pump and treat, drain tile collection systems (French drains), and biopolymer extraction/interception trenches used for the capture of contaminated groundwater, which are thus directly applicable in the collection of lixiviant in in situ leaching of waste repositories.

Injection of grout under pressure has been considered in the creation of impermeable barriers, for example underneath existing waste sites where vertical trench construction would not apply. Typically, this is only applied to coarse-grained soils (sands and gravels) where standard cement-based grouts are used but the use of ultrafine cements and other materials has allowed the use in finer materials. Alternative technologies such as using chemical grouts (where the reactants are entirely liquid or soluble) or jet grouting may extend this. In terms of use as a control of preferential flow, the technology has not previously been considered, as usually the aim is to make an entire soil mass impermeable. However, Dwyer [70] reports that heterogeneity in the finished product may be due to preferential flow of the injected material and therefore the use

of the technique (for creating an impermeable barrier) should be limited to homogenous soils. It should be self-evident that flow will occur in preferential flow paths at least initially and so there is potential for this technology to be adapted for use in in situ mining.

Bioclogging and Biomineralization

Biofilms occur in soils, sediments, and many other environments and basically comprise large amounts of biopolymers exuded by microorganisms. When conditions allow, they can grow and extend, leading to clogging of pore space and flow diversion. Reductions in hydraulic conductivity of several orders of magnitude have been observed in soils [71], leading to diversion of fluid flow [72]. Biofilm has been considered as a hydraulic barrier to prevent escape of contamination via pore fluid transport [73]. In shorter term applications, biofilms have been shown to be successful in diverting flow, for example in microbially enhanced oil recovery (MEOR) where biofilm growth in preferential flow paths within oil reservoirs diverts fluid flow and increases recoveries of oil by flushing [74]. The ability of biofilm growth to control and divert groundwater flow in heterogeneous aquifer materials is being explored for contaminant remediation in soils (Al-shiblawi and Harbottle, unpublished results), showing that water flow of water can be diverted through less permeable paths due to bioclogging of the preferential flow paths.

A number of bacterial species have the capability to generate minerals (biomineralization). The mechanisms involved are varied and result in a range of mineral types, such as carbonates, sulfides, and others. A common mechanism is calcite generation instigated by urea hydrolysis, which leads to an increase in carbonate ion concentration alongside an elevated pH. The production of such minerals in porous media reduces pore space, resulting in similar effects to those noted with biofilm—significant reductions in hydraulic conductivity have been noted [75].

Electrokinetic Methods

Application of electrokinetics (see "Contaminated Land Remediation Technologies" section) can overcome heterogeneity within the target media because it does not rely on hydraulic flow to bring about recovery or remediation. Flow of the electrical current occurs equally well, or better, through fine-grained materials as it does in coarser fractions due to potentially greater electrical conductivity of the pore fluid in such materials. It may be, therefore, that it is still affected by heterogeneity in that better recovery may come from fine-grained regions of waste, but it is often the case that materials of interest are associated with

the fines fraction [59], and so in this case heterogeneity may positively reinforce recovery. For similar reasons, the combination of electrokinetics techniques with hydraulic flow methods is likely to be necessary to bring about improved and sufficiently timely removal, with, for example, electrokinetic transport moving target metals out of fine-grained zones to preferential flow paths where they can quickly be removed using traditional hydraulic techniques.

Applicability

The techniques described in "Hydraulic Flow Control and Non-Idealities in Flow Processes" to "Bioclogging and Biomineralisation" sections directly control flow, and so could be coupled with any of the flow-based techniques described in "In situ technologies" section (any form of in situ leaching, as well as the very similar in situ and ex situ flushing). It would be necessary to ensure that the flow control method was compatible with the leaching method, however; particularly the lixiviants employed, as cementitious grouts, walls, and biomineralized materials such as carbonates in particular, are likely to be deleteriously affected by acidic lixiviants.

Electrokinetic methods for resource recovery essentially do away with the need for flow control either because the need for hydraulic flow is obviated entirely by relying on electrokinetic transport phenomena or because the electroosmotic flow is directly controlled by the electric field. Externally applied flow may not, therefore, be necessary. However, electrokinetic transport phenomena can be slow where distances extend beyond a few meters and so a combination of electrokinetic and flow-based techniques (as described in the preceding section) has considerable potential. Although not directly an issue of preferential flow, electrokinesis can enhance a number of resource recovery methods due to its highly controllable nature. Phytoremediation, for example, has previously been combined with electrokinetics to enhance the availability and rate of transport of contaminants to the rhizosphere, increasing the reach of the phytoremediation system [76].

Process Intensity and Metal Capture

Process Deintensification

Decisions on the intensity of the beneficiation process are based largely on economics related to the ore grade, mineralogy/metal deportment, and liberation. The same reasoning has been applied to the economics of remediation of contaminated land [77]. Because wastes are of ultra-low grade, lower intensity processing through in situ techniques

is attractive. This leads to the question of how far should and how far can the reduction in process intensity be engineered. The ultimate recovery and the recovery rate of the target species are clearly important as to the ownership of the waste liability. If the incentive for the in situ leaching is economic recovery of metals through leaching (direct recovery) or decontamination of the material (indirect recovery), then the process necessarily has to be fast enough to ensure economic viability, while if the driver is decontamination of orphaned industrial legacy sites (albeit with economic recovery of metals to offset remediation), then the required intensity might be extremely low. It is possible to picture the opposite end-points of a spectrum of process intensity for resource recovery processes: with active ex situ mining and intensive beneficiation of a waste material at one end, and completely passive leaching of a waste by rainwater and capture of metals at the other end over decades.

Metal Capture

The intensity of processing also dictates the intensity of metal capture from the aqueous stream once metals have been mobilized and recovered from the waste. The decision about process intensity will be based (as discussed above) on the various cost/benefits which inform metal recovery and decontamination of wastes. Inevitably, factors such as less-intensive leaching systems using non-aggressive, environmentally benign lixiviants and limitation of recovery by heterogeneous flow mean that the intensity of in situ processing of wastes will be much lower and the timescales will possibly be counted in years. Thus, the technology used to capture the metals leached from the system has to be of commensurate intensity. For example, in high-value process-intensive systems such as gold processing, the gold is captured by 'carbon in pulp' and the carbon stripped from the gold, whereas lower intensity leaching systems might use ion exchange resins.

Low-intensity leaching needs commensurate metal capture techniques. Useful analogues are being developed for the "passive" treatment of metalliferous mine waters. Such systems use a variety of (bio)geochemical engineering approaches to achieve immobilization of metals. Various processes of adsorption, cementation, (bio)precipitation through microbiological reduction and/or oxidation, and manipulation of pH can achieve the sequestration of metals. Thus, these technologies offer readily transferable options for low-intensity harvesting of metals which achieve (i) eventual decontamination of the mine waste, (ii) protection of the environment from metal pollution, and (iii) recovery of the metals. This could also be viewed as engineering the formation of future anthropogenic mineral deposits.

Sustainable Remediation Context: Resource Recovery, Gentle Remediation, and Ecosystem Services

An important consideration not taken into account in many cost/benefit calculations is ecosystem service benefits/dis-benefits as well as landscape services related to sociocultural aspects of wastes which are important in some cases, e.g., historic mines. Many waste repositories provide ecosystem services with respect to wildlife habitat and can be among other things important educational and heritage features of the landscape. Thus, when considering recovery of metals from waste repositories, these factors need to be taken into account. The lower the intensity of the process, the lower the impact (probably) on potential ecosystem services that the site provides and thus lower intensity approaches to resource recovery may add (if apportioned significant weighting in the decision making) considerably to the benefits in the cost/benefit analysis. Parallels can be drawn from contaminated land risk management which has evolved from a focus on containment, cover, and removal to landfill through to in situ and ex situ treatment technologies and more recently to the concept of gentle remediation options where soil functionality is retained (and thus the ecosystem services of that soil preserved) in addition to risk management [77]. These technologies use plant, fungal, and microbiological methods, with or without chemical additives, for reducing contaminant transfer to local receptors by in situ stabilization or extraction of contaminants and are considered as part of a new suite of sustainable remediation technologies. In situ recovery of metals from waste repositories where the dual purpose of decontamination and site reclamation is achieved also fits into this classification.

Toward Application

Technology Readiness Levels

In recent years, Technology Readiness Levels (TRLs) have been adopted by policy makers, scientists, and engineers in a number of areas as a way of quantifying an estimate of the maturity of a particular process or technology. While various TRL definitions exist, most range from a TRL of 1, where only the basic principles have been observed, to a TRL of 9, where the technology has been successfully applied in its operational environment. The TRLs adopted by the European Commission as part of its H2020 pro-gramme [78] are summarized in Fig. 1 alongside estimates of the current technological readiness of the various in situ technologies considered here. The estimates are based upon the assessment methodology adopted by Rybicka et al. [79]

with an initial first-stage allocation where TRL 1–3 was defined as being at the laboratory scale, 4–6 at the pilot scale, and 7–9 at the commercial scale followed by a second allocation to determine, where possible, a single TRL level.

It is acknowledged that these assessments are dependent on the authors' judgement and as such can only be classed as estimates. In some cases, the technologies are at different TRLs for different applications or in different environments. For example, bioclogging has been investigated for use in the recovery of oil from porous rock and in that context is close to actual proven implementation [80] so its TRL is perhaps 7–8, but as in the context of resource recovery this is a very different environment, and its TRL for waste repositories is judged here as being at 3–4. Similarly, permeation grouting has been used extensively for other purposes, and so practical issues regarding its implementation have been overcome, but in different situations, the TRL is therefore judged here as 4. Furthermore, electrokinetic methods are well established in the field of remediation [81] but again not in the context resource recovery resulting in the TRL of 6. It is clear that technologies derived from the mining sector are very close to full commercial implementation, as the environmental conditions are comparable, resulting in the TRLs of 8–9; however, a number of other technologies are a considerable way from even pilot-scale demonstrations.

Legislation

While national legislative structures vary, it is likely that there will be a number of legislative regimes that could potentially be applied when considering the application of in situ techniques for resource recovery. These regimes will largely be the consequence of legislation introduced to either regulate waste management practices, mineral resource extraction, or regulate the assessment and remediation of contaminated land. It seems reasonable to expect that this will initially be considered at a site-specific level, until a clear precedent has been set; for example, in the UK context this establishment of legal precedent is not an unusual process in planning and regulatory processes. Also as noted in Crane et al. [82] the European Union Water Framework Directive is likely to result in the requirement of more stringent pollution control-driven interventions, and in such cases in situ approaches may well be implemented to offset remediation cost.

Future Research Directions

There is much scope for the development of the in situ approach to technospheric mining, and fruitful avenues for future research include (i) in situ conversion of wastes to
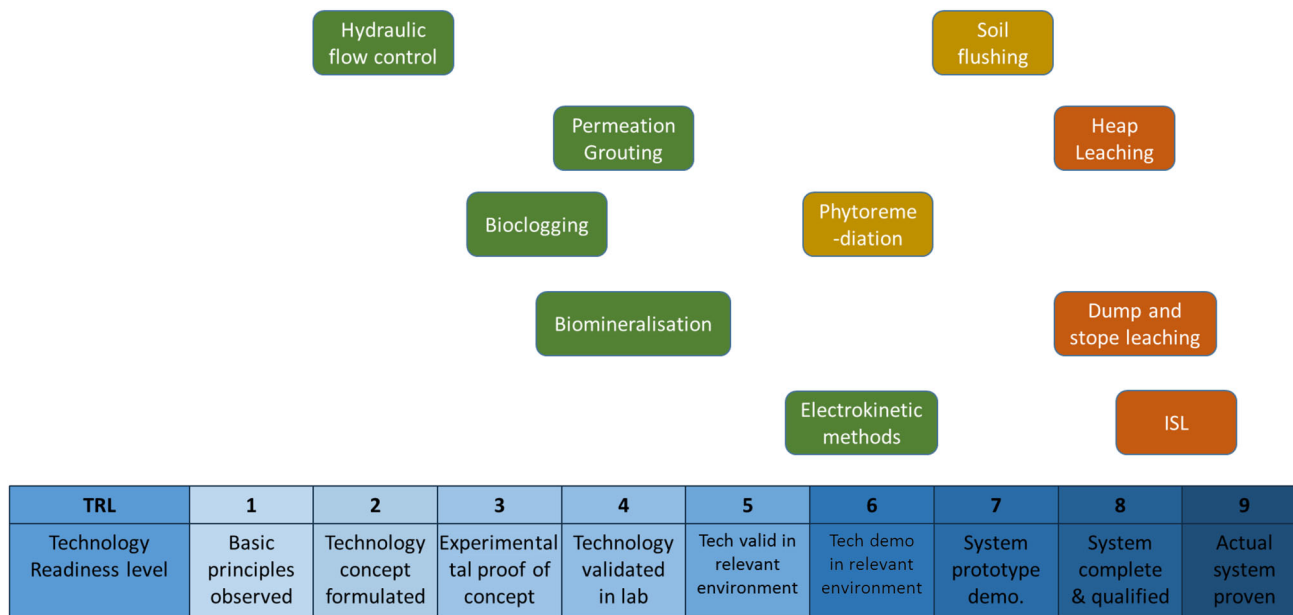
Fig. 1 Assessment of the TRL of technologies' applications to in situ recovery of resources from waste repositories

high-value products, (ii) the application of new metallurgical approaches that are highly selective to the target metals, (iii) novel technologies for the manipulation of biogeochemical environments within waste repositories to achieve dissolution of target species, and (iv) novel technologies for controlling flow in materials of low hydraulic conductivity.

International Perspective

Waste repositories are ubiquitous, both industrial and municipal, although the degree of engineering in such facilities varies significantly between countries and between waste types. In the UK, Europe, the US, and other areas where waste management is a mature industry, waste disposal in geological repositories is, in some cases, considered old-fashioned and there is a move away from such large-scale disposal of waste. An example is a shift toward greater incineration for municipal wastes. Resource recovery in these locations is therefore likely to focus on tackling residues from such processes alongside the large-scale industrial waste sources. The consideration of wastes as 'leakage' from the circular economy will therefore have to be bolted onto an existing industry, with the challenges and inefficiency that will likely result. In other parts of the world where waste management industries are developing, there may be more scope to intervene and to encourage the inclusion of a circular economy approach from the beginning, allowing these industries to leapfrog the current state of play elsewhere. For example, through design of waste repositories as temporary, rather than final, storage, and

allowing for low-intensity recovery processes, the concept of the circular economy may be thoroughly embedded within the waste management industry.

Conclusions

The in situ recovery of metals has been applied variously in mining and contaminated land environments although with different objectives. However, many aspects of both of the existing technologies are readily transferable to technospheric mining. This paper has outlined some of the key wastes where in situ leaching and related technologies could be applicable, outlined the similarities with existing processes and their geochemistry, and sought to outline the continuum of concepts between remediation and metal recovery and how this fits in with concepts of sustainable remediation. Future waste repositories from mining, metallurgical, and industrial wastes must be designed as "temporary storage" for future recovery of metal resources. Given the low grade of resources in typical wastes, these recovery techniques must necessarily be low intensity in terms of energy and reagent consumption. In situ recovery of metals involves mobilization, transport, and capture of the target metals in a concentrated form—the challenge is to devise various physical, chemical, biological, and biogeochemical engineering technologies to achieve this.

Compliance with Ethical Standards

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