1   # Circular economy landfills for temporary storage and treatment of mineral-

2   # rich wastes

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Abstract

42   Many countries face serious strategic challenges with the future supply of both aggregates and critical

43   elements. Yet, at the same time, they must sustainably manage continued multimillion tonne annual

44   arisings of mineral-dominated wastes from mining and industry. In an antithesis of Circular Economy

45   principles, these wastes continue to be landfilled despite often comprising valuable components, such

46   as critical metals, soil macronutrients and mineral components which sequester atmospheric CO_2. In

47   this paper, the authors aim to introduce a new concept for value recovery from mineral-rich wastes

48   where materials are temporarily stored and cleaned in landfill-like repositories designed to be mined

49   later. The time in storage is utilised for remediating contaminated materials and separating and

50   concentrating valuable components. It is proposed that this could be achieved through engineering the

51   repository to accelerate "lithomimetic" processes, i.e. those mimicking natural supergene processes

52   responsible for the formation of secondary ores. This paper summarises the concept and justifications

53 and outlines fundamental aspects of how this new concept might be applied to the design of future

54 repositories. The proposed concept aims to end the current "linear" landfilling of mineral-rich wastes

55 in favour of reuse as aggregates and ores.

56 **Keywords:** Waste management & disposal; Waste valorisation; Sustainability; Remediation; Recycled

57 material; Leaching; Landfills; Industrial wastes; UNSDG 9; UNSDG 11; UNSDG 12, UNSDG 13

1. Introduction

59 The aim of this paper is to introduce and explain a new technology concept for the management and

60 recovery of resources from high-volume mineral-rich wastes. There are recognised strategic challenges

61 with the future supply of aggregates, critical minerals and elements (Hayes et al., 2018; Gunn et al.,

62 2008; World Bank, 2017; Bazilian, 2018; MPA, 2018; EU, 2020; Sovacool et al, 2020). At the same

63 time, individual nations must sustainably manage the multimillion tonne annual waste arisings from

64 various economic sectors. Mineral-rich wastes are typically composed of a diverse range of mineral-

65 dominated materials that include waste rock and tailings materials from mining, residues and slags from

66 metal production, combustion ashes (fossil fuel, biomass, municipal solid waste incineration (MSWI)),

67 construction and demolition wastes, and contaminated dredgings (inland and coastal). Krausmann et al

68 (2017) estimate that the global solid waste flow was 9.7 Pg/yr (± 14%) for 2010, and that only 37% of

69 non-metallic minerals in end-of-life outflows from stocks were recycled. Recent mineral-rich waste

70 arisings statistics are not readily available but illustrative EU data (https://ec.europa.eu/eurostat/) reveal

71 some 1,537,090,000 t of mineral and solidified waste arisings in 2008 and 620,900,000 t of mining and

72 quarrying waste arisings in 2018. Although some of these materials do have accepted markets, these

73 are often limited to specific applications or the demand for the material may be small compared to

74 current (and projected increases in) production. Therefore, most of these materials are currently treated

75 as wastes, which continue to be disposed of in landfills or other engineered impoundments. This is

76 despite often containing valuable resources such as elevated concentrations of critical metals, soil

77 macronutrients and useful mineral components, some of which actively drawdown atmospheric CO_2

78 (Sapsford et al., 2017, 2019; Spooren et al., 2020; Antonkiewicz et al., 2020; Riley et al., 2020).

79   Although for many countries around the world, engineered sanitary landfill remains an aspiration for

80   solid waste management, other countries and supranational bodies such as the European Union are

81   looking towards ending reliance on landfill and are transitioning towards a circular economy (CE).

82   There are many definitions of CE and its precise meaning remains contested (see for example Kirchherr

83   et al, (2017); Korhonen et al (2018); Kalmykova et al (2019)), and critiqued (Corvellec et al., 2022).

84   However, the author's usage here is in line with common themes (particularly in the EU framing of CE;

85   McDowall et al, 2017) that include (i) closing materials loops through recycling and increasing resource

86   and materials efficiency and (ii) "using cyclical materials flows, renewable energy sources and

87   cascading-type energy flows" (Korhonen et al; 2018). A key part of this (following the waste hierarchy)

88   involves reduction in waste production, reuse, and recycling. Thus, opportunities to recycle, or upcycle

89   wastes are actively sought.

90   Despite enormous arisings, mineral wastes do not commonly feature prominently in public-facing CE

91   discourse. For example, despite England's "Our Waste, Our Resources" strategy focusing on the

92   development of CE and referencing the importance of tackling residual wastes and producer

93   responsibilities, many of the more tangible commitments relate to issues such as packaging, litter, fly-

94   tipping and residential recycling (HM Government, 2018). Another example is that extractive and

95   primary manufacturing industry wastes do not feature in the "butterfly diagram" used extensively in

96   communication of the CE principle (Ellen MacArthur Foundation, 2013). It is of note that mineral-rich

97   wastes are overlooked, considering the scale of production and the embodied carbon in such wastes

98   (Gomes et al., 2016; Zhai et al., 2021). This perhaps because in the popular imagination recycling of

99   post-consumer goods holds sway, whereas many pre-consumer wastes that result from the production

100   processes are 'out of sight' and therefore 'out of mind'.

101   Pre-consumer wastes split into two broad groups, clean unmixed materials, such as cardboard packaging

102   and metal swarf, which are easy to separate and have very high recycling rates; and, highly mixed

103   materials, often requiring decontamination, which are difficult to process, and therefore, are currently

104   uneconomic to recycle despite containing valuable components. Within many industries there are

105   efforts to view wastes as "secondary" or alternative raw materials, in keeping with the industrial

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symbiosis concept, where one industry's waste (or water/energy) serves as raw materials for the next or others in a cluster (Chertow and Ehrenfeld, 2012). Despite this, mineral-rich wastes are typically still landfilled and not used as secondary raw material. This can be attributed to three critical issues: (i) The technical or environmental constraints related to deleterious leachable components (e.g. Luo et al, 2019); (ii) The large volume of arisings and their low economic value as secondary materials; (iii) High-volume end-uses for the secondary material need to be (but are typically not) contemporaneous with their production. Expounding the latter points, bulk materials, whose unit value is low can be described as having a high "place value" (Shaw et al., 2013). This means that the financial and environmental costs incurred by transportation restrict their use geographically. Thus, if local end-use demand is not contemporaneous with waste production, storage becomes a solution to balance supply and demand. As a result of these constraints, landfill is currently the typical destination for most mineral wastes. Furthermore, there may be some reticence for use of secondary material where there may be (perceived or actual) variation in composition and uncertainty over geotechnical or geochemical performance leading to a preference for virgin materials (See Perkins et al (2021) and references therein).

The deportment of value (cf. "deportment" of metals in ore) is an important consideration for value recovery from wastes (Sapsford et al., 2019). The valuable target component(s) are either those that can be separated from the bulk material (direct recovery e.g. by leaching), or the residual bulk material, after removal of contaminants that prevent reuse (indirect recovery), or a combination of both (see Sapsford et al., 2017). There exists a very large body of research on hydrometallurgical, biohydrometallurgical and pyrometallurgical processes for extraction of valuable resource from mineral-rich industrial and mining wastes (see for example reviews by Jadhav and Hocheng, 2012; Sethurajan et al, 2018; Gunarathne et al, 2022). However, the sustainability (using the definition based on maintenance of genuine wealth, Arrow et al., (2004)) and economic viability of many of these processes requires careful consideration. To remove these metals, which are present at low concentrations, has consequences for increased energy demand, commensurate carbon footprint and economic cost over and above that for element recovery from primary ores (Sapsford et al, 2019). The energy demand and environmental cost of element recovery for low concentration ores (and by

extension of the same logic, wastes) is high. This can be illustrated by the example of platinum, where extraction from very low concentration primary ores is common and the embodied energy exceeds 100 GJ kg^{-1} and embodied carbon is 10,000 kg kg^{-1} (Gutowski et al., 2013). Thus, when considering direct and indirect value recovery from mineral-rich wastes, the energy required for the separation, be it of resource from residue, or contaminant from secondary raw material, needs to be from renewable sources to be sustainable.

2. ASPIRE: A new concept for temporary storage and treatment of waste

The authors propose a step-change in waste repository design for mineral-rich wastes, with a change in focus from solely environmental and health protection to one where environmental protection and resource recovery are designed in. The concept involves **A**ccelerated **S**upergene **P**rocesses **I**n **R**epository **E**ngineering (ASPIRE) and the authors refer to waste repositories following this paradigm as "ASPIRE repositories". Supergene processes are natural lithospheric weathering and pedogenetic processes involving downward percolation of meteoric water (i.e., derived from precipitation) and metal leaching from unsaturated metalliferous rock (Lelong et al., 1976) often enhanced by organic derived acids and ligands. Metals are then deposited as an enriched metalliferous secondary ore below the groundwater level during the transition to more chemically reducing conditions (Figure 1(a)).

The ASPIRE concept is a "nature-based solution" (Song et al., 2019), which involves mimicking these naturally occurring lithospheric mechanisms, such "lithomimicry" aims to achieve the same effect but with waste materials (Figure 1(b)). Interestingly, supergene alteration has been noted to occur in wastes such as mine tailings and smelter residues (Dill, 2015). Furthermore, the authors propose expanding the supergene concept to include other value propositions including phosphate recovery, carbon sequestration and the decontamination of the waste matrix. Because natural supergene processes can take millennia, there is a requirement to engineer the processes to accelerate them to anthropogenically relevant timescales. This will require innovative biogeochemical engineering such that processes

158 remove potential contaminants/resources from the bulk material matrix (leaving a cleaned residue) and

159 concentrate them within an anthropogenic ore zone over a prolonged period of waste storage.

160 ASPIRE repositories would be designed to significantly accelerate the ore-formation from the

161 geological timescales of natural supergene process to the order of years in the engineered system.

162 Critically, being a fully lined waste repository system, the external environment remains protected.

163 Inspired by research observations of revegetated industrial wastes (Bray et al., 2018), the ASPIRE

164 concept could also intersect with phytoremediation, phytocapping and phytomining concepts. Here,

165 environmental leaching agents ("lixiviants" in hydrometallurgical parlance) generated from plant roots

166 (which produce low molecular weight compounds that act as ligands for metals) accelerate mobilisation

167 of metals. Solar and self-powered electrokinetic and electrochemical phenomena are proposed for

168 acceleration of solubilisation and transport, followed by biogeochemical trapping of metals/elements,

169 potentially as new ores. Whilst it is impossible to "short-change" fundamental thermodynamics which

170 dictate the minimum energy requirement of separation of species from a parent material, it should be

171 possible to provide that energy in a sustainable way from the solar energy incident on the storage site

172 (insolation flux), directly via photovoltaic power systems or indirectly via photosynthesis (and the

173 environmental exergy cascade), albeit sacrificing the process intensity by prolonging the timeframe for

174 the process.

175 The central tenet of the ASPIRE concept is that the "time in storage" is used for material

176 cleaning/resource concentration. This has the dual benefits of facilitating future resource recovery and

177 defines the service-life for the barrier system. The dormant waste undergoes processes to (i) concentrate

178 valuable components (e.g. critical metals, phosphate) as an anthropogenic ore to facilitate their future

179 recovery, and (ii) concurrently decontaminate residual mineral material so as to make it available as a

180 bank of material to drawdown for "soft" end-uses in agriculture, forestry, greenspace, landscaping,

181 habitat creation (Song et al., 2019) and/or as a cement/concrete additive or replacement aggregate. As

182 such, the ASPIRE concept seeks to reconceptualise waste repositories as "temporary storage systems"

183 or "resource banks". This could involve regional ASPIRE repositories as hubs which import a range of

184 mineral-rich wastes arising in the region, in the UK context potentially contributing to the Managed

185 Aggregate Supply System. Alternatively, smaller site-specific repositories could be developed.

186 Importantly, the idea is not necessarily to displace any existing sustainable recycling activities for

187 mineral-rich wastes but to provide a practicable CE solution for materials that would otherwise go to

188 conventional landfill for lack of any other viable means to recover value. The authors suggest that this

189 concept thus fits into a modified waste hierarchy as shown in Fig 2.

190

191 3. The Case for Temporary Storage and *in situ* treatment

192 3.1 Existing concepts of temporary storage and 'End of Waste'

193 Temporary storage for MSW landfill has been proposed such that stored waste undergoes processes to

194 accelerate the "stabilisation" of the waste, to facilitate recovery of materials from the landfill mass or

195 the use of the land (Jones et al., 2013). The accelerated stabilisation is achieved either via active aeration

196 to promote the breakdown of organics (Ritzkowski and Stegmann, 2012), or the recirculation of landfill

197 leachate to accelerate anaerobic process and methane production (Reinhart and Al-Yousfi, 1996;

198 Warith, 2002), ultimately allowing a compressed timeline for land restoration or the recovery of

199 materials from the landfill mass. Temporary or 'interim' storage of municipal solid waste (MSW) has

200 been undertaken in Germany, in response to lack of a receiving market. Several million tonnes of

201 Mechanical Biological Treatment (MBT) sorted wasted was stored in interim landfills (Wagner and

202 Bilitewski, 2009). Temporary storage has also been mooted for e-wastes (Kahhat and Kavazanjian,

203 2010). Despite these examples, the practice of temporary storage has not taken hold more widely.

204 Functioning markets for recyclates are important so that there is contemporaneous demand for

205 materials. Yet even these relatively high-value recyclates are susceptible to market disruption. In 2017,

206 China banned the import of most plastic waste, this resulted in a sharp reduction in global plastic waste

207 trade flow (Wen et al., 2021). The fact that market volatility is commonplace in markets for relatively

208 high-value recycled resources emphasises the challenges of recycling low-value, high-volume and

209 contaminated mineral-wastes and why they are currently landfilled.

There has been some success in England and Wales in utilising secondary materials, this is also encouraged by a levy on virgin aggregates. "End of waste" (EoW) criteria have been developed for some mineral-rich wastes to facilitate reuse: notably, quality protocols developed in England and Wales by the Environment Agency and the Waste and Resources Action Programme (WRAP) for steel slag and pulverised fuel (coal) ash, furnace bottom ash, as well as a Code of Practice developed by MIBAAA (Manufacturers of IBA Aggregates Association in conjunction with the Environment Agency) for incinerator (MSWI) bottom ash. These approaches are framed around Article 6 (1) of the European Waste Framework Directive (2008/98/EC) which sets out end of waste status, and the conditions that if met enable waste which has been recycled or recovered to cease to be classed as waste. The substance or object must meet the following conditions: (i) It is to be used for specific purposes (ii) Market/ demand exists (iii) Achieves the technical requirements for the specific purposes and meets existing standards and legislation applicable to products (iv) Its use will not lead to adverse environmental or human health impacts. Thus, the materials referred to above need to demonstrate compliance with the quality protocol framework, as well as an appropriate standard such as BS EN 12620, Aggregates for concrete, and associated testing. In addition, markets may well be restricted, for example the Quality Protocol for PFA allows its use in bound or grout applications only.

Despite these examples, many mineral-rich wastes from mining and industry are still largely overlooked and thus poorly integrated into current CE strategies and developing policy (Cisternas et al., 2022). There is a lack of practicable, economically viable and sustainable technologies for returning these resources to the CE and the temporal and geographical dislocation between production and end-use (Cisternas et al., 2022). Providing a solution to the temporal dislocation is a key advantage of the ASPIRE concept, that once the material is cleaned, it stays in hibernation until end-user market arises. There are historical examples that demonstrate over a decadal scale, materials often thought as valueless waste later become highly valued resources, for example due to advances in recovery technologies, decreases in ore-grade or increase in value of specific components which were previously non-economic. For example, the re-mining of Pb/Zn spoil in 1970s / 80s in the northern Pennines (UK) for

236 fluorspar and Ba minerals, driven by demand for F in chemical industries and Ba for oil drilling fluids
237 (Dunham, 1985).

238 It is noteworthy that temporary storage and treatment as a concept is conceptually similar to the
239 "cluster" approach developed in the UK to facilitate the remediation of contaminated soils where a
240 number of sites are in close proximity. The sites share a "hub" for the central decontamination /
241 treatment of contaminated soils (CL:AIRE, 2021). This is similar to the concept proposed here, albeit
242 these remediation hub timescales are shorter, the relative intensification of remediation is economically
243 viable because of the value of land redevelopment.

244 **3.2 Future generations and conventional landfill**

245 Landfill remains the destination for wastes that cannot be combusted, composted or separated into
246 recyclates for which there is a current market value. Landfill philosophy has evolved from "dilute and
247 disperse" to a "store and contain" containment strategy, with the waste environmentally isolated by use
248 of engineered top and bottom liners (e.g. Cisternas et al., 2022). Allen (2001) critiques containment
249 strategies, raising concerns over the durability of liner systems and problems with clay liners.
250 Landfilling simply postpones the release of contaminants, as for future generations it is not a question
251 of "if" but simply "when" the containment systems for landfilled industrial wastes succumb to natural
252 degradation. This then leads to important questions about intergenerational equity for management of
253 wastes. Put simply, should the current generation be burdening future generations with pollution issues
254 from current waste production? This appears contrary to the sustainability agenda and UN sustainable
255 development goals (United Nations, 2015).

256 Whilst accepting that many of the drivers for diversion of mineral-rich wastes to landfill remain, the
257 authors contend that a sustainable and ethical philosophy would be "store, contain, clean and
258 concentrate" to allow future recovery. It is notable that future recovery of residues from the repositories
259 would likely be in the context of a decarbonised economy, thus future exploitation will likely be of low
260 carbon intensity.

261 It is interesting to consider temporary landfilling of waste in the context of a CE. A critical component

262 of a move to a CE is increased resource efficiency via the provision of a service-based economy. The

263 current landfill paradigm is a linear model, often with associated ownership/license surrender by the

264 operator. Where used, facility gate fees cover operational costs but neither the original waste producer

265 nor the landfill operator is paying (post-permit surrender) for the ultimate environmental impact of the

266 waste (i.e. these long-term impacts remain market externalities). Applying the ASPIRE concept changes

267 the landfill operator's function to the provision of a storage and remediation service, and the production

268 of secondary raw materials which will ultimately be removed from site, thus removing the long-term

269 environmental hazard.

270

271 **3.3 Alignment with current waste legislation**

272 To achieve a CE, it is essential to re-use and recycle materials from waste for future use in new

273 buildings, infrastructure, products etc, and to keep these in productive use as long as is feasible. Thus,

274 the treatment of waste materials to facilitate this is vital to deliver a CE. The Waste (England and Wales)

275 Regulations 2011 introduced a duty for waste importers, producers, waste carriers and waste

276 management facilities to "take all measures available to it as are reasonable in the circumstances" to

277 apply the waste hierarchy when transferring waste: (i) Prevention (ii) Preparation for re-use (iii)

278 Recycling (iv) Recovery (v) Disposal. The crux of this is the term "reasonable", which ultimately means

279 that aspects such as technical, economic and environmental aspects are considered in order to make a

280 judgement. Despite this legislation, landfill remains a commonly used option for mineral-rich wastes.

281 Whilst temporary storage and treatment may not be feasible for all landfilled wastes, the authors suggest

282 that it would be a practicable and sustainable solution for the many mineral-rich materials amenable to

283 processing via leaching. Because the ASPIRE concept involves waste materials being reprocessed into

284 products (clean residue and anthropogenic ore) it is in essence a recycling technology. However, the

285 confounding issues of the long-term storage and processing whilst stored in a "landfill-like" repository

286 will likely lead to challenge of its status as recycling technology in the eyes of the waste regulations.

287 At minimum, an ASPIRE repository could be viewed as providing a technology option at the base of

288 the waste hierarchy, replacing conventional landfill – and this may be a more regulatory acceptable

289 approach for early adoption of the technology. The Environment Act 2021 provides some potential

290 opportunities for the ASPIRE concept to gain acceptance in the UK. First, it confers power to the

291 Government to set regulations on producer responsibilities, including waste prevention and reduction.

292 It also enables regulations related to the re-use and recovery of materials.

293

4. Potential candidate materials for ASPIRE repositories

295 *4.1 Combustion Ashes*

296 Alkaline combustion ashes, bottom ash and fly ash/Air Pollution Control Residue (APCr) from

297 Biomass, MSWI, sewage sludge, co-firing of biomass/coal will be important wastes in the UK and

298 internationally for the conceivable future. By 2050, global arisings of biomass ash will be of the order

299 of 480 Mtpa (Vassilev et al., 2013). Furthermore, the UK's Sixth Carbon Budget (CCC, 2020)

300 highlights the important role of Bioenergy with Carbon Capture and Storage (BECCS) as one of the

301 key technology options for the UK in limiting the contribution to catastrophic global warming. BECCS

302 has been estimated to have a CO_2 removal potential of 20-70 $MtCO_2$ pa (Smith et al., 2016) and further

303 removal through carbonation of the ash arisings e.g., through enhanced terrestrial weathering. Biomass

304 ashes comprise major elements O, Cl, Si, Ca, K, P, Al, Mg, Fe, S, Na, Ti, Mn and are notably enriched

305 compared to coal ash in trace elements in Ag, Au, B, Be, Cd, Cr, Cu, Mn, Ni, Rb, Se, Zn (Vassilev et

306 al., 2013; Zhai et al., 2020). Heavy metals and readily soluble metal chlorides are the most recurrent

307 contaminants preventing ash reuse in many of its principal potential applications e.g. soil amendment

308 in silvi/agriculture and admixture in cements and mortars. Statistical analysis of numerous database

309 records shows that compositionally there are four main types of virgin biomass ash: hard wood ash,

310 softwood ash, and grass (straw) and non-grass type agricultural residues. Interestingly, wood ashes

311 have slightly higher trace metals, whereas agricultural residue ashes have notably higher levels of

312 persistent organic pollutants (POPs) (Zhai et al., 2021). Thus, the ASPIRE concept could offer a

313 practicable management/decontamination option for ash arisings from projected biomass combustion
314 and future BECCS with potential for further CO_2 mitigation through ash weathering, ash reuse as a
315 cement admixture and nutrient recycling to agri/silviculture whilst also recovering critical elements and
316 minerals.

317 There is also a clear trajectory for growth in MSWI ash arisings. Bottom ash is readily recyclable, but
318 market limitations mean that its reuse is restricted to prescribed applications, whilst MSWI fly ash/APCr
319 is considered hazardous which prevents recycling. Due to POP content, some 5% of MSWI APCr
320 requires further treatment before it can be disposed of to hazardous waste landfill. UK Energy from
321 Waste capacity was projected to be 15.4 Mtpa by 2020 (Tolvik, 2017), with estimated bottom ash and
322 APCr arisings of 3.1 Mtpa and 0.5 Mtpa respectively. MSWI ash can be enriched in Ca, Cl, K, Mg, N,
323 Na, P, Ti, trace elements Ag, Cd, Cr, Cu, Ho, I, Mn, Pb, Pr, Re, Sb, Sm, Sn, Zn (Lima et al., 2008; Tang
324 et al., 2015; Tang and Steenari., 2016; Zhai et al., 2021), the trace element compositions will be
325 susceptible to changes in waste composition.

326

327 *4.2 Dredgings*

328 The EU annually produces around 200 - 250 Mt of marine and 50-60 Mt of freshwater dredgings per
329 year, of which up to 5-10 % and <30%, for marine and fresh water respectively, are sent to "confined
330 disposal" (Mink et al., 2006). Example arisings from around the coast of England were 40 Mt of wet
331 sediment (Bolam et al., 2006). For inland UK contaminated dredgings data are sparse but the amounts
332 are likely in the range of hundreds of thousands of tonnes per annum. As with ash, the heavy metal
333 content of dredgings often prevents their reuse in applications such as bank protection, habitat creation
334 and conditioning agricultural land (Renalla, 2021). ASPIRE repositories could offer the practicable
335 means to create habitats and green corridors close to damaged waterways while also providing a means
336 to return metals that have escaped into rivers and bound up with sediment to the CE and bioremediating
337 any associated organic contaminants.

338

4.3 Alkaline Industrial Wastes

Industrial processes produce a very wide variety of different wastes, many of which are readily recyclable. However, several classes of mineral-dominated residues are produced during metal production that lead to high-volume, low-value materials that are uneconomic to recycle and commonly disposed of in land-based repositories. These include, steelmaking and metallurgical slags, bauxite processing residues, Solvay process wastes from the manufacture of soda ash, and chromite ore processing residues. In terms of global production, steelmaking slags and bauxite residues are particularly important and have rapidly increasing rates of production (170-250 Mtpa and 120 Mtpa respectively (Power et al., 2011)). These residues are often the product of high temperature processes that involve additions of alkali materials during extraction. Therefore, the resultant by-products are rich in alkaline mineral components (e.g. CaO, $NaOH$, Ca and Na – silicates) that readily hydrate and dissolve to produce a highly alkaline leachates (Gomes et al, 2016).

After uses for the bulk residues can be limited by a range of factors which include (i) extreme alkalinity and mobility of potentially hazardous metal(loid)s at high pH (e.g. As, Cr, V: Burke et al., 2012; Hobson et al., 2017), (ii) stability issues where weathering of residues can lead to expansion and limit aggregate after uses, (iii) the potential classification of some residues such as bauxite processing residue as Technologically Enhanced Naturally Occurring Radioactive Materials (TENORM) which can limit after uses in construction (e.g. Somlai et al., 2008). As such, virgin materials are often preferred over bulk reuse of alkaline residues given the additional costs associated with mitigating potential issues.

There is growing research in value recovery from alkaline industrial residues ranging from bulk material reuse to critical metal recovery and carbon capture (e.g. Gomes et al., 2016; Ujaczki et al., 2017; Pullin et al, 2019; Pan et al, 2020). However, relatively low fluxes of dilute valuable components (e.g. V) occurring in leachates with mixed contaminants makes separation and recovery challenging (e.g. Gomes et al., 2017). As such, there are few examples of large-scale critical raw material recovery from alkaline industrial wastes.

364   The ASPIRE concept brings a range of opportunities to overcome some of these current technical

365   constraints. The extended timescales on which ASPIRE is based provides scope for the gradual

366   leaching and accumulation of critical metals in enrichment zones that would be better candidates for

367   recovery than current technologies (e.g. Gomes et al., 2020). The decontamination of bulk material by

368   these leaching processes could produce more stable and less hazardous materials that could open up

369   more avenues for large scale reuse as aggregate (e.g. slags), in land reclamation or in ceramics / building

370   materials (e.g. bauxite residue). Alkaline wastes also have the potential to sequester atmospheric CO_2 -

371   See Section 8 below.

372

373   *4.4 Mine wastes*

374   Globally, arisings of mine wastes (including overburden, tailings and a range of related residues) likely

375   run to hundreds of billions of tonnes (Kinnunen and Kaksonen, 2019), for example there are an

376   estimated 39 billion tonnes of mine tailings in Chile alone at 0.2 wt.% Cu (Dold, 2020). Mine wastes

377   are deposited in a range of settings from uncontrolled storage in the landscape to highly engineered

378   impoundments, depending on their age and the environmental legislation relevant in the geographic

379   jurisdiction. Mine wastes deposited to land in an uncontrolled manner cause a substantial pollution

380   legacy through acid rock drainage and metal-leaching (e.g. Wolkersdorfer and Bowell, 2004), and

381   migration of fines (e.g. Fuge et al., 1989). Impoundments can pose risks through dust and failure of

382   dams (Dold, 2020). In the UK, an estimated 73 – 195 Mt of mine wastes were produced from the

383   historical working of lead, copper and tin, and cover an estimated 1960 – 4400 hectares of land

384   (Palumbo-Roe and Colman., 2010). The indicative estimated cost, over a 10-year life cycle, to

385   remediate solely the water-related environmental problems due to diffuse pollution caused by non-coal

386   mine wastes in England and Wales was estimated at £35 Million (Jarvis et al., 2012).

387   There is considerable interest in recovery of resources from mine wastes, and they are one of the more

388   obvious targets for sourcing of metals, particularly as ore grade continue to decrease and/or aggregate

389   resources decrease. Despite this, there has been very limited explicit adoption of the concept of the

390 circular economy amongst large-scale mining firms (Upadhyay et al., 2021). Large bottlenecks for

391 tailings valorisation identified by Kinnunen and Kaksonen (2019) include the low concentration and

392 mass of recoverable elements for economical processing, missing value chains, regulatory barriers, and

393 environmental risks of reprocessing. Applying the ASPIRE repository concept for future mine wastes

394 and tailings could address many of these concerns and at minimum a better option than the "do nothing"

395 option of continued landfilling.

396

397 *4.5 Temporary Storage as a Remediation Strategy for Legacy Wastes*

398 Where "polluter pays" legislation (see Stenis and Hogland, 2002) is in place then there is a compelling

399 case (from the perspective of environmental justice) for legacy mineral-rich wastes to be managed by

400 the generator. However, many legacy mineral-rich wastes remain where the waste generator has no

401 liability (due to lack of action-forcing legislation at the time), or no longer exist. In the UK, such legacy

402 wastes and orphan sites become the responsibility of local authorities and/or central government. The

403 ASPIRE concept could offer a potentially lower cost alternative to conventional site remediation, which

404 often leads to secondary landfilling of heavily contaminated materials in any case. In a sense, this would

405 be a revisitation of the "dig and dump" approach to site remediation that was common in the UK but

406 has become less common due to landfill tax (Hodson, 2010), with the critical difference that the "dump"

407 is into a temporary storage repository where the waste is remediated to allow future recovery.

408 Multi-dimensional value refers to the value as expressed by the range of measurable benefits and

409 impacts in the environmental, economic, social and technical domains (Iacovidou et al, 2017). Many

410 legacy sites in the UK, for example mine sites, display multi-dimensional value. For example, important

411 landscape and cultural designations, sites of special scientific interest and/or host rare and valued

412 species of flora and fauna. This contrasts with the negative value associated with sites that cause

413 environmental pollution (Crane et al, 2017; Sinnett, 2019). It is important to note that an ASPIRE

414 repository could feasibly be constructed *at* a legacy site, with some, or all, of the valuable aspects of

415 the legacy site (which tend to be associated with the upper surface) retained or recreated in the repository

416 cover.

417

5. Accelerating supergene processes: Lithomimicry and Biogeochemical Engineering

420

5.1 Storage time

422 The requisite duration of the storage of wastes within an ASPIRE repository is an open question but

423 one that dictates the necessary intensity/acceleration of the *in situ* supergene processes. Conceptually,

424 it is possible to envisage (i) shorter-term storge duration and processing of a few years (the system

425 resembling a contaminated land remediation project) (ii) medium term storage and treatment over one

426 to a few decades, through to (iii) long-term intergenerational storage and treatment on a timescale of

427 several decades to a century. It is envisaged that the *in situ* processes could be engineered to correspond

428 to the likely time in storage.

429

5.2 Upper surface and leaching zone

431 The primary hazards to humans and other vertebrates from industrial residues tend to result either from

432 direct contact with the residue, ingestion or inhalation of dusts, or ingestion of rainwater run-off

433 (Nathanail and Earl, 2001; Stange and Langdon, 2016). All these hazards can be significantly reduced

434 if vegetative cover is established on the residue surface, as plant growth will buffer the chemistry of the

435 surface layer and minimise dusts and suspended solids in runoff. When revegetation occurs naturally

436 at a site that is initially devoid of life (e.g. after deposition of volcanic ash) the process of initial

437 colonisation by pioneer species, and their subsequent replacement by a wider range of less hardy

438 species, is called primary succession (Breeze, 1973; Gemmel, 1972; Bradshaw, 2000; Wong, 2003;

439 Phillips and Courtney, 2022). Without intervention this process, which involves the slow build-up of

440   plant accessible nutrients with successive cycles of plant and microbial growth and decay, typically

441   takes decades and centuries to produce verdant vegetative cover (Ash et al., 1994; Lee and Greenwood,

442   1976; Bradshaw, 2000; Santini and Fey, 2013). However, many industrial residue disposal sites could

443   be transitioned from providing little benefit for people or nature to delivering multiple benefits, or

444   ecosystem services, in few years if this natural process is accelerated.

445

446   Revegetation and the potential for accelerated ecological succession has been demonstrated on acidic

447   coal mine (Wali, 1999; Fernandez-Caliani et al., 2021) and metalliferous mine wastes (O'Neill et al.,

448   1998; Bagatto and Shorthouse, 1999; Wong, 2003; Walker et al, 2004), circumneutral metalliferous

449   mine wastes (Shu et al, 2002; Yang et al, 1997; Sarathchandra et al, 2022; Peñalver-Alcalá et al, 2021),

450   and on alkaline wastes (Breeze, 1973; Courtney et al., 2009; Fellet et al., 2011; Lee and Greenwood,

451   1976; Ash et al., 1994). Interventions at industrial residue disposal sites that can accelerate the

452   succession process include conditioning treatments (to provide nutrients, create soil-like structure and

453   buffer extreme pH values) and seeding of plant mixtures (Ash et al., 1994; Courtney et al., 2009;

454   Kumpeine et al., 2008). Grass species appear to be good pioneer species, as species from calcareous

455   grassland appear readily established on alkaline wastes and species from acidic heathland on acidic

456   wastes (Ash et al., 1994), but early establishment of nitrogen fixing species (such as legumes) is

457   important for accelerated succession (Phillips and Courtney, 2022). Over the last 20 years Courtney and

458   co-workers have investigated the revegetation of bauxite processing residue (red mud); a very alkaline,

459   abiotic waste (Courtney and Timpson., 2004, 2005; Courtney et al., 2009; Courtney and Harrington.,

460   2012; Courtney et al., 2014). Their approach has been to augment the surface layer of the residue with

461   various combinations of sand-sized material to improve drainage, gypsum to buffer high pH, and

462   organic matter to provide a suitable substrate for plant growth, and they have shown that vegetative

463   cover can be established from seed and sustained for more than 20 years. A subsequent investigation

464   has shown that plant roots become established mainly in the amended surface layer, but vegetative cover

465   resulted in a pH reduction >2 pH units and a five-fold reduction in sodicity (compared with an untreated

466   and therefore largely unvegetated control plot) that extended to more than 3x the initial treatment depth

18

467   (Bray et al., 2017). Such beneficial effects from bio-rehabilitation have also been reported elsewhere

468   (Santini et al., 2015; Zhu et al., 2016; Sarathchandra et al., 2022).

469   Plants can alter the chemistry of any residue upon which they are grown through the chemicals they

470   secrete through their roots. These include CO_2 from respiration in roots; ions transported across the

471   soil-root interface during nutrient uptake, extrusion of H^+ by plant cells during energy metabolism, and

472   secretion of various organic species (termed "root exudates") (Hinsinger et al., 2003). Plants can secrete

473   a broad array of organic compounds into the rhizosphere, and while the flux is quantitatively quite

474   modest (Guo et al., 2010; Hinsinger et al., 2013), it represents 5% to 21% of the carbon

475   photosynthetically fixed by the plants (Helal and Sauerbeck, 1989; Marschner, 1995). Some root

476   exudates are produced in response to specific plant stresses, but the majority (including sugars, amino

477   acids, and organic acids) are believed to be passively lost from the root (Canarini et al., 2019). Organic

478   acids and phenolics released by plants (including those released to combat excessive aluminium in

479   acidic soils (Li et al., 2009; Mora-Marcias et al., 2017) and siderophores to that allow Fe uptake in

480   neutral to alkaline soils (Schenkeveld et al., 2014; Grillet and Schmidt, 2017), can mobilise toxic metal

481   contaminants by chelation of complexation. In healthy soil systems most plant exudates are consumed

482   by rhizosphere-dwelling microbes (resulting in production of CO_2), but in industrial residues, with a

483   thin, poorly established rhizosphere, plant exudates can migrate deeper into the deposit (Bray et al.

484   2018), see Fig 3.

485

486   **5.2 Capture Zone**

487   There are already many existing and developing technologies for the trapping of metals and other

488   contaminant species from water into a solid matrix that have variously been developed for treatment of

489   contaminated water (e.g. groundwater, municipal solid waste landfill leachate, mine water, highways

490   and urban runoff) and there are many parallels with the continuing development of such "passive"

491   technologies. These include permeable reactive barriers (PRBs), constructed wetlands, sustainable

492   drainage systems (SuDS), swales and bioswales (for examples see Scherer et al., 2000; Pat-Espadas et

493   al., 2018; Woods-Ballard et al., 2007 and Ekka et al., 2021 respectively). The mechanisms of trapping

494   variously involve biogeochemical process within the matrix that induce metal sequestration from

495   solution. These typically rely on changes in redox or pH, utilise precipitation by the common ion effect

496   or in situ sulphide production from microbial sulphate reduction, sorption, chelation or coprecipitation

497   (Sapsford et al., 2019). Less well understood are passive systems for removing metal and metalloid

498   oxyanions prevalent in alkaline wastes e.g. V, Al and As in bauxite processing residue (Burke, 2013).

499   For Cr(VI) reductive precipitation is important, for V(V) pH reduction by carbonation and sorption to

500   Fe-(oxy)hydroxides is key, whereas for As oxidation of As(III) to As(V) and then sorption to Fe-

501   (oxy)hydroxides is important (Ding et al., 2015; Hobson et al., 2018). In these cases, pH reduction and

502   carbonation are key mechanisms for metal capture. Additives such as gypsum and organic matter can

503   reduce the pH value (the former by promoting carbonation, the latter by dissolution of soluble humics)

504   and organic matter can capture metals directly by complexation or indirectly by supporting bioreduction

505   by microorganisms. Such mechanism should be effective in a trapping zone as they have been

506   demonstrated to reverse the mobility of elements including V in bauxite residues (Bray, 2018).

507   An underlying aim of the design and operation should be a significant concentration of the target metal

508   (or other species) compared to the leached residue. In achieving this, key challenges for capture zone

509   engineering include (i) maintenance of the long-term effectiveness of the biogeochemical removal

510   mechanisms (ii) maintenance and/or control of hydraulic conductivity (See below) (iii) concentration

511   achievable in the capture zone.

512

6. Hydraulic and Geotechnical Considerations

514   Many landfills (or in-ground impoundments) of wastes rely to some extent on isolation, preventing or

515   minimising the mobilisation of contaminants and their leakage into the surrounding environment. With

516   the ASPIRE concept of *in situ* processing during temporary storage, however, complete isolation is no

517   longer desirable as external environmental processes are required to be brought to bear on the waste

518   mass to enable no/low input processing. The most important of these is likely to be controlled hydraulic

519 flow into the waste repository. As a result, the repository design moves from a containment facility to

520 something more akin to a funnel-and-gate permeable reactive barrier, where water/lixiviant flow is

521 encouraged (or at least controlled) to pass through the waste mass prior to treatment of the resulting

522 liquor. This conceptual change leads to implications for both the hydraulic and geotechnical design of

523 in-ground temporary waste storage.

524

525 **6.1 Hydraulic processes in temporary storage**

526 Hydraulic flow is likely to be the major driver of waste processing under natural processes, permitting

527 the mobilisation and transport of both lixiviants and resource. Flow must enter the waste and travel

528 through the entirety of the mass to reach the region where mobilised species are deposited. It is therefore

529 vital to encourage water to flow through the waste mass – optimisation of this flow is dependent on-site

530 conditions, but general issues may be considered here. There are two major environmental sources of

531 water ingress, from rainfall / run-on (more transient) or groundwater (more consistent and predictable),

532 which could be extracted adjacent to, and introduced into the repository.

533 A second major issue is the volume and rate of flow – the optimal volume/rate will be determined by

534 properties of the waste mass including hydraulic conductivity, as well as the rate of resource deposition

535 and thus the minimum residence time in the capture zone. Mineral-rich wastes of interest in temporary

536 storage schemes have a wide range of potential hydraulic conductivities, for example municipal solid

537 waste incineration bottom ash is relatively coarse, with sand-like hydraulic conductivity ($5x10^{-6}$ m/s (de

538 Windt et al., 2011)) whilst finer fly ashes may vary from 10^{-7} to 10^{-10} m/s (Zabielska-Adamska, 2020).

539 Depending on the storage duration, rainwater alone may not provide sufficient water to fully mobilise

540 resource even on the long timescales considered here and where it does, the transient nature of rainfall

541 will potentially lead to periods of desaturation which in turn causes preferential flow and thus reduced

542 resource mobilisation. To overcome this, water retention in ponding schemes may be used to attenuate

543 flow transience, buffering water ingress by collection prior to gradual, more continuous release into the

544   waste. Alternatively, fluid exiting the storage facility may be collected and recirculated *via* autonomous

545   systems, for example powered by solar energy.

546   Flow in porous media, including mineral wastes, is impacted by preferential flow – certain paths through

547   the pore space offer less resistance to flow and so the majority of advective flow takes these paths

548   (Clothier et al., 2008) leaving the majority of the medium (the matrix) untouched. Causes may include

549   heterogeneities at a range of scales such as strata with varying hydraulic conductivity, pore sizes of

550   differing diameter or unsaturated pores where flow cannot occur. Such preferential flow leads to only a

551   proportion of the waste being treated by flushing / leaching, while decontamination of the remaining

552   waste is governed by the rate of contaminant diffusion to preferential flow paths from the matrix. Soil

553   flushing (or "pump and treat") of contaminated land is affected by this process and sometimes results

554   in active pumping continuing for decades to achieve the remediation objectives (Guo et al., 2019).

555   Without some kind of engineered system preferential flow will occur and will limit recovery from the

556   waste mass, so it is appropriate to question whether there exist autonomous natural processes that can

557   be engineered to alter fluid flow and/or diffusion. For example, this can, to an extent, be managed by

558   pumping techniques such as pulsed flushing where flow is intermittent to give time for diffusion within

559   the matrix (Cote et al., 2000); this doesn't significantly affect the overall remedial time but it does

560   reduce the active flushing time. Similar processes may be helpful in the case of temporary storage where

561   treatment time is not a significant issue, but diffusion alone is unlikely to be wholly effective in moving

562   resource, particularly when that resource may be bound tightly in mineral wastes and thus unavailable

563   to the pore fluid. More active technologies to enhance availability and diffusion, such as electrokinetics,

564   have been successfully employed to alter the mobility and availability of resources bound in mineral

565   wastes (Peppicelli *et al.*, 2018), though the challenge to engineer natural processes to autonomously

566   enhance resource availability remains.

567   Over time, the hydraulic behaviour of all aspects of a repository may alter, and the repository system

568   needs to be able to adapt to this. The whole waste body will undergo self-weight compaction to a degree

569   and pattern determined partly by compaction during placement and the rate at which self-weight

570   develops. With repositories that develop over time with the deposition of new waste, the process of

571　compaction and thus alteration of hydraulic flow in the original deposits will be a continuous process.

572　The surface ecosystem will be dynamic with the growth of established plants and potentially

573　successional growth of new plant species, potentially with new root architectures and behaviour which

574　could alter water infiltration, evapotranspiration and so on. Continued leaching may lead to erosional

575　processes, particle breakdown and aggregation, and clogging in the leach zone, whilst calcium-rich

576　wastes such as certain fly ashes may be susceptible to cementation with calcium carbonate (Zabielska-

577　Adamska, 2020), blocking the pore space and changing flow patterns and causing or preventing any

578　continued waste compaction. The capture zone is by definition an active biogeochemical zone with

579　deposition changing the pore structure and likely reducing the hydraulic conductivity, with challenges

580　then for hydraulic flow throughout the system.

581

582　**6.2 Geotechnical factors relevant to temporary storage**

583　The geotechnical stability of in- or on-ground waste repositories will be dependent on their site-specific

584　design, their profile and the surrounding ground conditions. The desire to have fully saturated wastes

585　(to maximise resource recovery and avoid preferential flow) is problematic for sloped surfaces. It is

586　likely that repository design will require non-sloping or modest-sloping surfaces as steeper slopes would

587　be susceptible to slip failure in the presence of large pore pressures, as well as seepage problems in a

588　similar manner to earth dams (Meyer et al., 1994), leading to failure mechanisms such as piping, heave

589　and internal erosion (as noted above). This may be exacerbated with enhanced water flow and pore

590　pressures should measures such as surface ponding and/or recirculation be employed. Settlement and

591　consolidation of the emplaced waste masses will also be impacted by variations in the development of

592　pore pressures, alongside factors such as internal erosion and changes on the structure of the waste

593　materials. These processes will require consideration to ensure flow systems and any installed rigid

594　infrastructure remain functional and to avoid adverse features such as differential surface settlement

595　occurring. Geotechnical liners employed to encapsulate the waste body will require redesign from

596　typical systems employed in traditional landfill. The inclusion of drainage layers should be avoided to

597　prevent preferential flow of influent water (rain or ground) around rather than through the waste. The

598 saturation of the waste without drainage, however, has the potential to create significant hydraulic
599 gradients across the liner which may lead to localised liner failure.

600

601 ## 7. Delivery of ecosystem services

602 In the proposed ASPIRE repository concept the upper surface can be vegetated to provide root exudates
603 that enhance leaching and drive capture zone biogeochemistry. In addition, the opportunity for
604 vegetated upper surface provides opportunities for considerable added value through the delivery of
605 ecosystems services. Restored landfills deliver a range of ecosystem services, or benefits to people,
606 following conversion to 'soft' land uses, including agriculture, forestry, amenity and nature
607 conservation (Li et al., 2019; Zalesny et al., 2020). There is an opportunity to tailor the design and
608 species selection to provide the necessary lixiviants as well as delivering ecosystem services. Using the
609 framework provided by Common International Classification of Ecosystem Services (CICES) V5.1
610 (Haines-Young and Potschin, 2018) it is possible to explore the opportunity to provide ecosystem
611 services in ASPIRE landfills. The CICES focuses on regulation and maintenance (e.g. mediation of
612 wastes, flood risk management, pollination), cultural (e.g. experiential and physical use, education) and
613 provisioning services (e.g. cultivated plants, energy generation, mineral resources), which are
614 underpinned by 'supporting' conditions (e.g. primary production; Haines-Young and Potschin, 2018).

615

616 ### 7.1 Regulation and maintenance services

617 Regulation and maintenance services include management of flood risk, temperature, and air, water and
618 soil quality, as well as pollination. It is well known that restored landfills can deliver positive outcomes
619 for nature conservation (MacGregor et al., 2022; Tarrant et al., 2012; Rahman et al., 2013). The sector
620 is already accustomed to designing restoration strategies for recreation (see below) and nature
621 conservation end uses, providing a variety of habitat types and amenities. For example, providing a
622 nutrient-poor soil and allowing natural colonisation of plants to facilitate the development of semi-

623   natural grasslands to provide for pollinators (Rahman et al., 2013). With ASPIRE repositories there

624   would also be a need to ensure that selected species also generated sufficient exudates/lixiviants.

625   New requirements for biodiversity net gain, introduced in the UK as part of the Environment Act 2021

626   create an opportunity to deliver biodiversity as an integral part of the system as well as the restoration

627   phase. For example, current landfills, or those planned for habitats of low distinctiveness, such as

628   quarries, or improved grassland, could be restored to provide mosaic habitats of medium or high

629   distinctiveness, including native flower-rich grasslands, to provide pollination services, open grassland,

630   mixed broadleaved woodland and ponds and wetlands (Natural England, 2021). Furthermore,

631   revegetation of landfills can provide many of the services of urban greenspaces, including improved air

632   quality, and reduced temperature and flood risk (Harwell et al., 2021; Li et al., 2019; Pereira et al.,

633   2018). As well as providing water storage, including wetlands and ponds will also allow the

634   recirculation of clean water contributing to the regulation of water pollution (Benyamine et al., 2004).

635

636   **7.2 Provisioning services**

637   In addition to cleaned residue, and a concentration of potentially valuable elements in the anthropogenic

638   ore zone, there are equally important potential applications for the upper surface of the ASPIRE

639   repository (as with other landfills) to provide food and energy. The establishment of agriculture and

640   forestry on restored landfills is a common end use and selecting species that can be used in food or

641   timber production whilst providing the necessary lixiviants could ensure ASPIRE repositories are also

642   able to contribute provisioning services. Landfills are also increasingly being used for energy

643   generation, through solar farms (Szabó et al., 2017) or biomass crops (Cervelli et al., 2020; Zalesny et

644   al., 2020) and there is also potential here for added value from ASPIRE repositories. This approach can

645   also achieve biodiversity benefits (Cervilli et al., 2020), and in the case of some energy crops, spaces

646   for recreation use for example through the creation of community woodlands. For example, the energy

647   crop miscanthus can provide a habitat for farmland birds (Bright et al., 2013) and the brown hare, whose

648   population is declining in areas of intensive agriculture (Petrovan et al., 2017). Creating solar farms on

649 landfills, particularly, can overcome some of the tensions with using agricultural land for this purpose

650 (Szabó et al., 2017). As a new concept, an ASPIRE landfill will be well placed to take advantage of the

651 latest developments in other fields, such as the dual use of land for cultivation and solar farms (Toledo

652 and Scognamiglio, 2021). These 'agrovoltaic' systems employ a range of technologies to ensure that

653 crop production and energy generation can work in unison, for example, by using vertical photovoltaic

654 (PV) panels, or elevating the panels several metres about the ground, allowing for vegetation growth

655 beneath (Toledo and Scognamiglio, 2021). Depending on the configuration and species selection this

656 approach can increase crop yields and improve the efficiency of the PVs (Toledo and Scognamiglio,

657 2021). There may also therefore be the potential to combine energy crops, such as miscanthus, with

658 solar farms to increase the energy generation from ASPIRE repositories.

659

660 **7.3 Cultural services**

661 Many landfills have been restored to high quality greenspaces, often close to where people live,

662 providing space for rest, relaxation, physical activity and contact with nature (Li et al., 2019). These

663 activities provide cultural ecosystem services including health and wellbeing benefits such as improved

664 mental health, physical activity (Li et al., 2019).

665

666 8 Carbon Capture

667 Any soil surface has the potential for capturing carbon through the biological colonisation and

668 development of the regolith. Thus, an ASPIRE repository, which deliberately includes a vegetated upper

669 surface would achieve carbon capture. Moreover, many mineral-rich wastes contain components which

670 will either directly react with CO_2 or will lead to capture of CO_2 as bicarbonate via mineral weathering.

671 The carbonation mechanism is particularly relevant to alkaline wastes, whereby soluble oxide and

672 silicate minerals react with CO_2 and form carbonate minerals (Renforth, 2019). Recent studies have

673 suggested that legacy iron and steel wastes have a carbon capture potential of up to 80 million tonnes

674   in the UK (e.g. Riley et al., 2020, and section 4.3), whilst global estimates suggest between 5-12% of

675   global CO_2 emissions could be mitigated through carbonation with alkaline residues (Renforth, 2019).

676   The sequestration process in these wastes is currently limited in disposal settings by low rates of

677   atmospheric CO_2 ingress into heaps and surface armouring of wastes by secondary deposits (e.g. Pullin

678   et al., 2019). An ASPIRE repository could accelerate carbonation rates in a controlled manner through

679   managed weathering of shallow piles (to encourage contact with atmosphere) and accelerated flushing

680   of residues (to minimise surface armouring and accelerate weathering rates). The deliberate engineering

681   of the surface to encourage downward percolation of water and root exudates may transport organic

682   materials into the waste repository where biological mineralisation should further enhance rates of

683   mineral weathering and carbonation. Interestingly, there may well be significant financial incentives

684   associated with this atmospheric CO_2 sequestration (e.g. in operating an auditable negative emissions

685   system: Renforth, 2019) which could assist with long-term management of ASPIRE repositories.

686

9 Technology Barriers and Risks

688   There are foreseeable barriers to the further development of the ASPIRE repository concept, including

689   both the technical engineering challenges and public or stakeholder perceptions. There could potentially

690   be enhanced risks of contaminant escape due to increased water movement, potential for the

691   development of hydraulic heads on liners and presence of agents that enhance contaminant

692   concentration and mobilisation. However, existing containment engineering for landfills and heap

693   leaching facilities (common in the mining industry) should find application in adequately mitigating

694   this risk. Furthermore, landfill monitoring technology is well established, and environmental

695   monitoring technologies continue to develop apace.

696   There are risks and barriers associated with public perceptions of the technology, similar to those

697   experienced during the remediation of contaminated sites and mining operations. For example, they

698   include concerns that there will be adverse impacts on the environment from the release of pollutants

699   and the time taken for resource extraction (Song et al., 2019; Tayebi-Khorami et al., 2019). There may

700 also be a lack of trust from local communities that operators are mitigating risks effectivity or acting in

701 their interests (Tayebi-Khorami et al., 2019; Sinnett and Sardo, 2020). Operators may also be resistant

702 to a new technology and any associated liabilities (Tayebi-Khorami et al., 2019; Song et al., 2019), and

703 uncertain that a market exists for the product generated by ASPIRE repositories (Tayebi-Khorami et

704 al., 2019) or the ownership of these products. Currently, there are also regulatory risks to adoption of

705 the ASPIRE concept as there is with an authentic shift towards a CE model for mining waste

706 management (Tayebi-Khorami et al., 2019; Cisterna et al., 2022), the proposed long timescales may

707 make it difficult to foresee future changes in environmental regulations that might impact on operation

708 or recovery. Furthermore, there may also be increased cost implications for the operators in terms of

709 long-term monitoring, which may result in additional running costs compared with a traditional landfill.

710

10. Conclusions

712 There are many reasons why mineral-rich wastes are currently landfilled. However, it is self-evident

713 that environmental containment offered by a landfill will eventually succumb to environmental

714 processes, potentially allowing pollution to escape to burden future generations. Thus, in a very real

715 sense, landfilled waste becomes "someone else's problem", removed in time, rather than

716 geographically, from the producer. Therefore, landfill cannot be considered as an intergenerationally

717 equitable waste management option. Temporary storage is an important concept that has been

718 considered but not adopted for MSW but may be more applicable for mineral-dominated wastes from

719 mining and industry. The case for temporary storage is more compelling, due to the disconnect between

720 potential end-users for residues and the waste production. The reuse of mineral-rich wastes is also often

721 limited because of the presence of leachable contaminants. Separating contaminants from clean residue

722 or concentrating valuable components from a waste where they are dispersed requires energy. The

723 ASPIRE concept looks to extend the concept of temporary storage to include in-built biogeochemical

724 engineering and the solar insolation flux to utilise the time in storage for the separation and

725 concentration of contaminants (or resources). Furthermore, there are clear opportunities for adding

726 value through ecosystem services and carbon capture. There are several important engineering and

727 legislative hurdles that remain to be solved. Importantly, recycling materials within ASPIRE

728 repositories is not intended to displace any existing economically viable and sustainable recycling

729 technologies for mineral-rich wastes. However, the concept could provide a potentially practicable

730 Circular Economy solution for materials that would otherwise go to conventional landfill, thus at

731 minimum replacing/displacing landfill with temporary storage and treatment for recycling at the base

732 of the waste hierarchy.

733

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737

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