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Ecological engineering to accelerate mineral weathering and transformation underpins sustainable tailings rehabilitation

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Abstract

Tailings are nothing like soil but are polymineral wastes containing residue economic metals (e.g. Al, Cu, Pb, Zn) and gangue minerals, exhibiting a range of geochemical reactivity in oxygenated and aqueous environments. Early colonisation of soil microorganisms and pioneer plants is inhibited by the bio-toxic geochemical conditions, as well as physical constraints, even with remediation inputs of organic matter and fertilisers. Geochemical conditions of the tailings are governed not only by chemical factors (e.g. acidity/alkalinity, soluble solutes and metal(loid)s) already formed in the soluble phase (i.e. porewater), but also the solid phase of reactive minerals. Tailing minerals undergo in situ weathering and replenish the soluble geochemical factors into the soluble phase over a prolonged and unpredictable period of time (e.g. decades). This makes short-term remediation ineffective in terms of sustaining long-term performance of reconstructed soil systems for vegetation cover. So far, the misperception of tailings as 'inferior/contaminated soil' and the adoption of 'soil remediation' approaches have largely failed in low-cost and direct phytostabilisation. Despite soil cover, opportunistic microbial bioweathering processes and associated hydrogeochemical dynamics in tailings have resulted in many ineffective conventional cover systems for rehabilitating tailings, such as sulfidic Pb-Zn tailings and red mud. Extensive weathering of these reactive minerals in the top layer of tailings (ca. 50–100 cm) is the prerequisite to hydrogeochemical stabilisation, abatement of acute toxicity, and colonisation of soil microbes and pioneer plants in reconstructed root zones covering the tailings. Bioweathering of reactive minerals (e.g. sulfides in sulfidic tailings, sodalites in red mud) can be readily catalysed by extremophiles (i.e. tolerant archaea, bacteria and fungi) upon provision of suitable conditions, such as moist conditions and relevant substrates (such as organic matter, phosphate). By using ecological engineering approaches combining engineering and geo-microbial ecology principles, the microbial processes would be enhanced by targeted and effective engineering inputs (e.g. water, organics) for achieving rapid exhaustion/depletion of reactive minerals, leading to long-term hydrogeochemical stabilisation (i.e. the completion of fast geochemical reaction phase). This would minimise risks of deterioration and failures of reconstructed soil and plant subsystems, due to minimal abundance of residual reactive minerals in the tailings underneath root zones. The present paper will draw on our recent research progress on sulfidic tailings, Fe-ore tailings and alkaline red mud for the purposes of (1) illustrating the importance of microbial driven weathering and transformation of key minerals in tailings rehabilitation and (2) introducing new technological pathways, that is, 'in situ soil (or technosol) formation' and 'mineral (bio)weathering, cementation and hardpan formation'. This aims to draw research attention onto translational research by adapting ecological engineering principles and practices to deliver cost-effective and feasible technologies for speeding up progressive and sustainable tailings rehabilitation in Australia and other mining countries.

Keywords: *tailings rehabilitation, bioweathering, mineral transformation, hydrogeochemical stabilisation*

1 Introduction

Increasing global demands for metals (e.g. Fe, Cu, Pb, Zn) and minerals (e.g. Fe-ores, bauxite) and much improved mining and mineral processing technologies have fuelled the exploitation of ores and mineral deposits of low grades that were considered uneconomic in the past, resulting in rapid accumulation of tailings in landscapes from tropical to temperate regions across the world (Mudd 2010; Mudd & Jowitt 2018; Northey et al. 2018; Prior et al. 2012). Australia is a leading producer of minerals, producing at least 19 minerals (in significant amounts) and the production of alumina (Al), copper (Cu), black coal, lead (Pb), zinc (Zn), and iron (Fe) ores has increased markedly over the last decade (Mudd 2010). From the mid-1800s to 2008, cumulative Cu, Pb and Zn production in Australia reached 20,473 kt, 37,945 kt, and 48,465 kt, respectively, in which their production in Queensland accounted for 50–60% (Mudd 2010). This trend of mining and processing ores and minerals of declining grades is generating tailings at much higher ratios to economic metals/minerals and at a much faster pace than in the past. As a result, technological breakthroughs are urgently required to commence cost-effective, progressive, and sustainable rehabilitation for lowering long-term liability and risks of environmental disasters.

Tailings are mineral residues containing gangue minerals and residual ores from flotation and/or mineral processing (Dold & Fontboté 2001; Lottermoser & Ashley 2006), for example, flotation for Cu, Pb and Zn (Lottermoser 2010), reverse flotation for magnetite (Xiong et al. 2015) and Bayer processing of bauxite for alumina (Power et al. 2011). Tailings have abundant levels of heavy metals (e.g. Cd, Cu, Ni, Pb and Zn), metalloids (e.g. As), radionuclides (e.g. U) and other pollutants (e.g. acidity, alkalinity, soluble salts), but low levels of life-forming macro-elements (e.g. C and N) (Li & Huang 2015). In Australia, the mining industry was estimated to produce more than 1,750 Mt of mine waste per year (Lottermoser 2010). Extraction of alumina from bauxite has so far generated more than four Gt bauxite residues (or red mud) worldwide, with about 50% located in China (Stanford 2016; Xue et al. 2016). In Australia, increasing interests in extracting magnetite-Fe-ore for balancing declining hematite Fe-ore grades have led to the generation of increasing volumes of finely textured and siliceous tailings in semi-arid regions of Western Australia (Wu et al. 2019). Unlike natural soil, tailings typically contain various hazardous chemicals and reactive minerals in significant quantities, which are biologically toxic and lack basic physical and chemical properties minimally required to support the colonisation of even extremely tolerant plant species (Huang et al. 2014; Huang & You 2018; Li & Huang 2015). The addition of large amounts of organic and inorganic materials or natural soils into the tailings concerned may accelerate the improvement of physical and chemical properties to enable colonisation of soil microbes and plant species, but which is economically unviable and unsustainable.

Past rehabilitation efforts using soil remediation approaches and agronomic practices has largely failed in achieving non-polluting and/or sustainable rehabilitation outcomes, as the tailings have been mistaken for 'degraded/contaminated soil'. From our own experience and consultation with relevant experts in the environmental engineering sector, the rehabilitation of Cu, Pb-Zn tailings and red mud dams at major operating mines and refineries in Australia would cost more than AUD 20 billion when using conventional cover methods, which require large volumes of clay, rock and topsoil. In many cases, such as sulfidic metallic tailings, unexpected seepage pollution could still occur in events of intensive rainfalls, even being covered with standard engineered cover systems. In addition, the lack of topsoil within a financially viable transport distance renders it unfeasible to implement conventional cover systems at remote mine sites. As a result, innovations are urgently required to speed up the progress of tailings rehabilitation for non-polluting and sustainable outcomes, which is critical to the 'social licence to operate' for the mining and minerals sectors. The rehabilitation of sustainable ecosystem to cover the tailings landscapes would also minimise or prevent legacy tailings dam failures associated with surface erosion and landslides caused by unpredictable and extremely intensive storm events in future, when production ceases and high degree of care and maintenance may become financially unaffordable or untenable. Unfortunately, game-changing innovation of cost-effective technologies for tailings rehabilitation according to modern legislative requirements has led to the slow progress of successful rehabilitation and the rapid accumulation of tailings liability and associated environmental risks worldwide.

The present paper aims to introduce a new paradigm of technological innovations for tailings rehabilitation, focusing on ecological engineering induced and systematic changes and evolution of tailings mineralogy, geochemistry and biogeochemistry, commencing with overcoming reactive mineralogical barriers, in order to achieve non-polluting and sustainable rehabilitation of tailings (Figure 1). The new paradigm is composed of two divergent technological pathways (Figure 2): (1) formation of growth substrates (i.e. technosol or eco-engineered soil) in tailings of low pollution risks (e.g. Fe-ore tailings, bauxite residues) (Wu et al. 2019; Huang & You 2018) and (2) formation of hydrogeochemically stable tailings with high degree of cementation and low risks of acidic/neutral metallic drainages (Liu et al. 2018), both of which share a fundamental phase of bioweathering of target minerals in the tailings for relevant purposes (Figure 2). In State 3 (Figure 2), there are two trajectories, depending on the type of tailings: (3a) formation of functional 'soil' or technosols for tailings of low pollution risks (e.g. Fe-ore tailings, red mud), and (3b) the formation of cemented and hydrogeochemically stable profile with low risks of metallic drainages, due to depletion of reactive minerals (e.g. sulfides) for tailings of high pollution risks (e.g. sulfidic Pb-Zn tailings) (adapted from Huang et al. 2014).

This paper will illustrate the importance and key mechanisms of microbial bioweathering of relevant minerals in tailings, for overcoming critical mineralogical barriers to hydrogeochemical stabilisation in tailings, by drawing on our research findings over a decade, on sulfidic Cu-Pb-Zn tailings, bauxite residues, and magnetite-Fe-ore tailings, in addition to relevant leading research cases in the literature. This paper by no means presents a comprehensive review of all aspects of tailings ecological engineering, but aims to stimulate research and industry interests in translational research to develop cost-effective and sustainable technologies based on ecological engineering principles. The new paradigm of technological innovation in tailings ecological engineering may help to speed up the progress of successful tailings rehabilitation in the near future.

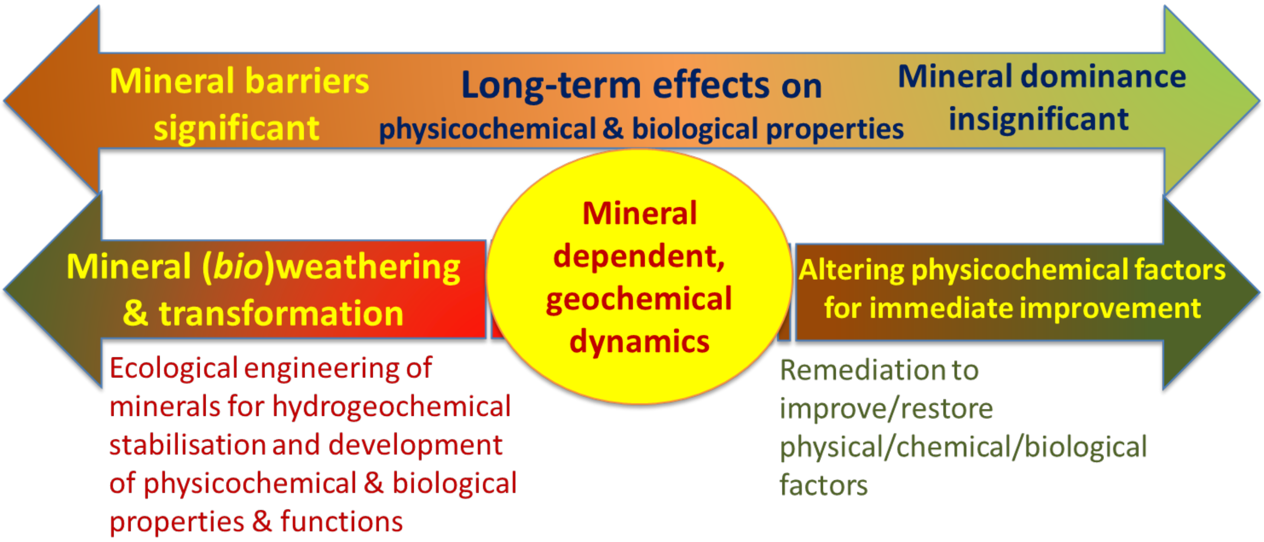

Figure 1 **A conceptual diagram illustrating the key roles of reactive minerals (such as sulfides, sodalities) that drive long-term geochemical dynamics in the soluble phase of the tailings and underpin long-term risks of pollution and ecological sustainability in the process of tailings rehabilitation. In 'ecological engineering' context, the weathering and transforming target minerals (e.g. sulfides) are essential to the cascaded improvements of physicochemical conditions by 'remediation', while 'remediation' focuses on instant adverse physicochemical factors not coupled with the 'hidden' minerals**

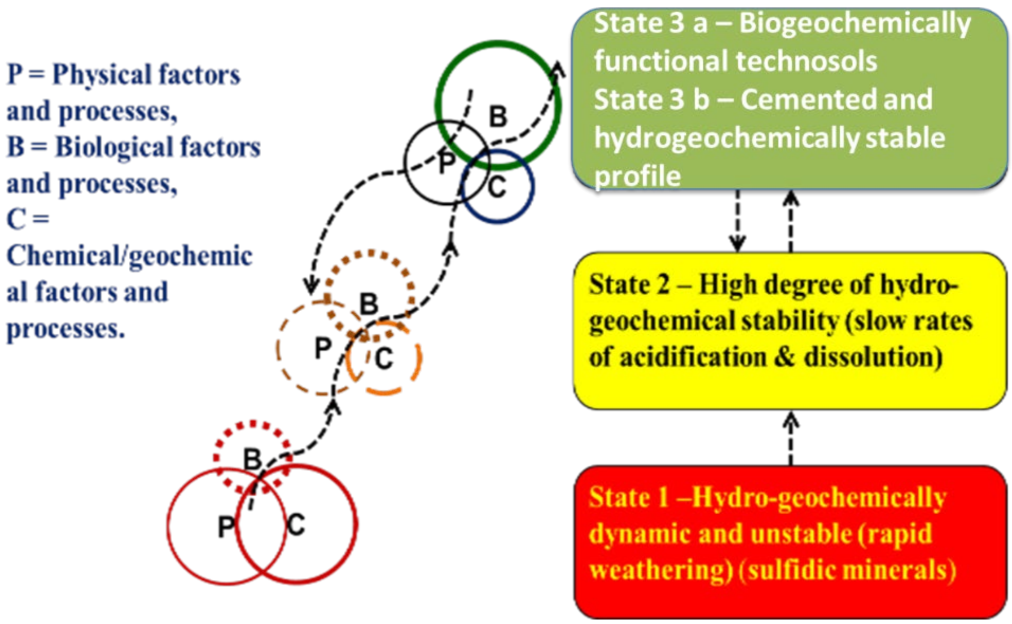

Figure 2 A conceptual diagram illustrating three key states of tailings undergoing mineralogical changes in response to ecological engineering inputs and practices: B = mainly microbial mediated biological factors and processes involved in mineral weathering and transformation; P = mainly physical factors and processes such as physical disturbance and manipulation of water relations; C = chemical factors and processes, such as inputs of mineral nutrient and chemicals/minerals for pH alteration, mineral dissolution/hydrolysis, ion exchange and formation of precipitate minerals. The solid line represents a relatively dominant process and the dashed line a relatively weak but growing process. The curved and dashed lines linking up the stages representing general direction of changes towards each of the transition stages

2 Importance of microbial bioweathering of minerals in ecological engineering of tailings for sustainable rehabilitation

Rehabilitation of tailings is a holistic and integrated process from ecological engineering of tailings' mineralogy, geochemistry and physicochemical properties, to whole soil-plant systems (Huang et al. 2012; Huang et al. 2014), which bear the characteristics of:

- Stable and self-sustaining biotic and abiotic structure; that is, physically and geochemically stable root zone (or soil subsystem) and healthy, diverse and self-recruiting plant subsystem.
- Dynamic linkages of functional processes, including:
  - Hydrological cycle (e.g. water infiltration, evapotranspiration).
  - Energy flows and material cycles (e.g. soil erosion, food/litter production, food for fauna).
  - Biogeochemical cycle (e.g. microbial decomposition and nutrient cycling).
- Capability of delivering sustainable ecosystem services.
- No polluting risks on- or offsite, such as the mobility and bioavailability of pollutants (e.g. metal(loid)s).

Tailings mineralogy underpins the progress and trajectory of tailings rehabilitation in the long-term. This has been why the initial emphasis has been placed on accelerating weathering of minerals governing geochemical reactions. This is critical to the rapid development of key physicochemical properties in tailings and fundamental to successful vegetation growth and tailings rehabilitation.

2.1 Mineralogical barriers in tailings

The primary physicochemical and mineralogical properties of tailings significantly differ from those of natural soils (Li & Huang 2015). Typical characteristics of tailings may be summarised into the three key aspects:

- Physical properties.
  - Fine/very fine texture without heterogeneity of particle size, highly compacted, lack of water-stable aggregates.
  - High hydraulic resistance.
  - Highly dispersive in water (e.g. red mud).
- Mineralogical imbalance.
  - Primary minerals – status of Gibbs energy.
    - Too high: e.g. pyrite, galena, chalcopyrite in sulfidic tailings.
    - Too low: e.g. magnetite, biotite in Fe-ore tailings.
  - Secondary minerals.
    - Too little: secondary minerals e.g. Al/Fe-oxyhydroxides, Al-Si-Fel minerals, which are required for aggregation (e.g. in Fe-ore tailings) and cementation (e.g. in sulfidic tailings).
    - Too much: abundant salts and alkaline minerals in red mud.
- Geochemical dynamics.
  - Low reactivity due to low Gibbs energy: very slow rates of weathering – e.g. biotite, magnetite, hematite in Fe-ore tailings.
  - High reactivity due to high Gibbs energy: readily oxidised or hydrolysed in oxygenated and aqueous environments, e.g. in sulfidic tailings and bauxite residues.

Conventional soil remediation approaches are ineffective in resolving mineral barriers in tailings for rehabilitation purposes, unless large amounts of inputs (e.g. water, fertilisers and/or oxidative/neutralising chemicals) are maintained over the long-term. Soil remediation focuses on improving chemical constraints in soluble phase, but not overcoming mineralogical barriers, such as sulfides in sulfidic tailings, sodalities in bauxite residues, and the lack of adequate secondary Al-Fe-Si minerals in Fe-ore tailings. For example, adding lime in sulfidic tailings may only neutralise acidic pH conditions already formed in soluble phase for a short-term as the alkalinity of lime (consortium of Ca-oxides and hydroxides) is quickly exhausted by concurrent neutralisation and formation of Ca-carbonates upon exposure to CO_2. However, sulfide oxidation may occur over a prolonged period of time (e.g. many decades) upon exposure to oxygenated and aqueous conditions. Organic matter may be used to improve physicochemical properties in highly weathered sulfidic Cu-Pb-Zn tailings, but not unweathered tailings containing abundant sulfides (You 2015; Yuan et al. 2016). In bauxite residues, the hydrolysis of sodalities and exchange of Na into the porewater causes the presence of strong alkalinity, and long-term neutral pH conditions cannot be resolved by simply neutralising soluble alkalinity in the porewater without depleting the alkalinity in the solid phase (i.e. sodalities) (Huang & You 2018; You et al. 2019). In contrast, under arid/semi-arid climatic conditions where Australian Fe-ore mines are located, Fe-bearing minerals in magnetite Fe-ore tailings possess low Gibbs energy and require very long weathering time (e.g. decades) to generate sufficient amounts of secondary Fe-Si mineral gels, which are critical to aggregation of fine particles and development of physical properties in the finely textured and densely compacted magnetite Fe-ore tailings (Table 1) (Wu et al. 2019).

Table 1 Typical characteristics of mineralogy and geochemical dynamics in sulfidic Cu-Pb-Zn tailings, magnetite Fe-ore tailings, and bauxite residues (or red mud)

Tailings	Minerals to be weathered	Hydrogeochemical dynamics
Sulfidic Pb-Zn tailings	Reactive primary minerals: pyrite (FeS_2), pyrrhotite (Fe1-xS), sphalerite (Zn[Fe]S), galena (PbS) and chalcopyrite ($CuFeS_2$) (Moncur et al. 2009)	Sulfide oxidation, acidification and metal(loid) dissolution, in oxygenated and aqueous environment
Magnetite Fe-ore tailings	Inert primary minerals: quartz, hematite, magnetite, biotite (Wu et al. 2019)	Resistant to weathering and hydrolysis, lack of secondary Fe-minerals for aggregation
Bauxite residues (red mud)	Crystalline sodalites and other Na-rich alkaline minerals (Gräfe et al. 2011; You et al. 2019)	Strong alkalinity buffered by hydrolysis of sodalites

Effective organic/inorganic inputs may be admixed into the tailings to prime, initiate and accelerate microbial driven processes of mineralogical changes, as bioweathering is much faster and more effective than chemical weathering when favourable conditions (e.g. most importantly, moisture) could be provided for the growth and functions of tolerant microbes in situ. Bioweathering enhanced by eco-engineering inputs would accelerate hydrogeochemical reactions in the short-term (i.e. the fast reaction phase) due to the presence of reactive minerals. This would be followed by subsequent slow reaction phases leading to hydrogeochemical stabilisation in the matrix profile. For example, acidification in sulfidic tailings would be intensified by activities of Fe/S-oxidising bacteria in the short-term, which results in co-dissolution and mineral transformation of gangue minerals (Fortin et al. 1995; Huang et al. 2014; Liu et al. 2018). The hydrolysis of sodalities and release of soluble ions into porewater favour effective leaching and removal in bauxite residues amended with biomass residues (You et al. 2018). The dissolution of Fe-bearing minerals (e.g. biotite, hematite and magnetite) is accelerated by microbial activities upregulated by organic matter in magnetite Fe-ore tailings, leading to the formation of secondary Fe-minerals, essential ingredients for aggregate formation in tailing-soil (Wu et al. 2019). These microbial driven geochemical reactions result in the exhaustion of reactive minerals, such as sulfides in sulfidic tailings and sodalites in bauxite residues, leading to long-term hydrogeochemical stabilisation, the foundation for subsequent development of cementation cap in sulfidic and metallic tailings or soil formation in bauxite residues and magnetite Fe-ore tailings (Liu et al. 2018; Wu et al. 2019; You et al. 2019). For example, geochemical reactions enhanced by functional microbes in sulfidic tailings involve coupled acidification, co-dissolution and/or precipitation, and in situ immobilisation/encapsulation of metal(loid)s, leading to the exhaustion of reactive minerals (e.g. sulfides) for hydrogeochemical stabilisation and formation of secondary mineral gels (e.g. amorphous Al-Fe-Si minerals and massive gypsum) required for tailings cementation (Huang et al. 2012; Liu et al. 2018). In bauxite residues, the hydrolysis of sodalities and release of soluble Na into the porewater are accelerated by microbial decomposition of biomass residues (You et al. 2019). Within the context of eco-engineered soil formation in tailings, systematic changes in mineralogy and associated hydrogeochemistry are critical to the development of physicochemical properties (physical structure, chemical buffering) and biological conditions (shifting from extreme microbes dominant to soil microbes dominant communities), to the state permitting the colonisation of pioneer plant species and target plant communities.

2.2 Microbial mediated bioweathering processes for mineral transformation in tailings

Tailings may be considered as engineered parent materials of geological origins for soil formation (Huang et al. 2014; Huang & You 2018; Li & Huang 2015). Microbial consortia are useful sparks to initiate and accelerate the changes in mineralogy and hydrogeochemistry in tailings. Direct and indirect roles of tolerant microbes in bioweathering of key minerals in tailings may be illustrated in the following scenarios in the three common (but by no means exclusive) types of tailings; that is, sulfidic tailings, Fe-ore tailings and bauxite residues:

- Bioweathering via bio-oxidation of reducing minerals, such as sulfides in sulfidic tailings, which are commonly catalysed by lithotrophic microbes (i.e. extremophiles – Fe/S-oxidising archaea and bacteria: *Thiobacillus, Thiohalobacter, Acidiferrobacter*) in tailings rich in sulfides (such as sulfidic Cu, Pb-Zn tailings) (Fortin et al. 1995).
- Bioweathering via bio-reduction of crystalline Fe-minerals (e.g. hematite, magnetite) in Fe-ore tailings (e.g. magnetite Fe-ore tailings), which are driven by organotrophs and Fe-reducing bacteria (Lovley 1987).
- Bioweathering via hydrolysis of minerals (e.g. sodalites) driven by organic acids generated in microbial fermentation of biomass residues in bauxite residues (You et al. 2019).

These microbial mediated mineral weathering processes play important roles in the ecological engineering of tailings by accelerating rapid geochemical reactions into slow reaction phase among reactive minerals in the tailings (Figure 2: State 1–2). The slow reaction phase represents a state of hydrogeochemical stabilisation in which the intensity of acute limitation and/or toxicity to soil microorganisms and plants have been significantly abated (Huang et al. 2014). Depending on the primary mineralogy and pollution risks, primary goals of ecological engineering of tailings may broadly include: (1) profile cementation and hardpan formation at the top layer (ca. 50–100 cm), for example, in sulfidic and metallic tailings, and (2) technosol (or engineered soil) formation to support the growth and development of plant communities, for example, in Fe-ore tailings and bauxite residues.

A hardpan cap at the top layer of sulfidic tailings is the surface layer of extensively weathered and highly cemented tailings possessing low geochemical reactivity due to the exhaustion of reactive sulfides and extremely low porosity and hydraulic conductance due to pore filling with mineral cements (Blowes et al. 1991; Liu et al. 2018). It is advocated to adopt this pathway to develop cementation and hardpan cap for sulfidic and metallic tailings with high pollution risks, though the weathered sulfidic tailings may also be eco-engineered into technosols (You et al. 2018; Yuan et al. 2016). This is because total (not bioavailable) metal(loid) levels in even highly weathered tailings remain excessively high and present high risks of metal(loid) dispersion and transport via air/water/food chain pathways. The hardpan cap may act as a hydraulic barrier separating the reactive tailings below and installed root zones above, thus substantially lowering pollution risks of tailings drainages and improving root zone sustainability for plant growth (Gravina et al. 2004; Liu et al. 2018). The soil system composed of hardpan and reconstructed root zones (i.e. resembling duplex soil system in nature) supported long-term growth of native plants phytostabilising the tailings landscapes in a field trial located at a decommissioned sulfidic Cu-Pb-Zn tailings dam at Mt Isa Mines, Queensland, Australia (Liu et al. 2018).

Technosol is a group of man-made soils and characterised by composition dominated by materials of technical origin (e.g. tailings), pedogenesis driven by ecological engineering (rather than just natural forces), and properties resembling natural soil but at a state of quasi-equilibrium (Schad 2018). The pathway of soil formation has been advocated for tailings of low pollution risks, such as Fe-ore tailings and bauxite residues, which is to eco-engineer soil formation for supporting the growth of diverse plant species, thus offsetting at least large proportions of topsoil volumes required for reconstructing root zones or soil cover.

The following sections will briefly introduce the conceptual pathways of hardpan formation by using recent research findings with sulfidic tailings and technosol formation in bauxite residues. It aims to attract research attention to translate these concepts into field-feasible technologies, for transforming tailings rehabilitation practices and improving economic and ecological sustainability in the future.

2.2.1 Microbial oxidation of sulfides, mineral co-dissolution/precipitation and cementation in sulfidic tailings

The formation of mineral cements or amorphous minerals (such as Fe-Si-gels, hydrous gypsum) is critical to the cementation of tailings particles and hardpan formation (Liu et al. 2018) (Figure 3). Depending on the mineral composition of the tailings concerned, mineral cements within hardpans may include secondary Fe (oxyhydr)oxides such as goethite (α-FeOOH), ferriyhydrite (Fe_2O_3•$0.5H_2O$), jarosite ($KFe_3(SO_4)_2(OH)_6$), schwertmannite ($Fe_8O_8SO_4(OH)_6$) and gypsum ($CaSO_4$•$2H_2O$) across different mine sites (DeSisto et al. 2011). Among these, secondary Fe (oxyhydr)oxides associated with dissolved silicate are considered as important mineral gels in maintaining the integrity of hardpans (Graupner et al. 2007; Liu et al. 2018). The structure of Fe-Si gels is likely to be the Fe (III)-Si or As-Si rich but S poor bonding structures, due to the replacement of surface S fractions (Graupner et al. 2007; Redwan et al. 2012).

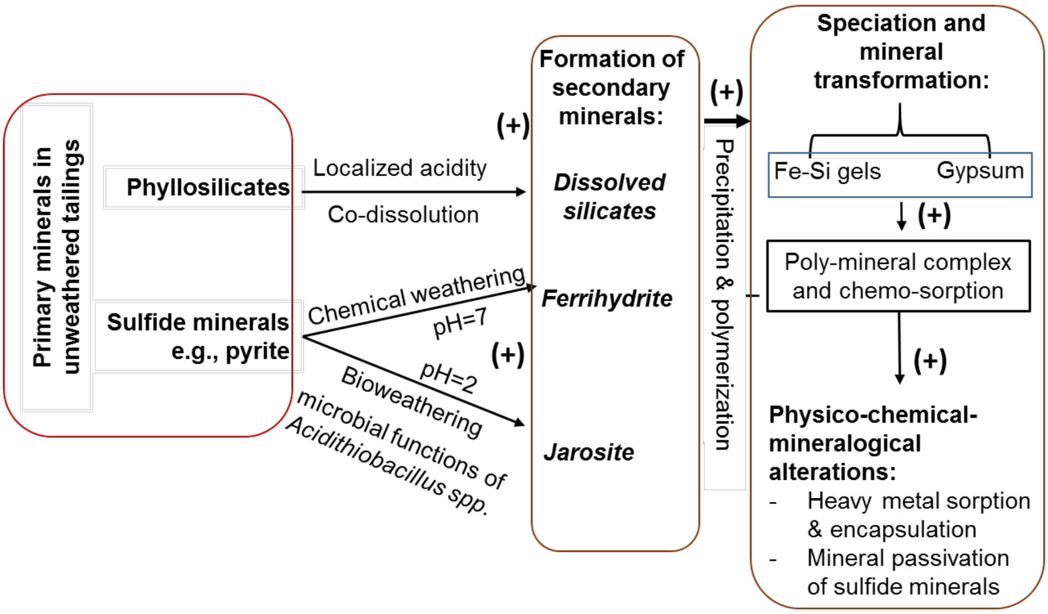

Figure 3 A conceptual diagram illustrating key mineralogical changes leading to the formation of secondary mineral gels, cementation and hardpan formation in sulfidic and metallic tailings. (+) = stimulating or accelerating

These secondary minerals may be formed in situ from the processes of sulfide oxidation, co-dissolution of gangue minerals (e.g. calcites, silicates) caused by localised acidification, and mineral transformation via redox reactions and precipitation, under cyclic wet and dry seasonal conditions (Liu et al. 2018; Mielke et al. 2003; Southam & Beveridge 1992). The bio-oxidation of sulfides by Fe/S-oxidising bacteria (e.g. *Acidithiobacillus*: *A. thiooxidans* (S oxidiser) and *A. ferrooxidans* (Fe and S oxidiser) (Schippers et al. 2010)) is the fundamental step to set off sulfide oxidation, cascaded geochemical reactions, and formation of secondary mineral gels (Figure 3). These extremophiles are commonly present in sulfidic tailings, which are highly tolerant of acidic and metallic geochemical conditions in acidic/neutral drainage water. The growth and activities of these extremophiles in the tailings will depend on growth conditions, such as water supply, acidic pH, and mineral nutrients (e.g. phosphorus). Water supply regimes in the tailings are not only critical to microbe survival, growth and functions, but also redox conditions driving mineral transformation and formation of mineral cements. Despite that the potential of hardpan in sulfidic tailings rehabilitation has been demonstrated (Liu et al. 2018), much research remains to be carried out before field trials may be implemented.

2.2.2 Microbial decomposition of organic carbon, bioneutralisation of alkalinity, and hydrolysis of sodalities in bauxite residues

The geochemical and physical properties of bauxite residues prohibit the colonisation of soil microbes and plants, even the highly tolerant halophytes, because of the extremely alkaline pH (10–13) conditions, acute toxicity of soluble solutes and highly compacted physical conditions (Buchanan et al. 2010; Gräfe et al. 2011; Meecham & Bell 1977; Power et al. 2011). Current closure strategies resort to conventional cover systems, which generally consist of a compressed clay layer (with the option of 1–2 layers of coarse gravel) or a geomembrane cover, and finally a top layer of growth media. This strategy to rehabilitate red mud dams would be far from acceptable regulatory standards for permanent closure. In addition, the high costs (up to AUD 1,000,000 per hectare) make this method unsustainable at industry scale (Huang 2018, unpublished data). These concerns may be addressed by the emerging technology of in situ ecological engineering of 'soil formation' from mine wastes (Huang et al. 2014; Huang & You 2018) (Figure 4).

Recent studies have revealed the potential of soil formation from bauxite residues amended with various combinations of organic (e.g. manure, mulch) and inorganic (e.g. gypsum) materials and managed with agronomic practices, after more than a decade of field treatments (Courtney et al. 2014; Courtney et al. 2013; Di Carlo et al. 2019; Wong & Ho 1994). However, this process of soil formation is too slow for rapid development of desired soil properties suitable for diverse plant species and sustainable plant communities.

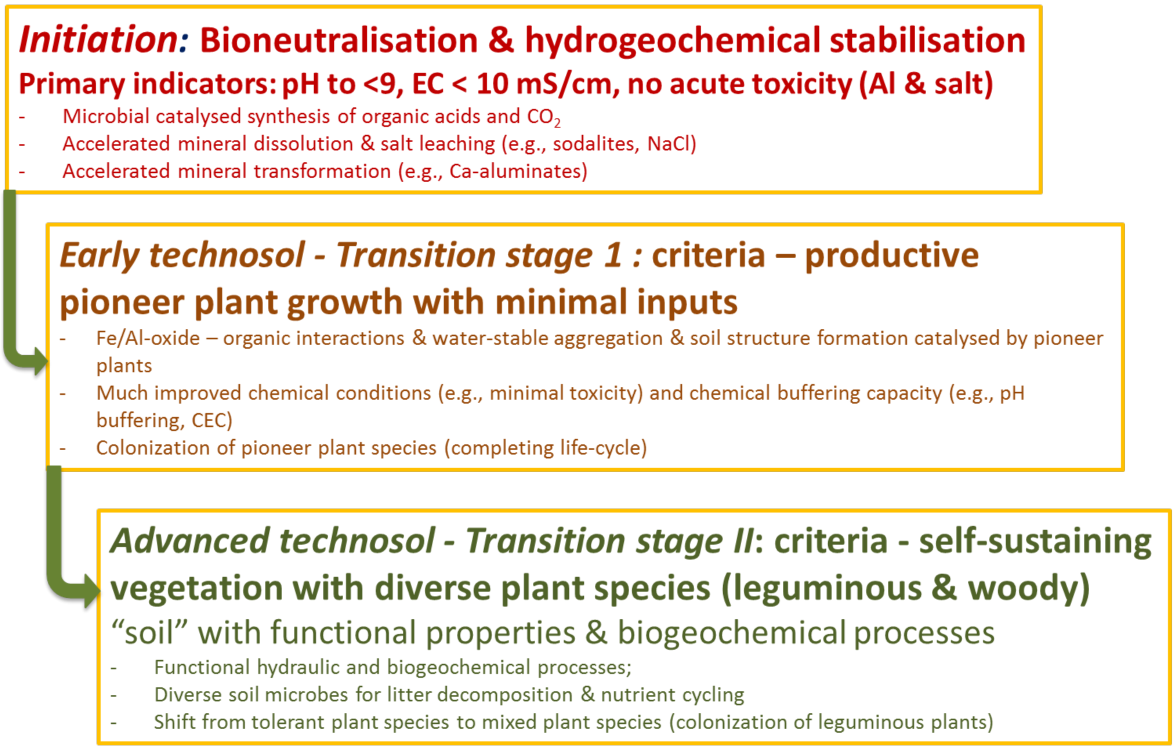

Figure 4 A conceptual diagram illustrating key transition stages of mineralogical, geochemical and physicochemical properties. The proposed transition stages may include (1) early technosol, which supports productive growth and completion of lifecycle of pioneer plant communities; and (2) advanced technosol, which supports the succession from pioneer plant communities into keystone plant communities

In the process of soil formation in bauxite residues, the fundamental mineralogical barrier is the Na-rich and alkaline minerals, such as sodalites, by-products from Bayer processing, which continuously buffer and replenish soluble alkalinity in the porewater (Gräfe et al. 2011). As a result, the hydrolysis of Na in sodalites is the prerequisite to neutralisation and subsequent improvement of physicochemical and biological conditions, resulting in soil formation (Huang & You 2018). The processes to accelerate hydrolysis of Na-rich sodalities should avoid strong acids that destroy the Al-Si mineral structures and require high solution/solid ratios, thus generating high secondary risks of waste water storage and management onsite. Alkaline tolerant and fermentative microbes have been found to decompose added organic matter and generate organic acids in bauxite residues under well-watered (but not flooded) conditions (You et al. 2019). The watering conditions may be maintained by using local greywater (e.g. in Western Australia) or dammed rainwater in (sub)tropical regions (e.g. in Northern Territory and Central Queensland).

The effectiveness of bioneutralisation and rate of pH reduction increase with the admixing rate of green organic matter, which was found to be modulated by addition of Ca-minerals (such as gypsum). The amount and rate of organic acid production in the amended red mud are closely affected by water relations, organic matter properties and ambient conditions, which are generally favoured by anaerobic or anoxic conditions. The resultant CO_2 from microbial activities also contribute to neutralisation of the extreme alkalinity by formatting carbonate minerals. However, the strong buffering effects of alkaline minerals in the (sea water neutralised) red mud may revert the bioneutralisation effects, leading to pH rise after a short-term pH reduction when the organic acids are exhausted in the porewater (You et al. 2019). Elevated levels of soluble Ca may stimulate cation exchange of the Na-bound in solid phase of the red mud, facilitating leaching process and removal of the high Na levels, and thus consolidating the effectiveness of bioneutralisation. The first stage of technosol formation could only be possible if sodalities have been largely hydrolysed, the strong alkalinity abated to <pH 9, and salinity decreased to the threshold for the growth of tolerant pioneer plant species. Further research under field conditions will be necessary before cost-effective technologies may be formulated, together with proven standards and criteria of key soil indicators (Huang & You 2018).

3 Conclusion

From the discussions above and relevant literature, it is realised that conventional remediation approaches are ineffective in resolving the mineralogical factors governing key geochemical conditions (e.g. oxidation of sulfides and acidification) and physical properties (poor aggregation and structure), unless the remediating materials and practices are provided in high quantity and frequency over the long-term. In some (but limited) situations where large volumes of local resources of topsoil and crushed rocks are reasonably available (though not up to 100%), tailings may be improved directly through dilution method by admixing high proportions of these materials into the tailings, such as Fe-ore tailings and bauxite residues, for the sake of rapid phytostabilisation with pioneer plant communities. However, these approaches are not economically sustainable for rehabilitating hundreds and thousands of hectares of tailings landscapes.

The ecological engineering approach represents a new way to rehabilitate tailings for cost-effective, non-polluting and sustainable outcomes. In this new approach, bioweathering of target minerals (such as sulfides and sodalites) is accelerated by microbial activities boosted by the provision of suitable conditions (such as water and mineral nutrients). Geo-microbial processes involved in mineral weathering and transformation have been well established in laboratory experiments. However, to translate these fundamental concepts and knowledge into field-based technologies requires substantial field-based research efforts. A close partnership between industry-users and researchers is the key to the progress of field-oriented research and investigations, leading to breakthroughs of innovative and cost-effective technologies, which should have high operational feasibility and minimal secondary risks. The comprehensive understanding of complex ‘real world’ problems in the tailings landscapes and mining operations are essential to the success of this kind of translational research.

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