



Recovery of strategically important critical minerals from mine tailings

Shuronjit Kumar Sarker^a, Nawshad Haque^b, Muhammed Bhuiyan^a, Warren Bruckard^b,
Biplob Kumar Pramanik^{a,*}

^a Civil and Infrastructure Engineering Discipline, School of Engineering, RMIT University, VIC 3001, Australia

^b CSIRO Mineral Resources, Clayton South, Melbourne, VIC 3169, Australia

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ABSTRACT

Society's hunger for commodities is leading to an increased consumption of minerals considered critical or strategic. A range of minerals containing elements such as lithium (Li), cobalt (Co), rare earth elements (REEs) are considered critical and more important for strategic uses than others. In this paper we describe these as strategically important critical minerals (SICMs). However, their continuous depletion from primary sources coupled with supply risks due to geopolitical issues and geographical segregation is a major concern. As a consequence, recovering these valuable elements from non-conventional sources such as abandoned mine tailings has recently gained increased worldwide attention. In some part this is due to the fact that the potentially recoverable amount of these elements in abandoned mine tailings is often higher than the concentration in some primary ores. A review of the scientific literature reveals the use of modern recovery techniques such as tailored made hydrometallurgical and bio-hydrometallurgical processes can lead to effective recovery of these elements from low grade sources such as mine tailings. However, there remain some technical, economic and environmental challenges associated with recovering SICMs from mine tailings. This review critically analyzes these challenges and discusses the opportunities available for recovering SICMs from abandoned mine tailings using conventional hydrometallurgical techniques as well as bioleaching methods, which can offer significant advantages in reprocessing. This paper also concludes by providing an outlook of an integrated approach to the reprocessing of mine tailings where the recovery of SICMs as well as clean water production should be the combined overall reprocessing and recovery goal, helping to realize the full economic potential of the tailings.

1. Introduction

Engineered application of some chemical elements of metals, non-metals and minerals into high-tech products have made our modern lives much easier than ever before. These chemical elements include REEs (e.g., cerium, lanthanum, neodymium, dysprosium, praseodymium, scandium, erbium, europium, terbium and yttrium), precious metals (e.g., rhodium, palladium, and platinum), radioactive metals (e.g., uranium and radium) and alkaline metals (e.g., magnesium, potassium, and Li). Their applications are widespread in high-tech industries which are critical to modern society and sustainable development. Such industries include telecommunications, renewable energy, electric vehicles, aerospace, medical, agriculture and defense technologies. Moreover, the uses of these minerals in these industries are expected to grow significantly in the coming decades. This is likely due to increasing population with increasing standards of living for the vast majority of

the world's population, and meeting the targets of the low-carbon society to contain the impacts of climate change. For example, the demand for Li, Co, manganese (Mn) and aluminum (Al) is expected to be increased by 12 times in 2050 compared to 2013 [1]. However, the supply of these minerals may be at risk due to various reasons including geological scarcity, geopolitical issues and trade policies [2]. The geopolitical and trade policies pose the greatest threat to the supply disruptions of these minerals as their production is concentrated only in few countries. For example, the production of REEs, Co and Li are concentrated mainly in China (59%), the Democratic Republic of Congo (DRC) (68%) and Australia (49%), respectively [3]. Furthermore, for most of the critical minerals, there are no true substitutes, meaning that the consumers, economies and deployment of low-carbon technologies could be significantly affected if they are subject to supply restrictions. Due to the perceived unreliable supply of these minerals against their multi-sectoral importance, they are known as critical elements or critical

* Corresponding author.

E-mail address: biplob.pramanik@rmit.edu.au (B.K. Pramanik).

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raw materials (CRM) or critical minerals in many of the world's largest economies such as the United States of America (USA), European Union (EU) including United Kingdom (UK), and Australia.

The definition of critical minerals varies from one country to another depending upon the need for a particular element for a particular time period, and some other criteria such as supply risk, environmental implications and vulnerability to supply restrictions [4]. The EU defines CRM as "raw materials of high importance to the EU economy and of high risk associated with their supply". However, the USA defined critical minerals in the Presidential Executive Order No. 13817 as "a mineral (i) identified to be a nonfuel mineral or mineral material essential to the economic and national security of the United States, (ii) from a supply chain that is vulnerable to disruption, and (iii) that serves an essential function in the manufacturing of a product, the absence of which would have substantial consequences for the U.S. economy or national security". Similarly, Geoscience Australia defines critical minerals as "metals and non-metals that are considered vital for the economic well-being of the world's major and emerging economies, yet whose supply may be at risk due to geological scarcity, geopolitical issues, trade policy or other factors". Moreover, the number in the list of critical minerals varies over time. For example, the list of CRM of EU consists of 30 elements in 2020, while it contained 14, 20 and 27 elements in 2011, 2014 and 2017, respectively. Table 1 below shows

Table 1

List of critical minerals in USA [5], EU including UK [6,7], and Australia (Geoscience Australia, [8]).

USA	EU (including UK)	Australia
Aluminum (bauxite)		
Antimony	Antimony	Antimony
Arsenic		
Barite	Barite	
	Bauxite	
Beryllium	Beryllium	
Bismuth	Bismuth	
Cesium	Borate	
Chromium		
Cobalt	Cobalt	Cobalt
	Cooking Coal	
Fluorspar	Fluorspar	
Gallium	Gallium	Gallium
Germanium	Germanium	Germanium
Graphite (natural)	Graphite (natural)	
Hafnium	Hafnium	
Helium		
Indium	Indium	Indium
Lithium	Lithium	Lithium
Magnesium	Magnesium	
	Natural Rubber	
Manganese		
Niobium	Niobium	Niobium
	Phosphorus	
	Phosphate rock	
Platinum group metals	Platinum group metals	Platinum group metals
Potash		
Rare earth elements group	Rare earth elements (Heavy)	Rare earth elements group
	Rare earth elements (Light)	
Rhenium		Rhenium
Rubidium		
Scandium	Scandium	
	Silicon metal	
Strontium	Strontium	
Tantalum	Tantalum	
Tellurium		
Tin		
Titanium	Titanium	
Tungsten	Tungsten	Tungsten
Uranium		
Vanadium	Vanadium	
Zirconium		

critical minerals in USA, EU and UK and Australia.

Not all critical minerals are equally important when it comes to some strategic usages. The critical minerals that are used in strategic uses often called as strategic minerals. The term "strategic minerals" was used first in the USA in early nineteenth century. Though the definition of strategic minerals is ambiguous [9] and there is no common definition in the literature, some tried to come with a definition based on their assumptions. For example, the Colorado Geological Survey (CGS) defines strategic minerals as "commodities essential to national defense for which the supply during war is wholly, or in part, dependent upon sources outside the boundaries of the U.S.". Similarly, in a study material on Geology and Economics of Strategic and Critical Minerals by McLemore at New Mexico Institute of Mining and Technology, it has been defined as the critical minerals that are used in defense technologies called strategic minerals. However, there is no clear list of the critical minerals that are considered strategic in either of the sources and in the current literature. Using the term "strategic metal*" or "strategic mineral*" in Scopus yielded 163 search results, in which 133 documents were in English. Some articles mentioned Li, Ni, Co, and Mn [10], Scandium (Sc) [11], Cr, Mb and V [12], and Co [13] as the strategic metals. It is to be noted from these literatures that the authors had the liberty of calling these elements "strategic" may be due not having any established definition for it and their key role in certain key industries. Taking these two points into account, the authors of this article would like also to have the liberty to call some industries, such as renewable energy industry, including wind turbine and solar energy, and electric vehicle industry, as strategic in which Li, Co and REEs are vital ingredients. Acknowledging the roles of Li, Co and REEs in these industries, we would like to call this trio the strategically important critical minerals (SICMs) in this review article.

Generally, metals are extracted from ores collected from hard rock mines. Recent advancement in mineral extraction technologies and declining ore grades in primary deposits have led to the idea of reprocessing of mine tailings as the potential secondary source of some of the critical minerals [14-17]. The mine tailings are mixture of fine-grained solid materials remained after extraction of recoverable metals and minerals, and water used in the process [18]. There are two main reasons for extracting critical minerals from mine tailings, particularly from old and abandoned tailings. The first reason is that the mining cost is significantly reduced as the ores have already been collected and partly processed and ground. The second reason is that the likelihood of finding valuable minerals at economic concentration is higher in older tailings than in newer ones. This is because the technologies used in the past were less efficient than they are now. Moreover, many critical minerals are generated as byproducts or companion products of major commodities during ore processing [19-21]. In addition, these by-products minerals had very little uses in the past, which resulted in a little desire to extract them regardless of their concentration. However, with an increased application of these critical minerals in rapidly diversifying high-tech industries and ever declining discoveries of new mineral deposits with economic ore grades, recovering the critical minerals from mine tailings is increasingly gaining attention. It is noted that the concentration of some valuable minerals in old mine tailings may surpass some primary ores ([22], Edraki et al., [23]). This indicates that old mine tailings could be a secondary source of critical minerals. Moreover, reprocessing of tailings for minerals recovery, including some heavy metals such as copper (Cu) and zinc (Zn), can help reduce the volume of the tailings to be managed, and concentration of some potentially toxic elements such as Cu and Zn. The processed tailings, which become less harmful to the environment, can be used as backfills for open pits and underground mines [24,25], building materials and agriculture fertilizer (Hu et al., 2017). Repurposing tailings these ways have the potential for both economic and socio-environmental benefits (Vitti & Arnold, [26]).

However, there remain challenges in the recovery of critical minerals from mine tailings. The main challenges are due to technological

inefficiencies and low economic output. Commonly, most mineral recovery technologies are designed to target primary ores and, using them on mine tailings, which are mostly low grade, can often lead to low mineral recovery outcomes. This may make investment for critical minerals recovery from mine tailings risky [14]. However, a number of technologies have been developed and used successfully to recover critical minerals from mine tailings. Principally, most of these technologies are hydrometallurgical based such as froth flotation, solvent extraction and bio-hydrometallurgy or bioleaching. In some studies phyto-extraction, which is an in situ method employing certain kind of plants to bioconcentrate metals or metalloids in their shoots to remove from their biomass ([27], Vitti & Arnold, [26], [28]). However, solvent extraction has been one of the most commonly used methods for extracting minerals from both primary and secondary sources. In this method, acid leaching is a key step to dissolve minerals by use of H_2SO_4 , HCl, HNO_3 acids to leach the concentrates after physical beneficiation [29,30]. The leached liquor is then processed further using solvent extraction to separate the pure compounds [29]. Unlike solvent extraction, bio-leaching processes use various microorganisms to solubilize followed by solvent extraction to separate metal elements from solid phase [31].

A number of studies have reported recovery of minerals including some SICMs from mine tailings. For example, recovery of REEs from mine tailings was investigated and found promising results by a number of researchers such as Peelman et al., [32]; Tunsu et al., [16]; Sutterlin, [33]. Similarly, several studies have been conducted to recovery Co from mine tailings by Zhang et al., [34]; Mäkinen et al., [35,36]; Chen et al., [37], and reported promising results. Recovery of Li, however, from mine tailings is less common in the literature than recovering it from other secondary sources such as spent Li-ion batteries [38-41] and brine solutions [42-44]. More recently, a study by Zhang et al. [17] investigated Li recovery from bauxite mine tailings using a mixed acid leaching method, and they found 96.35% leaching efficiency.

A number of review articles on resource recovery from mine tailings are available in the literature. Notably, a review article by Park et al. [45] discussed about different strategies of recycling of the wastes generated by the mining, mineral processing, and extractive metallurgical industries. They have suggested preventing AMD generation rather than remediation due to cost, time and sustainability issues. The article discussed some studies that recommended tailings recycling and reprocessing, and recommended further studies on environmental impacts. In another review article by Lyu et al. [46] discussed on environmental implications, recycling strategies, and ecological remediation of mine tailings. However, none of the articles discussed about the challenges of reprocessing and recycling of mine tailings for critical mineral recovery. Similar gap has also been noticed in some other studies. For example, a review article by [47] has presented a wide range of information on global supply-demand, market flow and status of Li resources including a range of both primary and secondary sources. Nonetheless, the discussion did not cover the challenges of recycling of the Li sources, especially from non-conventional sources such as tailings. In a recent study by Meng et al. [48] discussed Li production and recovery from different primary and secondary sources such as minerals, brines and Li-ion batteries. Their discussion consisted of application of a number of processes to recover Li from these sources and recommendation for future research on sustainable and simplified Li recovery processes. Though the article discussed about Li recovery from secondary sources (Li-ion batteries), but it lacked in considering mine tailings as one of such potential sources. More recently, a short review article has been published by Vitti and Arnold [26] on reprocessing and revalorization of mine tailings for critical minerals recovery for both socio-economic and environmental wellbeing. The article discussed about some environmental friendly methods to recover critical minerals from tailings. Yet again, the review also does not have any critical discussion on the challenges in reprocessing of mine tailings for resource recovery as well as recycling.

There is significant gap in current literature of considering the potential of recovering other critical minerals such as REEs and Co, and critically discussing the challenges and opportunities present in reprocessing of mine tailings for resource recovery and recycling. Therefore, the purpose of this review is to fill the gap by discussing a systematic understanding of REEs, Co and Li recovery potential from mine tailings, and discuss the challenges and opportunities present in such recovery projects. This article will also conclude by providing an outlook of the future perspective and research needs in regards to mine tailings as a secondary source of SICMs. The recovery methods currently in practice for recovering SICMs from both primary and secondary sources were thoroughly reviewed and critically analyzed for their recovery technical efficiency, environmental footprints, and socio-economic impacts.

2. Strategically important critical minerals

2.1. Lithium (Li)

Li has been listed as a critical mineral by several major economies such as the USA, EU, UK and Japan. It has many industrial uses, ranging from glasses, ceramics, pharmaceuticals, and polymer production. But the single biggest market is the battery industry in which it is used in disposable batteries and Li-ion rechargeable batteries. The use of Li in rechargeable batteries began in 1992 due to its greater charge-to-density ratio and its uses in the battery industry have kept growing ever since. Now the battery industry accounts for 46% of the global consumption of this mineral [49]. The consumption of Li-based batteries has also increased significantly in recent years, owing to an increase in the market size for the renewable energy sector and electric and hybrid vehicles [50]. The increase in demand of Li-based batteries can be attributed to their qualities of long cycle life, high energy density and low environmental impacts [51,52]. The energy density, also referred to as “driving range per charge”, is the most important quality for rechargeable batteries in electric cars. At present Li-based batteries have one of the highest energy densities (100–265 Wh/kg or 250–670 Wh/L) of any battery technology [53]. Energuide (2021) calculated that most electric automobiles consume 15 kWh of power to travel 100 kilometers. Another study by Gaines and Nelson [50] estimated that the amount of Li required to travel a distance of 65 km on the road without any interruption, and found that such a trip in an electric car would require 1.4–3.0 kg of Li before requiring recharge. A typical electric car uses 1.4 kg Li carbonate for its battery with a capacity of 5 kw [54]. As the demand for electric cars is soaring, the demand for Li is also expected to increase. The demand for Li rose from 34.6 kt in 2015 to 49.0 kt in 2019 [55]. Moreover, this progressively growing demand for Li is expected to continue in the future with a view that the demand will exceed the supply in the next 10 years [56].

The production of Li has also grown proportionately to meet demand. The global production of Li has increased from 14.0 kt in 2000 to 43.0 kt in 2017 [49]. However, the production of Li decreased to 77 kt in 2019 from 95 kt in 2018 [55]. This was due to excess production relate to consumption and reduced prices of Li. The global production of Li stayed at 82,000 tons at the end of 2020, 5000 tons increased from 2019 [3]. The production of Li is predicted to rise further, and it is expected that the production will be triple in size by 2025 from 0.5 million tons in 2019–1.5 million tons in 2025 [57]. A report by Goonan [54] expressed concerns that this demand rate is unsustainable because of geochemical issues in extracting Li from known sources. Despite the concerns that Li production will become unsustainable, Greim et al. [58] optimistically suggests that there is a limited risk of Li gets runs out in this century. Many major economies around the world have considered Li as a critical metal, based mainly on its geographical segregation. Both production and reserves of Li are concentrated in only a few countries, notably Australia and Chile. Chile has the largest reserves of Li but is currently the second-largest producer after Australia which has a reserve of only

33% of that of Chile [55]. Despite Australia being the largest producer of Li, it consumes a relatively small amount. Being a small consumer while being the largest producer with the second largest reserve, Australia possesses great economic potential for supplying Li to the world. By some estimate, it was suggested that Australia could account for around 80% of global Li supply from hard rock deposits during 2019–2020. This makes Li a SICM for Australia. Currently, the worldwide reserve of Li is estimated to be 21 million tons in total [3] and a majority of this reserve is concentrated in Chile, Australia, Argentina and China. Li is mainly produced from two distinctive sources: Li containing minerals, mostly from spodumene ($\text{Li}_2\text{O}\cdot\text{Al}_2\text{O}_3\cdot 4\text{SiO}_2$) (Geoscience Australia), and brines, which is a highly saline solution with an average concentration of 170–330 g/L [59]. There are significant resources of Li containing minerals in Australia, China, Canada, but brine is the most significant source for Li production in Chile, Argentina, China, and USA (Geoscience Australia, [8]).

2.2. Rare earth elements (REEs)

According to the International Union of Pure and Applied Chemistry (IUPAC), REEs are a group of seventeen chemical elements that include fifteen metallic elements of the lanthanides, and yttrium (Y) and scandium (Sc). Both yttrium and scandium are included in the REEs group because they exhibit similar geochemical properties to lanthanides [60, 61]. The REEs are subdivided into two sub-groups: light REEs (LREEs) and heavy REEs (HREEs). The LREEs are from lanthanum (La) to europium (Eu) and HREEs are from gadolinium (Gd) to lutetium (Lu) including yttrium. Both LREEs and HREEs are found in the same deposits, scandium is an exception [62]. This is the reason why scandium is not included in either of the subgroups of LREEs and HREEs.

The REEs are unique in their electrical, magnetic and optical properties. Therefore, their applications in technologies have grown significantly. These technologies include clean energy technologies (solar panels and wind turbines), defense technologies and advanced high technology devices, such as flat-screen displays, computer hard disks, smartphones, digital cameras, fluorescent and light-emitting-diode lights. A large quantity of REEs is used in renewable energy technologies such as wind energy. For example, a 3.5 MW wind turbine requires nearly 500 kg of neodymium, molybdenum, and dysprosium to produce permanent magnets [63]. With increasing demand for wind energy worldwide, the demand for REEs in permanent magnets will increase. Apart from wind turbine, electric vehicle is another industry in which a large amount of REEs is required. It is estimated that an average electric vehicle uses between 2 and 5 kg of REEs depending on the design [64]. As electric vehicle is increasingly becoming the future of road transport with prediction of 7.2 million vehicle sales worldwide in 2020, this would demand for 14 times more REEs in this sector compared to 2015 [65]. A study by Alonso et al., [66] reported that the demand for REEs was nearly 100,000 tons in 2010, which would reach to nearly 1.5 times more by the end of 2020. But the growth in rare earth demand is expected to fall back in 2020 due to COVID19 related disruptions impacting industrial production. However, the demand graph is climbing again in 2021 and if this trend continues, the demand for REEs will be increased by 10% of 2020 by the end of 2021 [67]. A study by Alonso et al., [66] estimated that the demand will reach to about 400,000 tons by the end of 2035.

The supply and demand of REEs were in well balanced during the period from 2010 to 2015. The supply of REEs was 200,000 tons in the form of rare earth oxides (REOs) in 2015, leaving a surplus of 50,000 tons [68]. But the demand is expected to exceed the supply in the near future as a result of more countries with large populations needing to technologies using REEs. Despite the progressive demand of REEs, their future supply cannot be guaranteed due to mainly geological scarcity. Moreover, the occurrences and production of REEs are geographically segregated. The REEs are currently produced by a few countries only, notably China, which produced 59% of these minerals in 2020 [3].

Given their future supply uncertainty due to geological scarcity and geopolitical policies, their essential role in the latest technologies, they are regarded as critical minerals by many countries.

The REEs are relatively abundant in the earth crust and they are found in the form number of minerals, such as oxides, carbonates, silicates, phosphates and halides [69,70]. But the occurrences of economic ore deposits are rare, and mostly the REEs are in low concentration [62, 71]. The current global minable reserve of REEs is estimated to be approximately 120 million tons. However, they are not uniformly distributed across the world rather a large portion of it which is 44 million tons is located in China [3]. There are other countries that have a significant amount of REEs reserve after China. These countries are, in descending order, Vietnam, Brazil, Russia, India and Australia with reserves of 22, 21, 12, 6.9 and 4.1 million tons respectively [3].

The current annual global production of REEs is about 240 kilotons [3], yet the demand for REEs is expected to grow further as the renewable and electronic industries continue to expand. The annual growth rate of global REE demand is estimated to be about 5% [72,73]. A similar prediction was made by Dutta et al. [74] who noted that global REE demand is expected to grow at an annual rate of 5% by 2020. Graedel et al., [19] indicated that there would be scarcity and supply risk issues for some elements of REEs such as europium (Eu), dysprosium (Dy) and erbium (Er), in addition to several others in the near future. This prediction was based on their scores in three areas: supply risk, environmental implications, and vulnerability to supply restrictions. Hence, the global availability of REEs appears to be at substantial risk for several reasons. These include the introduction of production quotas, export quotas and export taxes, enforced environmental legislation, and low rates of granting new rare earth mining licenses [75]. Furthermore, the production of REEs is concentrated only in few countries, notably China which accounts for more than 80%. This makes REEs probably the critical minerals of greatest concern. As a result, there has been increased interest and investigation into non-traditional REEs resources, extraction, separation and purification processes.

2.3. Cobalt (Co)

Co has unique combination of properties which make it ideally suited to many varied applications. Many of the Co applications are also critical and strategic. These include commercial, industrial, and military uses. A large amount of Co is used in high temperature, high-wear applications including superalloys for jet engines; magnets; carbides; and diamond tools. Co is also used in bulk quantities in batteries, catalysts and pigments. For example, a Li-ion rechargeable battery in a cell phone requires 190 kg of Co to generate 1 megawatt (MW) of power [76-78]. The number of cell phones sales has increased dramatically in the last decade. The number of cellphones sold globally was 122 million in 2007 and the sales gradually rose to over 1540 million in 2019. But the number of sales was slightly less in 2020 due to the COVID-19 pandemic. However, the number is on the rise again and expected to come back to a normal progressive trend by the end of 2021 [79]. With the increased number of sales of cellphones predicted, the demand for Co used in the cellphone batteries will also increase. The demand for Co in the rechargeable battery industry was 45000 tons in 2015, which accounted for 49% of total Co consumption [80,81]. An electric vehicle is another industry in which Co is used in bulk quantities in rechargeable batteries. The electric vehicles are powered by Li-ion rechargeable batteries containing Co as an essential element. It is estimated that as many as 130 million electric cars will be running on the road worldwide in 2030, which is 40 times more than what was in 2017 [80]. A study by Fu et al., [82] estimated that the demand for the Li-ion battery will increase by 300% throughout the next decade. This means that the demand for Co, which is an integral part of Li-ion batteries, would also increase accordingly. It is estimated that the demand for Co would be in the range of 235–430 kilotons in 2030 [82].

The production and demand of Co remained well balanced during

the last decade with production at 89,500 tons in 2010. In 2017, global Co production increased by 68% [49]. The production gradually rose to 144,000 tons in 2019, but the COVID-19 pandemic caused the production to decline a bit to settle at 140,000 tons in 2020 [3]. However, the production is predicted to increase again and will continue to meet the demand until 2025. After 2025, the Co supply may not be able to meet the growing demand. With projected demand exceeding supply in the future this will escalate the Co price, which was tripled between 2016 and 2018 (Elves et al., 2018). Further increases in the Co price may elevate the price of aligned commodities, such as electric cars, which are considered critical to achieving global temperature targets to combat the large-scale impacts of climate change. Restricting supply to raise the Co price is voluntary and cannot be controlled as the production is geographically concentrated. Currently, the DRC is the largest source of mined Co, supplying approximately 68% of Co mined production [3].

The total global reserve of Co is estimated to be around 7.1 million tons, of which the DRC holds the largest reserve of 3.6 million tons [55]. Australia has the second largest reserve of Co, but it only produced 4971 tons in 2020 [55]. The global production of Co was 140 kt in 2020 and the DRC produced highest 68% of Co [3]. Co is primarily produced as a by-product of copper and nickel ore processing and production. As a result, Co supply is closely tied to the copper and nickel markets. Examples of SICMs' demand (kg/MW) in electric cars, wind turbine and rechargeable cellphone batteries are shown in Fig. 1.

2.4. Impacts of Covid-19 on SICMs

Without warning Covid 19 pandemic has impacted global economy rapidly, including whole supply chain of critical minerals. The pandemic has increased Li use due its widespread applications in medical devices, such as ventilators, which has been critical during this covid time. On the contrary, due to decreased sales in electric vehicles (EVs) Li-ion battery industry experienced slow growth in last couple of years [85]. Nonetheless, the industry is predicted to grow by many folds from USD 46.8 billion in 2021–168 billion in 2026 [86]. The pandemic has also impacted REEs market too due to being key materials for many consumer electronics, magnets, and medical devices. Due to social distancing and lockdown during the pandemic the demand for consumer electronics, such as computer, smart phones, increased significantly. But shut down of manufacturing plants resulted in decreased in supply. However, the market for REEs is predicted to rise again due its application in production of magnets-key items in various industries such as EVs, electronics, power generation, and medical. It was estimated that the market for consumer electronics was USD 384 billion in 2020, which is expected to grow to USD 415 billion in 2021 and USD 487 billion in 2026 [86]. Similarly, the Covid-19 had a number of impacts on Co market. Average annual prices of Co declined in 2020 compared to the year before due to reduced demand during the peak of Covid 19 pandemic [3]. There was temporary suspension of Co operations in some production facilities in DRC, the largest producer of the mineral, but the production was not much affected by Covid, as other production facilities exceeded yearly target of the production. However, most of the

production facilities were temporarily shut down to prevent the spread of the virus in some countries, such as Madagascar and South Africa, which resulted in the traders turned into other suppliers from Mozambique, Tanzania and Namibia (Cobalt Institute, [87]).

3. Characteristics of tailings containing SICMs

Characteristics of tailings play a key role in evaluating the SICMs recovery potential. The tailings characteristics differ with ore deposits which determine what critical minerals can be recovered. The ores containing valuable minerals are typically mined and processed to extract the valuable minerals. The extraction of the desired minerals, which often are the host metals, such as copper, iron, zinc, can be accompanied by other metals and minerals as by-products. These by-products, also called companion products, often are the critical minerals such as Li, REEs and Co [20], some of which are strategically important [84]. Principally, the SICMs are hosted by a wide range of minerals. For example, REEs can be found in more than 200 minerals, but only 3 REE-bearing minerals, such as monazite [(La–Gd, Th)PO₄], bastnäsite [(Ce, La, Y)CO₃F], and xenotime (YPO₄), are considered to be economically significant [88–90]. In commercial REEs production, they are recovered as major elements from its monazite deposit, which is a phosphate mineral [(Ce, La, Nd, Th)PO₄], which contains around 50% of the REEs and around 25% phosphate in oxide form. Though Co can be found in more than 100 minerals, but only 30 of them are known to have Co in recoverable amounts. The most common rock-forming minerals containing Co in concentrations high enough to support economic extraction are spinel, olivine, chloride in lateritic and hydrothermal deposits (Cobalt Institute, [87]). Moreover, Co can always be found in association with some metal minerals, for example, pyrite, chalcopyrite, carrollite and pentlandite [91,35,36,34]. As opposed to REEs extraction, more than 50% of the Co is produced as by-product of extracting of other metals, such as Cu and Ni, which are hosted by a wide variety of deposits. The deposits mostly consist of Cu-Co sediment bearing deposits, Ni-Co laterites, Ni-Cu-Co sulfides or hydrothermal and volcanogenic deposits [92]. Among these, the Cu-bearing minerals found in porphyry deposits is considered to be one of the major sources of Co production as a by-product. Though porphyry Cu deposits can be potential sources of Co as a by-product, the Co separation processes complex due to the nature of the porphyry deposits whose formation is also highly complex. A number of mine sites in the world process Cu-bearing porphyry ores and produce tailings that could be studied for potential Co extraction. For example, the Cu mine sites in Chile, such as Escondida Cu mine, which processes porphyry ores to produce Cu, Au, and Ag. Currently, Escondida Cu mine produces the largest quantity of Cu, 1400 kg ton per annum, in the world [93,94]. There are several others Cu-bearing porphyry ore processing mine sites, such as Collahuasi in Chile, Buenavista del Cobre (former Cananea) in Mexico, and Morenci in USA [93,94], which may be explored for Co recovery.

Like Co, Li can be found in nearly 145 minerals. However, only a few minerals, for example, spodumene, petalite, and lepidolite, are economically worthwhile to process [95,17,96]. The spodumene, which

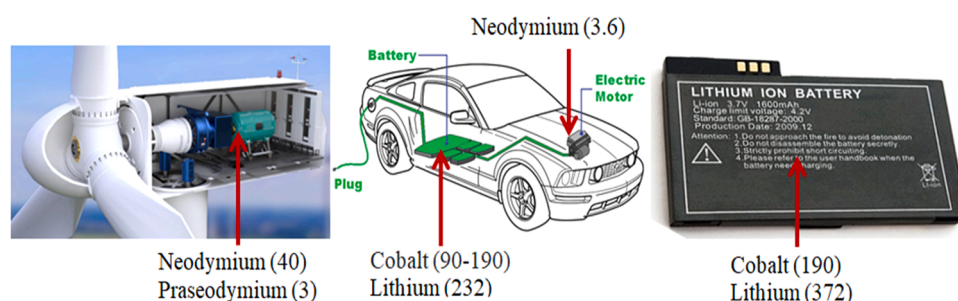


Fig. 1. The demand (kg/MW) of Li, Co and REEs are in electric cars, wind turbine and rechargeable cellphone batteries [83,76,77,84,78].

is pyroxene mineral consisting of lithium aluminium inosilicate, LiAl (SiO₃)₂, deposits contain an average of 1–3% of lithium oxide [97]. Tailings produced from the abovementioned ore deposits have the higher chances of SICMs recover potential due to their geochemical properties. Therefore, understanding the mineralogical information is vital in determining which tailings are worth exploiting for resource recovery. Moreover, the concentration of target minerals in tailings varies due to presence of other non-valuable elements, also called gangue. Some critical minerals can be disseminated and intergrown with gangue minerals. This can make recovery of critical minerals complicated and often may not be cost effective [98]. The studies listed in Table 2 conducted on tailings samples collected from different countries and a brief description of the mine sites is given. A study by Peelman et al. [32] investigated the REEs recovery from tailings samples collected from Luossavaara-Kiirunavaara Aktiebolag (LKAB) mine in Kiruna, Sweden. The LKAB mine site processes iron ore and produces tailings rich in REEs, which has REEs in concentration of 1200–1500 ppm. The tailings at the LKAB mine site is one of the largest sources of REEs in Europe. In an article by Reynier et al. [99] reported the recovery of REEs from samples of tailings collected by filed campaign from two tailing storage facilities in a restored uranium mine site located in the Elliot Lake area, Ontario, Canada.

Similarly, for Co recovery, a study by Mäkinen et al., [35] conducted on the tailings supplied by a material owner, but detail information of the mine site was not provided. In another similar study by Zhang et al. [34] for Co recovery from tailings, the samples were collected from the tailings storage facilities from an abandoned mine site named Ramelsberg mine located in the historic Harz Mountains mining district in central Germany. The mine site was operated till 1988 and has stored flotation tailings in the Bollrich tailings pond. The German Federal Ministry of Education and Research (BMBF) investigated the tailings and reported that the tailings contain Cu, Co, Zn, Ag, Ga, In, and other valuable metals. A process scheme for Co recovery was developed and tested at a laboratory scale. [36] studied Co recovery from tailings collected from an old Cu mine in the district of Kasese located 420 km west of Kampala, Uganda. The mine site produced around 16 million tons of Cu and Co-rich pyrite concentrate. The concentrate contained about 80% pyrite and 1.38% Co. It was estimated that the concentrate contained over 11000 tons of Co. For Li recovery from tailings, Zhiqiang et al., [96] investigated tantalum–niobium mine tailings collected from Yichun Tantalum and Niobium Mine (Jiangxi Province, China) tailing dam.

4. Technologies for recovering SICMs from mine tailings

The recovery processes of minerals from secondary sources often differ from the processes used for primary sources. This is because the concentration of minerals in secondary sources is less than that of primary sources and the nature of the minerals/materials is different. A number of processes have been used to extract minerals from mine tailings. Most of these processes belong to hydrometallurgical techniques. These include solvent extraction, acidic leaching, liquid-liquid extraction and bioleaching, all of which are hydrometallurgical in nature. These hydrometallurgical processes rely on the use of aqueous chemistry to recover metals from ores, concentrates and recycled or residual materials at relatively low temperature. The hydrometallurgical techniques for minerals recovery from mine tailings is discussed in the following section.

4.1. Hydrometallurgical extraction

Hydrometallurgical extraction is a commonly used method to extract minerals. The method has proved to be cost-effective for mineral recovery from mine tailings that have a relatively higher concentration of target minerals [100]. A number of hydrometallurgical processes have been used to extract the critical minerals from mine tailings. Fig. 2 shows

Table 2
Chemical and mineralogical composition of some selective tailings containing SICMs.

Strategic element	Major chemical composition (in wt%)											References					
	Ca	P	TREE (ppm)	Fe	Mg	Si	Al	K	S	C	Mn		Ni	Zn	Co	Cu	Li
REE	37.6	15.7	5300	1.15	0.84	0.59	0.29	–	–	–	–	–	–	–	–	–	Monazite, apatite Peelman et al. [32]
	0.13	–	1330	2.2	0.34	31.8	1.31	0.61	0.26	6.0	208	263	–	–	–	–	Quartz, illite, gypsum, pyrite, microcline, calcite and muscovite. Reynier et al. [99]
	0.09	–	1760	0.089	0.06	42.2	1.07	1.07	0.18	4.7	67.9	7.25	–	–	–	–	Quartz, illite, gypsum, pyrite, microcline, calcite and muscovite. Reynier et al. [99]
Co	1.13	0.01	–	35.6	0.73	2.53	0.15	0.25	43.6	–	0.03	0.37	4.46	1.17	0.41	–	Pyrite, Pyrrhotite, Quartz, Sphalerite, Oxidized Fe-sulfide Mäkinen et al., [35]
	–	–	–	10.2	–	–	–	–	9.28 (TS)	0.16 (org.)	–	–	1.49	0.02	0.12	–	Quartz, pyrite Zhang et al. [34]
	–	–	–	27.9	–	–	–	–	34.25 (TS)	1.44 (org.)	–	–	4.85	0.06	0.57	–	Quartz, pyrite Zhang et al. [34]
	1.4	–	–	38.3	–	3.5	1.16	–	0.3	–	0.03	0.12	0.016	1.38	0.2	–	Pyrite [36]
	2.11	–	–	27.79	1.69	4.76	–	–	33.06	–	–	–	–	1.06	18.82	–	Carrollite, chalcocopyrite, chalcocite, bornite, covellite, and pyrite Chen et al., [37]
Li	–	–	–	–	0.09	17.5	23.7	0.27	–	–	–	–	–	–	–	0.098	Kaolinite, diaspore, boehmite, anatase and illite. Zhang et al., [17]
	–	–	–	–	–	32.7	10.5	2.47	–	–	–	–	–	–	–	0.28	Feldspar, quartz, sericite, kaolinite, lepidolite, pyrite, Zhiqiang et al., [96]

TS: total sulfur content; org: Organic carbon content, TREE: Total rare earth elements

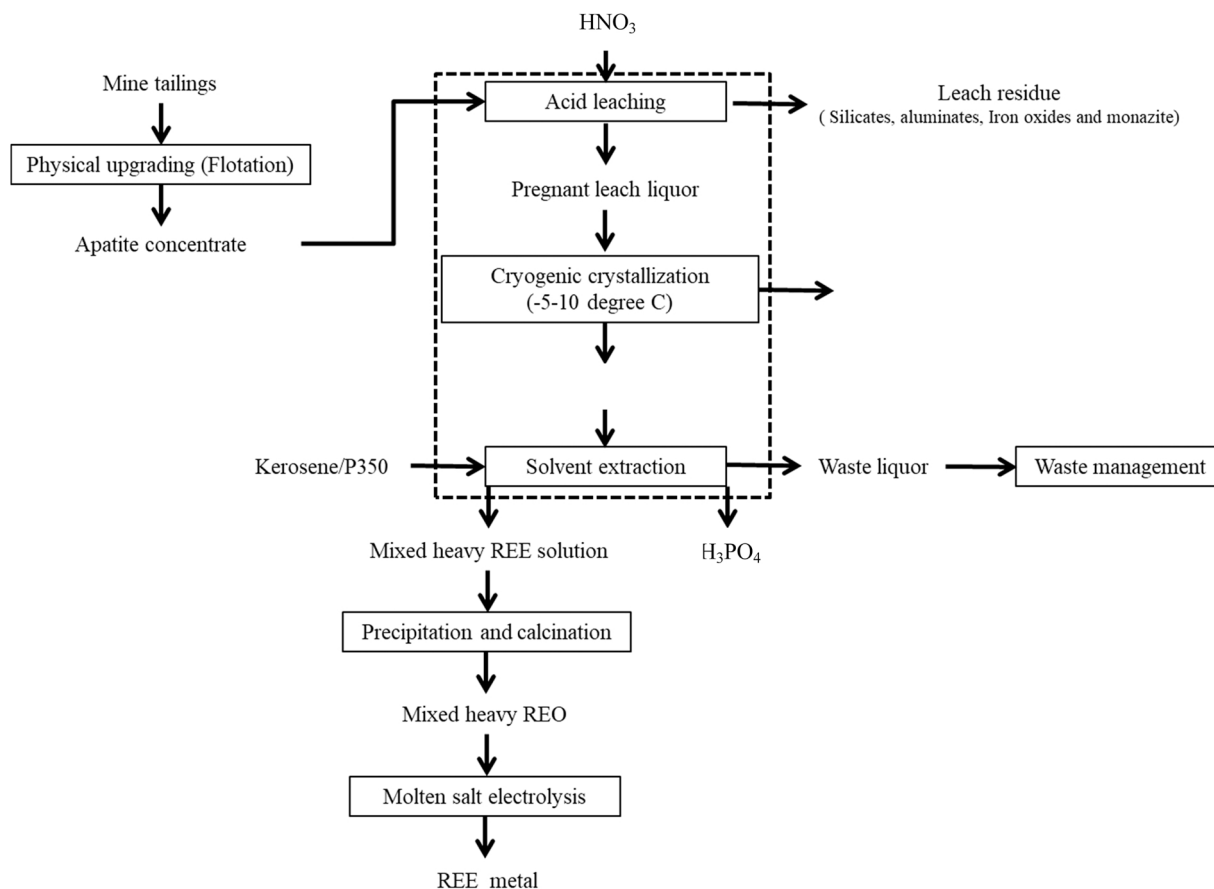


Fig. 2. Proposed recycling flowsheet of hydrometallurgical methods for REEs recovery from mine tailings in Kiruna, Sweden. The process steps highlighted by the dashed box are the focus of this study [30].

a proposed recycling flowsheet of hydrometallurgical methods for REEs recovery from mine tailings in Kiruna, Sweden [30]. A typical hydrometallurgical process employs leaching, followed by separation of metal ions in the resulting leachate either using precipitation or solvent extraction or a combination of these two.

Recovery of Li from mine tailings is a relatively new concept. The increasing demand for Li in diverse high-tech applications is now warranting exploration of all viable sources including some non-conventional sources such as mine tailings. Primarily Li is extracted for commercial production from Li chloride salts found in brine pools and from minerals found in igneous rocks such as spodumene [101]. The most commonly used methods of Li extraction from ores/minerals include roasting followed by leaching, while the use of evaporation, precipitation, adsorption and ion exchange methods are common for Li extraction from brines [102]. Since mine tailings are partly processed ores/minerals, leaching without roasting can be used to extract Li. Such technique has been investigated by Zhang et al., [17]. The researchers used a mixed acid leaching method without roasting to recover Li from bauxite mine tailings in Guiyang, China. The mineralogical analysis of the tailing sample revealed that Li was contained in the kaolinite and illite, which are clay minerals [103]. The study found that the tailings samples were enriched with Li at a concentration 0.21% in the form of Li_2O , which is considered to meet the grade of industrial ores [104]. They used mixed acid leaching with sulphuric and phosphoric acid, which resulted in the highest 96.35% leaching rate of Li. The study showed that leaching of Li was strongly influenced by the particle size of the tailings. It was found that 74 μm was the best particle size for Li extraction, larger and smaller particle sizes than this reduced the leaching rate. The particle size can also affect other methods of minerals recovery, such as flotation. The efficiency of flotation process

dramatically drops when the particle size is too small. Studies have shown that the efficiency of flotation decreases to 50% from 95% when the particle size is reduced from 20 μm to 3 μm . This reduction may be due to lees contact potential of particle with air bubbles [105,106]. To prevent this from happening and to enhance the efficiency of flotation process, Hornn et al. [107] applied anionic emulsifier and had excellent result. It was found that the flotation efficiency rose from 68% to 97% in chalcopyrite and quartz system.

There are a number of other factors that played a significant role in achieving high Li recovery from the bauxite mine tailings in this study. The factors were acid concentration of 60%, liquid-solid ratio of 4 mL/g, reaction temperature of 100 $^\circ\text{C}$ and a reaction time of 3 h. The shortcoming of the acid leaching method for Li recovery from bauxite mine tailings is that the technique consumes a significant amount of energy and chemicals, which have the potential to cause environmental issues. Though not enough studies have been conducted on Li recovery from mine tailings, it is evident from the above mentioned study that Li could also be sourced from mine tailings and other opportunities will exist. Some new leaching processes have been developed by a number of researchers and these processes could be applied to leaching of Li from tailings after some modification. These leaching methods include electrochemical leaching for Li recovery [108] and redox potential-dependent chalcopyrite leaching [109].

A number of hydrometallurgical processes have been used to recover REEs from secondary sources. Usually, recovery of REEs involves several steps. The steps consist of beneficiation to produce concentrate by flotation or gravity or magnetic processes. The concentrate then undergoes hydrometallurgical processes such as acid leaching and solvent extraction. The concentrate is acid leached with HCl, H_2SO_4 , or HNO_3 followed by filtration of the pregnant leach solution. Then individual

elements of the REEs are separated or a REEs mixed solution is produced by solvent extraction (Gupta and Krishnamurthy, [110]). Other methods for REEs recovery include leaching and co-extraction, in-situ leaching, bioleaching [111]. The solvent extraction process is one of the most commonly used processes to selectively recover REEs from leach solutions. This process was investigated by Tunsu et al., [16] to evaluate REEs recovery performance in presence of high amount of impurities, such as interfering anions and cations, phosphate, iron and copper, of two different tailings collected from two mine sites: New Kankberg in Sweden and Covas in Portugal. The study compared extraction of REEs from acid leached solutions with different solvating and acidic extractants. The study found that the co-extraction of some impurities, such as iron, was reduced by blending of solvating and acidic extractants. In recent studies researchers have developed some promising and selective recovery techniques for critical minerals, Co and REEs, from leach solutions, which could be applied and/or reconfigured in tailings reprocessing. The methods include application of galvanic interactions between zero-valent aluminum and activated carbon to recover Au ions [112], application of Zn powder to cement Co ion from Mn ion in leach solution to separate Co as precipitate by magnetic separation [113]. Technologies have also been developed by a number of researchers recently to remove potentially toxic metals, such as Pb, Co, from solutions [114-116]. Possible application of these processes in combination to selectively recover critical minerals and removal of toxic metals present as impurities in leach solution of tailings could enhance tailings reprocessing prospect.

4.2. Bio-hydrometallurgy technique (bioleaching)

The hydrometallurgical method of mineral extraction using bioleaching is known as bio-hydrometallurgy. The biological reactions and their interplays with metals in minerals have resulted in the exploitation of these natural phenomena in a wide range of applications including metals and minerals recovery. Mineral recovery using bio-hydrometallurgy offers some advantages over conventional methods. These include less operational and energy cost, and lower environmental impacts which can make mineral recovery from low-grade sources effective.

Bio-hydrometallurgical techniques for metal or mineral extraction using microorganisms from low-grade ores is a rapidly evolving technology that has proven to have both environmental and economic advantages over conventional hydrometallurgical methods such as acid leaching solvent extraction. The bio-hydrometallurgical technique makes use of the interaction between microbes and minerals, which can trigger chemical transformation and mineral transport phenomena. This mineral transformation mechanism is exploited to bioprocess minerals from low-grade sources such as mine tailings. The most commonly used bio-hydrometallurgical method to recover minerals from mine tailings is bioleaching, in which heavily soluble metal sulfides are converted into water-soluble metal sulfates by bacteria via biochemical oxidation reactions [117]. The bioleaching method was first used by Hallberg and Rickard [118] to recover copper from a copper deposit of the Falun Mine in Sweden. With the improvements in biotechnology, bio-hydrometallurgical techniques have become interdisciplinary promising techniques for metal extraction from low-grade sources and remediation of waste generated in industries. These techniques have now become an industrial reality as many companies are now using bioleaching methods to recover valuable metals from low-grade secondary sources such as industrial wastes, sewage sludge, soil, coal as well as fossil fuels [119-121]. One of the major benefits of using these techniques is that the bioleaching minerals extraction can be conducted under mild conditions in which the use of the toxic chemicals is avoided. Bioleaching by a microbial consortium of mesophilic and acidophilic bacteria was used to recover Co from polymetallic mine bulk tailings at the Rammelsberg sulfide deposit, Harz Mountains, Germany. The study also used archaea, single-celled microorganisms with a structure similar

to bacteria, in shake flasks and stirred tank reactors to bioleach Co, copper and other valuable metals from flotation tailings concentrate. The study achieved 91% Co and 57% copper extraction from the bulk tailings, Co and copper concentration of 0.02% and 0.12%, respectively in the studies after 13 days in stirred tank reactors using an adapted mesophilic microbial consortium. The study also showed that bioleaching with microbial consortium provided a higher Co and copper recovery.

Several studies have used bioleaching methods to extract REEs from mine tailings. REEs are abundant in the earth crust but low in concentration, and their concentration is often even lower in secondary sources such as mine tailings and ion adsorbed clays. Generally, the concentration of REEs in secondary sources is often below 1% (around 0.5%) even after physical upgrading [30]. Because of the low concentration of REEs in mine tailings other leaching methods such as total leaching by strong mineral acids or bases will generate a large amount waste which makes the recovery processes ineffective from an environmental standpoint. Due to this issue, recovering of REEs by bioleaching methods has been investigated in several studies which showed promising results. Reynier et al. [99] investigated REEs recovery from mine tailings collected from restored uranium mine sites in Ontario, Canada. The tailing samples were bioleached with a mix of native sulfur- and iron-oxidizing bacteria to solubilize REEs, uranium and thorium. The bioleached solutions were then processed for selective recovery of REEs using the ion-exchange resins. The resins used were Lewatit TP272 and Lewatit SP112. The Lewatit TP272 resin was used for the extraction of scandium and uranium, which resulted in the extraction of 94% and 99%, respectively. Lewatit SP112 resin was used to extract thorium and REEs, which yielded 57% and 73% (average), respectively. The experiments showed that bioleaching of REEs from these mine tailings could be effective.

Bioleaching was used in a study in Egypt where a low-grade gibbsite ore was used as a source for extracting REE and uranium using *Acidithiobacillus ferrooxidans* bacteria, which is normally used to bioleach copper [122]. The study carried out by Ibrahim and El-Sheikh [123] showed that the leaching rates for REE and uranium were 55% and 49%, respectively. The study also used other two types of bacteria: *Aspergillus ficuum* and *Pseudomonas aeruginosa*, to investigate the effect on leaching rate. The leaching rate improved for REE to around 75%, using these bacteria, but there was a concern for human safety as these bacteria are not as harmless as *Acidithiobacillus ferrooxidans*.

The literature on hydrometallurgical extraction of Co from mine tailings is limited only to bioleaching. Bioleaching has been used for extracting Co from mine tailings at laboratory scale, pilot scale study and commercial application. The bioleaching method has been proved to be suitable for Co extraction compared to other traditional pyrogenic methods. This is mainly due to the difficulty in recovering Co from low-grade sources using conventional pyrogenic method where co-occurs with other metal minerals such as chalcopyrite, pyrite, carrollite, pentlandite [124], and associated environmental concerns. In contrast to conventional methods, the biohydrometallurgy method of bioleaching offers a range of economic and environmental benefits [125,126]. Recognizing these benefits, Co was extracted using bioleaching methods from tailings of a former copper mine site located in Kasese, Uganda. It was the first commercial application (began operation in 1998) to bioleach Co from mine tailings containing Coiferous pyrite. In the full circuit running, the Co yield was nearly 80%.

Though bioleaching is widely used for the recovery of the strategically important critical minerals Co and rare earths from mine tailings, the literature on Li recovery by bioleaching from low-grade sources are limited to only spent Li-ion batteries, spodumene and lepidolite. Rezza et al., [127] investigated the recovery of Li from spodumene using bioleaching. Bioleaching of Li in this study was carried out using heterotrophic micro-organisms previously isolated from the minerals. Three different types of micro-organisms: *Penicillium purpurogenum*, *Aspergillus niger* and *Rhodotorula rubra*, were used separately. The

experiments used two different bioleaching media; one of them was highly limited in Mg^{2+} , Fe^{2+} and K^+ . The experimental results showed that *P. purpurogenum* and *R. rubra* were effective for Li, but that *A. niger* was less effective. It is possible that *P. purpurogenum* and the yeast could extract more Li together than individually [127].

A study by Sedlakova-Kadukova et al. [128] used three different biological systems to investigate Li extraction from lepidolite. The biological systems consisted of consortium of autotrophic bacteria *Acidithiobacillus ferrooxidans* and *Acidithiobacillus thiooxidans*, heterotrophic fungus *Aspergillus niger* and heterotrophic yeast *Rhodotorula mucilaginosa*. The study found that the bacterial consortium was the most effective where 11 mg/L of Li was dissolved in the absence of nutrients within 336 days. However, fungal and yeast bioleaching were faster (40 days) but with a lower extraction performance. Bioaccumulation represented the main mechanism of Li extraction with *R. mucilaginosa* and *A. niger*, with 92% and 77% of the total extracted Li accumulated in the biomass, respectively. The study concluded that a two-step process; bioleaching by heterotrophic organisms followed by autotrophic bioleaching, could lead to an increase in process kinetics and efficiency. Bioaccumulation of Li could be a viable technique for Li extraction from mine tailings and is worth of future investigation. Although there are a number of advantages of bioleaching, the method is slow compared to other conventional methods. Bioleaching processes often generate toxic chemicals such as sulphuric acid and H^+ . These toxic chemicals can leach into the ground and surface water causing a range of environmental issues, notably AMD. Another disadvantage of bioleaching is that the bacteria die when excessive heat is generated in the system.

4.3. Membrane processes for SICMs recovery

Pressure-driven membrane processes by reverse osmosis (RO), nanofiltration (NF), ultrafiltration (UF), and microfiltration (MF), have been used for the SICMs recovery. These advanced separation methods offer a number of advantages over conventional hydrometallurgical separation processes. These include higher separation rate, material selectivity, low energy requirement, simpler operation, low waste generation, and easy combining with other processes [129]. Several studies have reported Li recovery from brine using NF membrane (Wen et al., 2006; Li et al., [130]; Pramanik et al., [131]). Separation of REEs, especially LREEs such as Ce and Nd, was studied by Murthy and Choudhary [132] and found that they can be separated by the NF membranes. The study showed that the rejection of Nd-ions increased when the applied pressure was increased, but the rejection decreased when the feed concentration was increased. Notably, the rejection of Nd was influenced by pH, which may have been caused by variation of charge characteristics of NF membrane under different pH.

Li recovery from salt lake brine using NF90 and low-pressure reverse osmosis (LPRO) membrane was studied by Somrani et al., [133], and found that NF90 was more effective in separating Li than LPRO. This may have been caused by an elevated hydraulic permeability of the LPRO and high rejection factor for monovalent ions such as Li. Separation of both monovalent ions (Li) and divalent ions (Mg) by NF from brine solution was studied by Pramanik et al., [131] and Sun et al., [134]. The studies reported that separation of both Li and Mg was dependent on several factors, such as pH, applied pressure, and ration of Mg/Li. Li recovery by modified NF membrane was studied by Li et al., [130], which showed that the selectivity of Li^+/Mg^{2+} and water permeability improved when the NF membrane was modified by changing its metal-coordinated structure.

Recovery of Co by using membrane processes has been investigated in a number of studies. For example, separation of Co and Li by using polymer inclusion membrane electrodialysis membrane was studied by Wang et al. [135]. The study reported significantly a higher transport flux of Co (II) and higher selectivity was achieved using the polymer inclusion membrane electrodialysis membrane than commercially available membranes. The result sustained in 10 repeated transport

cycles. The high separation of Co compared to Li was likely due to higher surface area of the membrane which favoured the transport of the divalent metal ions than monovalent ions of Li. Notably, separation of Co(II) was influenced by several factors such as solution temperature, types of carrier used, concentration of feed solution and current density.

Despite many advantages of the pressure-driven NF membrane for metal recovery, the process also comes with some limitations. The process requires high amount of energy in operation which leads to higher operational cost, and elevated environmental concern if the energy demand is not met from the sustainable sources. Another major drawback of this process is impaired performances of the membrane due to excessive fouling which reduces the long-term usability of the membrane. These limitations may partly be overcome by using this NF membrane processes in combination of other conventional hydrometallurgy such as flotation, and magnetic or gravity separation as the beneficiation processes of the tailings samples. Using the beneficiation processes may reduce the fouling issues of the membrane as the processes can reduce the concentration of gangue materials in the feed solution significantly [69].

5. Key challenges in SICMs recovery from tailings

The key challenges in the recovery of strategically important critical minerals from mine tailings are technical, economic and environmental. These challenges have some underlying factors that can have direct influences with the recovery processes. The challenges and their inter-dependending factors are shown in Fig. 3. The technical, economic and environmental challenges are discussed below.

5.1. Technical challenges

The low concentration of critical minerals in mine tailings is one of the major technological constraints to cost-effective recovery of critical minerals. This is mainly because the mainstream technologies are designed for primary ores which are, mostly, higher in grades than mine tailings. Using these technologies to recovery minerals for mine tailings in the first place often results in low recovery. In a study by Araya et al., [14] to produce rare earth oxides (REO) for REEs recovery from mine tailings in Chile, the research found that the production was significantly reduced as a result from low concentration of REO in tailings. Low concentrations of SICM in mine tailings relative to the gangue minerals or elements have been reported in a number of studies. For example, Zhang, et al. [17] found that whilst the concentration of Li_2O_2 (0.21%) in mine tailing was similar to conventional ores it was considerably low compared to other components present such as Al_2O_3 (44.79%). Zhang et al., [34] showed that the concentration of Co was quite low in bulk samples of mine tailings, from 0.02% to 0.06%, whereas the concentration of other elements such as Pb and Zn were higher at 1.35% and 1.49% respectively.

SICMs are generally produced as a by-product or companion product of other minerals with higher concentration. This poses an issue of recovery of minerals that often occur in the least concentration. The recovery of minerals occurring in the least concentration can often be limited due to the competition of other metals occurring in higher concentration in mine tailings. An example of low-grade ore is brine solution, in which minerals such as Li can be found in a concentration similar or higher than mine tailings. Since the literature concerning the recovery of Li from brine solution is more common than the recovery of it from mine tailings, the challenges faced by the former case can help explaining the potential challenges of later one. For example, a study by Hu et al. [136] showed that the scandium recovery was limited because of the presence of REEs, aluminum and iron. It is not only base metals that affect the recovery of critical minerals, there are other unwanted competing ions such as calcium, magnesium, zinc, iron and aluminium can hinder separation processes for critical minerals. A study by Callura et al. [137] reported that these ions can inhibit the recovery of REE from

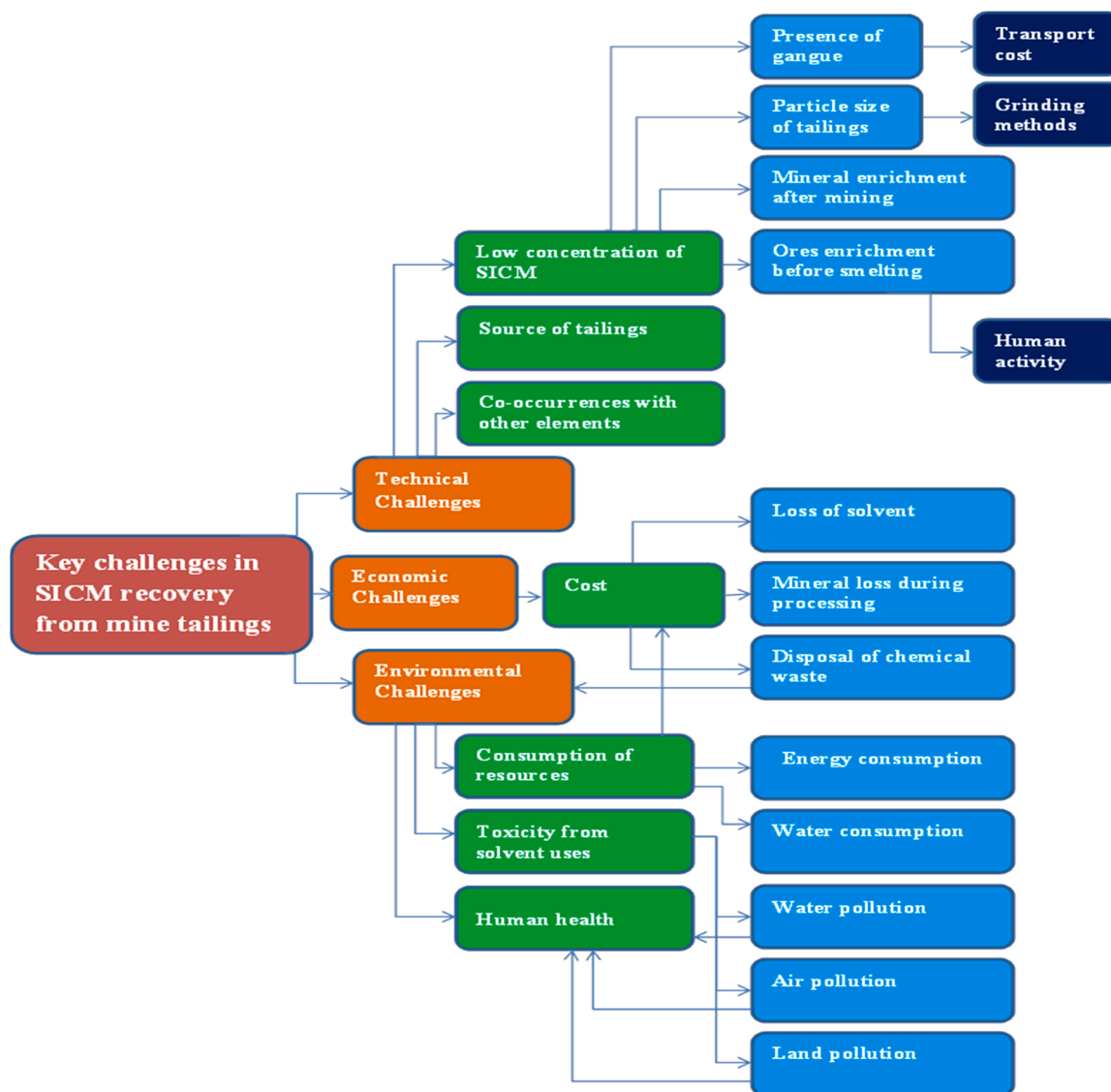


Fig. 3. Key challenges and their interdependencies in the recovery of SICMs from mine tailings.

brine solutions. To achieve high extraction rates of the target minerals these impurities need to be separated from the tailings first. A similar strategy was followed in one study that separated the impurities such as phosphate, iron, aluminum, nickel, chromium, manganese, zirconium, uranium and copper contained in the brine solution using oxalic acid as weak acidic medium to achieve subsequent high efficiency of REE recovery [138]. Following a similar approach Safarzadeh et al., [139] found that the recovery of platinum group metals (PGMs) from copper residues can be significantly improved following impurities removal, and they reported a 99.9% recovery of PGMs.

5.2. Environmental challenges

Success to SICMs recovery processes from mine tailings has a critical connection to number of environmental challenges. These challenges are due to a number of interconnected factors that have negative environmental impacts. These environmental impacts are caused by the tailings reprocessing processes such as tailings concentrate production, beneficiation, and separation of target minerals. These processes cause impacts which include urban land occupation, natural land transformation, fossil depletion, climate change, ecosystem change, and human health [140]. One of the most negative environmental impact

factors is the fossil depletion due to consumption of resources in tailing reprocessing. The reprocessing of tailings requires a large amount of water, energy and chemicals. The uses a large amount of chemicals, specifically different types solvents, which are often toxic to the environment and ecosystem. Toxicity from solvent used in the mineral recovery processes affects water, air, land and human health [141] and standards of living [142]. Moreover, mining-related activities may affect the traditional way of living of indigenous peoples in nearby communities [143]. It may even create conflicts of land use and other social issues related to public health and public wellbeing [143-146]. For example, Yang et al. [147] observed that large polluted areas have been created as a result of mining and production processes in China. Though reprocessing of tailings for resource recovery is considered to be less polluting than mining and production, still significant pollution can occur from dust generation and emission, wastewater production and disposal, and the production of radioactive wastes. Hurst [148] estimated that a ton of REEs production generate approximately 8.5 kg fluorine, 13 kg of dust, about 75 cubic meters of acidic wastewater, and approximately 1 ton of radioactive waste residue. Apart from these, mine tailings can often contain some potentially toxic elements such as arsenic (As), cadmium (Cd), lead (Pb), copper (Cu), zinc (Zn), Co (Co), iron (Fe), aluminium (Al), manganese (Mn), molybdenum (Mo),

selenium and chromium (Cr) ([149]; Edgardo et al., 2020; [150,151]). These elements may be present in tailings as impurities. There are two main issues relating to these toxic elements in tailings. One issue is that they can be transported through various media, such as wind, surface water and ground water, to end up in soil and sediment [152,153]. The other issue is that they can cause the recovery process of some of the SICMs, such as Li, incomplete [154]. Studies have also shown that the presence of calcium (Ca), Fe and other heavy metals can inhibit the efficiency of ion exchange process to recover REEs from secondary sources [155]. Moreover, some tailings are known to contain pyrite (iron sulphides) minerals in amounts sufficient to trigger the generation of amount of acid mine drainage (AMD). This AMD often gets enriched when they come in contact with the heavy metals. Another environmental challenge of tailing reprocessing is that some mineral deposits contain naturally occurring radioactive materials (NORMs) which end up in tailings impoundments after processing to recover target metals and minerals. Notably, most economically significant mineral deposits containing REEs host NORMs in variable concentrations. The most common NORMs found in REEs mineral deposits are thorium and uranium [156]. Some studies have reported that around 80% of thorium remained in the tailings concentrate of REEs after flotation process [157]. Exposure to high doses of these radioactive materials for extended period of time is an environmental and safety hazards. Therefore, reprocessing of tailings containing the radioactive elements such as tailings from REEs and uranium mine sites, come with environmental challenges due to the need for special consideration of radioactivity [158].

Reprocessing of tailings using the hydrometallurgical processes for SICMs recovery generate highly polluting wastes such as waste solid, wastewater, and off-gas, as they generate from processing of ores for minerals recovery by the hydrometallurgical processes (Qi, [159]). The generated wastes are hazardous in nature as they contain acidic, alkaline, ammonium, radioactive elements, and heavy metal ions. They have also the potential for contributing to generation of AMD. A number of studies, such as Tabelin et al. [160], have reported how AMD is formed and As mobilize under strong acidic condition. Since only a fraction of the tailings is recovered as valuable minerals through reprocessing, the rest goes back to the tailing storage facilities again, unless a part of it is recycled for other purposes such as construction materials, soil amendment. The leaching process in conventional hydrometallurgy generates waste solid which primarily contains the precipitations of impurities and residues of concentrates. To regulate these pollutions from damaging the environment, the amount of wastewater, off-gas, and waste solid generated while tailings reprocessing needs to be reduced. One way of doing through reducing of toxic elements, such as As, in the processes tailings by using some treatment technologies. Tabelin et al. [161] investigated As immobilisation by using hematite-catalysed scorodite formation under ambient conditions. The study showed that the hematite acts as a catalyst on the formation of scorodite at room temperature (25 °C) in material rich in pyrite. A study by Igarashi et al. [162] investigated the treatment of AMD in a two-step neutralization fertile-formation process. The results suggested that this method could further be improved by dissolving Si concentration before fertile formation step.

The environmental challenges of SICMs recovery from tailings are complex and site specific, and therefore they may vary from one recovery project to another due to varying tailings' characteristics. One way of knowing more about the site specific environmental challenges is through conducting detailed life cycle assessment (LCA) of the recovery project. Despite having the LCA standards, LCA studies may vary from one study to another for a given project. This is due to consideration of varying system boundaries and parameters, incomplete inventories, choices of impact assessment methods and different or incorrect allocations or calculation errors [163]. One of most common methods of conducting an LCA is called "cradle to gate" method, in which partial life cycle of a product from resource extraction to factory gate is considered.

An LCA generally consists of four phases: setting up goal and scope of LCA, inventory analysis, impact assessment and interpretation (ISO 14040:2006). An LCA of tailings beneficiation for REEs recovery from two different tailings facilities was conducted by Grzesik et al. [140]. The study considered two different system boundaries: one included 3 stages and another with 4 stages of beneficiation processes, and different major impact categories. The reason for these variations was not explained in the literature, but it may have been due to variation in tailings characteristics: one from gold mine and other one from tungsten mine. According to the results, the gold containing tailings has the highest negative impact caused by disposal of tailings residues, while tungsten containing tailings has highest impact caused by depletion of fossil fuels for electricity production and for transportation [140]. Another LCA was conducted by [152,153] for exploitation of tailings for resource recovery from concentrate generated after beneficiation by flotation of iron, niobium, and rare earth at Bayan Obo mine site in China. The study found that rare earth flotation has the second highest impact on human health after iron flotation. Due to tailings reprocessing for resource recovery being a relatively new idea, literature availability of LCA on this area is very limited.

Although LCA of Co and Li recovery from tailings is not reported in literature, there are number of studies have been conducted on LCA of Co and Li extraction from primary sources. For example, Rinne et al. [164] studied LCA of Co-sulphate extraction from unused Co-and Au-bearing ores. In this LCA, cradle-to-gate approach was considered with a range of impact categories such as global warming, acidification, eutrophication, ozone depletion, photochemical smog creation and water use. The result showed that the main mineral types (cobaltite, linnaeite) had minor impacts on the studied categories. The study also reported that the concentrate with high grade of Co can significantly lower the impacts due to hydrometallurgical processes. Similarly, LCA on Li recovery from brine, mineral deposit and spent Li-ion battery is available [165,166]. The findings of LCA obtained from these primary sources can be used to assume the potential impact of recovering these minerals from tailings.

5.3. Economic challenges

Conventionally, mining industry follows a linear economic model which would keep escalating the issue of mining waste-related problems [167]. To alter the continued issue of mining waste, a circular economy approach, in which reuse, recycle and reprocessing of mining waste, along with a reduction of mine waste generation by using advanced technologies, is needed. However, there remain some challenges of these aspects. For example, reprocessing of tailings for mineral recovery relates to mainly to the cost of resources used, such as water, energy (electricity for plant and fuel for vehicle transport), chemicals, labors, incurred losses of mineral and solvents, and waste disposal cost. Studies suggest that consumption of energy increases exponentially with decreasing ore grades. Koppelaar and Koppelaar [168] calculated that energy required to produce 30% Cu concentrate increased from 60 MJ/kg for 0.5% Cu up to 450 MJ/kg for 0.3% Cu. With lower concentration of target minerals in tailings requires larger plant, which increases the capital cost. It was also estimated in bench-scale tests that resources and power costs for copper recovery from tailings can be nearly 80% and 16%, respectively, of the total cost [169]. The costs in the recovery processes are inversely proportional to the concentration of the target minerals. This is because of low concentration of target minerals in tailings demands more resources against less economic output, which may even lead to negative balance. The cost of some chemicals (such as strong acids used in the recovery processes) is high and a large amount of these chemicals are required for processing of low-grade sources such as tailings. Use of the strong acids has both negative environmental and economic impacts [170]. Furthermore, SICMs recovery from mine tailings is typically a multi-step processes which increase the overall cost of minerals recovery. Losses of the target

minerals in multi-step processes can be costly. Some studies, such as Virolainen et al., [171], estimated that the typical losses of Li in multi-step processes could go well over 10%. The multi-step recovery processes for solvent extraction of SICMs can lead to an increase in solvent losses and unfavourable economics. Market prices of SICMs play an important role in determining the recovery success. Since some SICMs are produced as by-products or companion products, variation in market prices of their host metals can also affect the prices of the SICMs, and hence overall economics.

6. Opportunities and Perspectives

The major processes associated with the resource recovery start with a carefully selection of mine sites with tailing storage facilities which have the potential to contain the minerals of interest. The initial selection of tailing storage facilities should be based on the historical records containing data on the type of ores mined and processed, major metals extracted, a type of extraction method used, and the nature and volume of the tailings. Taking these factors into consideration can help identify tailings facilities with enhanced economic potential. The initial selection of a tailing storage facility should be followed by a detailed characterization of the tailing samples. The characterization of tailing samples should seek to determine chemical, physical and mineralogical properties. These properties of the tailing samples are key to evaluating the economic potential for SICMs recovery. Therefore, an extensive sampling campaign should be carried out covering all possible locations at various depths of the tailings storage facilities. The detailed characterization provides the concentration of the target minerals of interest, which can be used to evaluate the economic potential of the recovery project. Several protocols for assessing sulphide containing tailings in respect of contained value have been developed and these provide a useful framework for assessment [172-174]. However, the economic potential is not solely dependent on the concentration of minerals, it also depends on market prices for the recoverable minerals [175,176]. Moreover, the economic feasibility of mineral recovery from mine tailings can be improved by co-recovery of other companion minerals present in the tailings. The economic benefit of mineral recovery from mine tailings should not be the only goal, social and environmental benefits should also be taken into consideration. In order to achieve the combined goals of economic, social and environmental benefits of a mineral recovery project from mine tailing, an integrated approach is

needed. In that respect, bioleaching which is a proven low-cost and environmentally friendly method, should be given strong consideration in flowsheet design [177]. Water recovery and re-use should also be considered in the mineral recovery system design [99,178,34]. Fig. 4 shows a roadmap of an integrated approach of SICMs recovery and water production from mine tailings. The wastewater produced in tailing reprocessing could be treated with advanced technologies such as membrane processes [179] and virtual curtain (a new treatment method developed by CSIRO) [180]. The membrane process comprising of NF and RO which are the proven methods of treating variety of wastewater. Virtual curtain treatment process uses hydrotalcites, which are minerals sometimes found in stomach antacids. The virtual curtain treatment of mine wastewater is rather simple and does not require complex infrastructure and chemicals to treat the wastewater. The produced water is compared to rainwater, making it safe to discharge to the environment [180]. However, a detailed techno-economic assessment is recommended as an integral part of the feasibility study on mineral recovery project from mine tailings project.

Although a range of challenges are associated with SICMs recovery from mine tailings, there are a number of obvious opportunities too. The first and the foremost opportunity is the economic benefit which could be created through effective recovery of these valuable minerals by applying advanced technologies to recover the SICMs from much lower grades of tailings. To achieve this there is a need for developing tailored-made technologies in accordance with the needs of varied tailings' characteristics and mineral separation complexity [16]. The tailored-made technologies can be designed to address some of the most urgent issues such as environmental impacts from the use of resources and chemicals, and the cost effectiveness of any mineral separation processes. Being able to address these issues can lead to both socio-economic and environmental benefits. The socio-economic benefits of reprocessing of abandoned mine tailings to recover SICMs can largely be attributed to the tailing characteristics itself. Since the mine tailings are partly processed and there is generally no need for further processing, the overall cost of mineral recovery can be significantly reduced. This reduction of cost is due to mainly to lower amounts of water and electricity needed for the recovery of minerals from tailings compared to conventional mining. In contrast to conventional mining, which requires a lot of upfront capital investment, reprocessing of tailings can be done in the existing ore processing plants, saving a significant amount of capital investment related to equipment, labour, fuel and

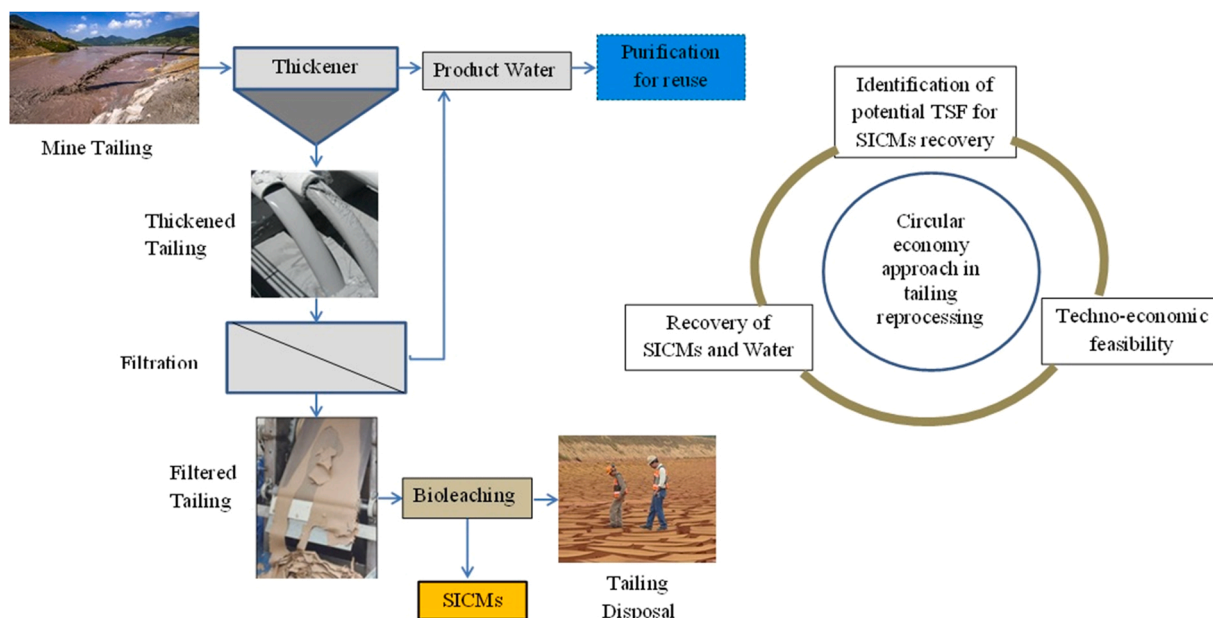


Fig. 4. A possible roadmap of an integrated approach for SICMs recovery and water production from mine tailings.

maintenance. In addition, the reprocessing of mine tailings for minerals recovery can bring back new life to abandoned mine sites through creating employment opportunities for the local communities.

There are a number of environmental benefits of reprocessing of mine tailing for mineral recovery. For example, the tailings-related pollutions, such as air and soil contamination by dispersion of dust, and groundwater and surface water pollution by seepage from tailings, can be reduced. In addition, reprocessing of tailings can reduce the volume of tailings, which can result in reduced risk of tailing dam failures and less land requirement. Apart from this, dewatering of tailings and dry stacking after minerals recovery will help reduce the economic, environmental and societal risks associated with tailings stored as slurry. Finally, the reprocessing of tailings will also allow additional production of valuable minerals without increasing the environmental footprint as could be the case with a new mine.

7. Conclusions and recommendations for future research

Critical minerals such as Li, Co and REEs have a number of strategic applications for society, ranging from meeting our simple daily demands to playing a role in solving global problems relating to energy, water, the environment, communications etc. Therefore, they have been referred to as Strategically Important Critical Minerals in this paper. Though their demand is expected to increase many folds in the coming decades, their future supply is at risk because of the low number of new deposits containing them being found, the gradually declining ore grades of the current deposits, and evolving geopolitical tensions and developments. Therefore, it is important to recover these critical minerals where we can from non-conventional sources, such as mine tailings, in which they can often be found in higher concentration than in some primary deposits. The findings from this review and future research recommendations are summarized in the points below.

- There are technical, economic and environmental challenges associated with the recovery of critical minerals from tailings where they are mostly generated as by-products or companion products of host metals such as Zn, Pb, Cu, Au, Ni, Fe and Sn. One key challenge relates to the form of the SICMs in the tailings is their concentration, grain size, liberation, and association with other gangue constituents, and moreover the influence or impact of these characteristics on the reprocessing method and efficiency. Therefore, detail characterization, chemical and mineralogical, of tailings with techno-economic assessment is necessary, which needs more research.
- Traditional hydrometallurgical methods, such as solvent extraction and acid leaching have been used in a number of studies investigating the recovery of critical minerals from mine tailings. These methods can be resource intensive, costly and complex, and can often lead to adverse environmental impacts as a result, for example, of the use of particular chemicals in reprocessing. Therefore, more research is needed to develop methods for selectively recovering and/or removing valuable and gangue minerals, including potentially toxic minerals, from tailings.
- Bio-hydrometallurgical methods such as bioleaching, which is one of the cheapest and most environmentally friendly methods for metal recovery from low-grade resources [181], offers a more promising reprocessing option for the treatment of mine tailings and we believe it deserves greater attention and consideration. However, slow recovery rate and excessive heat generation are the main downsides of this method, which need further investigation to improve its efficacy.
- The use of bioleaching processes to recover REEs and Co from mine tailings has been investigated in number of studies and showed good results but to date there have been no studies on Li recovery from tailings using bioleaching. Therefore, investigating the potential of Li recovery by bioleaching can be a new research area.
- Finally, it is to be noted that many tailings impoundments are in the form of dams or ponds contain large quantities of water. Application

of integrated approach combining resource recovery and clean water production, where possible, needs more research to help realize the full economic potential of tailings reprocessing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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