Opportunities for Mineral Carbonation in Australia's Mining Industry

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Abstract: Carbon capture, utilisation and storage (CCUS) via mineral carbonation is an effective method for long-term storage of carbon dioxide and combating climate change. Implemented at a large-scale, it provides a viable solution to harvesting and storing the modern crisis of GHGs emissions. To date, technological and economic barriers have inhibited broad-scale utilisation of mineral carbonation at industrial scales. This paper outlines the mineral carbonation process; discusses drivers and barriers of mineral carbonation deployment in Australian mining; and, finally, proposes a unique approach to commercially viable CCUS within the Australian mining industry by integrating mine waste management with mine site rehabilitation, and leveraging relationships with local coal-fired power station. This paper discusses using alkaline mine and coal-fired power station waste (fly ash, red mud, and ultramafic mine tailings, i.e., nickel, diamond, PGE (platinum group elements), and legacy asbestos mine tailings) as the feedstock for CCUS to produce environmentally benign materials, which can be used in mine reclamation. Geographical proximity of mining operations, mining waste storage facilities and coal-fired power stations in Australia are identified; and possible synergies between them are discussed. This paper demonstrates that large-scale alkaline waste production and mine site reclamation can become integrated to mechanise CCUS. Furthermore, financial liabilities associated with such waste management and site reclamation could overcome many of the current economic setbacks of retrofitting CCUS in the mining industry. An improved approach to commercially viable climate change mitigation strategies available to the mining industry is reviewed in this paper.

Keywords: mineral carbonation; greenhouse gas (GHG) emission; climate change; Australian mining; mining waste; mine reclamation

1. Introduction

Global concerns about climate change have been growing over the last few decades. Greenhouse gas (GHG) emissions with anthropogenic origins are recognised to be the major driver of these climate change effects [1]. The increased industrial developments and human activities which have been traditionally reliant on burning fossil fuels have resulted in a critical accumulation of CO_2 in the atmosphere. Largely anthropogenic GHG emissions have raised CO_2 concentrations significantly during the last century, with up to 408 ppm in November 2018 [2] in comparison to under 300 ppm in the 19th century. During the last three decades, the surface temperature on earth has increased progressively [3]. The increasing trend of global warming is attributed to the increased rate of CO_2

emissions in the last few decades. All these data indicate that disastrous climate change effects can occur by the end of the 21st century if the current trends continue. It is important that countries with major emissions take a leading role to address GHG emissions. Australia contributes about 0.33 percent of the world population, but is accountable for nearly 1.3 percent of the global emissions [4]. As a country with access to advanced technologies and developing industries, Australian industrial sectors have the opportunity to improve toward long-term decarbonisation.

The Australian mining industry, as a major industrial sector, has been one of the contributors to the GHG emissions and climate change effects. This has been through energy consumption and fugitive GHG emissions associated with mining operations and processing of minerals and metals, as well as burning coal, one of Australia's most important commodities, in power stations. Australia's GHG emissions for the year to June 2018 were 547.0 Mt CO_2-e. Emissions from the electricity sector account for 35% of Australia's greenhouse gas emission [5], and mining accounts for approximately 10% of Australia's energy consumption, and it has increased 6% per year during the last decade [6]. The demand for electricity production has been growing with the development of industries. In fact, Australia's expanding resource sector relies heavily on electricity consumption to operate mines, mineral processing plants, refineries, chemical manufacturing facilities, and its infrastructure. Under The Paris Agreement [7] Australia made the commitment to reduce its GHG emissions by 26 to 28% of 2005 levels by 2030 [4,8]. In order to be on track to meet this target, it is crucial that all sectors in Australia improve their emission mitigation strategies. Therefore, it is important for the mining industry to adopt appropriate technologies, which support offsetting GHG emissions and decarbonisation of the entire industry in the long-term.

Many attempts have been made to address the effect of climate change and reduce GHG emissions from Australian industries. It is widely accepted that a portfolio of strategies needs to be applied to reduce CO_2 emission in the long-term. Considering the energy sector as the source of the largest GHG emissions, replacement of fossil fuels with renewable energies has been suggested repeatedly as a major climate change mitigation strategy. However, there is a general consensus that in the short-term it is not possible to stop using fossil fuels and switch to renewable energies without major economic disruption [9]. Therefore, transition to a portfolio of energy supply chains with a larger proportion of renewable energies, and hence reducing GHG emissions, also requires the development of cleaner fossil fuel energy production. Due to the increasing demand for minerals and metals production, resource mining worldwide should develop such decarbonisation strategies, and particularly in countries such as Australia, with globally significant mining activities.

Carbon capture, utilisation, and storage (CCUS) is an emerging strategy, which has proven to be a promising tool as a long-term solution to address CO_2 emissions [10]. Estimates by the International Energy Agency (IEA) suggest that carbon capture and storage (CCS) deployed to coal-fired power stations globally might remove 10% of the CO_2 emissions from the energy sector by 2050 [11,12]. However, recent estimates by IEA, at the 24th Conference of the Parties to the United Nations Framework Convention on Climate Change (COP24), [10] suggest that CCUS remains well off-track to meet the IEA's Sustainable Development Scenario (SDS) [13], which is aligned with the energy-related concerns of the Sustainable Development Goals (SDGs) [14].

All of these recent trends show the importance of the application of CCUS in various industrial sectors, including the mining industry. Successful application of CCUS in the mining industry has the potential for substantial carbon offsetting of mining operations and minimising CO_2 emissions from coal-fired power stations. Global acceptance and successful integration of CCUS in large-scale industries, such as mining, requires successful trials. Australia as a country with an advanced resources industry is a perfect place for trial and development of mineral carbonation for climate change mitigation in the mining industry, which can then be utilised globally.

Economic drawbacks have been the major barrier to the deployment of CCUS technologies at a large-scale [15]. However, opportunities exist which can assist in reducing the deployment cost of mineral carbonation. Magnesium and calcium silicate mineral sources, which are generally considered

for mineral carbonation, can be expensive to pretreat and energy-intensive to activate. However, industrial wastes are usually more reactive and require minimum pretreatment and activation. Furthermore, industrial wastes are liabilities which are available at no or minimum cost and are often costly to dispose [16]. Successful utilisation of alkaline industrial wastes, including furnace slag, cement kiln dust, red mud, coal combustion by-products, and mine tailings as the feedstock of mineral carbonation has been reported [15–21].

Recent investment and interest by major mining companies [22–24], governments (i.e., UK council for CCUS [25]), and research institutions for deploying CCUS at industrial scale, indicate the significance of these technologies. In particular, investigating the potential role of mining waste in mineral carbon sequestration, for example, investigation of mineral carbonation in diamond tailings at Venetia Mine in South Africa and the Gahcho Kué Mine in Canada [24] and process optimisation toward more economically viable technologies with reduced energy requirements [26], more efficient process routes [27,28], and recycled industrial waste feedstock for mineral carbonation [29,30] will contribute to the low carbon future. Rehabilitation of mined land often includes various factors related to the risks associated with residue during and after mining operations, for example, management of acid mine drainage (AMD). Even where high standards of remediation have been applied, there is a requirement for ongoing monitoring and management of residual risk. Nevertheless, remediation poses significant expenses to the mining industry as shown, for example, in the reports by Rio Tinto [31], BHP [32], and Glencore [33].

This paper discusses decarbonisation in Australian mining by recycling waste as a viable climate change mitigation strategy. The potentials that exist for the utilisation of mineral carbon sequestration in the Australian mining industry are reviewed. The drivers of and barriers to the deployment of the mineral carbonation, and an integrated mineral carbonation model for Australia's mining industry are discussed. Some of the many potential and nearer-term opportunities and synergies that exist in using mining waste for climate change adaptation are identified, in this paper.

2. Mineral Carbonation as a Route for Reducing CO_2 Emission

Geological sequestration [34–37], ocean disposal [38–41], and biological fixation [42–44] have been reported extensively in the literature, while mineral carbonation, also called CO_2 mineralisation, a less explored method of sequestering CO_2, in a disposal option that has great potential due to the availability of large quantities of suitable mineral materials [45]. Mineral carbonation involves the chemical conversion of CO_2 to solid inorganic carbonates. It commonly refers to the fixation of CO_2 using alkaline and alkaline-earth oxides, such as calcium oxide (CaO) and magnesium oxide (MgO) [46], for which the respective basic reactions are [47]

$$CaO + CO_2 \rightarrow CaCO_3 + 179 \text{ kJ/mol} \tag{1}$$

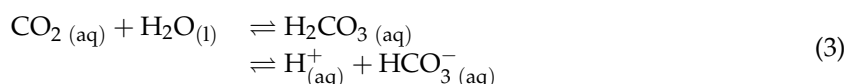$$MgO + CO_2 \rightarrow MgCO_3 + 118 \text{ kJ/mol} \tag{2}$$
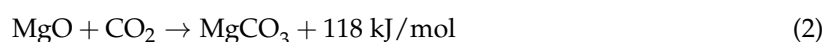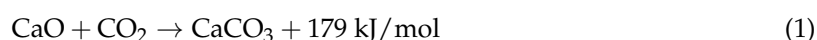
Pure calcium and magnesium oxide minerals are rare in nature, but abundant quantities of minerals containing chemical combinations of these oxides occur naturally as silicate minerals [45]. For example, ultramafic igneous rocks (primarily peridotites and serpentinites) contain large amounts of MgO bound into a silicate structure.

The process of mineral carbonation occurs naturally, where it is known as one of the weathering phenomena [48,49]. Silicate minerals form approximately 92 wt.% of the earth's crust which makes the weathering of silicate minerals a quantitatively important process [50]. Weathering of serpentine is known as a primary source of natural magnesium carbonates and is responsible for the majority of all large-scale sedimentary magnesite ($MgCO_3$) deposits [51]. However, the kinetics of chemical weathering reactions under natural conditions are not sufficient to absorb all current anthropogenic CO_2 emissions [52]. Therefore, the employment of ultramafic rocks for industrial-scale mineral carbonation processes was proposed for the first time in 1995 by Lackner et al. [47]. As mentioned
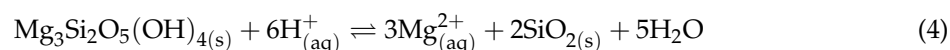
above, the precursor material for mineral carbonation must be rich in alkaline earth metals, either Mg or, more preferably but less abundant, Ca. The alkaline earth metals are present as oxides in silicate minerals such as wollastonite $CaSiO_3$, olivine Mg_2SiO_4, or the more abundant ultramafic rock serpentine $Mg_3Si_2O_5(OH)_4$, which is hydrolysed olivine [47,53]. Dunite is a rock made up entirely of olivine. Serpentinites and dunites are attractive feedstock for the mineral carbonation process because of their great abundance and the high Mg content by weight (35 to 49 wt-% MgO) [47,54,55]. There are large deposits of these minerals available worldwide and, in particular, in Australia, USA, Canada and parts of Europe [45]. Serpentinised peridotite reserves are estimated to be in the hundreds of thousands of gigatonnes [56]. In an industrial application between 1.6 to 3.7 tonnes of ore (depending on the magnesium content of the ore) are typically required to fix one tonne of CO_2 [57]. In comparison, Brent and Petrie [58] showed the mining practice of serpentinite required for mineral carbonation is of a similar scale of coal-based mining operations for coal-fired power generation.

Mg/Ca-rich precursors can be converted into carbonates using two methods: directly via the gas-solid reaction and in aqueous media. The aqueous route has clear advantages in terms of the speed of reaction and process flexibility. Therefore, the majority of research on CO_2 mineralisation focuses on the aqueous route. Ex situ aqueous mineral carbonation, which is the accelerated form of natural weathering of silicate rocks through above-ground processes, involves three steps: (1) the dissolution of CO_2 in the aqueous phase, (2) the extraction of the alkaline earth metal from the mineral structure while leaving a SiO_2 rich layer, and (3) the precipitation of stable metal carbonate. The sequence of the reactions is illustrated through Equations (3)–(5).
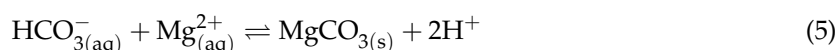
CO_2 dissolution:

$$CO_{2\,(aq)} + H_2O_{(l)} \quad \rightleftharpoons H_2CO_{3\,(aq)}$$
$$\rightleftharpoons H^+_{(aq)} + HCO^-_{3\,(aq)} \qquad (3)$$

Serpentine dissolution:

$$Mg_3Si_2O_5(OH)_{4(s)} + 6H^+_{(aq)} \rightleftharpoons 3Mg^{2+}_{(aq)} + 2SiO_{2(s)} + 5H_2O \qquad (4)$$

Carbonate formation:

$$HCO^-_{3(aq)} + Mg^{2+}_{(aq)} \rightleftharpoons MgCO_{3(s)} + 2H^+ \qquad (5)$$

CO_2 is first dissolved in water to form carbonic acid (H_2CO_3), which is then dissociated to protons (H^+ or H_3O^+) and bicarbonate ions (HCO_3^-). The protons hydrolyse the mineral, resulting in the liberation of magnesium cations and the formation of free silica and water. The magnesium cations in the solution react with the bicarbonate ions to form solid magnesium carbonate ($MgCO_3$) [56]. The Albany Research Centre (ARC) conducted the best-studied case of the aqueous mineral carbonation including over 700 kinetic tests [57]. They investigated carbon mineralisation in a single step mode, where the steps in Equations (3) to (5) take place in the same reactor simultaneously. The overall reaction sequence in Equations 3 to 5 is exothermic, the enthalpy of reaction of the given example being -64 kJ mol^{-1} of CO_2. However, the kinetics of the reactions involved are very slow [48]. Therefore, different strategies to increase the speed and efficiency of the carbonation reaction by, among others, Los Alamos National Laboratory (LANL) [47,59], Åbo Akademi University [60,61], and ARC [57], have proved at least partially effective. Table 1 summarises the main methods used by the researchers to date.

Table 1. Strategies and methods used to increase the speed and efficiency of the carbonation reaction.

Strategy	Reference	Method	Benefit
Mechanical pretreatment of serpentine rock	[62–66]	High-energy crushing and grinding	Increases the overall specific surface area of the minerals, i.e., the surface per unit mass available for Mg/Ca-extraction
Thermal activation of the hydrated Mg-silicate serpentine	[67–73]	Heating the mineral to 630 °C or above	Destabilises the crystal lattice, thus increasing the reactivity of the mineral Heat activation also creates an even higher specific surface area [46]

Table 1. *Cont.*

Strategy	Reference	Method	Benefit
Increasing reaction temperatures	[74,75]	Applying operating temperatures above 100 °C	Accelerate the kinetics of the reaction
Increasing reaction pressure	[76,77]	Applying operating CO_2 pressure above 150 bar	Increases the activity of protons needed for metal extraction. Also counteracts the low solubility of CO_2 at high temperature
Using organic and inorganic chemicals	[61,78–80]	Adding different substances as catalysts and additives to the reactants	Enhances the kinetics of the carbonation process and precipitation of magnesium carbonate
Applying New double-step and multistep process designs	[81]	The operating conditions are changed between the different stages	Allows to control and promote extraction and precipitation separately
Direct capture of CO_2 from flue gas stream	[81,82]	Capturing CO_2 directly from a flue gas stream by mineral carbonation	Provides a simple and straightforward process route

In addition to the minerals mentioned above, MgO and CaO can also be found in alkaline industrial residues, i.e., in steel slags, fly ashes, or cement wastes, as well as in waste brines and slurries such as alumina production residue (red mud). The sequestration of carbon using industrial and commercial waste is an attractive option and offers the potential to utilise residues. Steel manufacturers can benefit significantly from use of a carbonation process to improve the environmental and mechanical properties of the large amounts of alkaline solid residues that are generated during steel manufacturing such as basic oxygen furnace (BOF) slag, a by-product of the conversion of iron into steel [83–86]. This research has recently been tested on a pilot scale and resulted in a successful outcome [87]. In other studies, coal combustion fly-ash, an industrial waste that contains about 4.1 wt.% of lime (CaO), was used to sequester CO_2 by aqueous carbonation [88–90]. All in all, while in many industries these waste materials are available only in small quantities [48] which can be a barrier to commercial viable CO_2 fixation at the large-scale, they can be found in large quantities as by-products of many mining-related operations in Australia. This can be an opportunity that may help simultaneously to mature the technology towards commercial application and solve the problem of massive waste management in mining and other industries. Mines with mineral carbonation capabilities could use their waste rock as a value-added product. This could effectively lower the grade of material that is economical to mine. This additional revenue stream could also lower the strip ratio of a surface mining operation by virtue of a greater proportion of the material being mined, having value [91]. As a result, there may be the potential to enhance a marginal project to a level in which that mining project could become economically feasible and environmentally attractive [92].

3. Mineral Carbonation in Australia

One of first applications of mineral carbonation at an industrial scale in Australia was in Alcoa's Kwinana aluminium refinery, Western Australia [93]. A carbonation plant was developed by Alcoa for bauxite residue treatment in order to reduce long-term storage risk and adverse environmental effects. Deployment of the mineral carbonation plant assisted neutralising red mud by reducing its alkalinity from pH 13 to 10.5 and by sequestrating CO_2 from the nearby CSBP ammonia plant [93].

Another case for mineral carbonation in Australia was outlined in 2008 by Brent and Petrie [58]. They focused on carbonation of serpentine as feedstock for CCS via mineral carbonation, based on the fact that large outcropping deposits of serpentinite are located in close proximity to existing large emission sources in Australia [94]. This study showed the feasibility of achieving 80% net sequestration of CO_2 using Aspen (Aspen Technology Inc., Bedford, MA, USA, 2007) to model a standalone mineral carbonation process based on the ARC/NETL aqueous process [57]. Following this research, Mineral Carbonation International (MCi), an Australian-based company, has been developing technology for carbon capture and utilisation (CCU) since 2013. Their research and technical facilities are centred at the University of Newcastle, Australia. MCi's suite of projects includes fundamental theoretical and laboratory research [66,95–100] as well as one of the world's first mineral carbonation pilot plant

facilities [101,102]. MCi's project has primarily been studying the carbonation of serpentine, which as mentioned above is an abundant mineral in New South Wales (NSW), Australia and globally [102]. It is believed that the future feedstock supply needs will be available on the gigatonne scale. For example, the size of only one resource in the north of the town of Barraba, NSW, Australia, is sufficient to sequester all NSW stationary emissions for over 300 years [94]. The MCi's project would be the first to encompass the full CCUS chain from mineral mapping, characterisation, processing, and pretreatment through to integration with power generation and CO_2 capture processes, carbonation, value-adding and final product storage. Another aim of the MCi project is determining the potential for utilising the carbonated product in next-generation "green" building products [102].

Other research groups at the University of Queensland and Monash University, Australia have been working on different aspects of mineral carbonation, field studies and the use of waste materials [103–110]. Recent research investigated ultramafic mine tailings as feedstock because they contain abundant Mg-rich silicate and hydroxide minerals and have a smaller grain size and increased reactive surface area due to ore processing [106,107]. Table 2 presents a summary of research on mineral carbonation in Australia.

Table 2. Summary of the mineral carbonation research undertaken in Australia institutes and industries.

Institute/Industry	Research Focus	Goal
Alcoa's Kwinana Aluminium Refinery	Bauxite residue treatment	To reduce long-term storage risk and adverse environmental effects
Mineral Carbonation International (MCi)	Carbonation of serpentine as an abundant mineral in New South Wales (NSW), Australia and globally	integration with power generation and CO_2 capture processes, carbonation, value-adding and final product storage
The University of Queensland	field studies	use of waste materials and ultramafic mine tailings as feedstock
Monash University	field studies	use of waste materials and ultramafic mine tailings as feedstock

4. Drivers of Utilisation of Mineral Carbonation in the Australian Mining Industry

Mining is a primary industry that forms one of the pillars of Australia's economy [111]. Due to Australia's role as a world leader in resource mining and production, it is crucial to integrate climate change adaptation technologies in mining to address the ever-growing environmental awareness and the increasing demand for minerals and metals in the future for Australia simultaneously. Integration of mineral carbonation in the Australian mining industry is one of the climate change adaptation strategies which will benefit sustainable development initiatives of the mining sector by addressing GHG mitigation, improved mining waste management and mine reclamation. Australia has a large and diverse portfolio of mineral commodity production and is one of the world's biggest producers of minerals and metals.

Table 3 lists Australian mineral resources and the commodities, indicating the potential of tailings and by-products for mineral carbonation, according to the mineralogy of the waste. It also shows how critical some commodities are by presenting Australia's production levels and rankings in the world. Table 3 also highlights the potential of tailings and waste by-products which can be further investigated to evaluate their potential for mineral carbonation, according to the mineralogy of tailings and available research in the literature. This is in accordance with the fact that mineral carbonation can occur as the result of the reaction between CO_2 with elements such as calcium, magnesium, and iron [112]. The rankings and production data were extracted from the Australian Atlas of mineral resources, mines, & processing centres, Australian Government as of December 2016 [111].

Table 3. Australia's mineral resources and potential of its mining waste for carbon capture and storage (CCS) by mineral carbonation.

Australia's Mineral Resources (Alphabetic Order)	Can Be Found in Mafic-Ultramafic Ores	Mineral Carbonation Potential Elements in Waste Products	Examples of Mineral Phases Prone to Carbonation	Australia's World Ranking for Resources	% of World Resources	Australia's World Ranking for Production	% of World Production	Australia's Production (Mt)	World's Production (Mt)
Antimony				4	9	4	4	5.5 (kt)	130 (kt)
Bauxite		Ca, Mg, Fe, Na	Magnetite (Fe_3O_4), Sodium aluminosilicate ($Na(AlSiO_4)$)	2	22	1	31	82.152	271.5
Black Coal		Ca, Mg	Hydrocalumite ($Ca_2Al(OH)_6Cl \cdot 2H_2O$), Ettringite ($Ca_6Al_2(SO_4)_3(OH)_{12} \cdot 26H_2O$), Portlandite ($Ca(OH)_2$)	4	10	4	7	566.3	7795
Brown Coal		Ca, Mg	Hydrocalumite ($Ca_2Al(OH)_6Cl \cdot 2H_2O$), Ettringite ($Ca_6Al_2(SO_4)_3(OH)_{12} \cdot 26H_2O$), Portlandite ($Ca(OH)_2$)	2	24	5	6	63.3	783.3
Chromium	X	Mg		n.a.	n.a.	n.a.	n.a.	n.a.	30.4
Cobalt	X	Ca, Fe		2	14	5	4	5.47 (kt)	123 (kt)
Copper	X	Ca, Mg, Fe		2	12	5	5	0.948	19.4
Diamond	X	Ca, Mg, Fe	Serpentine-Group minerals ($Mg_3(Si_2O_5)(OH)_4$), Forsterite (Mg_2SiO_4)	3	18	2	24	13.958 (Mc)	127 (Mc)
Flourine				n.a.	n.a.	n.a.	n.a.	n.a.	3.1
Gold	X	Ca, Mg, Fe		1	17	2	9	288 (t)	3255 (t)
IImenite				2	19	3	13	1.4	11.6
Iron Ore	X	Fe, Ca, Mg	Magnetite (Fe_3O_4), goethite ($FeO(OH)$), biotite ($K(Mg,Fe)_3AlSi_3O_{10}(F,OH)_2$)	1	29	1	38	858	2230
Lead				1	40	2	9	0.45	4.82
Lithium				3	18	1	41	14 (kt)	34.7 (kt)
Magnesite		Ca, Mg, Fe		5	4	8	2	0.554	27.7

Table 3. *Cont.*

Australia's Mineral Resources (Alphabetic Order)	Can Be Found in Mafic-Ultramafic Ores	Mineral Carbonation Potential Elements in Waste Products	Examples of Mineral Phases Prone to Carbonation	Australia's World Ranking for Resources	% of World Resources	Australia's World Ranking for Production	% of World Production	Australia's Production (Mt)	World's Production (Mt)
Manganese Ore				4	13	4	9	3.2	44
Molybdenum	X			7	1	n.a.	n.a.	n.a.	227 (kt)
Nickel	X	Mg, Ca,	Enstatite ($MgSiO_3$), Diopside ($MgCaSi_2O_6$), Talc ($Mg_3Si_4O_{10}(OH)_2$), Serpentine-Group minerals ($Mg_3(Si_2O_5)(OH)_4$)	1	24	5	9	0.204	2.25
Niobium				2	6	minor	minor	minor	64 (kt)
Phosphate				10	2	minor	minor	1.54	264
PGEs (Platinum-group elements)	X	Mg, Ca, Fe, Na	Enstatite ($MgSiO_3$), Talc ($Mg_3Si_4O_{10}(OH)_2$), Bytownite [(Ca, Na)[Al(Al, Si)Si_2O_8], Diopside ($MgCaSi_2O_6$)	n.a.	n.a.	n.a.	n.a.	678 (kg)	380 (t)
Potash				minor	2	minor	minor	minor	39
Rare Earths (REO & Y_2O_3)				6	3	2	11	0.014	0.126
Rutile				1	50	1	42	0.3	0.7
Silver				2	16	5	5	1.418 (kt)	27 (kt)
Tantalum				1	n.a.	n.a.	n.a.	183 (t)	1.1 (kt)
Tin				4	10	7	2	6.635	278
Tungsten				2	12	n.a.	n.a.	0.11	86.5
Uranium				1	29	3	10	6.314	62 (kt)
Vanadium	X	Ca, Mg, Fe		4	11	minor	minor	minor	76 (kt)
Zinc				1	28	3	7	0.884	11.9
Zircon				1	67	1	31	0.6	2.4

Notes: t: tonne, kt: kilotonnes, Mt: million tonnes, Mc: million carrat, n.a.: not available.

4.1. Integrated Mineral Carbonation Model for Australia's Mining Industry

A major barrier of deploying mineral carbonation, at an industrial scale, is the financial requirements necessary for obtaining appropriate feedstock for the process [15]. Integration of mine waste recycling with mineral carbonation provides a distinctive opportunity for the mining industry to overcome some of the economic barriers in adopting an effective climate change adaptation technology.

Calcium- and magnesium-rich mining wastes, particularly tailings, are capable of reacting with CO_2 to form stable carbonate minerals. Mineral carbonation of alkaline mining waste under controlled conditions not only accelerates this process for CO_2 sequestration, but it can also neutralise the alkalinity and potentially immobilise and fix heavy metals and other mine waste contaminants in stable mineral carbonate forms. The research on mineral carbonation has been focused mainly on magnesium and calcium silicate minerals, while iron-containing minerals have not been investigated as much, to be used for the feedstock of mineral carbonation, due largely to their value for steel production. However, as shown in Table 3, Australia holds the world's largest resources and is the biggest producer of iron ore. That also includes large deposits of low-grade magnetite that can have the potential to be used for mineral carbon sequestration. Magnetite can react with carbon dioxide to form siderite according to the basic reaction below [112].

$$Fe_3O_4 + CO_2 \rightarrow FeCO_3 + Fe_2O_3 \tag{6}$$

However, during magnetite carbonation, formation of hematite as a passive layer can limit the carbonation kinetic, resulting in a slow reaction rate [113].

In addition, many of the Australian minerals are found in ultramafic rocks, which are rich in magnesium, iron, and calcium, as shown in Table 3. Alkaline mining wastes provide several economic advantages for CCUS by mineral carbonation. Mineral carbonation can reduce the financial burden associated with the disposal of mine waste materials. The availability of these wastes, such as red mud and fly ash, reduces the cost of sorbents which are currently used as the feedstock for mineral carbonation. Furthermore, mining wastes, tailings, and by-products are often fine and comminution energy for pretreatment of minerals is not required. In addition, neutralising the mining waste, which can be potentially hazardous to the environment, by mineral carbonation makes it safe for recycling and disposal which reduces the cost of waste management, and carbonated minerals can be used as locally available, environmentally benign materials for mine reclamation.

Figure 1 shows the schematic model of climate change adaptation technology for the Australian mining industry. Integrated mineral carbonation of alkaline mining waste in Australia brings forth the chance for waste management of some of the critical commodities' by-products, such as red mud and fly ash, at the same time reducing Australia's GHG emission and mine reclamation. This is particularly significant for countries such as Australia with many active and legacy mines. There is an estimate of over 1 million legacy mines worldwide [114], and more than 60,000 in Australia, many of which require remediation to solve and mitigate environmental and social issues such as AMD associated with past mining activities in those regions [115].
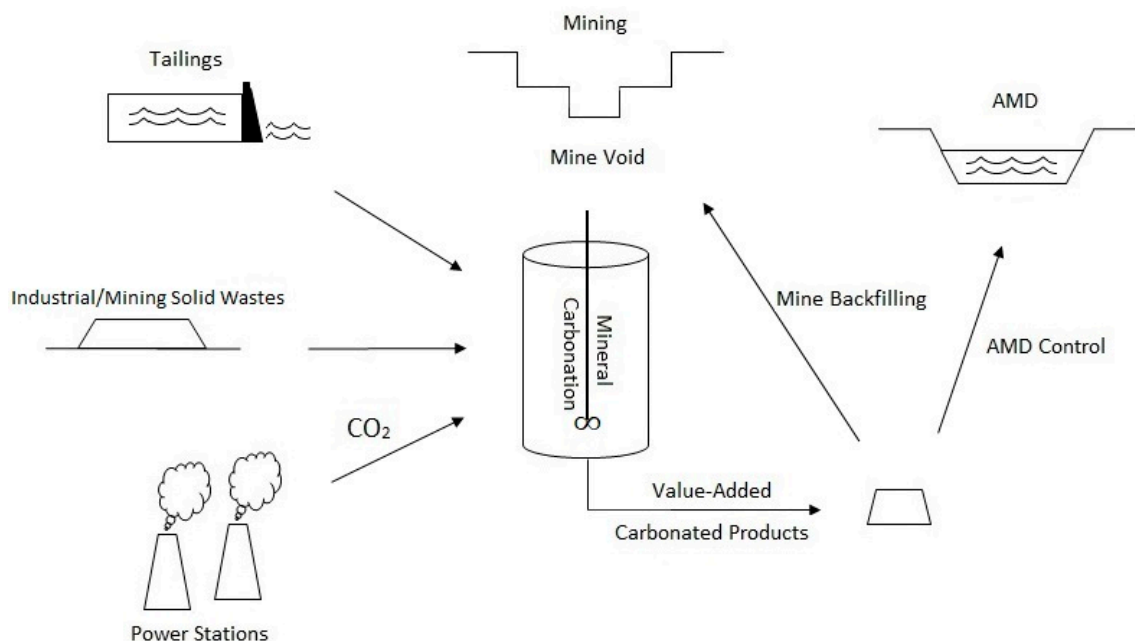
Figure 1. Integrated mineral carbonation of mining waste for waste management and mine reclamation.

The integrated mineral carbonation model will contribute to the development of an innovative climate change adaptation technology by addressing three critical issues at the same time including GHG reduction (CO_2 sequestration), mine waste management (neutralisation and reuse) and mine reclamation (i.e., mine backfilling and AMD control using the environmentally benign products of mineral carbonation).

According to Table 3, the magnitude of Australia's mineral production is very significant considering Australia's ranking for production in many critical commodities. Hence, there are accordingly large amount of mining waste, by-products and GHG production. Integration of mineral carbonation provides the opportunity to offset some of the GHG emission of the mineral's life cycle in a sustainable way. The outcome of this approach will have the potential to provide national economic, environmental and social benefits for Australia by contributing to Australia's adaptation to climate change.

4.2. Synergies between Coal Mines and Coal-Fired Power Stations in Close Proximity

One-third of Australia's GHG emissions are from the electricity generating sector, 80% of which is based on coal-fired power generation, which has been critical for the operation of many Australian industries [116]. Furthermore, it has created many jobs and is central to the Australian economy. For instance, one out of eight employment in the whole of Queensland and one out of four in Central Queensland, are dependent on the resource industry [116]. Therefore, because of the effect of coal industry on Australia's economy it is crucial that adaptation technologies such as CCUS be integrated into the industry to support coal use while offsetting its emissions, especially due to the fact that fossil fuels will still remain a significant portion of Australia and world's energy portfolio in the short-term.

Coal has historically been the primary source of electricity generation in Australia, and as the result, numerous coal-fired power stations have been built to support this demand, mainly in proximity of some of the major coal mines. The shorter distance between coal mines and power stations reduces the cost of coal transport between the source and end use. However, it also provides a unique synergy between the two for coal mine reclamation via the integrated mineral carbonation model. Figure 2 shows the locations of coal-fired power stations in proximity to coal mines in Australia.
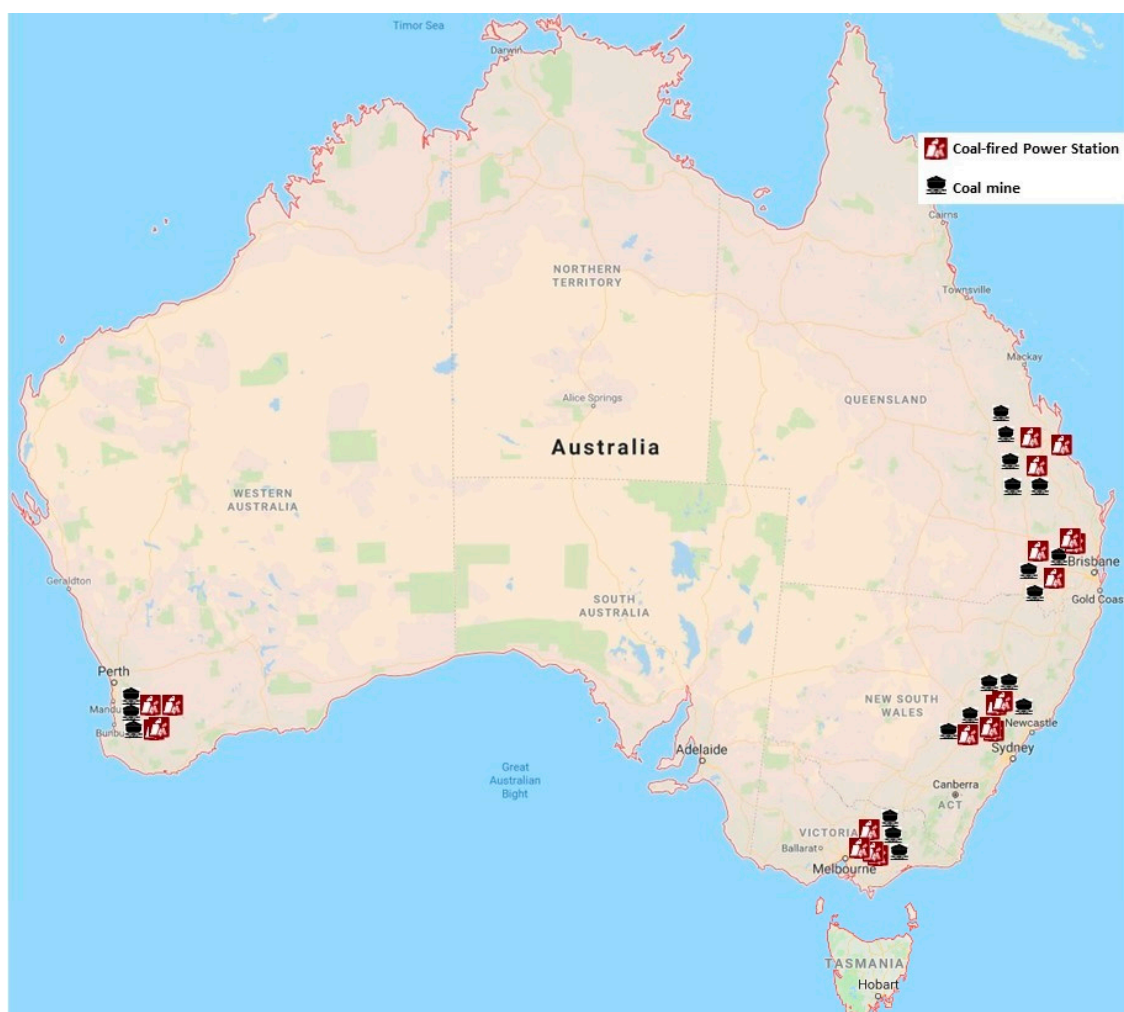
Figure 2. Coal-fired power stations in proximity of coal mines in Australia.

Fly ash is the primary by-product of coal combustion in power stations. Approximately 777.1 million tonnes (Mt) of coal combustion by-products (CCBs) are produced globally every year as the result of burning coal in power stations [117]. The global average of fly ash reuse and utilisation is 53.5% [117]. In the National waste report 2018, the Australian Government reported, for the financial year of 2016–2017, the generation of 12.3 Mt of ash by coal-fired power stations, 43% of which was recycled [118]. It was emphasised in this report that there are opportunities for fly ash utilisation if the contamination issues are addressed. Comparison of Australia's CCBs recycling ratio with other regions such as Japan (96.4 %) and Europe-EU15 (90.9 %) [117] points out the existing opportunity of fly ash utilisation in Australia. However, fly ash can be a hazardous material due to the presence of potentially toxic trace elements in the flue gas [119]. This is a major liability in Australia due to the scale of coal-fired electricity production, as it has the potential to cause serious environmental issues if it is not managed properly.

The scale of which fly ash is produced in Australia provides the opportunity for its recycling as the feedstock for mineral carbonation and, consequently, mine reclamation. Mineral carbonation has the potential to neutralise the hazardous effect of fly ash, turning it into environmentally benign minerals. This can be achieved by neutralising the alkalinity of fly ash due to the reaction with CO_2, and potentially immobilisation of trace elements due to carbonate formation. The carbonated minerals can then to be used for reclamation of the coal mines, such as backfilling and acid mine drainage treatment.

Short distances between mines and coal-fired power stations not only reduces the cost of coal transport between the coal mines and power stations, but it also provides the opportunity to transport the carbonated fly ash back to the coal mine, at low cost. For instance, Stanwell Corporation, which is owned by the Queensland Government and is the state's largest electricity generator, operates coal-fired power stations at Central and South East Queensland. The 1843 MW Tarong Power Stations are located in the South Burnet in South East Queensland [120], only a few kilometres away from Meandu Mine that provides their coal. Several other coal-fired power stations in Australia are similarly located in close proximity with coal mines (Figure 2) providing a unique synergy and the opportunity to overcome the economic barrier of deploying mineral carbonation technology in Australia's mining industry, as an economically viable climate change adaptation technology.

4.3. Red Mud

Australia produced 1.49 Mt of aluminium, as the world's 6th largest producer, in 2017 [121]. Consequently, large quantities of red mud, a by-product of the Bayer process in the alumina industry, are generated every year in Australia. Red mud is regarded as highly hazardous, with significantly high alkalinity and pH values up to 13. It often contains considerable amounts of trace elements and poses critical hazards to the environment. For instance, the dam wall of Ajka Alumina plant's red mud reservoir in Hungary, collapsed on October 4th 2010. As the result, 10 people were killed, over 100 were injured, and the lakes, rivers and surrounding ecosystems were significantly polluted [122–124]. In Australia, disposal of 26.4 Mt of red mud was reported in 2018, at sites in Queensland and Western Australia. Over the last 50 years, 812 Mt of red mud has been deposited in Australia [118]. While many methods have been trialled, such as red mud neutralisation with sea water, associated waste management concerns still exist, and millions of dollars are spent every year for rehabilitation.

Mineral carbonation can be used to reduce and neutralise the alkalinity and contaminations in red mud. The reaction pathways of red mud carbonation are reported in the literature [125,126]. It reduces the environmental risk of long-term red mud storage and utilisation, as well as offsetting CO_2 emission associated with aluminium production [127]. Treatment of bauxite residue with mineral carbonation has the advantage of not introducing impurities to the refining process [93]. It also reduces the impact of residue leachate on clay seal material and its long-term deterioration at storage facilities. It also alleviates the effect of red mud residue leachate on groundwater contamination. Furthermore, carbonated red mud requires reduced drying area due to higher shear strength and potentially faster drying time. In addition, the surface of carbonated red mud is less susceptible to aerosol emissions, besides from the fact that reduced drying area limits the exposure of red mud to dust emissions [128–130].

4.4. Ultramafic Mine Tailings

Many of the mineral commodities in Australia are mined from ultramafic ores. The growing demand for the development of infrastructure for low carbon technologies has resulted in increased ultramafic metals' mining operations such as nickel and cobalt. For example, BHP has recently shown interest in increased cobalt and nickel production driven by the market call for rechargeable batteries. BHP's plan for increasing the production of nickel sulphate to 200,000 metric tons per year is reported, which will make Nickel West in Western Australia the largest plant in the world [131]. Furthermore, a number of other commodities are mined in Australia from ultramafic rocks such as PGEs (platinum-group elements), diamond and legacy asbestos mines.

Many concerns exist for the climate change impacts of mining wastes and by-products of ultramafic mine tailings. They can be hazardous due to containing many toxic elements and mineral groups, and in the case of asbestos tailings, pose serious human health risks if not managed properly [132].

Ultramafic mine tailings are often very rich in magnesium, which makes them a suitable feedstock for CO_2 sequestration by mineral carbonation. In addition, they are often ground and do not require

high activation energy and grinding, which reduces some of the related costs. More importantly, the carbonation process can fix and immobilise hazardous elements and mineral groups in carbonate forms, and in some cases, produce environmentally benign minerals which can be used for mine site rehabilitation. In case of legacy asbestos mine tailings, mineral carbonation can potentially reduce contamination and dust emissions, as well as providing the benefit of capturing CO_2 permanently in a stable form. Table 4 lists the potential mining wastes in Australia to be used for the integrated mineral carbonation.

Table 4. Applicability of Australian mining waste for integrated mineral carbonation for mine waste management and mine reclamation.

Mine Waste	Carbonation Advantages	Product Applicability	Carbonation References
Red mud	• Neutralising the high alkalinity for safe waste management • Immobilisation and fixation of heavy metals and contaminations • Reduced the risk of groundwater contamination • Reduced drying area • Reduced hazardous dust emissions • Large-scale availability of red mud due to Australia's extent of aluminium production	• Mine backfilling • Acid mine drainage (AMD) control	Cooling et al. 2002 [130] Tran 2016 [125] Revathy et al. 2017 [133] Liang et al. 2018 [134] Sahu et al. 2010 [135]
Fly ash	• Produced at power stations as a major source of CO_2 emission • Reduced aerosol emissions • Large-scale Availability due to Australia's extent of coal-fired power generation • No requirement for grinding	• Mine backfilling • Acid mine drainage (AMD) control • Soil amendment	Liu et al. 2018 [136] Jaschik et al. 2016 [137] Tamilselvi Dananjayan et al. 2016 [138] Ukwattage et al. 2015 [139]
Ultramafic mine tailings (nickel, diamond, PGE and asbestos tailings)	• Neutralising the alkalinity • Immobilisation of environmentally hazardous metals in tailings • No requirement for grinding • Reducing hazardous/fatal dust emissions	• Acid mine drainage (AMD) control	Nickel tailings: Teir et al. 2007 [140] Teir et al. 2009 [141] Diamond tailings: Mervine et al. 2018 [142] PGE tailings: Meyer et al. 2014 [143] Vogeli et al. 2011 [144] Asbestos tailings: McCutcheon et al. 2017 [145] Oskierski et al. 2016 [146]

5. Barriers to Utilisation of Integrated Mineral Carbonation

The initial capital investment is one of the primary barriers for utilisation of mineral carbonation in Australia's mining industry. The range of the cost estimates of implementing mineral carbonation technology reported in the literature varies in a wide range of values [57,147–149]. Although some of these initial cost estimations (i.e., \$69/ton CO_2 [57]) are too high to be economically feasible, the fact that this method has the potential for permanent storage over geologic time makes it desirable [45].

Despite the fact that many strategies used to improve the mineral carbonation process, they have been successful in terms of increasing the overall yields of carbonation process; each method has its own disadvantages. The energy cost associated with fine grinding of the feedstock directly contributes to decreasing the process viability [149–152]. Thermal activation of serpentine, for instance, requires a considerable amount of energy that results in a not economically feasible process unless methods of thermal activation can be optimised or reconsidered [69,153]. Obviously, high operating temperatures and pressures also add significantly to the cost of operation. Therefore, studies have been investigating the extraction of Mg from minerals under low temperature and pressure conditions using mineral acids [61,64,154]. Also, any chemical that is added to the reactants must be fully recoverable in a real process to make it commercially viable. The rate-limiting step for serpentine carbonation is the dissolution step [96]. Many previous investigations have reported the potential passivating quality of silica layers during the dissolution of serpentinite minerals [74,155]. Other research has shown

reprecipitation of amorphous silica in the solution that causes a precipitate layer to form on the surface of the mineral particles and significantly limits further extraction of magnesium [96]. The removal of silica layers is possible through a concurrent mechanical exfoliation process could enhance carbonation yields [66,156].

Obtaining a constant source of high-concentration CO_2 can also be a major issue to deploy mineral carbonation plant in Australian mining industries. There might not always be a reliable source of CO_2 in the vicinity of mining waste storage facilities to support mineral carbonation plants. Therefore, the transportation of CO_2 to the carbonation plant can potentially add to the cost of the operation. However, this is not always the case and for example Alcoa used the CO_2 from the waste stream of an ammonia plant near to the Alcoa Kwinana Refinery for the operation of their mineral carbonation plant [93].

Mineral carbonation can reduce the alkalinity of the mining waste, by the reactions between CO_2 and alkali and alkaline earth metals. However, in some cases there is risk of changing the alkalinity back to higher pH again after carbonation. Alkali elements such as sodium, which can be found in waste streams like red mud, can react with CO_2 and generate bicarbonates that are less stable for long-term storage of CO_2 above the ground due to their solubility in water [16,47,157]. The carbonation reaction of CO_2 with the sodium hydroxide, which is found in red mud, is relatively rapid, while the reaction with the alkaline earth metals can be slower. In addition, the presence of tricalcium aluminate (TCA6) in red mud is reported to cause technical issues by increasing the pH in the residue over time [130].

6. Conclusions

The effects of climate change have been observed on a global scale. CCUS via mineral carbonation appears to be a promising emerging technology to be included in the portfolio of climate change mitigation options, as an effective long-term strategy. Barriers mainly due to the cost of deployment and the lack of a reliable source of alkaline waste have hindered its application at an industrial scale. Utilisation of mining waste provides the opportunity to overcome this challenge. The abundance of mining wastes and processing by-products such as fly ash, red mud and ultramafic mine tailings in Australia creates a unique opportunity for retrofitting mineral carbonation in its mining sector. The high environmental and financial liabilities of mining wastes in Australia justify investments in detailed investigation into mineral carbonation opportunities. Integrated mineral carbonation of mining wastes contributes to mining waste management, mine reclamation, and GHG emissions. Development of climate change adaptation technologies within the mining industry in Australia, as a country with an advanced resource industry, can also initiate global models for utilisation in developing countries.

This review discussed the opportunities and barriers that exist for unlocking the value of mining waste by mineral carbonation. Changing CO_2 policies, further research for improved technology through better understanding of the mineral carbonation process, the development of pilot-scale operations and controlling mining waste contamination could collectively promote the progression of ex situ mineral carbonation to make it applicable at industrial scales.

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