Research progress on carbon dioxide mineralization sequestration technology by tailings

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ABSTRACT

Carbon dioxide capture, utilization, and storage (CCUS) has become a focal point of research for scientists worldwide to address the global warming issue. Among these studies, utilizing mine tailings as CO_2 mineralization materials has emerged as a new research approach in recent years. This study reviews the research progress of using mine tailings for CO_2 mineralization and sequestration. The reaction mechanisms of CO_2 mineralization using tailings have been introduced first. Notably, ultramafic tailings containing divalent cations, such as Ca^{2+}, Mg^{2+}, and Fe^{2+}, can react with CO_2 to produce carbonate precipitates. Furthermore, factors including the size of mineral particles, reaction temperature, and pressure will influence the mineralization reaction rate. The activation methods of tailings to accelerate mineral carbonation have been summarized. The activation of tailings for mineralization generally includes acid leaching, ammonium salt pH swing, and physical grinding. Newly developed methods, such as a combination of acid digestion and electrolysis of olivine, have also been introduced. Thereafter, the processes and technologies for CO_2 sequestration through tailings mineralization have been analyzed. The *in-situ* and *ex-situ* mineral carbonation processes have been introduced. The last part explores the technical approaches of synergistic mineralization and storage of CO_2 by cemented tailings backfill in underground mined-out areas, which is considered to be one of the key technological pathways for CCUS in the future. However, critical technical challenges must still be addressed for this process, including low-cost multiphase CO_2 storage in backfill plants, synergistic transportation of backfill slurries and CO_2, and *in-situ* injection processes of CO_2 in mined-out stopes to produce suitable carbon curing environments. The research content of this article can provide valuable insights for studies on the mineralization and storage of CO_2 using tailings.

1. Introduction

Natural disasters due to global warming have become increasingly frequent and severe. From the report by the United Nations (UN) Environment Programme, using the average temperature from 1901 to 2000 as the baseline, global temperatures have accelerated since the 21st century and have now risen by nearly 1°. The climate warming trend is established; without intervention, the global average temperature is expected to rise by 3.4 to 3.7 °C relative to pre-industrial levels by the year 2100 [1]. Greenhouse gas emissions, primarily carbon dioxide emissions from fossil fuel combustion, are considered to be the main cause of global warming [2]. Since the 26th UN Climate Change Conference, the “Glasgow Climate Pact” has been adopted by 200 countries to address the aforementioned issue; they unanimously agreed to solve the problem of global warming and jointly control and reduce greenhouse gas emissions [3].

In this background, the application of new types of energy, such as solar and wind power and technologies for carbon emission reduction, have become the focus of scientific investigations. Carbon dioxide capture, utilization, and storage (CCUS) have been considered an effective and economically viable method to reduce CO_2 [4–6]. At present, different kinds of CCUS technologies have been established. The commonly used carbon capture technology for fossil fuel combustion includes pre- and post-combustion capture. Pre-combustion capture refers to the process of using gasification and reforming reactions to separate carbon-containing components from the fuel before combustion, thereby achieving the goal of carbon capture [7]. Post-combustion capture refers to the technology of capturing and separating carbon dioxide from the flue gas produced by the combustion of fossil fuels. The main technologies currently include chemical absorption, membrane separation, and solid adsorption. These technologies are widely used in industries such as power generation and metallurgy due to their long development history and effectiveness [8]. A type of direct air carbon dioxide capture technology (DAC) is also available [9]. However, this technology generally has high energy consumption and is not widely applied.

The utilization pathways of carbon dioxide can be categorized into physical utilization, chemical utilization, and biological utilization. In terms of physical utilization, carbon dioxide can be directly used to produce dry ice, carbonated beverages, and beer. Chemical utilization involves using carbon dioxide to synthesize hydrocarbons and organic acids [10]. In recent years, electrochemical methods have been employed to use carbon dioxide in the production of materials such as carbon nanotubes or graphene [11]. Biological utilization of carbon dioxide, such as microalgae adsorption, can be used to produce fertilizer and food additives or utilized for greenhouse cultivation. This method is currently considered to be a green and ideal approach to utilizing carbon dioxide.

Carbon sequestration technology refers to the safe storage of captured carbon dioxide to facilitate its isolation from the atmosphere. This technology mainly includes geological and ocean sequestration. In addition, carbon sequestration technologies include mineralization and liquefaction sequestration [12]. Among these technologies, mineralization carbonation sequestration offers advantages such as process controllability, high sequestration efficiency, and stable carbon fixation structures. Therefore, since its introduction by Seifritz in "Nature" in 1990 [13], this concept has become a research focus for an increasing number of researchers [14–16]. Existing studies [17,18] have indicated that metal mine tailings typically have high alkaline calcium–magnesium silicate content, small particle size, and high specific surface area, which provide considerable potential for mineral carbonation. More specifically, tailings with high contents of calcium and magnesium oxides, silicates, or hydroxides in their mineral composition generally have high carbon mineralization potential [19]. In the carbon dioxide curing environment, these minerals can easily dissolve or undergo ion exchange on the surface of the tailings when they encounter water, reacting with carbon dioxide to produce corresponding carbonates (such as calcium carbonate) and forming precipitates, thereby achieving carbon mineralization and sequestration [20]. Notably, tailings from copper–nickel and ultrabasic rock mines, such as diamond mines, can react with carbon dioxide in the deposit, achieving a considerable amount of carbon dioxide sequestration [18,21].

For the carbon dioxide mineralization sequestration by tailings (CMST), the bibliometric analysis was conducted based on the data obtained from Scopus. As shown in Fig. 1, since 2008, the mineralization of tailings for carbon dioxide sequestration has gradually become a research focus, and the number of related publications has been steadily increasing. As shown in Fig. 2, on an international scale, China has been the country most interested in the topic of tailings mineralization for carbon dioxide sequestration in recent years, followed by Canada and the United States. From the perspective of research keywords, this phenomenon can reflect the focus of the studies. As shown in Fig. 3, the hot topics in related research include keywords such as pH, mining, phase interface, mineral carbonation, and waste management. Therefore, researchers studying the mineralization of carbon dioxide in tailings mostly focus on theoretical research related to the mining process, studies on mineral carbonation reactions, and research on solid waste disposal.

In terms of the specific content of studies, Wilson *et al.* conducted measurements on the carbonation of ultramafic rocks from the Clinton Creek and Cassiar chrysotile deposits [22], validating the feasibility of using tailings for mineral carbonation and CO_2 sequestration. Liu *et al.* realized accelerated regulation of the carbonation rate of magnesium–calcium oxides and silicate minerals in the CO_2 loading and release cycle by adding amine-containing solvents such as monoethanolamine [23]. This advancement promotes the development of controlled mineral carbonation technology. Ragipani *et al.* developed an experimental framework and data analysis method for studying mineral dissolution and mineralization behaviors based on molecular simulation techniques [24], establishing the foundation for related CMST research. Kelemen *et al.* reviewed the processes for capturing and mineralization sequestering atmospheric CO_2 using ultramafic rocks [25], providing valuable insights for CMST research. Moreover, the University of British Columbia established the CarbMin Lab, dedicated to developing technologies for carbon sequestration using mine wastes. The representative results include the following: Oskierski *et al.* launched a series of experiments on the mineralization sequestration of carbon dioxide from atmospheric air using ultrabasic tailings from a

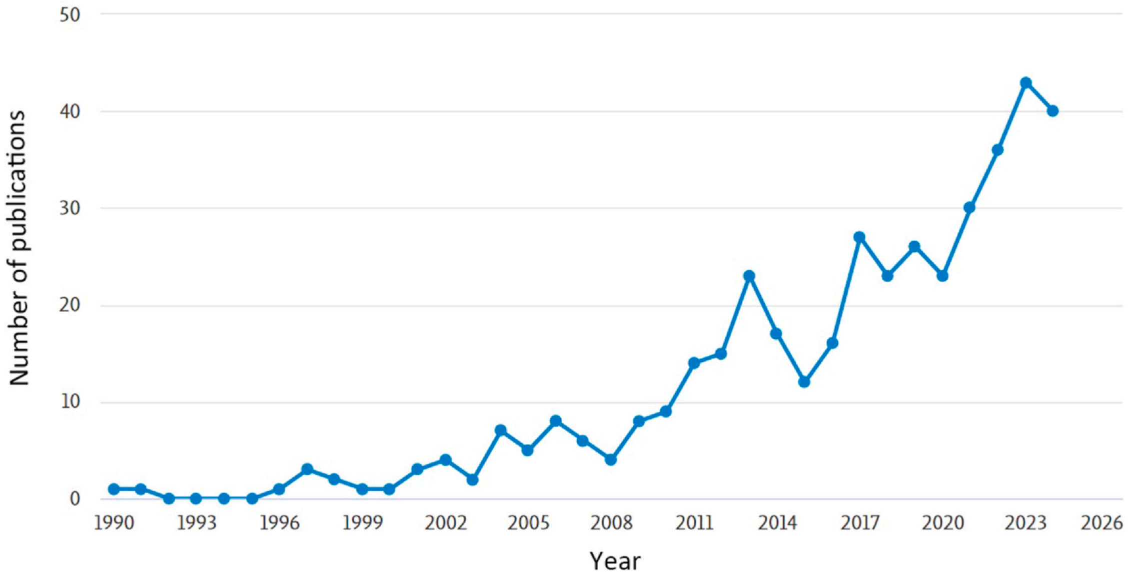

Fig. 1. Number of publications about CMST in different years.

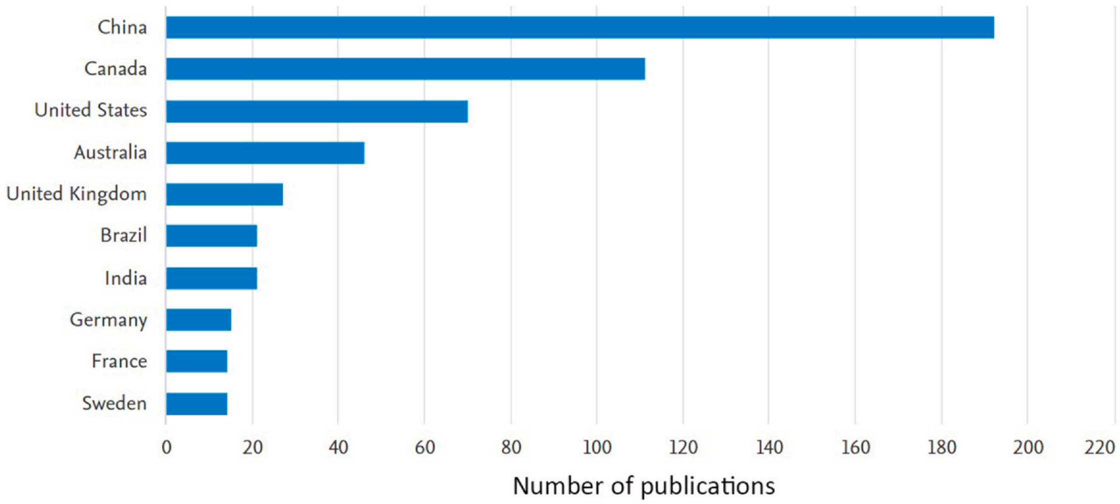

Fig. 2. Number of publications about CMST in different countries.

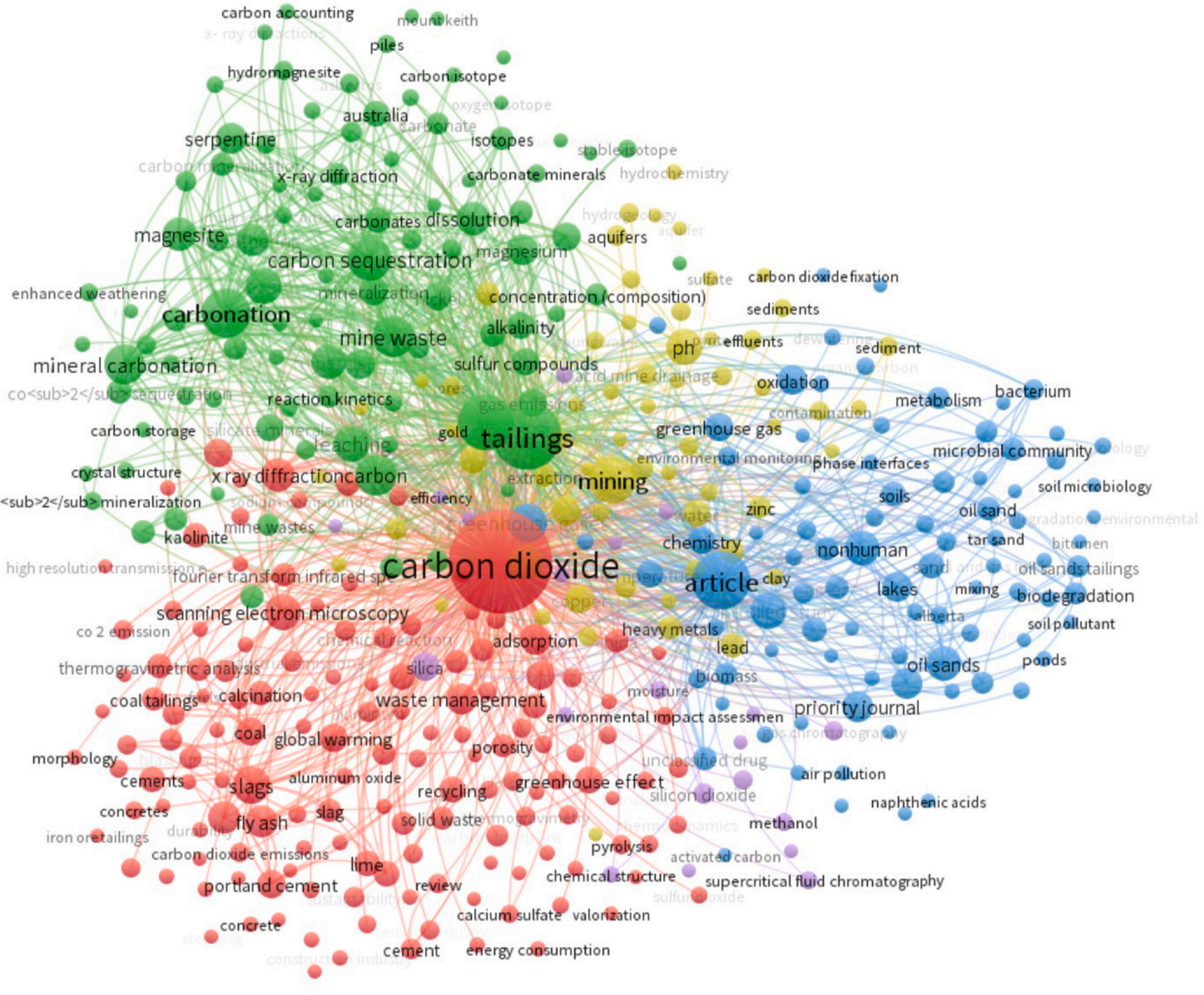

Fig. 3. Word cloud analysis of research keywords about CMST.

mine [26] and analyzed the related tailings activation method. Woodall *et al.* evaluated the carbon mineralization potential of the tailings from a platinum group metal mine in North America, performed carbon dioxide mineralization sequestration experiments, and summarized the processes and technologies [27].

The aforementioned information briefly summarizes the purpose and importance of CCUS, as well as the advantages and recent studies regarding the use of tailings for CO_2 mineralization and sequestration. Using mine tailings for carbon dioxide mineral sequestration can solve the problem of tailings storage while leveraging the carbon fixation potential of the tailings to achieve carbon sequestration. Thus, this method is considered an ideal, low-cost, and convenient approach for carbon sequestration. The following section will introduce the technologies and processes related to CO_2 mineralization and sequestration in tailings to provide a reference for related research.

2. Reaction mechanism of carbon dioxide mineralization in tailings

The CO_2 mineralization and sequestration by tailings mainly occur through the reaction between silicate minerals containing divalent cations such as Ca^{2+}, Mg^{2+}, and Fe^{2+} and CO_2 to produce stable carbonates. The reaction equation can be summarized as Eq. (1), and the process is presented in three steps: first, gaseous CO_2 is dissolved to form CO_3^{2-} and HCO^{3-}. Subsequently, the divalent metal cations of mineral tailings are dissolved (such as Ca^{2+}, Mg^{2+}, and Fe^{2+}). Finally, metal cations react with acid anions to produce stable carbonate minerals, such as $CaCO_3$, $MgCO_3$, and $FeCO_3$, and the reaction process is shown in Fig. 4 [28].

$$(\mathrm{Ca,\ Mg,\ Fe})\mathrm{SiO_3} + \mathrm{CO_2} \rightarrow (\mathrm{Ca,\ Mg,\ Fe})\mathrm{CO_3} \rightarrow \mathrm{SiO_2} \tag{1}$$

2.1. Mineralization mechanism of different minerals

Some rocks in nature contain a large number of calcium–magnesium silicate minerals, such as calcium-rich wollastonite ($CaSiO_3$), magnesium- and iron-rich olivine (Mg_2SiO_4 and Fe_2SiO_4), serpentine ($Mg_3Si_2O_5(OH)_4$), and talc ($Mg_3Si_4O_{10}(OH)_2$). Others, such as pyroxene ($Mg_2Si_2O_6$ and $CaMgSi_2O_6$) and feldspar ($CaAl_2Si_2O_8$, $NaAlSi_3O_8$), and metal oxides such as magnetite (Fe_3O_4) and hematite (Fe_2O_3), can react with CO_2 to produce the carbonate phase [29]. These minerals can be theoretically mineralized by CO_2, and the equations are presented as follows:

(1) As one of the most abundant minerals in nature, olivine commonly includes forsterite (Mg_2SiO_4) and fayalite (Fe_2SiO_4). In particular, forsterite is a key component of rocks in the upper mantle and lower crust. Forsterite reacts with CO_2/H_2O fluid to form magnesium carbonate due to its relatively fast dissolution rate, attracting considerable attention [30].

$$\text{Forsterite:}\ \mathrm{Mg_2SiO_4} + 2\mathrm{CO_2} \rightarrow 2\mathrm{MgCO_3} + \mathrm{SiO_2} \tag{2}$$

$$\text{Fayalite:}\ \mathrm{Fe_2SiO_4} + 2\mathrm{CO_2} \rightarrow 2\mathrm{FeCO_3} + \mathrm{SiO_2} \tag{3}$$

(2) Pyroxene, a common silicate rock-forming mineral, widely exists in igneous and metamorphic rocks. Many kinds of pyroxene, diopside, hedenbergite, augite, aegirine, and spodumene exist. Among them, diopside ($CaMgSi_2O_6$) has attracted considerable attention because it can produce various carbonate products once exposed to CO_2. This phenomenon is mainly attributed to the abundance and complex multi-ion composition of diopside [31].

$$\text{Enstatite:}\ \mathrm{Mg_2Si_2O_6} + 2\mathrm{CO_2} \rightarrow 2\mathrm{MgCO_3} + \mathrm{SiO_2} \tag{4}$$

$$\text{Ferrosilite:}\ \mathrm{Fe_2Si_2O_6} + 2\mathrm{CO_2} \rightarrow 2\mathrm{FeCO_3} + \mathrm{SiO_2} \tag{5}$$

$$\text{Diopside:}\ \mathrm{CaMgSiO_3} + \mathrm{CO_2} \rightarrow (\mathrm{Ca,\ Mg})\mathrm{CO_3} + \mathrm{SiO_2} \tag{6}$$

$$\text{Hedenbergite:}\ \mathrm{CaFeSiO_3} + \mathrm{CO_2} \rightarrow (\mathrm{Ca,\ Fe})\mathrm{CO_3} + \mathrm{SiO_2} \tag{7}$$

(3) Serpentine is a magnesium-rich silicate mineral formed by the alteration of olivine and pyroxene in nature, which mainly includes lizardite, chrysotile, and antigorite. Serpentine did not retain the nesosilicate and chain silicate structure (olivine and pyroxene) during the alteration process but still changed into a phyllosilicate structure, retaining the form of parent minerals [32]. The reaction between serpentine and CO_2 is as follows:

$$\text{Serpentine:}\ \mathrm{Mg_3Si_2O_5(OH)_4} + 3\mathrm{CO_2} \rightarrow 3\mathrm{MgCO_3} + 2\mathrm{SiO_2} + 2\mathrm{H_2O} \tag{8}$$

(4) Studies have shown that using alkaline earth materials, including iron oxides, may be a related method for the long-term storage of CO_2 [33]. However, iron oxide needs certain conditions when

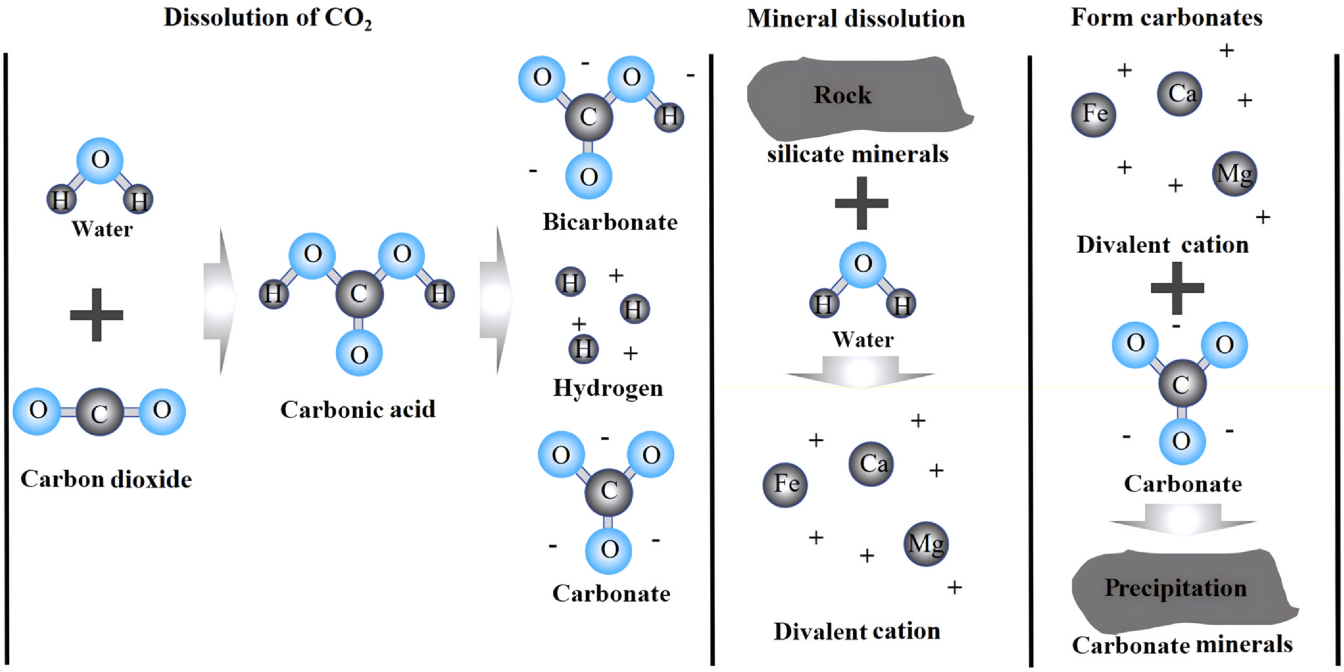

Fig. 4. Carbon mineralization process modified from [28].

capturing CO_2 and reacting with it to generate iron carbonate. Therefore, iron oxide is not conducive to the storage of CO_2 under natural conditions.

$$\text{Hematite: } Fe_2O_3 + 3CO_2 + Fe \rightarrow 3FeCO_3 \tag{9}$$

$$\text{Magnetite: } Fe_3O_4 + 4CO_2 + Fe \rightarrow 4FeCO_3 \tag{10}$$

(5) Feldspar, a collective term for a common type of aluminosilicate mineral in nature, contains calcium, sodium, and potassium. This mineral is also the most common in the Earth's crust, accounting for approximately 60% of the total mass. Among feldspar, sodium and calcium feldspar are the two most common main minerals. The equation of their mineralization reactions with CO_2 is as follows:

$$\text{Albite: } NaAlSi_3O_8 + CO_2 \rightarrow NaAl(CO_3)(OH)_2 + 3SiO_2 \tag{11}$$

$$\text{Anorthite: } CaAl_2Si_2O_8 + CO_2 \rightarrow CaCO_3 + Al_2O_3 + 2SiO_2 \tag{12}$$

2.2. Reaction equation and influence factors of mineral carbonation

2.2.1. Reaction equation of mineral carbonation

The carbonation speed of minerals is extremely slow under natural conditions; thus, different means are necessary to improve its reaction speed, including but not limited to raising temperature, increasing pressure, and using wet carbonation. Compared with dry carbonation, wet carbonation helps CO_2 and silicate minerals to dissolve in solution in ionic form; therefore, its reaction rate is markedly improved compared with dry carbonation and is relatively more economical [34]. The specific reaction equation can be expressed as

$$\text{Dissolution of carbon dioxide: } CO_2 + H_2O \rightarrow H^+ + HCO_3^- \tag{13}$$

(1) Dissolution and carbonization of forsterite:

$$Mg_2SiO_4 + 2HCO_3^- \rightarrow 2MgCO_3 + 2OH^- + SiO_2 \tag{14}$$

$$CO_2 + OH^- \rightarrow HCO_3^- \tag{15}$$

(2) Dissolution and carbonization of serpentine:

$$Mg_3Si_2O_5(OH)_4 + 6H^+ \rightarrow 3Mg^{2+} + 2SiO_2 + 5H_2O \tag{16}$$

$$Mg^{2+} + 2HCO_3^- \rightarrow MgCO_3 + CO_2 + H_2O \tag{17}$$

(3) Dissolution and carbonization of wollastonite:

$$CaSiO_3 + 2H^+ \rightarrow Ca^{2+} + H_2O + SiO_2 \tag{18}$$

$$Ca^{2+} + 2HCO_3^- \rightarrow CaCO_3 + CO_2 + H_2O \tag{19}$$

2.2.2. Factors affecting carbonation efficiency

Some researchers have shown that the mineralization efficiency of carbon dioxide is not only affected by the richness of divalent cations that can react with CO_2 in minerals but also depends on the dissolution rate of divalent cations in minerals [35]. Several factors affect the dissolution rate of mineral ions, including the size of mineral particles, temperature and CO_2 pressure during carbonation, pH value, stirring speed of solution, reaction time, and solid–liquid ratio. The specific reasons for the impact are presented as follows:

(1) Particle size. When solid wastes, such as tailings or metallurgical slags, are crushed and ground, the reduction of particle size not only increases the specific surface area of the material but also enhances the activity of the material due to the destruction of crystal structures (physical and chemical effects) during particle crushing and grinding. Therefore, grinding not only increases the reaction surface area but also destroys the mineral structure to some extent, thus promoting the mineral carbonation reaction [36].

(2) Temperature. The increase in temperature can raise the leaching rate of metal cations in the solution to some extent, increase the contents of Ca^{2+}, Mg^{2+}, and Fe^{2+} in the aqueous solution, and accelerate the reaction. However, if the temperature is too high, then relative negative benefits will emerge because the dissolution rate of CO_2 will decrease with the increase in temperature [37,38]. Therefore, determining the best balance conditions of reaction and dissolution rates is necessary to improve the final conversion rate.

(3) CO_2 pressure. Notably, carbonation efficiency will also vary with CO_2 pressure. When CO_2 pressure is higher than atmospheric pressure, ${CO_3}^{2-}$ in solution is easy to leach out, which promotes the carbonation reaction [39]. If the dissolution of CO_2 becomes the limiting step in the carbonation process, then the carbonation efficiency can be promoted by increasing the pressure. However, increasing the pressure will also raise the cost; therefore, the pressure increase must be appropriate.

(4) pH. In carbonation, pH has an important influence on the precipitation of carbonate: when $pH < 7$, that is, under acidic conditions, carbonation is beneficial to the dissolution of Ca^{2+} and Mg^{2+} ions; when the $pH > 7$, that is, under alkaline conditions, carbonation will be beneficial to the precipitation of carbonate [40]. Moreover, with the advancement of the reaction, the pH of the solution will gradually decrease with that of the reaction. Therefore, adding alkaline substances such as NaOH and $NaHCO_3$ is necessary to promote the precipitation of carbonate.

(5) Reaction time and stirring speed. Reaction time is an important factor in determining the size, shape, and crystal structure of carbonate products during the reaction. Different additives also contribute to the polymorphism of carbonate during the carbonation process [41]. Additives can change the recrystallization process for different surfaces as well as adjust the pH value. The dissolution and recrystallization of crystals can be transformed into a relatively stable carbonate crystal structure by prolonging the reaction time. The stirring rate affects the particle size and morphology of the crystals; thus, a fast stirring speed leads to a smaller size of the obtained crystal product [42]. Taking the carbonate of Ca as an example, calcite is the main phase at a low stirring rate. The final product will change to vaterite with the increase in stirring rate.

(6) Solid–liquid ratio. The solid–liquid (S/L) ratio refers to the ratio of solid powder to water. In the process of mineral carbonation, water, as a medium for dissolving CaO, MgO, and Fe_2O_3 metal oxides in water, reacts with CO_2 dissolved in the solution. When the S/L ratio increases, the concentration of metal cations in the slurry increases, which promotes the carbonation reaction. However, the viscosity of the solution is further increased with the S/L ratio, which affects the interfacial mass transfer between liquid–solid–gas and hinders the carbonation process [38].

3. Activation methods for accelerated tailings carbonation

The direct gas–solid reaction of CO_2 with tailings has relatively low efficiency and yield. Serpentine or olivine minerals generally have a highly stable structure, and the magnesium atoms are linked to hydroxyls inside the structure. The slow dissolution kinetics of Ca/Mg-silicates from minerals of tailings limit their carbonation reactivity. Numerous physical, chemical, and thermal activation methods can be employed to break down the stable structure of serpentine.

3.1. Chemical activation

Chemical activation aims at polarizing and weakening the magnesium bonds within the Mg-silicate structure. In chemical activation, carbonation reactivity could be improved by adding chemicals such as acids, bases, and ammonium salts.

3.1.1. Acid leaching

Literature shows that acids can extract substantial amounts of calcium and magnesium from feedstocks to be subjected to carbonation experiments in the following steps. Notably, H_2SO_4 was the most efficient for extracting magnesium from serpentinite at room temperature, followed by HCl, HNO_3, HCOOH, and CH_3COOH [43]. The main reactions are as follows [44]:

$$Mg_3Si_2O_5(OH)_4(s) + 6HCl\,(aq) \rightarrow 3MgCl_2(aq) + 2SiO_2(s) + 5H_2O(l),\ \Delta H = -236\ \text{kJ/mol} \quad (20)$$

$$Mg_3Si_2O_5(OH)_4(s) + 6HNO_3(aq) \rightarrow 3Mg(NO_3)_2(aq) + 2SiO_2(s) + 5H_2O(l),\ \Delta H = -393\ \text{kJ/mol} \quad (21)$$

The extraction of magnesium in deionized water was as low as 0.2wt %. Even dilute acid concentrations (0.1 M) notably increased the extraction of Mg and Fe in serpentinite compared to that of pure water [44]. A remarkable increase in Mg recovery during the acid leaching process of serpentine was realized using sulfuric acid with an additional 5wt% fluorite powder. The complexation of fluoride ions (F^-) with Si in serpentine promoted a distorted tetrahedral orientation, which led to a loose crystal structure of serpentine and contributed to the exposure of additional Mg. A high leaching efficiency of approximately 94% for Mg was achieved, representing a 36.42% increase compared to the system without fluorite [45].

3.1.2. Ammonium salt pH swing

Two barriers to the development of CO_2 mineral carbonation are the low efficiency of mineral dissolution and the unrecyclable use of additives [46]. A pH swing process was developed; magnesium was extracted from serpentine under low pH conditions in the first stage, and the solution pH was then raised to improve the carbonate precipitation [47]. In this process, recyclable ammonium salts were proposed to overcome these barriers.

The main reaction in the dissolution step involves using ammonium salt aqueous solutions, which include $(NH_4)_2SO_4$, NH_4Cl, and NH_4HSO_4. By contrast, NH_4HSO_4 has the highest efficiency in extracting magnesium from serpentine [46]. The reaction mechanism between serpentine and the ammonium salt can be described as follows [48]:

$$Mg_3Si_2O_5(OH)_4 + 6NH_4HSO_4 \rightarrow 3MgSO_4 + 2SiO_2 + 5H_2O + 3(NH_4)_2SO_4 \quad (22)$$

The second stage of the process involved the pH swing from acidic (pH 0.2) to neutral (pH 7) and then basic (pH ≥ 8) through additional ammonia water to remove all the impurities (Fe, Al, Zn, Ni, Cu, and Mn) from the filtrate by precipitating them as hydroxides, as shown below:

$$NH_4HSO_4 + NH_3{\cdot}H_2O \rightarrow (NH_4)_2SO_4 + H_2O \quad (23)$$

$$(Fe, Al)(SO_4)_3 + 6NH_3{\cdot}H_2O \rightarrow 2(Fe, Al)(OH)_3(\downarrow) + 3(NH_4)_2SO_4 \quad (24)$$

$$(Mn, Ni)SO_4 + 2NH_3{\cdot}H_2O \rightarrow (Mn, Ni)(OH)_2(\downarrow) + (NH_4)_2SO_4 \quad (25)$$

In the pH swing process, temperature exerts a direct effect on the rates of olivine dissolution to permanently sequester the CO_2 into stable carbonates. The high dissolution efficiency of olivine with particle size from 75 to 150 µm was 77% at 100 °C for 3 h due to a limitation of product layer diffusion with an apparent activation energy of 31 $kJ{\cdot}mol^{-1}$. The CO_2 sequestration efficiency was 70% [49].

3.2. Mechanical activation

Mechanical activation has been achieved through the application of mechanical force (high-energy attrition grinding), which disorganizes the crystal lattice around Mg ions of olivine or serpentine. This process stores excess energy on the particle surface and bulk, thereby facilitating CO_2 adsorption under ambient conditions [50]. The changes in the surface properties and crystal structures of minerals result in enhanced reactivity in carbonation or dissolution. The practice has proven that compared to direct aqueous carbonation, using a stirred mill to first grind the minerals as mechanical activation results in the highest reaction efficiency, followed by the planetary and vibratory mills, respectively [51–53].

Dry grinding has an effective activation limit; when this limit is not exceeded, high grinding intensity will lead to crystallites with small particle sizes. Conversely, if such a limit is exceeded, the particles tend to agglomerate as the milling time increases [54]. Particle reduction by grinding generally enhances serpentine dehydroxylation and yields highly disordered materials. In addition to accelerated dehydroxylation, the recrystallization rate of the reaction products also decreases due to the topotactic nature of serpentine transformation [51]. Moreover, stirred milling in wet conditions produces the largest specific surface area, while vibratory milling in dry conditions generates the most disordered materials [52].

As the pretreatment method within an integrated mineral carbonation process for mine waste (e.g., olivine, serpentine, forsterite, and lizardite), mechanical activation is feasible in a direct aqueous carbonation process, effectively reducing particle size, enlarging the surface area, distorting the crystal structure, and thereby enhancing the efficiency of CO_2 sequestration [53,54].

Fig. 5 shows the concept of an integrated mineral carbonation process in a type of nickel tailings with mechanical activation as pretreatment [53].

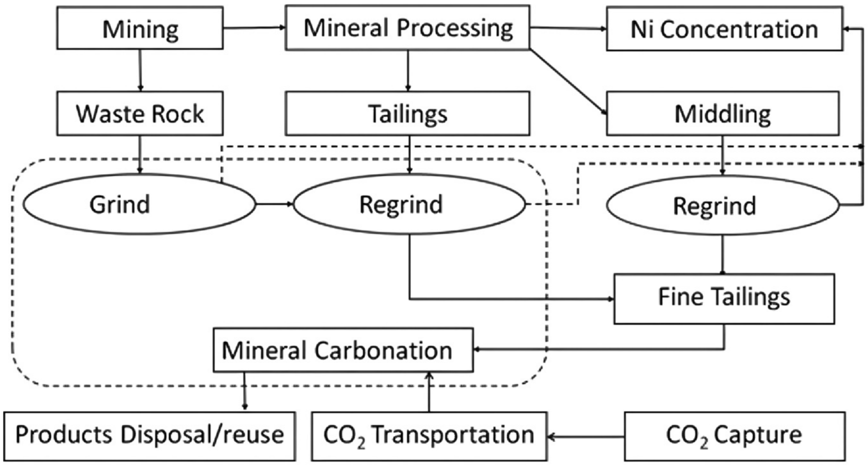

Fig. 5. Concept of an integrated mineral carbonation process with mechanical activation as pretreatment method. Reproduced with permission from Ref. [53]. Copyright (2018) Elsevier.

A case study of a real mine shows that for an optimum CO_2 sequestration efficiency of 60%, the mine must spend 210 and 260 kW·h/t of ore on mechanical activation costs and carbonation reaction energy consumption, respectively. This condition results in a CO_2 sequestration cost ranging from $104.1 to $107.1 per ton. Based on the productivity of the integrated mineral carbonation plant of the mine, 14.62 Mt of CO_2 has been sequestered per year using mechanical activation on waste rock and tailings during the 28-year life of the mine [53].

3.3. Thermal activation

Thermal activation can be realized by heat, steam, or microwave. This method is appropriate only for serpentine group minerals, which contain up to 13wt% of chemically bound water [50]. During thermal pretreatment, water is released in accordance with Eq. (26), disorganizing the compact and resistant structure of serpentine. The intermediate, namely $Mg_3Si_2O_7$ (s), which is formed in Eq. (26) and defined as dehydroxylate I, is more reactive than the original (uncalcined) serpentine, thus displaying faster magnesium dissolution in acidic solutions. At temperatures above 800 °C, dehydroxylate I is converted into forsterite (Eq. (27)). The overall reaction involves the conversion of serpentine to forsterite at temperatures between 800 and 1000 °C, as shown in Eq. (28) [55].

$$Mg_3Si_2O_5(OH)_4(s) \rightarrow Mg_3Si_2O_7(s) + 2H_2O(g) \tag{26}$$

$$2Mg_3Si_2O_7(s) \rightarrow 3Mg_2SiO_4(s) + SiO_2(s) \tag{27}$$

$$2Mg_3Si_2O_5(OH)_4(s) \rightarrow 3Mg_2SiO_4(s) + SiO_2(s) + 4H_2O(g) \tag{28}$$

Aqueous serpentine carbonation reactivity can be enhanced by heat activation. Based on the study on the influence of heat treatment temperature on the extent of the carbonation reaction and weight loss from the serpentine, the optimum temperature range for heat treatment is 600–650 °C. The hydroxyl groups are removed by heating serpentine minerals to 600–650 °C, destabilizing the crystal lattice into a pseudo-amorphous structure [56].

The implementation of the current process is too expensive. Thus, understanding the characteristics and mechanisms of materials that enhance carbonation is crucial for further cost-reduction efforts.

High-temperature calcination, mainly performed at elevated temperatures higher than 873 K, is generally associated with expensive costs and intensive energy consumption. Compared with the high-temperature roasting process, the microwave heating process reduces the roasting time, but the equipment requirements are typically high, and its industrial utilization is also notably difficult [57,58].

3.4. Hybrid activation

Hybrid activation is performed using a combination of different methods to increase the effects of activation methods on tailings. Hybrid activation methods include mechanochemical, chemical–thermal, mechanical–thermal, and mechanochemical–thermal.

3.4.1. Mechanochemical activation

Indirect carbonation introduces one or more additional steps by extracting the reactive compound from the matrix of raw samples by chemical activation.

Olivine was milled with high energy intensity using a laboratory planetary mill, presenting a high Mg extraction potential of 0.01 M HCl solution at room temperature and pressure. The samples after the dry mill had the lowest measured specific surface areas ($< 4\ m^2/g$) with the highest rate constants. Mechanical activation was beneficial to the initial specific reaction rates by approximately three orders of magnitude for a sample dry milled for 60 min compared to an unactivated sample [59].

3.4.2. Mechanical–hydrothermal activation

When applied concurrently with heating, grinding substantially improves the activation process and reduces its severity. This approach is denoted as thermomechanical activation. Heating, grinding, and shocking serpentine minerals lead to their dehydroxylation, resulting in the amorphization of their structures [49].

When serpentine is ground into particle sizes below 125 µm and thermally activated at 610 °C, even in the absence of additives, the dissolution rate is two to three orders of magnitude faster than that of olivine [60].

3.4.3. Chemical–thermal activation

The process of producing pure magnesium compounds from serpentinite starts with acid leaching. However, serpentinite calcination before leaching not only facilitates fast magnesium dissolution but also notably mitigates issues related to the corrosion of the leaching equipment. This phenomenon is due to thermally activated serpentine, which allows the use of less aggressive leaching agents and/or at low leaching temperatures and pressures [61].

The process involves a solid–solid reaction between Mg-silicates and ammonium sulfates, resulting in the formation of reactive compounds, primarily $Mg(OH)_2$, which offers a remarkably higher conversion rate. The mechanical activation of the reactants is achieved through high-energy magneto ball milling with a precisely controlled energy input. The optimal milling energy input for enhancing the reaction kinetics of $MgSO_4$ extraction is approximately 27.6 $kJ \cdot g^{-1}$, leading to a reduction of approximately 34% in the activation energy required for $MgSO_4$ solid-state extraction [62].

Farhang *et al.* [63] investigated the precipitation of magnesium carbonate during single-step high-pressure, high-temperature carbonation of thermally activated serpentine in an aqueous bicarbonate solution. Subsequently, they investigated the dissolution rate of heat-activated serpentine (lizardite polymorph) across a wide range of pH, S/L ratio, and particle size at room temperature, which determined the effect of the variables on the dissolution kinetics of heat-activated lizardite [64].

Raschman *et al.* [61] examined the enhancement of serpentinite calcination on the magnesium dissolution rate in solutions with three distinct leaching agents. They found that the initial magnesium dissolution rate of calcined serpentinite was up to 30, 125, and 165 times higher than that of uncalcined serpentinite in solutions of hydrochloric acid, acetic acid, and ammonium chloride, respectively.

The dissolution of thermally activated serpentine (75% dehydroxylated) was investigated through direct flue gas mineralization under far-from-equilibrium flow-through operating conditions. Thermal activation enabled the dissolution of serpentine at mildly acidic conditions. Magnesium and silica were released upon dissolution [65].

3.4.4. Mechanochemical–thermal activation

Breuil *et al.* [66] determined the influence of particle size distribution on the dehydroxylation process and dissolution rates in aqueous mineral carbonation reactions by comparing ground and unground serpentine with thermally activated serpentine. Regarding Mg dissolution efficiencies, grinding samples before thermal activation leads to large Mg^{2+} extraction.

3.5. Newly developed processes

Pretreatment of feedstock, such as magnetic separation and sonication, was found to increase the reaction rate [51]. Cauterization with brine solution and biological enhancement have also been used for tailing waste residues to enhance the mineral carbonation rate.

A combination of acid digestion and electrolysis of olivine was used to produce $Mg(OH)_2$ in a fully recoverable system, which is highly reactive and capable of forming stable carbonates [67]. The synthesis of

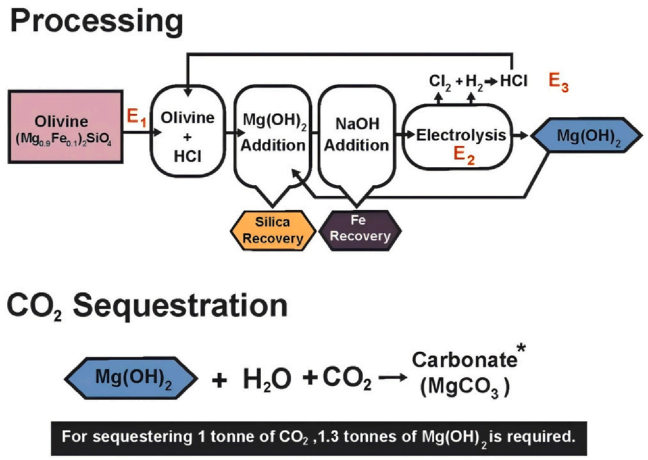

Fig. 6. Simplified mineral extraction and carbon sequestration [67].

$Mg(OH)_2$ uses olivine from ultramafic rocks and/or olivine-rich basalts as the raw material and source of magnesium, as shown in Fig. 6 (E stands for the energy requirements for each tone of $Mg(OH)_2$ produced. $E_1 = 0.32$ GJ, $E_2 = 6.6$ GJ, and $E_3 = -0.64$ GJ).

The detailed comparison table is presented as Table 1 to summarize and compare various aforementioned activation methods.

4. Tailings mineral carbonation process

Mineral carbon sequestration technology mimics the natural weathering process of rocks, where carbonic acid, formed from CO_2 dissolved in rainwater, reacts with mineral alkalinity to produce carbonate minerals [68,69]. This method can use various feedstocks, including natural minerals and industrial byproducts, such as steel and blast furnace slag, as well as waste rocks and mine tailings [70].

Mineral sequestration is particularly effective for carbon capture in areas where geological sequestration is impractical due to risks of underground leakage or the lack of suitable cavities. The products of mineral carbonation remain solid, ensuring the absence of CO_2 release after sequestration. As shown in Table 2, mineral carbon sequestration offers several benefits over other carbon storage methods [71–74]. This method provides a permanent solution for CO_2 disposal; a substantial amount of energy would be required to release CO_2 from carbonates, yielding environmentally benign products that may even have commercial value. Moreover, this method can be employed in regions where other techniques, such as geological carbon sequestration, are not viable. Therefore, carbon mineralization is emerging as a promising technology for the permanent and secure storage of CO_2 [75].

Alkaline mining and mineral processing wastes are highly suitable as feedstock for mineral carbonation because they have already been mined and processed to fine particle sizes during metal extraction [76,77]. Several types of deposits, mainly found in ultramafic rock, produce tailings that are ideal for mineral carbonation. These tailings include asbestos deposits, copper–nickel-PGE (platinum group elements) deposits hosted by dunite, serpentinite, and gabbronorite, as well as diamondiferous kimberlite pipes and podiform chromite deposits [78]. Ultramafic and mafic rocks, which constitute the majority of the Earth’s mantle, are rich in magnesium and iron, making them excellent candidates for mineral carbonation. Mining operations that extract valuable metals from these deposits generate substantial quantities of carbon-sequestering tailings, demonstrating remarkable potential for carbon capture and material repurposing.

Bauxite residue, or red mud, from alumina processing is another type of mineral processing waste suitable for mineral carbon sequestration. The carbonation of these residues, particularly asbestos tailings and red mud, not only sequesters CO_2 but also improves the properties of the wastes, increasing their storage safety or suitability for secondary uses [79–81].

In recent decades, many researchers have studied the mechanisms of mineral carbonation in processed tailings, focusing on laboratory-scale activation methods and factors influencing the rate of carbon mineralization. However, limited research has been directed toward the practical application of achieving industrial-scale carbon mineralization using mine tailings and the associated processing methods [82–85]. Despite successes with the carbonation of residues and promising process ideas for the carbonation of rock material, concepts for the large-scale implementation of natural mineral carbonation have not yet matured. However, recent developments and trends indicate rapid progress and increased support for promising concepts. CO_2 mineralization applications with a clear plan for the use of the resulting products appear most viable.

4.1. In-situ mineral carbonation

Mineral carbon sequestration can be performed either *in situ* (within geological formations) or *ex situ* (in a chemical processing facility) [73]. *In-situ* mineral carbonation processes require CO_2 injection into reservoirs and stockpiles for feedstock of mineral carbonation to initiate the reaction between minerals and CO_2 in geological formations. *In-situ* mineral carbonation offers the potential for long-term, safe CO_2 storage at a reasonable cost. However, optimizing this technology requires a multifaceted effort, including field-scale pilot studies to effectively characterize the rates of mineral carbonation reactions and the fate and consequences of injecting CO_2 into reactive waste rocks or tailings storages [86].

Baidya *et al.* [87] designed a system utilizing perforated pipes installed in a series of equal-sized tailings blocks to inject diesel exhaust from power plants operating in remote, off-grid mines for carbon sequestration purposes (Fig. 7). A financial analysis comprising various operating parameters under this concept was conducted. The analysis revealed that operational expenses, particularly energy costs, are influenced by the permeability of the tailings. For instances of low permeability, large injection pipes are necessary. The findings indicate that achieving viable operating costs for sequestering 1 t of carbon dioxide depends on appropriate pipe sizing and engineering. Additionally, the

Table 1
Comparison table of different activation methods of tailings (summarized from [43–67]).

Activation method	Advantages	Disadvantages	Application scope
Acid leaching	Can extract substantial amounts of calcium and magnesium, high efficiency	High cost, possible leakage	Only in laboratory
Ammonium salt pH swing	Permanently sequester the CO_2 into stable carbonates, high efficiency	High cost, possible leakage	Only in laboratory
Mechanical activation	Less environmental issues, relatively low cost, ease of use in industries	Relatively low efficiency, poor performance for some tailings	In laboratory, used in mines
Thermal activation	Less environmental issues, high efficiency	Energy intensive, only appropriate for serpentine group minerals, high cost	Only in laboratory
Hybrid activation	More flexible, high applicability	Complicated process	In laboratory, used in mines

Table 2
Summary and comparison of methodologies for CO_2 storage (modified from [71–74]).

CO_2 storage method	Advantages	Disadvantages	Cost (USD/t CO_2 stored)
Ocean sequestration	Large storage capacity, no monitoring	Temporary storage, potential harmful effects on aquatic microbes and biota	6–31 (pipeline),12–16 (tanker)
Geologic sequestration	Feasible on a large scale, substantial storage capacity known, extensive experience, low cost	Monitoring necessary, leakage possible	0.5–8.0 (storage), 0.1–0.3 (monitoring)
Biological sequestration	Less environmental issues, high potential	Monoculture plantations can imbalance the ecosystem, insufficient capacity, risk of disruption of the food chain	60–160
Mineral sequestration	Only known form of permanent storage, minerals required available in quantities capable, environmentally benign carbonation products, utilization of industrial wastes	Energy intensive, slow reaction kinetics, high cost	50–100

study estimates that maintaining a reasonable ratio (approximately 1%) between the decarbonized power and the required power for the carbon sequestration operation is crucial.

4.2. *Ex-situ mineral carbonation*

The *ex-situ* mineral carbonation processes involve rock mining and comminution of the mineral ore as pre-processing for carbonation. Various methods have been reported in carbonation studies for performing *ex-situ* CO_2 sequestration, which is classified as direct carbonation and indirect carbonation. Direct carbonation is the most direct method and comprises a single-process stage for carbonation. Direct carbonation is further classified into two categories: (1) gas–solid reactions, in which CO_2 gas directly reacts with solid minerals; (2) mineralization in aqueous solutions, where CO_2 is dissolved in water and reacts with minerals in solution [70,88].

The indirect mineral carbonation process utilizes multiple stages. First, the ions of Mg or Ca are extracted from the mineral or waste materials through the use of acids or other solvents and then converted to their respective oxides or hydroxides. Subsequently, these oxides or hydroxides are allowed to react with CO_2 to form carbonates. Pure carbonates can then be produced from indirect methods, given the impurity extraction before the carbonate precipitation stage. Most indirect processes deliver faster carbonation than direct methods [86,89–91]. A schematic of *ex-situ* mineral carbon sequestration showing the material fluxes is given in Fig. 8.

The primary challenge for *ex-situ* mineral carbonation is enhancing the reaction kinetics to develop an economically viable commercial process. Regardless of other manageable issues, such as managing the environmental impact of large-scale mining operations, considerable research is still needed. Despite substantial progress in increasing carbonation rates, further scientific efforts are also essential to increase their economic and technological feasibility. The main limitations of large-scale mineral carbonation deployment include the consumption of vast quantities of carbonation products, high process costs, and environmental concerns. Strategies for the disposal and reuse of carbonation products must be developed to mitigate these challenges. Additionally, detailed cost analyses and innovative, efficient methods are crucial for industrial feasibility. Reliable assessments of the environmental impacts associated with mining, transportation, processing, and disposal are also necessary due to increasing environmental concerns.

5. Technological approach for CO_2 mineralization sequestration through underground tailings backfill

Underground tailings backfill refers to the process where a mine prepares tailings slurry with a certain concentration, which is then transported through a pipeline system to fill underground stopes and/or areas designated for backfill. A certain amount of cementitious materials must be added to ensure the stability of the tailings backfill mass. Underground tailings backfill must generally be coordinated with

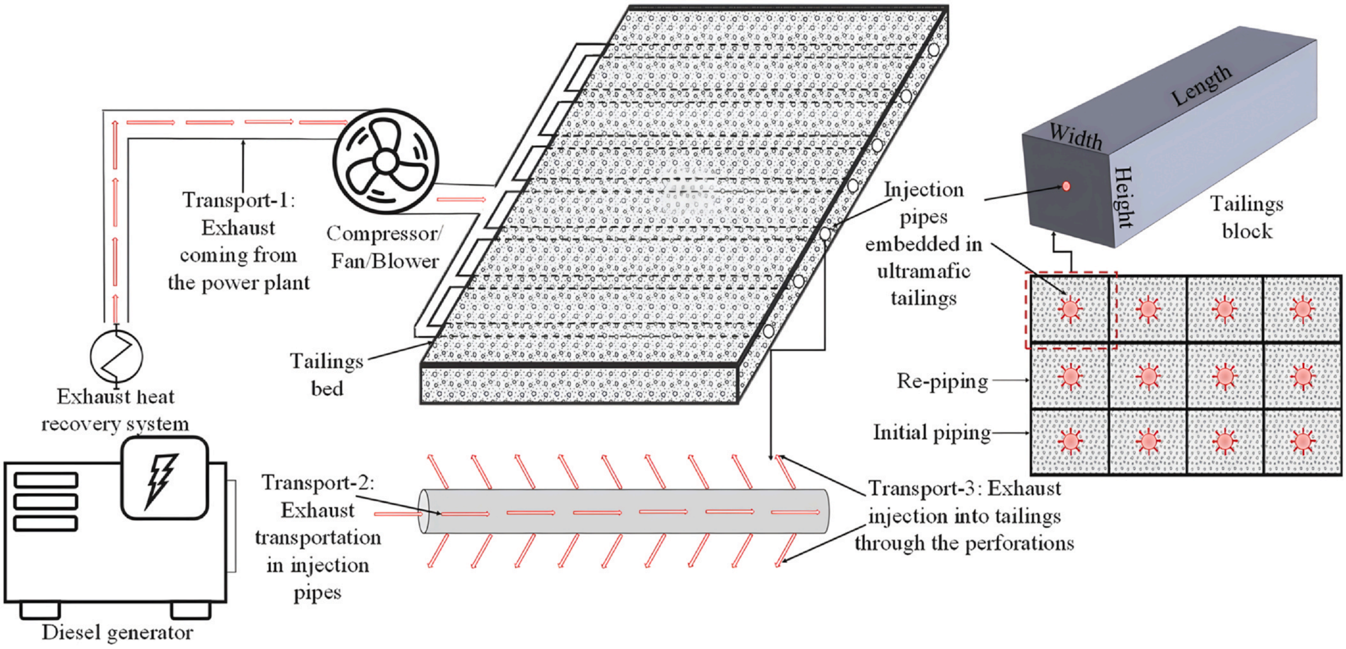

Fig. 7. Illustration representing the proposed carbon sequestration approach tailored for off-grid mining sites [87].

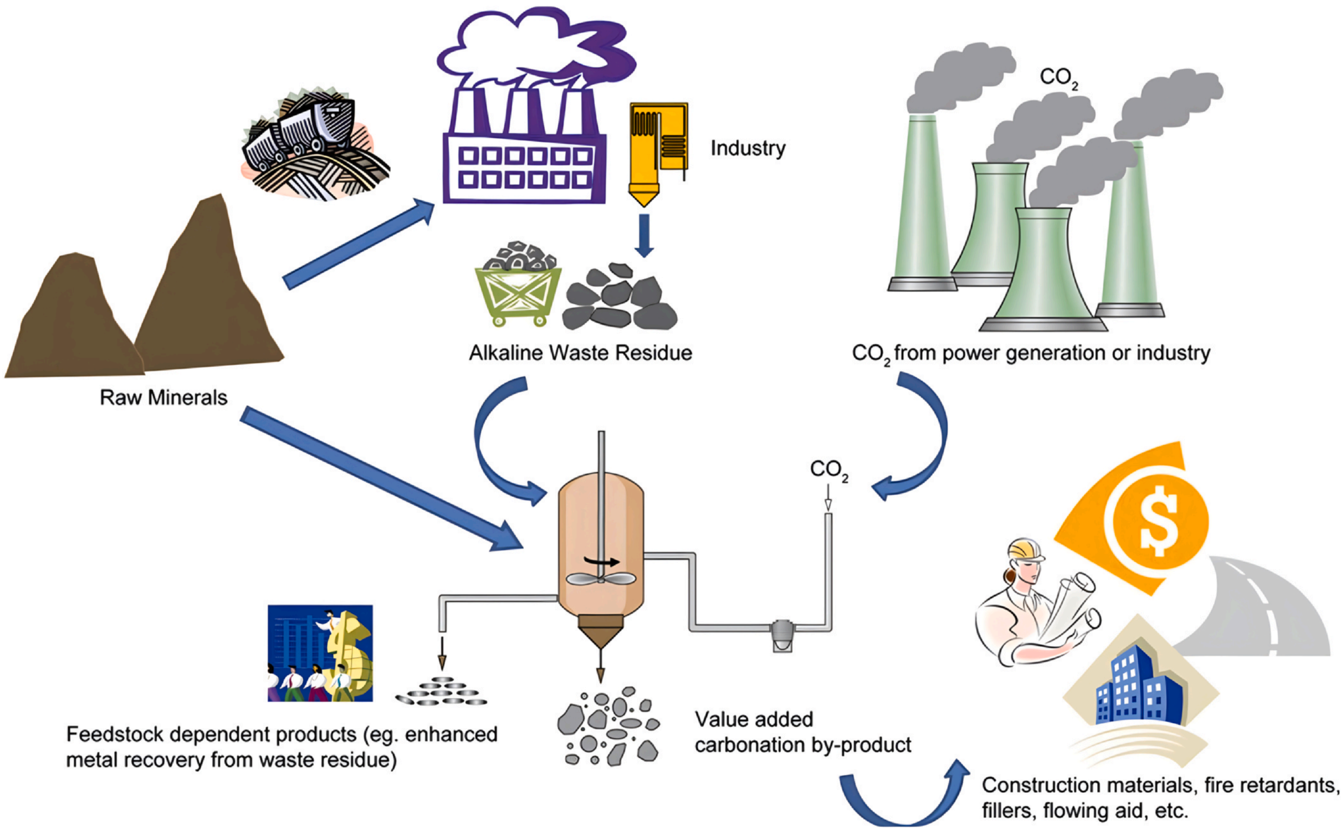

Fig. 8. Material fluxes associated with mineral carbonation of minerals and industrial residues. Reproduced with permission from Ref. [21]. Copyright (2012) Elsevier.

mining operations. Mines use the tailings backfill to dispose of the mined-out areas and/or to provide a platform for mining operations [92–94], thereby improving the recovery rate of underground ore. This platform not only improves the safety of mining operations and prevents surface disasters but also effectively disposes of surface solid waste, making it a safe, efficient, economical, and environmentally friendly mining method for metal mines. Based on the aforementioned advantages, researchers from the BGRIMM Technology Group have proposed the idea that using mined-out areas as the sites for CO_2 mineralization and sequestration, combined with the concept of filling these voids with tailings to solidify CO_2 (Fig. 9), can achieve large-scale disposal of tailings, ensure the proper filling and disposal of mined-out stopes, and address the goal of large-scale CO_2 sequestration. This ideal carbon sequestration method achieves multiple benefits. Some technical problems must be addressed to achieve the proposal.

5.1. Transportation and surface storage of CO_2

5.1.1. Pipeline transportation of CO_2

The capture of CO_2 typically occurs in areas with high carbon emissions, such as smelters or coal-fired power plants. The captured CO_2 must then be transported to a storage site. Considering the long distance and large volume of the gas, the earliest method involved injecting CO_2 into a liquid to form a medium similar to a carbonated solution for pipeline transportation [95]. Afterward, the transportation of carbon dioxide through pipelines is primarily conducted in three forms: gaseous, supercritical flow, and subcooled liquid transport [96–98]. Among these forms, liquid transport offers higher and more stable flow rates but is more sensitive to transport pressure and ambient temperature, increasing its suitability for long-distance transportation. By contrast, direct gaseous CO_2 transport has a relatively slower rate

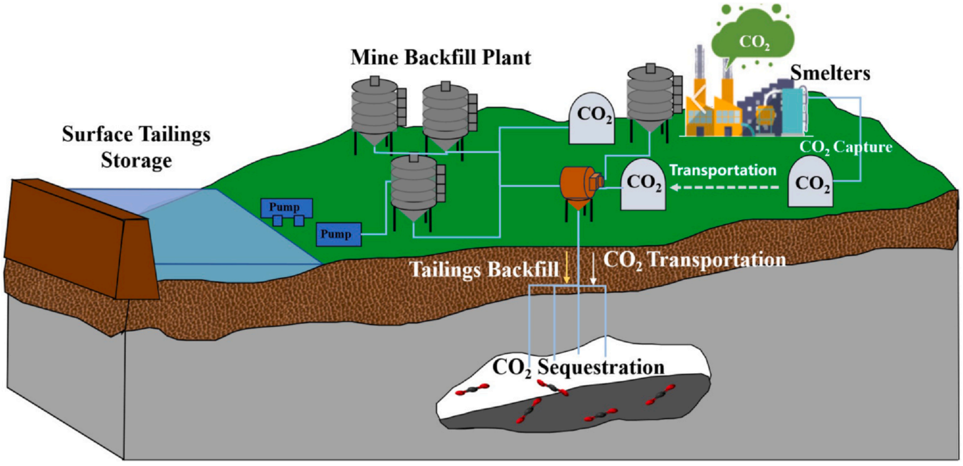

Fig. 9. Concept of CO_2 mineralization sequestration through underground tailings backfill.

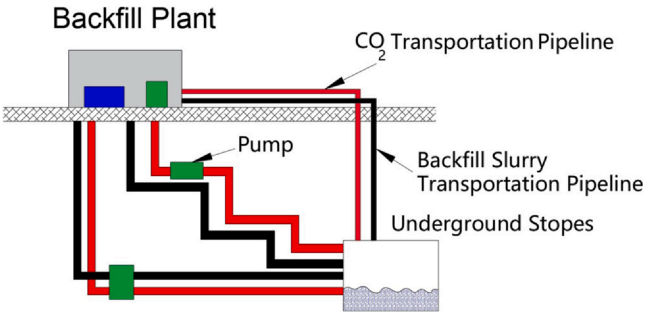

Fig. 10. Concept of CO_2 transportation with backfill slurries.

and requires larger pipeline diameters due to the greater gas volume. In actual transportation processes, comprehensively considering factors such as transport distance, efficiency, cost, and external environment is necessary to formulate a reasonable transport proposal. For CMST, mines are typically not the capture sites; however, the CO_2 gathered in some other place can be transported to surface storage facilities in liquid form through pipelines. CO_2 must react with tailings filling materials in the form of gas to achieve mineralization and storage purposes in the underground mined-out area. Thus, the transportation of CO_2 from the surface to the underground stopes can be conducted in gaseous form. The schematic of the concept is presented in Fig. 10.

5.1.2. Surface storage of CO_2

Carbon dioxide storage includes short-term storage and long-term sequestration. As aforementioned, long-term sequestration includes geological and ocean sequestration [5,6]. Short-term storage is mainly used in scenarios such as industrial utilization of carbon dioxide or sea–land combined transportation [99–101]. In terms of CMST, carbon dioxide must be transported in liquid form from the capture site, stored in surface facilities, converted to gaseous form, and then transported again to the underground stopes to create a carbon-rich curing environment for tailings filling slurry in underground mined-out areas. Therefore, the surface storage facility must contain independent storage units for gaseous and liquid phases of CO_2, as well as equipment for phase conversion (from liquid to gas). The process plan design uses a carbon dioxide storage tank as an intermediate storage facility. The commonly used storage tanks are currently either vertical or horizontal. Among them, horizontal tanks are easier to control and more stable but require extra space. By contrast, vertical tanks occupy less space [102,103]. Considering the limited space at the backfill plant, vertical intermediate storage tanks can be used. Additionally, a dedicated anti-liquefaction plant is necessary to gradually convert the transported liquid carbon dioxide into the gaseous phase through heating and depressurization [104,105] and then temporarily store it in the tanks (Fig. 11).

5.2. Proposal of CO_2 mineralization with tailings backfill

The general process of the carbon dioxide mineralization reaction includes the dissolution, carbonation, and mineralization reaction with target compounds, leading to the gradual precipitation of the products until the reaction is complete [106–108]. In this process, the dissolution of carbon dioxide typically requires the target compound to have suitable activity (the activation methods have been discussed in Chapter 3 of this paper). Additionally, this process is related to the concentration of carbon dioxide, ambient temperature, and the specific surface area of the mineralization materials [109–111]. Existing studies have shown that while tailings in the dams can react with environmental carbon dioxide to some extent, the reaction rate and volume are both relatively low [112,113]. This finding is mainly due to the low concentration in the environment, increasing the difficulty in the occurrence of passive mineralization reactions. Numerous practices have proven that the passive mineralization reaction rate on the surface of tailings can be improved by increasing the supply rate or raising the ambient concentration of carbon dioxide [114,115]. Moreover, the contact area between the carbon dioxide gas and tailings particles can be effectively increased by directly injecting carbon dioxide into the tailings deposit, extending the contact time, and enhancing the dissolution of carbon dioxide, thereby actively improving the mineralization reaction rate [116–118].

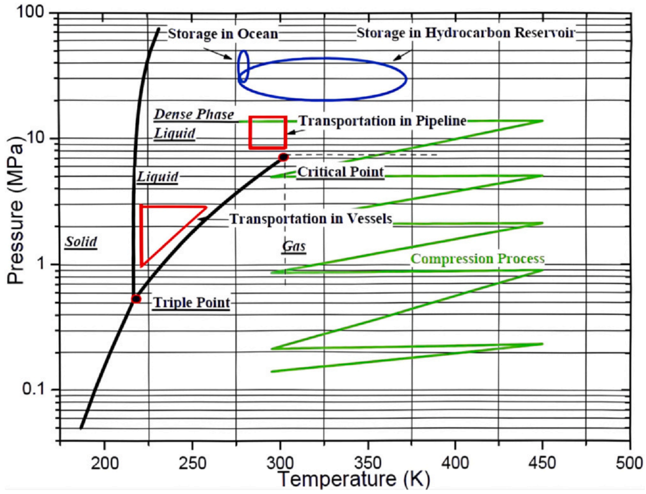

Fig. 11. CO_2 phase diagram and application to transportation and storage. Reproduced with permission from Ref. [104]. Copyright (2017) Elsevier.

The CMST can be promoted by utilizing carbon dioxide injection. However, some necessary parameters that must be studied for carbon injection into tailings backfill materials include injection pressure, rate, method, and carbon dioxide concentration [113–115,118]. The impact of carbon injection on the working performance of the mine backfill, such as tensile strength [119] or the capability of stope roof support [120], must also be considered. Mature process schemes for CMST are also currently lacking. Moreover, different carbon injection techniques must be adopted for different mining conditions. For instance, in metal mines with upward cut and fill mining, the mined-out areas are distributed in uniform layers. In such cases, similar to the injection method shown in Fig. 7, a central pipeline layout with uniform injection can be used. Meanwhile, for the mining method, such as sublevel open stoping with subsequent backfill mining method, meeting the operational requirements using uniformly straight pipelines for carbon dioxide injection is difficult due to the large size of the stopes [121–123] (in some circumstances, the possibility of stope heights can exceed 60 m). Under this condition, the filling process may last for several weeks or even months due to the large size of the stope. Therefore, CO_2 injection aims to ensure a carbon curing environment within the stope and allow the tailings backfill slurries to have sufficient long-term contact with CO_2 and undergo mineralization reactions, thereby achieving the goal of carbon dioxide mineralization and sequestration.

As shown in Fig. 12, BGRIMM proposed a concept of synergistic mineralization and CO_2 sequestration technology based on a pre-embedded branched CO_2 injection pipeline network for sublevel open stoping with a subsequent backfill mining method to solve the above technical challenges. In this concept, the branched pipeline network can achieve uniform multipoint CO_2 injection at different stope heights to accommodate the size of the stopes. Additionally, the pre-embedded pipes can facilitate the dewatering of backfilled stopes, achieving multiple benefits.

These technical proposals still require further investigation to increase their applicability in industrial trials. For example, dedicated studies on grouting and reinforcing the stope walls to prevent CO_2 from escaping are currently lacking. Additionally, the change in the working performance of the tailings backfill material after the mineralization

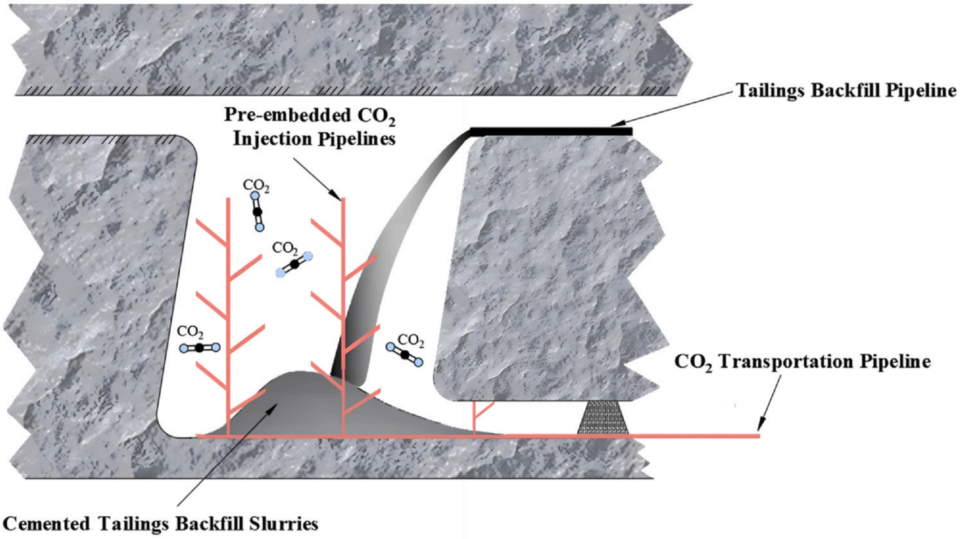

Fig. 12. Proposal for CMST in sublevel open stoping with the subsequent backfill mining method.

reaction with CO_2 is a topic requiring in-depth studies. These areas all merit future investigations.

6. Conclusions and future perspectives

This study systematically summarizes the research work on tailings CO_2 sequestration technology. An in-depth analysis of the progress and development trends in tailings CO_2 mineralization sequestration technology is provided from four aspects: the reaction mechanism of tailings mineralization with CO_2, methods to accelerate CO_2 mineralization, the process of tailings mineral carbonation with CO_2, and the synergistic CO_2 sequestration method with tailings backfill. The following conclusions are drawn.

(1) Metal mine tailings are typically subjected to multistage crushing and grinding, resulting in relatively fine particle sizes, and may contain a certain amount of metal silicate minerals, making them potential candidates for carbon dioxide mineralization and sequestration. In particular, ultrabasic rock tailings, which contain calcium and magnesium oxides, silicates, or hydroxides, have superior carbon mineralization potential. Current studies have demonstrated that tailings from a certain copper–nickel mine can mineralize and sequester carbon dioxide, mainly due to the chemical properties of olivine minerals in the tailings, which can dissolve and undergo exchange reactions with carbon dioxide, producing carbonate precipitates. However, the reaction rate between tailings and carbon dioxide is slow in natural environments. Therefore, pretreatment to activate the reactivity of the tailings is necessary to accelerate the carbon mineralization reaction rate and meet the demand for large-scale carbon dioxide mineralization and sequestration.

(2) For CMST, the pH value from the dissolved minerals in the tailings and the fineness of the tailings particles substantially affect the reaction rate. Currently, chemical and physical activation methods to enhance the reactivity of tailings in carbon mineralization, such as further grinding of the tailings or using acid leaching to increase the specific surface area and improve the surface dissolution efficiency of silicate minerals, have been successfully applied in laboratory settings. However, for large-scale utilization, the challenge of balancing the cost with the effectiveness of activation remains unresolved, limiting the industrial application of CMST. Establishing an economically feasible and standardized process for reactivity activation of tailings at the laboratory level will be the next requirement to advance the industrial application of CMST.

(3) Two essential technical pathways for the large-scale and efficient sequestration of CO_2 by tailings involve the mineralization of CO_2 through natural weathering of mine tailings in tailings ponds and the synergistic CO_2 sequestration with the tailings backfill process in mines. Enhancing tailings weathering is crucial for the surface stockpiled tailings in mineral sequestration of CO_2, and core technologies for the synergistic and efficient *in-situ* sequestration of CO_2 through the mine tailings lie in the establishment of an optimized CO_2 injection network and a mineralization reaction environment in the underground mining areas.

(4) One of the key technological pathways to achieving CMST lies in using mine backfill while synergistically mineralizing carbon dioxide. Critical technical challenges must still be addressed for this process. First, the development of low-cost carbon dioxide phase transition storage technology at mine backfill plants is fundamental to combining carbon dioxide sequestration with backfilling processes. Second, the technology for the co-transport of tailings slurry and carbon dioxide is crucial for achieving the mixing and mineralization of tailings and carbon dioxide in underground stopes. Additionally, methods and processes for carbon dioxide injection into *in-situ* mined-out areas must still be developed. Thus, creating a carbon-rich curing environment while maintaining the strength of the *in-situ* cemented backfill is necessary. In the future, additional investigations on these challenges and relevant theoretical studies applicable to backfill mining practices are also necessary to drive industrial-scale applications.

CRediT authorship contribution statement

Lijie Guo: Conceptualization, Writing – review & editing. **Xiaopeng Peng**: Supervision, Writing – original draft. **Qianqian Wang**: Writing – review & editing. **Yue Zhao**: Writing – original draft. **Linglin Xu**: Writing – original draft. **Shan Wu**: Supervision, Project administration.

Declaration of Competing Interest

Lijie Guo is an editorial board member for this journal and was not involved in the editorial review or the decision to publish this article. 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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