

Review

High-value recycling of industrial solid wastes using mineral carbonation: A review of recent developments

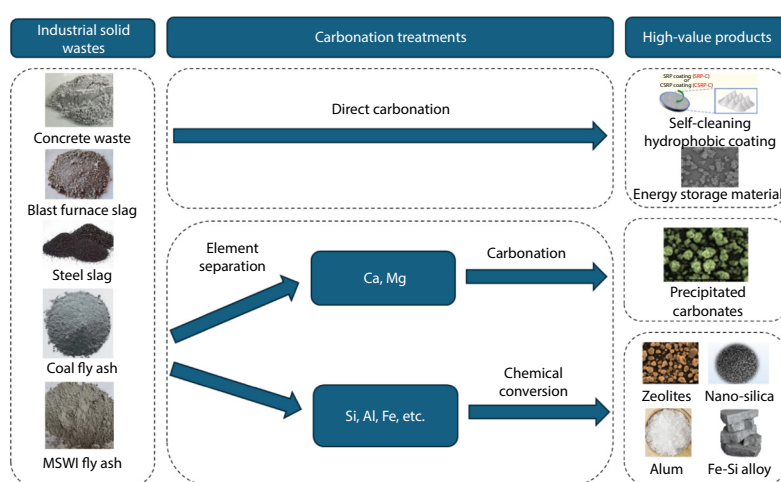
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Abstract: Mineral carbonation of certain types of industrial wastes allows CO₂ sequestration and recycling of the original waste as a new material. These processes require energy input and normally produce relatively low-value products such as aggregates, supplementary cementitious materials, and fillers. This paper reviews recent progress in producing higher-value products by mineral carbonation of waste concrete, blast furnace slag, steel slag, coal fly ash, and municipal solid waste incinerator fly ash. Zeolite and nanosized silica are potential higher-value products, while other materials such as superhydrophobic coatings, glass-ceramics, energy storage materials, and CaCO₃ ceramics have also been produced. The production of

higher-value products transforms the economic viability of mineral carbonation. Further work is required to comprehensively evaluate specific applications of carbonation and determine the overall CO₂ balance.

Keywords: mineral carbonation, waste recycling, low-carbon technology, high-value products

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1. Introduction

Mineral carbonation is used to recycle certain types of inorganic industrial solid wastes. In these processes, CO₂ reacts with Ca-bearing components in the waste to form stable carbonate minerals, typically CaCO₃^[1]. Direct mineral carbonation of granular wastes can produce aggregate particles that are bound together by CaCO₃ and have low water absorption and improved compressive strength^[2,3]. Indirect carbonation

involves leaching Ca from the waste prior to carbonation of the leachate. This can produce high-purity CaCO₃ that can be used as a filler in cement that refines particle packing and promotes hydration^[4].

CaCO₃ exists in three crystalline forms under normal conditions, namely vaterite, aragonite, and calcite^[4]. The mineral carbonation conditions determine the type of CaCO₃ formed. In the presence of Mg²⁺ and temperatures above 60 °C, aragonite is produced. This has been used to reinforce cement

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paste^[5,6]. Aragonite is also reported to act as a rheology regulator, as it increases the static yield stress and reduces the dynamic yield stress of cement pastes^[7].

Vaterite is produced in the presence of NH_4^+ when the pH is less than 10^[8]. It is metastable and forms spherical particles with high porosity and solubility^[9]. It is the major component in calcium carbonate cements, which develop strength through the polymorph transformation from vaterite to aragonite^[10,11]. CaCO_3 cement pastes have low strength (<30 MPa) and high porosity (>40%) compared to Portland cement paste^[10].

Mineral carbonation of industrial solid wastes can also generate silica-alumina gel^[12]. This is amorphous and highly pozzolanic, and it can be used as a substitute for up to 40% of cement clinker^[4]. Nevertheless, the large specific surface area and water absorption of this gel reduce the workability of concrete and lead to an increased superplasticiser demand^[13,14].

Although mineral carbonation can permanently sequester CO_2 and produce useful products from industrial solid wastes, the commercial viability is a major barrier to large-scale application^[12]. Mineral carbonation normally produces low-value construction products such as carbonated aggregates and fillers^[15], as the revenue from these may not offset processing costs^[12]. Recently, emerging technologies have investigated the production of zeolites (\$540–\$1012 per tonne), nanosized silica (\$500–\$1000 per tonne), and other products with higher value, and these offer great potential to improve the economic viability of mineral carbonation processes^[16,17].

There have been numerous reviews of mineral carbonation of industrial solid wastes, but none have specifically addressed opportunities for producing higher-value products. In this work, recently developed mineral carbonation processes of industrial solid wastes that produce higher-value products are reviewed. Various industrial solid wastes are suitable for mineral carbonation. The review covers the types of high-value products that can be produced, the mechanisms involved, the controlling factors for optimum product production, and the application of the products addressed. Scale-up issues are also discussed.

2. Suitable industrial solid wastes for carbonation

Major industrial solid wastes suitable for carbonation are concrete waste, blast furnace slag (BFS), steel slag (SS), coal fly ash (FA), and municipal solid waste incinerator fly ash (MSWIFA).

Concrete waste is generated from the demolition of end-of-life concrete structures and the washing of concrete mixing facilities. This consists of waste concrete aggregates (WCA), waste concrete powder (WCP), and concrete slurry waste (CSW)^[4,18,19]. Billions of tonnes of waste concrete are generated globally each year. WCA contains primary aggregate, sand, and small amounts of cement paste. It is normally downcycled as a road base material^[4]. WCP contains more cement paste with more gel porosity than WCA. Due to the high porosity and low pozzolanic reactivity of WCP, the

workability and strength of the WCP-blended cement paste are reduced compared to pure CEM I paste^[4]. CSW is produced when excess concrete is produced or when concrete fails to meet specific construction standards^[19]. After washing the excess fresh concrete, clean sand and aggregates are recovered. The remaining wastewater containing cement paste is CSW. This is normally dewatered and sent to landfill^[19].

BFS is a by-product from the production of crude iron. Approximately 0.3 to 1.0 tonnes of BFS is produced per tonne of crude iron^[20]. BFS contains 30%–50% CaO, 5%–15% MgO, and 7%–18% Al_2O_3 ^[12]. Some types of BFS contain ~10%–25% TiO_2 , which results from the use of vanadium titanomagnetite ores^[21]. BFS is quenched in water to generate small granules with a high amorphous content^[20,22]. Ground granulated blast furnace slag (GGBFS) is extensively used as a supplementary cementitious material (SCM) to reduce cement use. However, the availability of GGBFS is ~400 million tonnes per year, and this cannot satisfy the total demand^[12,20,23]. The low alkalinity and stable structure make the carbonation rate of granulated BFS low^[24].

SS is generated during steel refining using a basic-oxygen furnace or electric arc furnace. Globally, around 240 million tonnes of steel slags are produced every year^[12], with chemical compositions of 23%–60% CaO, up to 10% MgO, up to 38% Fe_2O_3 , 3%–4% Al_2O_3 , and 7%–20% SiO_2 ^[25]. The major mineral phases are C_2S , C_3S , C_2F , RO phase (solid solution of MgO, FeO, and MnO), free CaO, and MgO^[26]. Due to the amorphous SiO_2 and limited C_3S contents, steel slags have poor hydraulic properties. Consequently, when used as an SCM, it does not make a significant contribution to compressive strength development under standard curing conditions^[27]. The high free CaO and MgO content leads to excessive expansion, cracking, and volume instability over time^[27]. Moreover, steel slags often contain heavy metals such as V and Cr, which may lead to health and environmental concerns^[12]. Alternatively, the presence of above-mentioned mineral phases in steel slags results in higher alkalinity, thereby enhancing reactivity in mineral carbonation compared to BFS.

FA is generated from the combustion of coal in coal-fired power plants at 0.75–1 billion tonnes per year^[28]. The major components of FA are SiO_2 , Al_2O_3 , and Fe_2O_3 present in mulite, quartz, and magnetite phases^[12]. The content of CaO varies with the chemical composition of the coal. FA may also contain heavy metal elements such as Pb, Cd, As, and Hg that can leach into groundwater from landfills^[12]. FA is widely used as a cement replacement, but low pozzolanic reactivity and unstable quality restrict its effective application as an SCM^[23]. Therefore, finding novel approaches to turn FA into value-added products is urgently needed.

MSWIFA is the solid residue formed from the incineration and air pollution control in the waste-to-energy process. The Ca, Si, Al, Cl, and LOI contents of MSWIFA are 12%–24%, 4%–8%, 2%–8%, 3%–12% and 11%–35%, respectively. The major mineral phases of MSWIFA are portlandite, calcite, quartz, and chloride salts (NaCl, KCl, and CaClOH)^[29]. It is usu-

ally considered hazardous because it contains heavy metal elements such as Pb, Cd, Hg, and Mo and toxic compounds such as dioxins and furans^[29]. Therefore, MSWIFA is mainly stabilised using cement and landfilled, which leads to excess land consumption and soil pollution risks^[30].

Generally, these industrial solid wastes have small particle sizes and contain considerable levels of alkalinity, which makes them suitable for mineral carbonation. Using industrial solid wastes in mineral carbonation avoids the extra cost of mining and grinding natural silicates. To develop an efficient and cost-effective mineral carbonation process of industrial solid wastes, the carbonation degree needs to be promoted, the value of the carbonation products should be improved, and the environmental risk should be minimised.

3. Carbonation technologies developed to produce high-value products from waste materials

3.1. Concrete waste

WCA mainly contains the natural aggregates. The major component of natural aggregates is quartz, which is chemically inert and difficult to transform into high-value-added products by carbonation. CO₂ can react with the attached cement paste, and the microcracks and micropores of WCA are filled^[4]. Therefore, carbonated WCA is mainly used as recycled aggregates in concrete, which is a low-value application. In contrast, CSW and WCP contain higher amounts of cement paste and are more reactive, which presents an opportunity for chemical conversion into high-value-added products.

3.1.1. Synthesis of phosphorus adsorbent

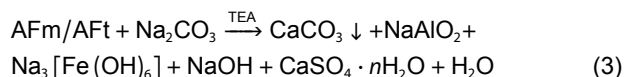
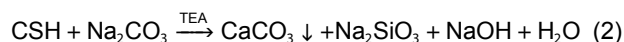
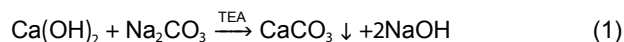
Iizuka et al. proposed an indirect carbonation process of CSW using water as the Ca extractant (Fig. 1). In the first step, CSW was mixed with water for 1 h to generate a Ca(OH)₂ solution. Large water/CSW ratios (5–15) were used to avoid hardening of the slurry and improve Ca extraction^[31]. In the second

step, flue gas containing 6%–13% CO₂ was introduced to the Ca(OH)₂ solution to precipitate 99% pure CaCO₃^[31]. The solid residue generated from the Ca extraction step was used to absorb phosphorus in the form of hydroxyapatite [Ca₁₀(PO₄)₆(OH)₂]. The highest phosphorus absorption capacity was 49.6 mg g⁻¹ with the residue formed at a water/CSW ratio of 10 and 105 °C drying^[19]. This was probably because higher water increased the Ca(OH)₂ consumption through carbonation, and drying accelerated the dissolution rate of Ca(OH)₂. This process did not use expensive chemical extractants, and the wastewater after carbonation can be reused in a new extraction step. However, only 30% of the Ca in CSW was extracted, and the phosphorus absorbent contained significant inert impurities such as CaCO₃ and SiO₂, which limited the CO₂ sequestration and phosphorus removal potential of CSW^[31].

3.1.2. Synthesis of nanosized silica-alumina gel

Alkali dissolution and carbonation of WCP produce nanosized silica-alumina gel. This process includes extracting silica and alumina from the WCP using Na₂CO₃ solution and carbonating the resulting Si-rich solution to form silica gel^[33,34].

In the first step, cement hydration products (portlandite, CSH, AFm, and Aft) react with Na₂CO₃ in the presence of triethanolamine (TEA), forming CaCO₃, NaOH, Na₂SiO₃, and NaAlO₂, as shown in Equations (1)–(3)^[34]. Because Ca²⁺ dissolution is unfavoured under alkaline conditions, TEA is used as a chelating agent to promote Ca²⁺ dissolution.



The resulting filtrate is rich in NaOH, Na₂SiO₃, and

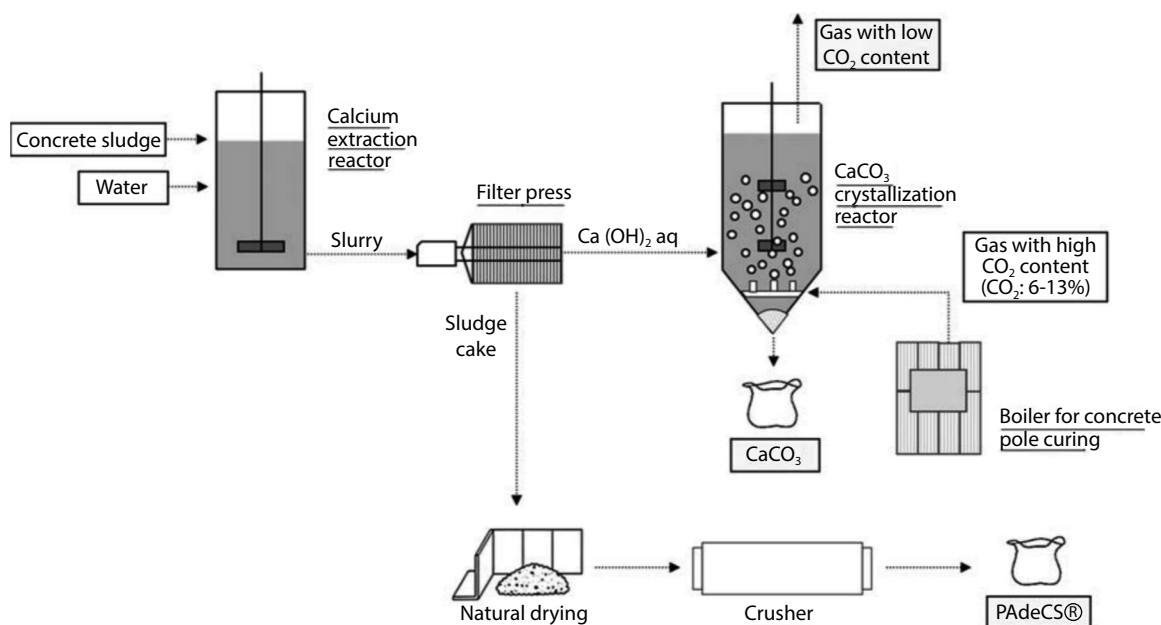
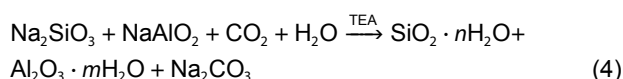


Fig. 1. Indirect mineral carbonation of CSW with the production of a phosphorus adsorbent (PAdeCS®). Reproduced with permission from Ref. [32].

NaAlO₂. In the second step, CO₂ is bubbled to this filtrate to reduce the pH, precipitate the silica-alumina gel, and regenerate Na₂CO₃, as shown in Equation (4).



The alkali extraction and carbonation processes are influenced by the particle size of WCP, the Na₂CO₃ concentration, and the extraction time. Using WCP smaller than 150 μm exposed more surface to the extractants and improved the amount of Ca-rich residue and silica-alumina gel by 6% and 260%, respectively^[33]. Increasing the extraction time from 1 h to 24 h did not change the amount of Ca-rich residue but increased the amount of silica-alumina gel by 50.6%^[33]. Increasing the concentration of Na₂CO₃ from 1% to 10% increased the amounts of Ca-rich residue and silica-alumina gel. A further increase resulted in excess alkalinity and a prolonged time for carbonation, which reduced the efficiency of this process^[33]. The carbonation process is additionally influenced by the CO₂ concentration. A CO₂ concentration higher than 50% accelerated the drop in pH, hence the precipitation of silica-alumina gel finished within 1.5 h^[33]. The final solution after carbonation contained regenerated Na₂CO₃ and NaHCO₃. After raising the pH to around 12 using a 30% NaOH solution, it was suitable to be used in a new leaching-carbonation cycle^[33]. The main product was an amorphous Na-bearing silica-alumina gel with 71.4% SiO₂, 10.1% Al₂O₃, and 11.3% Na₂O^[33,34].

To purify the nano-silica, a modified process was employed. WCP was first carbonated in water, forming CaCO₃ and silica-alumina gel; then, silica was extracted from the solid residue by NaOH; then, the resulting solution was carbonated to precipitate the Na-bearing silica-alumina gel; and

finally, the gel was washed by a 0.5 mol L⁻¹ HCl solution to remove the Na and Al impurities^[35]. The product was 98.8% pure amorphous silica as agglomerates from less than 10 nm nanoparticles (Fig. 2). It was mesoporous with a large specific surface area (662.47 m² g⁻¹) and a narrow pore size distribution^[35]. This material can be used in catalyst, adsorption, sensor, insulation, and as an ultra-fine additive to generate high-performance concrete^[35,36].

3.1.3. Synthesis of superhydrophobic self-cleaning concrete coating

A combined treatment of direct carbonation and silane modification of WCP can be used to generate a carbon-negative superhydrophobic self-cleaning concrete coating^[37]. In this process, WCP was mixed with deionised water at a liquid-to-solid ratio of 20 mL g⁻¹ and reacted with pure CO₂ for 2 h. Then, the carbonated WCP was reacted with octyltriethoxysilane, deionised water, and anhydrous ethanol at a ratio of 5:2:6:8 for 1 h. The resulting carbonated and silane-modified recycled powder (CSRP) coating was sprayed onto the fresh cement paste and cured for 28 d^[37].

The mechanism of this treatment is presented in Fig. 3. The calcite on the surface of carbonated WCP was hydrolysed, forming Ca-OH groups and releasing CO₃²⁻. The surface Ca-OH groups condensed with the Si-OH groups of the silanol from the hydrolysis of silane, forming Ca-O-Si groups and binding the silane to the carbonated WCP^[37]. The alkyl groups on the rim of CSRP provided superior hydrophobicity. The hardened cement paste with CSRP coating had a contact angle and sliding angle of (160.1±0.8)° and less than 5°, respectively^[37]. The CSRP coating was mechanically strong and chemically stable due to the similarity and strong bonding to the concrete matrix. It also improved the self-cleaning property and weathering resistance of concrete^[37].

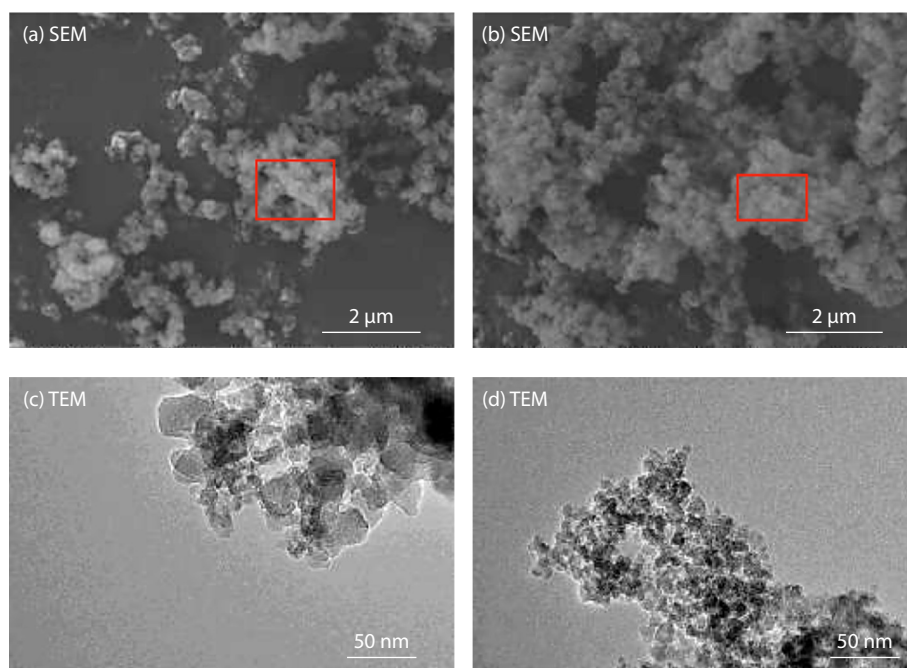


Fig. 2. SEM and TEM images of the silica-alumina gel from the alkaline dissolution and carbonation process. Reproduced with permission from Ref. [35].

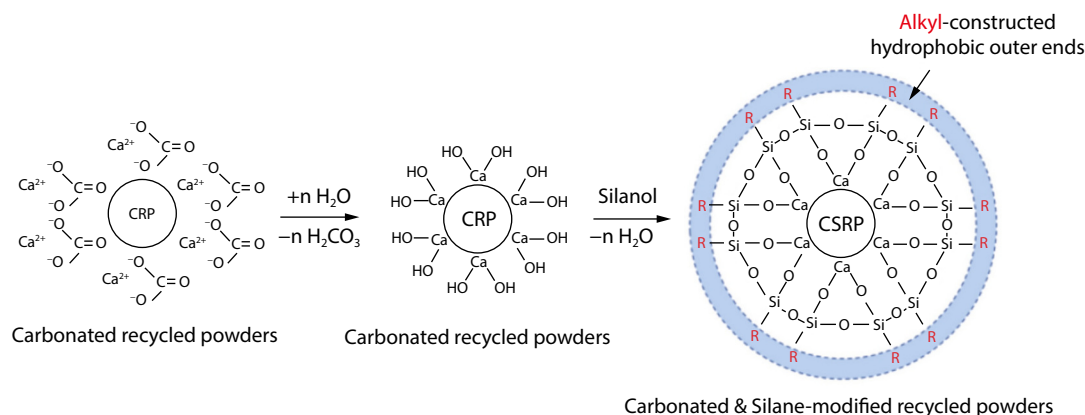


Fig. 3. Mechanism of developing self-cleaning hydrophobic coating from carbonation and silane modification of WCP. Reproduced with permission from Ref. [37].

3.1.4. Synthesis of zeolite

Zeolites are crystalline aluminosilicate minerals which are built from corner-sharing $[\text{SiO}_4]$ and $[\text{AlO}_4]$ tetrahedrons. The structure of zeolite contains a high level of porosity and connected channels, making it suitable for high-value applications such as contaminant adsorption, ion exchange, and catalysis[38]. Concrete waste contains SiO_2 and Al_2O_3 , which are fundamental precursors of zeolite, but its high CaO content inhibits the formation of zeolite[39,40]. The indirect carbonation of concrete waste includes acid or alkali leaching and carbonation[4,33,34,41]. The acid leaching of concrete waste dissolves the Ca-bearing components and produces an amorphous Si- and Al-rich residue, which is potentially a feedstock for zeolite. The solid residue of WCP leached by HCl was reacted with NaOH solution in a hydrothermal process. Under conditions of an NaOH concentration of $>2 \text{ mol L}^{-1}$, a temperature $>160^\circ\text{C}$, and a reaction time $>12 \text{ h}$, analcime-type zeolite with polyhedral morphology was synthesised. This was effective in absorbing methylene blue from its water solution[39].

3.1.5. Summary

The state-of-the-art carbonation technologies of concrete waste to produce high-value products include synthesising phosphorus adsorbent, superhydrophobic self-cleaning concrete coating, nanosized silica-alumina gel, and zeolite, as summarised in Table 1. Producing phosphorus adsorbent and superhydrophobic self-cleaning concrete coating uses direct carbonation, which is a simple process. Producing nanosized silica-alumina gel and zeolite involves indirect carbonation, where concrete waste is pre-treated by acid or alkali, making

the process more complicated and increasing the cost of chemicals. Although limited data are available in the literature, producing high-value-added products from concrete waste by indirect carbonation presents higher CO_2 sequestration potential compared to direct carbonation. Nevertheless, the production efficiency through indirect carbonation is lower because of the chemical composition of cement paste (high content of CaO , low content of SiO_2 and Al_2O_3) and the loss of materials during multiple reaction stages.

3.2. Blast furnace slag

Due to the slow carbonation kinetics of BFS, indirect carbonation is preferred to improve the carbonation efficiency. Numerous acids and acidic salts including HCl , CH_3COOH , NH_4Cl , NH_4NO_3 , $\text{CH}_3\text{COONH}_4$, and $(\text{NH}_4)_2\text{SO}_4$ have been used in the indirect carbonation of BFS[42–44]. The Ca content in BFS is activated and carbonated to form CaCO_3 at an elevated rate. However, the recovery of other elements in BFS, such as Si, Al, Mg, and Fe, is less investigated in these works. Besides, these extractants are usually not recyclable, increasing the cost and wastewater production of this process. The leaching residue is Si- and Al-rich and forms potential feedstocks for value-added materials, including silica-based nanomaterials, aluminium-based materials, and zeolites. Some types of slag contain other valuable metals such as Ti and V, which can also be recovered in the indirect carbonation process.

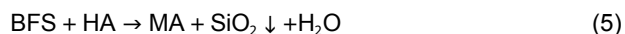
3.2.1. Synthesis of silica-based nanomaterials

Several studies have extracted nano-silica from BFS. Because silica is less soluble in acidic solutions and precipitates as polysilicic acid, while the metal elements are dissolved form-

Table 1. Summary of carbonation technologies of concrete waste towards high-value products.

Product	Production efficiency	CO_2 sequestration potential	Advantages	Disadvantages
Phosphorus adsorbent[31,32]	48.9%	0.39 kg t^{-1}	Simple operation, no chemical additives	Low efficiency
Nanosized silica-alumina gel[35,36]	33.6%	270 kg t^{-1}	High CO_2 sequestration potential	Low yield, long process
Superhydrophobic self-cleaning concrete coating[37]	-	-	Simple operation, carbon negative	Unclear long-term durability
Zeolite[39]	-	-	Quartz can be transformed	Complicated process, large chemical consumption

ing a salt solution, as shown in Equation (5).



where M is the metal elements (Ca, Mg, Al, Fe, etc.) and A is the anion of the acid.

Bang et al.^[45] used aqua regia to dissolve the metal elements in BFS and synthesise nano-silica. Under an aqua regia concentration of >20% and a liquid/solid ratio of $\leq 10 \text{ L kg}^{-1}$, pure amorphous silica with trace amounts of impurities was produced after 120 min of leaching. This silica was agglomerated from around 20-nm-sized nanograins with a specific surface area of $690\text{--}750 \text{ m}^2 \text{ g}^{-1}$ and was suitable for applications in semiconductors and solar cells^[45]. In a similar study^[46], HNO_3 was used to leach BFS to extract pure silica. The yield of silica was affected by acid concentration, temperature, acid/BFS ratio, and time. Acid concentration up to 50% promoted the dissolution of SiO_2 , and temperature up to 120°C promoted the hydrolysis and precipitation of SiO_2 , hence increasing the yield of silica. However, a further increase in these parameters decreased the yield of silica because the mixture becomes more viscous, and the diffusion rate of reactants is slower. The yield of silica increased with an acid/BFS ratio up to 2.3 and a reaction time of 90 min and plateaued afterwards, because the acid was in excess, and the precipitated silica formed a protective layer on the unreacted raw material^[46]. The acid-precipitated silica was then converted to spherical nano-silica particles of around 100 nm by treating with a surface modifier, cetyltrimethylammonium bromide (CTAB) (Fig. 4). The resulting nano-silica had an increased specific surface area from 304 to $1506 \text{ m}^2 \text{ g}^{-1}$ and uniformly distributed mesopores with an average pore size of 2.5 nm^[46].

Instead of using non-recyclable strong acids, Chu et al.^[47] developed an indirect carbonation process to separately recover the Ca, Mg, Al, and Si in BFS using a recyclable solvent, $(\text{NH}_4)_2\text{SO}_4$. The process is sketched in Fig. 5. BFS was pro-

cessed by (i) leaching with NH_4HSO_4 formed by stoichiometrically mixing H_2SO_4 and $(\text{NH}_4)_2\text{SO}_4$; (ii) cryogenic cooling of the leachate under $0\text{--}30^\circ\text{C}$ to crystallise $\text{NH}_4\text{Al}(\text{SO}_4)_2 \cdot 12\text{H}_2\text{O}$; (iii) heating the Al-depleted leachate at $70\text{--}100^\circ\text{C}$ to precipitate the silica gel; (iv) carbonation of the Mg^{2+} -rich solution using excess NH_4HCO_3 ; and (v) carbonation of the CaSO_4 -rich solid residue generated in step (i).

The pH of the Al-depleted leachate was around 1.7, at which the dissolved silica was stable. Increasing the pH of the leachate led to co-precipitation of silica with Fe and Al, so temperature was varied to selectively precipitate the silica gel. The gelation ratio increased from around 77.5% at 70°C to 98.1% at 100°C . The silica gel contained impurities such as adsorbed $(\text{NH}_4)_2\text{SO}_4$ and hydrolysed TiO_2 , which were removed by water washing and H_2O_2 and $\text{NH}_3 \cdot \text{H}_2\text{O}$ leaching, respectively. The final product was 97.7% pure silica gel^[47].

Although highly pure silica gel was successfully synthesised, the complicated purification process increased the cost and hindered the scale-up application. In a follow-up study, Liu et al.^[48] calcined the unpurified BFS silica with $\text{LiOH} \cdot \text{H}_2\text{O}$ or Li_2CO_3 at 800°C for 5 h to synthesise Li_4SiO_4 as a high-temperature CO_2 absorbent. The properties and CO_2 absorption performance of BFS silica-derived Li_4SiO_4 were compared to those synthesised from commercial quartz powder and Na_2SiO_3 . Because of the amorphous nature and finer size of the silica gel, the BFS silica- and Na_2SiO_3 -derived Li_4SiO_4 were purer. Using BFS silica and Li_2CO_3 resulted in the largest specific surface area and the highest CO_2 absorption rate among all products. The Li_4SiO_4 absorbent derived from BFS silica and Li_2CO_3 showed a high CO_2 absorption capacity (329 kg CO_2 per tonne of adsorbent) and good cyclability. This was because the BFS silica contains Ti^{4+} and Al^{3+} ions, which exchanged with the Li^+ in the crystal lattice of Li_4SiO_4 (Fig. 6). The free Li^+ formed more surface Li_2CO_3 with CO_2 , hence the CO_2 absorption was enhanced. Besides, they functioned as spacers in the crystal lattice, which retained the mesoporous structure, improved ion diffusion, and prevented sintering dur-

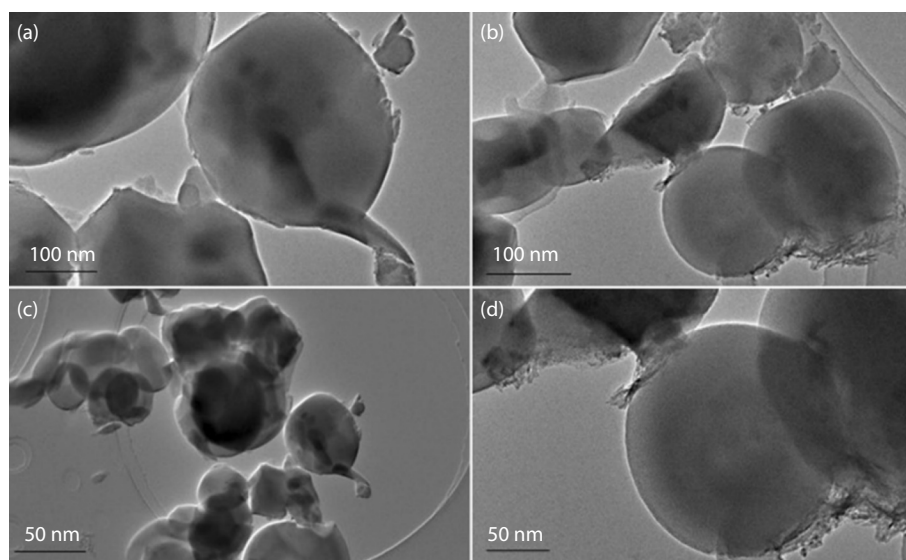


Fig. 4. TEM images of (a, c) acid-precipitated silica before surface modification and (b, d) nano-silica after surface modification. Reproduced with permission from Ref. [46].

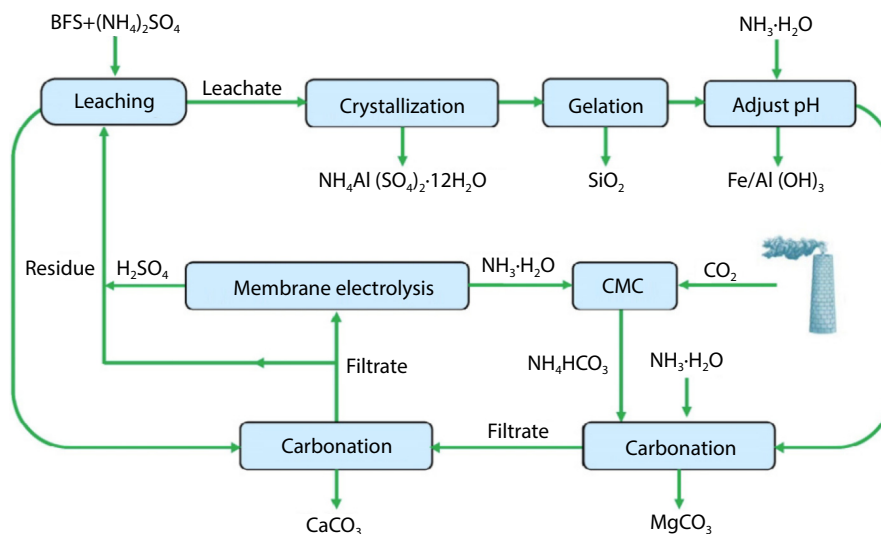


Fig. 5. Process diagram of the indirect carbonation process of BFS using recyclable $(\text{NH}_4)_2\text{SO}_4$. Reproduced with permission from Ref. [47].

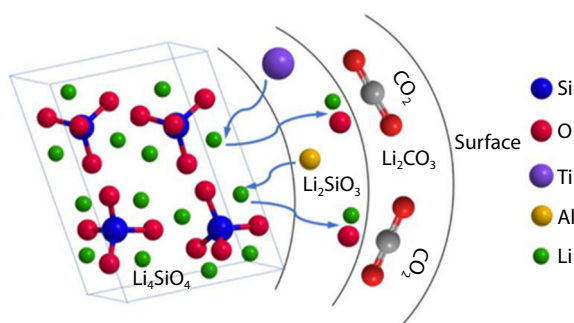


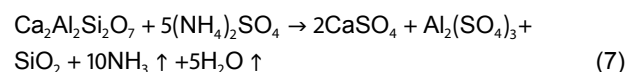
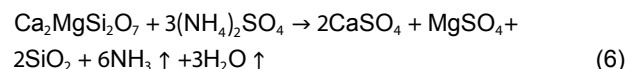
Fig. 6. Mechanism of improving CO_2 absorption performance of Li_4SiO_4 by ion exchange. Reproduced with permission from Ref. [48].

ing the absorption-desorption cycles^[48].

3.2.2. Synthesis of aluminium-based materials

BFS contains 7%–18% Al_2O_3 , which represents an opportunity for high-value products. Currently, aluminium is produced by electrolysis from natural bauxite, whose price is 5 times that of limestone^[49]. Alternative Al precursors can be produced from the indirect carbonation of BFS. The process flow

diagram is shown in Fig. 7. BFS was first roasted with $(\text{NH}_4)_2\text{SO}_4$ at 370 °C. $(\text{NH}_4)_2\text{SO}_4$ decomposed to molten NH_4HSO_4 and gaseous NH_3 at 250 °C. NH_4HSO_4 reacted with BFS, producing Ca-, Mg-, Al-sulfates, and silica, as shown in Equations (6)–(7)^[50].



An $(\text{NH}_4)_2\text{SO}_4$ /BFS ratio of 3:1 achieved 100% extraction of the metals. The roasted solids were leached with deionised water at an L/S ratio of 4 at 80 °C for 1 h, resulting in an Mg- and Al-rich solution. This was acidic due to the residual $(\text{NH}_4)_2\text{SO}_4$ and NH_4HSO_4 . The NH_3 gas released from the roasting process was bubbled into this solution to raise the pH and selectively precipitate the Al into an amorphous aluminium hydroxide sulfate, which could be used in producing

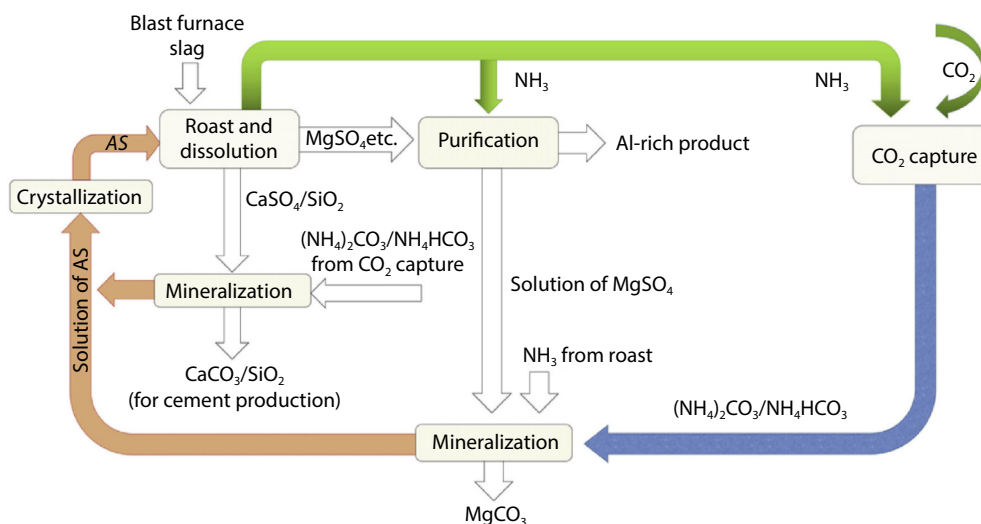


Fig. 7. Process flow diagram of the indirect carbonation of BFS by roasting with $(\text{NH}_4)_2\text{SO}_4$. Reproduced with permission from Ref. [50].

aluminium by electrolysis^[49]. The CaSO_4 -rich leaching residue and the Mg-rich solution were carbonated to produce CaCO_3 and MgCO_3 .

A follow-up work extracted ammonium alum $[\text{NH}_4\text{Al}(\text{SO}_4)_2 \cdot 12\text{H}_2\text{O}]$ from the above-mentioned Al-rich solution. After roasting and leaching, the solution was kept at low temperatures for 12 h to crystallise ammonium alum. By decreasing the temperature, the crystallisation ratio of Al increased from 57% at 30 °C to 93% at 0 °C^[47,50]. Around 99.6% pure ammonium alum was obtained^[47,50], which can be used in manufacturing nanosized alumina with a high economic value^[51].

3.2.3. Synthesis of zeolite

Leaching BFS in acidic aqueous solutions extracts the Ca and Mg contents, and the amorphous Si- and Al-rich residue is a suitable source of zeolites. Acetic acid was found to selectively leach Ca and Mg while limiting the dissolution of Al to less than 40%. A hydrothermal treatment of the residue in NaOH solution at 150 °C produced tobermorite and zeolitic phases, including sodalite, lazurite, and analcime^[52]. The phase composition of zeolite was controlled by the $\text{SiO}_2/\text{Al}_2\text{O}_3$ and $\text{H}_2\text{O}/\text{NaOH}$ ratios, which were changed by varying the concentration of acetic acid and NaOH. A higher acetic acid concentration (2 mol L^{-1}) reduced the amount of Ca in the leaching residue and suppressed the formation of tobermorite. Lower NaOH concentrations (0.5–1 mol L^{-1}) promoted the formation of analcime, showing potential value in selective adsorption and heterogeneous catalysis. The Ca- and Mg-rich leachate was blended with the excess NaOH from the hydrothermal reaction and carbonated, as shown in Fig. 8^[52]. By controlling the $\text{SiO}_2/\text{Al}_2\text{O}_3$ ratio at 2 with NaAlO_2 and the $\text{H}_2\text{O}/\text{NaOH}$ ratio at 20, high-purity (93.5%) NaA zeolite was synthesised and applied in water treatment. The result showed it had a high NH_4^+ absorption capacity of 83.3 mg g^{-1} ^[53].

Amorphous Si- and Al-rich gel was also precipitated by neutralising the pH of the leachate from $(\text{NH}_4)_2\text{SO}_4$ -roasted BFS^[50,54,55]. Gismondine-type zeolite (zeolite P) was synthesised through hydrothermal reaction in 1.5 mol L^{-1} NaOH at 120 °C for 2–4 h^[55]. To avoid the generation of alkaline wastewater, a solvent-free process was developed. The Si- and Al-rich gel was milled with NaOH and heated at 90–120 °C for 4 h to synthesise 99.7% pure hydroxycancrinite. The product was washed by deionised water, and zeolite P was further syn-

thesised from the washed solution via hydrothermal reaction at 120 °C for 2 h. Therefore, a zero-waste process was achieved^[54].

3.2.4. Recovery of titanium-based materials

The same approaches as described in previous sections apply to Ti-bearing BFS. A TiO_2 -rich product was obtained by hydrolysis of the leaching solution from roasted Ti-bearing BFS at 102 °C for 4 h. Its crystallinity was improved by calcining at 1000 °C, resulting in a mixture of anatase and rutile^[56]. This mixture contained around 10% SiO_2 impurity due to co-precipitation. The impurity was removed by flocculation with polyoxyethylene ether 9 before hydrolysis and calcination, generating 99.1% pure anatase TiO_2 with uniform particle size distribution^[57]. To avoid the NH_3 emission during $(\text{NH}_4)_2\text{SO}_4$ roasting, waste copperas ($\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$) was used in the roasting of Ti-bearing BFS. Copperas decomposes at 550–750 °C and released SO_2 , which reacted with CaTiO_3 and formed CaSO_4 , as shown in Equations (8) and (9)^[58].



Adding 10% Na_2SO_4 improved the conversion of CaTiO_3 to TiO_2 from 53% to 98% because of the formation of molten $\text{Na}_3\text{Fe}(\text{SO}_4)_3$, which penetrated the particles, exposing the inner layers of CaTiO_3 , and partly decomposed into SO_2 , reacting with CaTiO_3 . After carbonation, TiO_2 was separated from Fe_2O_3 and carbonates by flotation^[58].

3.2.5. Summary

The state-of-the-art carbonation technologies of BFS to produce high-value products include synthesising silica-based nanomaterials, Al-based materials, zeolite, and Ti-based materials, as summarised in Table 2. These technologies require indirect carbonation to separate and purify different elements, and this usually results in a complicated process and large chemical consumption. Despite these operational challenges, the technologies demonstrate notable performance, achieving high production efficiency (>60%), high product purity (>90%), and a significant CO_2 sequestration potential exceeding 180 kg t^{-1} .

3.3. Steel slag

3.3.1. Production of glass ceramics

SS is a potential feedstock material for preparing CaO-MgO- Al_2O_3 - SiO_2 glass ceramics. However, its high Ca content requires external SiO_2 and Al_2O_3 sources to improve the properties of glass ceramics^[59]. Indirect carbonation can reduce the Ca content and benefit the production of glass ceramics (Fig. 9)^[60]. NH_4Cl was used to selectively leach Ca from SS due to its low acidity. A carbon thermal reduction process was applied, where the leaching residue was mixed with analytical silica powder and coal powder and heated up to 1500 °C to recover the iron and prepare the parent glass. The parent glass was heated up to 1350 °C and annealed at 600 °C to generate the glass ceramics. The final product was composed of melilite and diopside, which had high mechanical strength,

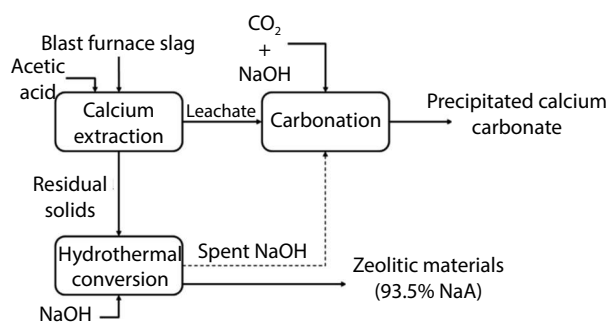
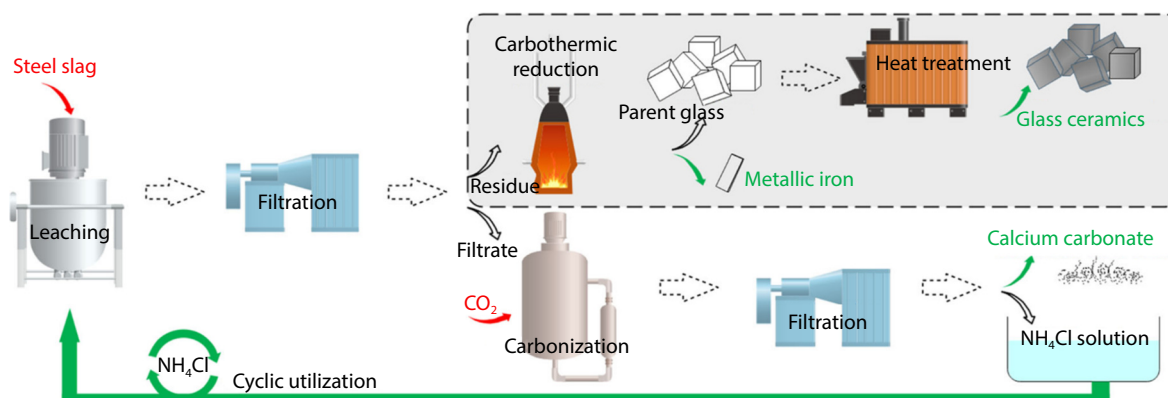


Fig. 8. Flow diagram of indirect carbonation of BFS producing zeolite and calcium carbonate. Reproduced with permission from Ref. [52].

Table 2. Summary of carbonation technologies of BFS towards high-value products.

Product	Production efficiency	CO ₂ sequestration potential	Advantages	Disadvantages
Silica-based nanomaterials ^[46]	60%–78%	-	High quality product ($S_{\text{BET}} > 1400 \text{ m}^2 \text{ g}^{-1}$)	Large chemical consumption
Al-based materials ^[49,50]	73%	303 kg t ⁻¹	Highly pure product (99%), reactants can be recycled	Energy consumption (400 °C), complicated process
Zeolite ^[52]	-	368 kg t ⁻¹	Highly pure product (93.5%)	Complicated process, difficult to scale up
Ti-based materials ^[58]	Up to 98%	187 kg t ⁻¹	High production efficiency	Energy consumption (550–750 °C)

**Fig. 9.** Schematic process showing the indirect carbonation of SS producing glass ceramics. Reproduced with permission from Ref. [60].

resistance to abrasion, and stability^[60].

3.3.2. Synthesis of zeolite

Similar to BFS, the acid leaching residues from the indirect carbonation of SS are also suitable as a zeolite precursor. A 4-hour leaching in 5 mol L⁻¹ HCl extracted 96.3% Ca in the SS. The residue was roasted with NaAlO_2 at 350 °C, controlling a $\text{SiO}_2/\text{Al}_2\text{O}_3$ ratio of 2.5. Then the roasted solid was dissolved in water and hydrothermally reacted at 100 °C for 24 h. Well-crystallised FAU-type zeolite was synthesised, and this can be used as a support for TiO_2 catalyst^[40].

3.3.3. Summary

The state-of-the-art carbonation technologies of SS to produce high-value products include synthesising glass ceramics and zeolite, as summarised in Table 3. Because the formation of glass ceramics and zeolites is inhibited by excess CaO, acid leaching is a necessary pre-treatment step^[40,60]. Despite the increased chemical consumption, calcium is accumulated in the solution, which improves the kinetics of carbonation and the purity of the carbonation product. The acid leaching residue can be converted to glass ceramics and zeolites via heating and hydrothermal processes, respectively. As the crystallisation of glass ceramics requires temperatures above 1000 °C, the energy consumption of heating is high. The hydrothermal treatment requires a long reaction period and

excess NaAlO_2 to adjust the Si/Al ratio. Therefore, the processing conditions of extracting high-value products from SS still need optimisation to improve the economic performance.

3.4. Coal fly ash

3.4.1. Synthesis of nanosized silica

To improve the value of FA, indirect carbonation co-producing mesoporous silica was developed, and ultrasonication was used to facilitate this process^[61]. As shown in Fig. 10, this process involved NaOH leaching of FA and gas-liquid carbonation of the resulting Na_2SiO_3 solution. From 1 tonne of FA in this process, 283 kg of mesoporous silica with a high specific surface area (up to 355.5 m² g⁻¹) and an average pore size of 7.67 nm was synthesised. A Na_2SiO_3 concentration of 10 g L⁻¹ and an ultrasound power of less than 270 W increased the surface area of silica. Additionally, 1.02 tonnes of 95% Na_2CO_3 was generated and can be used as an industrial raw material^[61].

3.4.2. Synthesis of zeolite

Synthesising zeolite from FA involves indirect and direct carbonation pathways. Fig. 11 shows the indirect carbonation of FA with HCl, NH_4Cl , and seawater, which produced a solid residue by-product containing predominantly mullite and quartz^[62]. It had a $\text{SiO}_2/\text{Al}_2\text{O}_3$ ratio of 2.0–2.6, which favours

Table 3. Summary of carbonation technologies of steel slag towards high-value products.

Product	Production efficiency	CO ₂ sequestration potential	Advantages	Disadvantages
Glass ceramics ^[60]	-	30 kg t ⁻¹	Iron can be extracted from the leaching residue	High energy consumption (crystallization activation energy=660.7 kJ mol ⁻¹)
Zeolite ^[40]	-	-	Complete extraction of calcium	Long processing time, pure chemicals need to be added

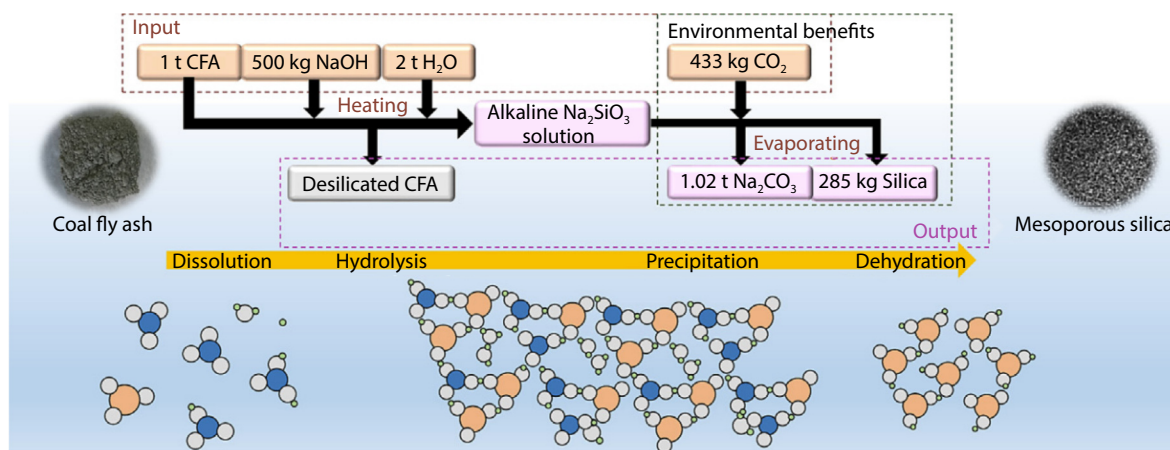


Fig. 10. Indirect carbonation of FA producing mesoporous silica. Reproduced with permission from Ref. [61].

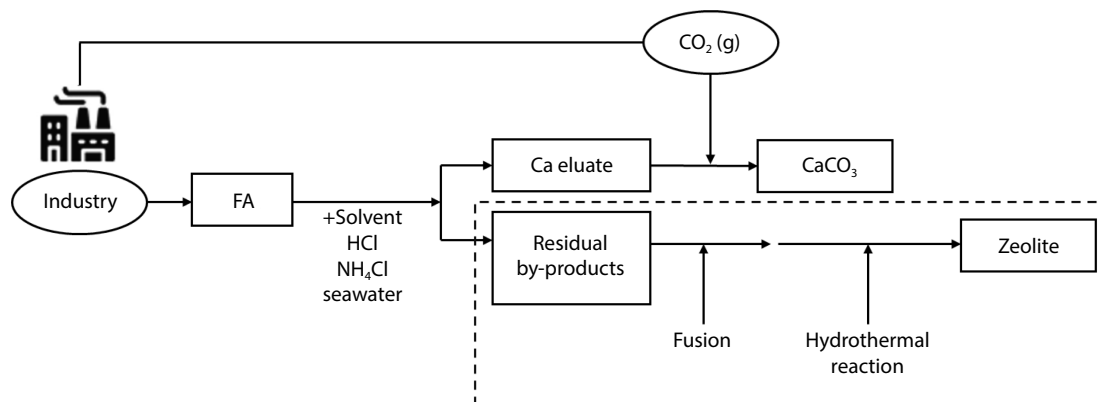


Fig. 11. Synthesis of zeolite from indirect carbonation of FA. Reproduced with permission from Ref. [62].

the formation of zeolite P, hence no external SiO_2 or Al_2O_3 was needed. The residue was treated by fusion with NaOH at 600 °C for 1 h and hydrothermal reaction at 100 °C and 180 °C. A higher temperature improved the conversion ratio by 19%–33%. Zeolite P, synthesised from HCl-leached residue at 180 °C, had the highest surface area ($111.1 \text{ m}^2 \text{ g}^{-1}$) and cation exchange capacity ($139.9 \text{ cmol kg}^{-1}$) and had huge potential in applications such as contaminant adsorption and molecular sieves^[62].

Another study investigated zeolite synthesis from FA using direct carbonation with carbonate and bicarbonate solutions at 150 °C for 72 h^[38]. The process with carbonate solutions had a higher carbonation efficiency than that with bicarbonate solutions because the dissolution of FA was enhanced by a higher pH. Using Na_2CO_3 improved the carbonation efficiency by 76% and the zeolite yield by 82% compared to K_2CO_3 . This was because Na^+ acted as a structure builder, driving the formation of secondary crystalline phases including zeolites and carbonates^[63]. After treatment, the leaching of toxic elements (As, Sb, V, Cu, Mo, and Zn) in an alkaline environment was reduced compared to untreated FA^[38].

3.4.3. Recovery of aluminium- and iron-based materials

Although most work focused on extracting high-value silica or aluminosilicate materials, recent research has investigated the possibility of recovering Al_2O_3 and Fe-based materials during the mineral carbonation of FA. Because class-F FA had a

low CaO content (less than 7%), it was co-processed with BFS to improve the CO_2 sequestration capacity^[64], as shown in Fig. 12. FA, BFS, Fe_2O_3 (adjusting the Fe/Si ratio), and coking coal (reducing agent) were mixed and pressed into pellets. The pellets were subjected to vacuum thermal reduction at 1200–1300 °C for 4 h. Fe_2O_3 was reduced, and the aluminosilicate phases in FA and BFS were decomposed, forming Fe-Si alloy and calcium aluminates. In the next step, the residue was carbonated with Na_2CO_3 and NaOH solutions at 90 °C for 1 h. The Fe-Si alloy was resistant to alkali reaction, while the calcium aluminates were transformed to CaCO_3 and NaAl(OH)_4 . The carbonation products were magnetically separated to produce 80.8% pure Fe-Si alloy and 79.4% pure CaCO_3 . Moreover, by further carbonating the NaAl(OH)_4 solution, pure Al(OH)_3 was formed and Na_2CO_3 was regenerated, achieving a cyclable process^[64]. Fe-Si alloy finds value-added applications as deoxidizers in steelmaking and as electrode materials in batteries^[65,66].

3.4.4. Summary

The state-of-the-art carbonation technologies of FA to produce high-value products include synthesising nanosized silica, zeolite, and Al- and Fe-based materials, as summarised in Table 4. Alkali dissolution and carbonation produce nano-silica with a high specific surface area ($355.45 \text{ m}^2 \text{ g}^{-1}$) but with low production efficiency due to the low Si extraction ratio^[61]. For synthesising zeolite, the direct method features a

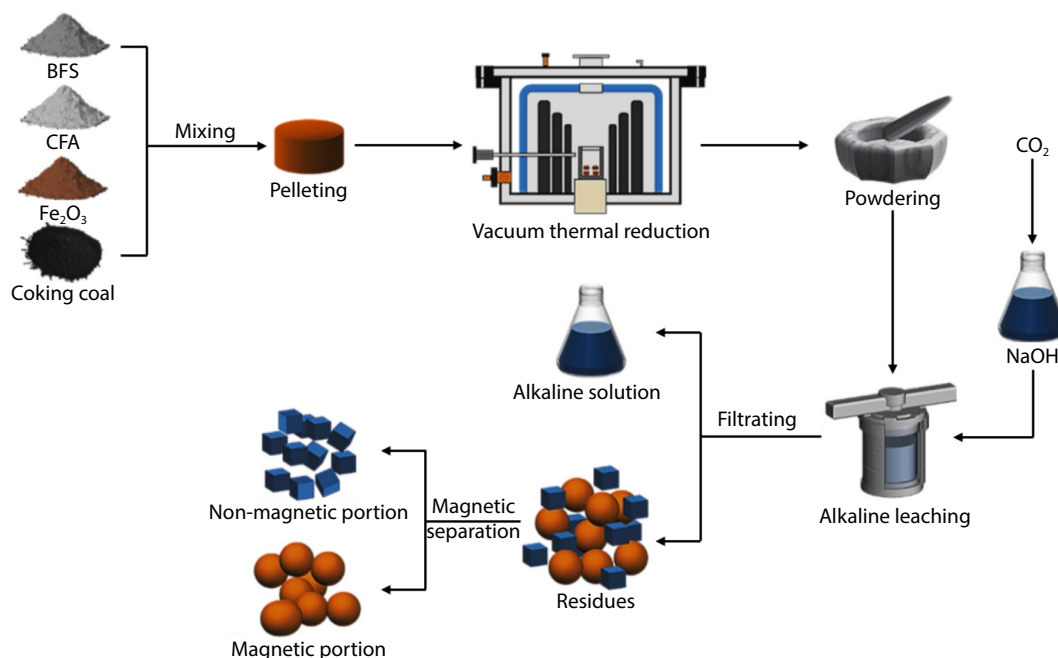


Fig. 12. Flow diagram of mineral carbonation of FA and BFS with recovery of Al_2O_3 and Fe-Si alloy. Reproduced with permission from Ref. [64].

Table 4. Summary of carbonation technologies of coal fly ash towards high-value products.

Product	Production efficiency	CO_2 sequestration potential	Advantages	Disadvantages
Nanosized silica ^[61]	57.3%	433 kg t^{-1}	High product purity and quality	Low Si extraction efficiency (50%)
Zeolite ^[38,62]	Up to 60% ^[36]	45 kg t^{-1} ^[38]	Direct method: Simpler process; Indirect method: No need for external chemicals, high product purity	Direct method: Large chemical consumption, low product purity; Indirect method: Energy-intensive (600 °C), large chemical consumption
Al_2O_3 and Fe-Si alloy ^[64]	80.6% (Al_2O_3)	$167.8 \text{ kg (BFS+FA) per tonne}$	High alumina extraction rate, efficient product separation, zero waste	Energy-intensive (1300 °C), complicated process

simpler process. However, the product contains a mixture of different zeolitic phases with a relatively low specific surface area ($20.4 \text{ m}^2 \text{ g}^{-1}$). The indirect method, although less competitive in energy and chemical consumptions, yields products containing a single zeolite P phase with a higher specific surface area ($111.1 \text{ m}^2 \text{ g}^{-1}$)^[38,62]. Although co-processing of BFS and FA is energy-intensive and consists of multiple stages, it improves the CO_2 sequestration potential of low-Ca FA. Effective production of Al_2O_3 and Fe-Si alloy can be achieved with zero waste generated^[64].

3.5. MSWI fly ash

Due to its high CaO and low SiO_2 and Al_2O_3 contents, large amounts of external SiO_2 and Al_2O_3 sources are needed to control the Si/Al ratio in the hydrothermal reaction of MSWIFA or acid-leached MSWIFA, which limits its valorisation through mineral carbonation with zeolite synthesis^[67]. Therefore, current research is focusing on maximising the value of carbonation products of MSWIFA by developing energy storage materials and ultra-high strength functional materials^[68,69].

3.5.1. Production of thermochemical heat storage materials

Thermochemical heat storage (TES) is a promising way to pro-

vide a stable, efficient, and sustainable energy alternative to fossil fuels. The CaO/CaCO_3 system involves storing thermal energy during the decomposition of CaCO_3 to CaO and CO_2 and releasing energy during the reaction of CaO and CO_2 forming CaCO_3 . It has gained much attention because of its high energy density of 1764 kJ kg^{-1} ^[70]. However, sintering of CaO during the heating/carbonation cycles causes pore closure and reduced exposure to CO_2 , and this effect increases with the increase in cycles. This can cause significant reactivity losses and decrease the energy efficiency of this system^[71,72].

To overcome this issue, the CaO/CaCO_3 system was modified by 5% SiO_2 or 20% aluminates to resist the mitigation of the CaO grain boundary and prevent sintering^[70,73]. MSWIFA is a cheap and largely available alternative resource of SiO_2 and Al_2O_3 . Direct carbonation of MSWIFA yields a product containing high amounts of CaCO_3 , which is suitable for use in thermochemical energy storage materials. Raw MSWIFA was washed with water to remove the chloride salts. Then it was carbonated in 0%–20% ammonia solution with a CO_2 flow rate of 200 mL min^{-1} and at a stirring speed of 500 rpm for 15 min^[68]. Increasing ammonia concentration from 0% to 8% improved the alkalinity of the solution, decomposed CaSO_4 , and promoted the carbonation efficiency from 65.2% to

94.7%. A further increase marginally improved the carbonation efficiency but caused leaching of toxic elements such as As and Cr due to chelation with NH_4^+ ^[68]. The energy density of the product carbonated in 8% ammonia solution was increased to $1193.5 \text{ kJ kg}^{-1}$ compared to that carbonated in water (665.8 kJ kg^{-1}). When used in multi-cycle thermochemical energy storage, it maintained the morphology of fine porous powder after 3 cycles. In contrast, MSWIFA carbonated in water sintered after the first cycle, as shown in Fig. 13. This was due to the removal of CaSO_4 by adding ammonia solution^[68]. This indicated that the carbonation product of MSWIFA in 8% ammonia solution had superior thermal stability and was suitable for thermochemical energy storage.

3.5.2. Production of CaCO_3 ceramics

Cold sintering involves pressing a mixture of CaCO_3 and water under low temperature ($<250^\circ\text{C}$) and high pressure ($>80 \text{ MPa}$) to generate densified sintered bodies through pressure solution creep (Fig. 14). Vaterite is deformed and dissolves under high pressure, then reprecipitates at the contact point between particles upon heating and forms a bond between particles^[74,75]. This process uses carbonate minerals sintered at a lower temperature than in the traditional ther-

mal sintering process (over 1000°C), representing an energy-saving approach to develop carbon-negative ceramics^[74].

In a recently reported process, indirect mineral carbonation of MSWIFA was conducted by leaching in a 2.4 mol L^{-1} HCl solution, and the resulting Ca-rich leachate was carbonated with the addition of 10% ammonia solution for 5 min to obtain pure vaterite^[69]. This product was cold sintered under 100, 300, and 500 MPa at room temperature to produce CaCO_3 ceramics. The compressive strength of the CaCO_3 ceramics increased with pressure and reached 265.9 MPa under 500 MPa pressure^[69]. Given its ultra-high strength, CaCO_3 ceramics have great potential for high-value applications as functional ceramic materials in energy, mechanical, electronics, and aeronautics sectors.

3.5.3. Summary

The state-of-the-art carbonation technologies of MSWIFA to produce high-value products include synthesising thermochemical heat storage materials and CaCO_3 ceramics, as summarised in Table 5. Although producing CaCO_3 -based thermochemical heat storage material from direct carbonation of MSWIFA uses ammonia solution as a reaction medium, which may cause odour problems, this process is simple and can be

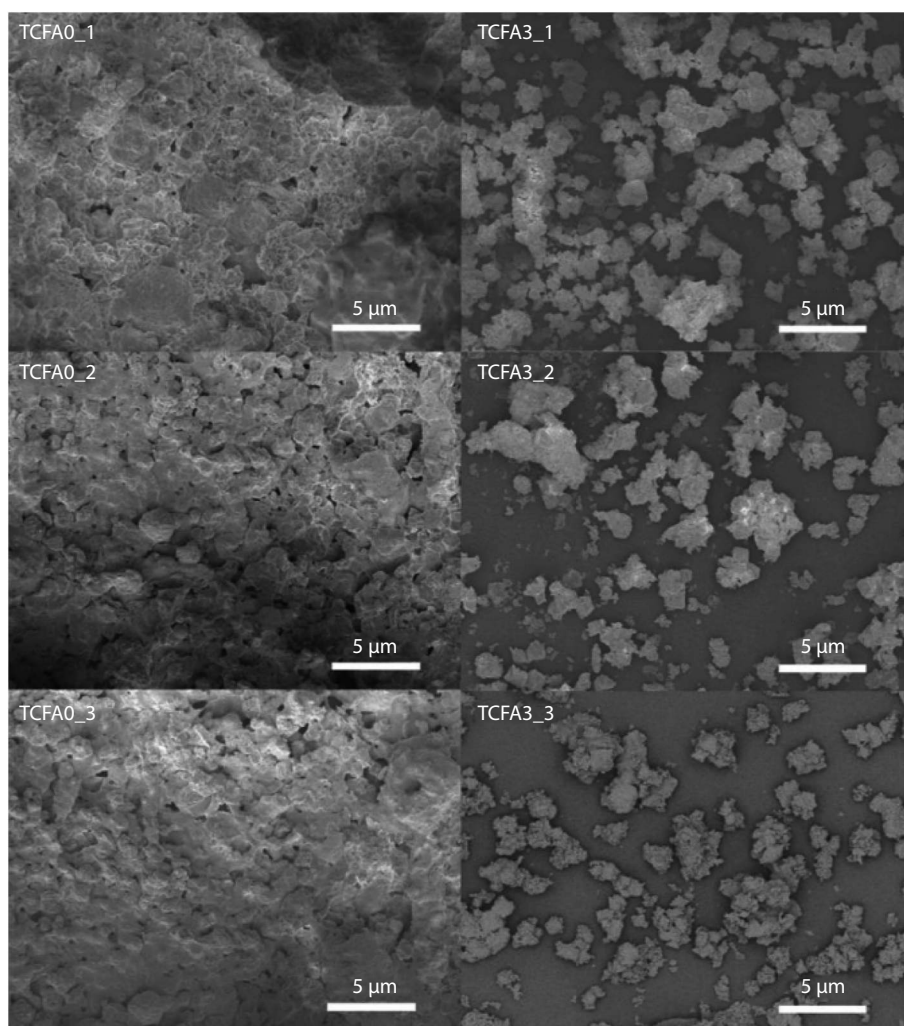


Fig. 13. Morphology of thermal-treated carbonated MSWIFA in water (TCFA0) and 8% ammonia solution (TCFA3) after 1, 2, and 3 cycles. Reproduced with permission from Ref. [68].

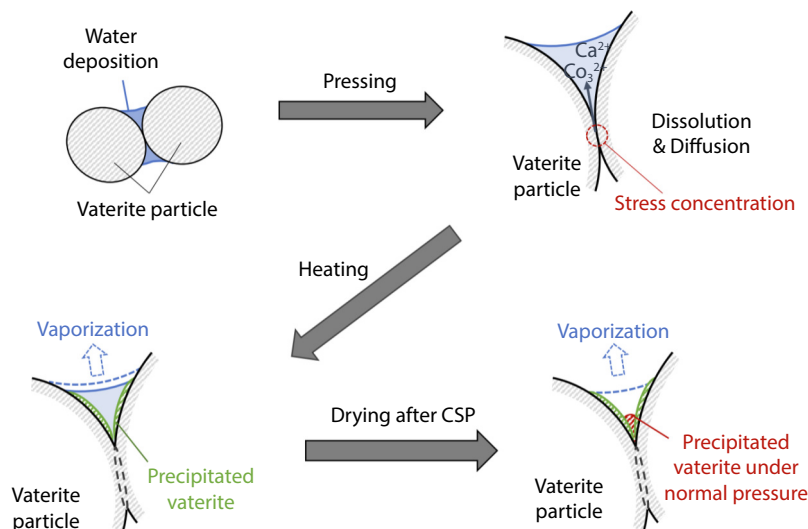


Fig. 14. Mechanism of cold sintering of vaterite. Reproduced with permission from Ref. [74].

Table 5. Summary of carbonation technologies of MSWIFA towards high-value products.

Product	Production efficiency	CO ₂ sequestration potential	Advantages	Disadvantages
Thermochemical heat storage material ^[68]	-	211.9 kg t ⁻¹	Simple and fast process, stabilisation of heavy metal elements	Using ammonia water may cause odour problems
CaCO ₃ ceramics ^[69]	-	-	Production of ultra-high strength ceramics, significant energy saving	Large chemical consumption, complicated process

completed within 15 min. Furthermore, heavy metal elements are stabilised through conversion into carbonates and the formation of a dense CaCO₃ coating on the particles^[66]. Indirect carbonation of MSWIFA produces vaterite, which can be converted into ultra-high strength ceramics (>250 MPa) by cold sintering. The energy consumption of cold sintering is 80 kWh per ton of MSWIFA^[69]. Compared to conventional sintering of similar types of fly ash (around 1000 kWh per ton), this technology achieves an energy saving of 92%^[76]. However, the same as other indirect carbonation processes, this pathway consists of multiple stages and consumes large amounts of chemicals (>2M HCl and concentrated ammonia solution)^[69]. It is claimed that HCl and ammonia can be recycled from the waste NH₄Cl solution after carbonation, but the recycling efficiency and energy consumption have yet to be verified.

4. Scale-up mineral carbonation processes

Recently, several pilot-scale mineral carbonation processes using industrial solid wastes including WCA and SS have been established^[77–79]. However, most of them focus on producing low-value products including CaCO₃ powder and silica-rich SCMs. Some scale-up production of high-value-added products from concrete waste and blast furnace slag has been explored in recent studies.

4.1. Scale-up applications of concrete waste

Based on the process described in Section 3.1.1, a pilot-scale process for producing phosphorus adsorbent and CaCO₃ using indirect carbonation of CSW with water was established in a study^[32]. The extraction step used a lower

water/CSW ratio of 1 than the bench-scale operation (19) to increase the Ca²⁺ concentration in the carbonation step^[31]. The reaction mixture was mechanically stirred at 17 rpm in a reactor with an inner volume of 40 m³ for 6 h to compensate for the lower reaction degree generated by the low water/CSW ratio. Then the mixture was introduced to a filter press with a treatment capacity of 80 tonnes per day to separate the phosphorus absorbent and the Ca²⁺-rich solution. Flue gas containing 6%–13% CO₂ was reacted with this solution at a flow rate of 2 m³ min⁻¹ for 3 h to produce 97% pure CaCO₃^[32]. This purity is lower than that of CaCO₃ produced at bench-scale (99%) due to the generation of CaSO₄·2H₂O under longer carbonation time^[31,32].

The pilot-scale process was operated 5 times within 1 week. The mass balance is shown in Fig. 15. It should be noted that 344.7 tonnes of undiluted CSW was filtered to improve the yield of CaCO₃. Overall, 174.6 tonnes of potential phosphorus absorbent and 0.319 tonnes of CaCO₃ can be produced from 356.7 tonnes of CSW, and the water used in Ca extraction is recyclable. The authors claimed that the process consumed 54.4 kWh of power and could achieve a net CO₂ reduction of 0.33 kg per tonne of CSW^[32].

This scale-up route presents several challenges that stem from the intrinsic properties of the material and its interaction with process equipment. CSW contains fine cement particles with high water retention capacity and forms a compressible, low-permeability filter cake. This can increase the filtration resistance, leading to extended cycle times, high energy consumption, and difficulty in achieving a low-moisture-content filter cake for disposal or further use. To mitigate this, a flocculation step before filtration and an optimised solid-to-

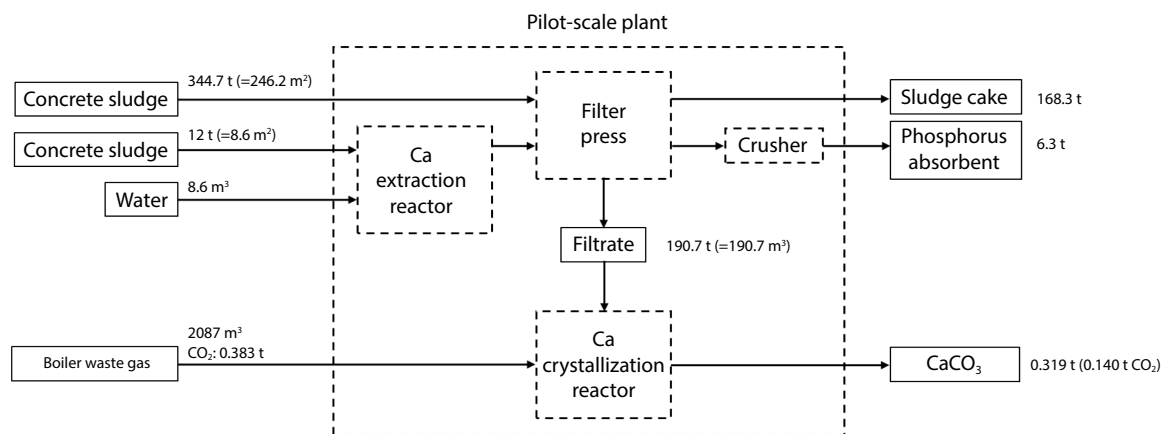


Fig. 15. Mass balance of the pilot-scale plant of indirect mineral carbonation of CSW. Reproduced with permission from Ref. [32].

liquid ratio can be applied. Besides, in a simple stirred tank reactor, the poor solubility and slow diffusion of CO_2 into the slurry limit the gas-liquid contact. Furthermore, the high viscosity of the slurry impedes bubble breakup and dispersion, reducing the effective gas-liquid interfacial area. This leads to a large fraction of CO_2 passing through the reactor unreacted. An improved reactor design, such as those generating nano-sized bubbles and greater turbulence, is important for overcoming this problem^[80]. Additionally, the quality of the product and the recyclability of wastewater are affected by the impurities in CSW (e.g., soil, clay, organic additives, or other construction debris). Careful source control and pre-sorting of CSW and purification of wastewater before recycling are potential solutions to this issue.

4.2. Scale-up applications of blast furnace slag

Based on the process described in Section 3.2.1, a preliminary investigation of the scale-up mineral carbonation of BFS by roasting has been conducted^[81]. BFS was continuously roasted in a rotary kiln, as shown in Fig. 16. The roasting conditions were an $(\text{NH}_4)_2\text{SO}_4/\text{BFS}$ ratio of 4, a temperature of 440 °C, and a duration of 1.5 h. The roasted products were leached with 2% H_2SO_4 at a liquid/solid ratio of 4 at 60 °C for

40 min to dissolve all the Ca, Mg, and Al^[81].

In this process, 1.4 tonnes of aluminium alum, 0.16 tonnes of MgCO_3 , and 0.62 tonnes of CaCO_3 can be produced from 1 tonne of BFS. By evaporating the solution, 95% of the $(\text{NH}_4)_2\text{SO}_4$ was regenerated. The authors claimed that most of the heat required during roasting could be circulated, and this process could achieve a net CO_2 reduction of 36 kg per of BFS^[81].

A significant operational challenge arose from the low melting point (147 °C) of NH_4HSO_4 , which causes it to become liquid at the process temperature of 340–440 °C. Due to the high viscosity of liquid NH_4HSO_4 (around 30 Pa·s), it adhered to the kiln walls, forming kiln rings. These deposits obstructed the feedstock flow and disrupted the continuous operation. For process intensification, this issue can be resolved by adding 30% CaSO_4 , which increases the melting point of the reaction mixture and serves as a reinforcing skeleton to improve the structural deformation resistance^[81].

5. Limitations and outlook

Mineral carbonation of most industrial wastes (concrete waste, BFS, SS, and FA) represents a promising route for the co-production of zeolites and nanosized silica. While zeolites

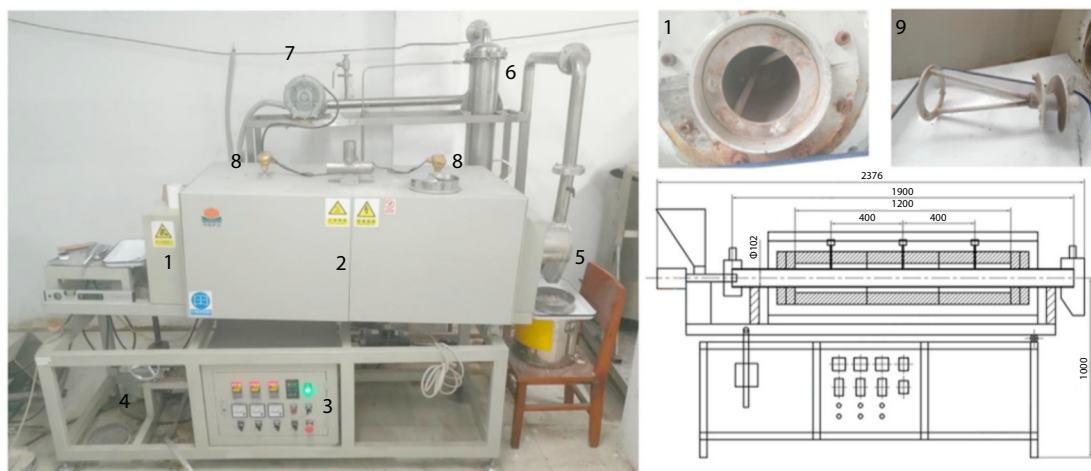


Fig. 16. Image showing the self-designed rotary kiln in the scale-up mineral carbonation process with continuous roasting of BFS. 1-inlet, 2-kiln body, 3-temperature and rotational speed controller, 4-tilting angle controller, 5-outlet, 6-absorption tower, 7-draught fan system, 8-thermocouples, 9-spiral propeller. Reproduced with permission from Ref. [81].

represent a significantly high-value product synthesized through the mineral carbonation of industrial solid waste, their performance in key applications such as catalysis, adsorption, ion exchange, and molecular sieving still requires comprehensive evaluation. Specifically, comparisons to commercially available zeolites synthesized from pure silica and alumina need to be conducted to assess their competitiveness and industrial potential. Also, the processes for synthesizing zeolites, aluminium alum, and Ti-based products are complex, involving multiple pH and temperature changes, which complicates operations. Additionally, some of these methods demand high temperatures, resulting in increased energy consumption and CO₂ emissions, which offset some environmental benefits of the recycling process. Large-scale filtration of the reaction mixture containing nanosized silica and alumina gels, which have high viscosity and may block the cavities of the filter, remains a technical challenge that must be addressed for industrial scalability. Another aspect requiring further investigation is the recyclability of ammonium salt extractants; while they show potential, a detailed assessment of their recovery and reuse must be carried out to ensure the sustainability of the process.

For CaCO₃ ceramics, particularly those produced from vaterite, current research has demonstrated the potential for ultra-high strength materials. However, these results have only been achieved with small-sized samples, and the size effect needs to be considered before industrial application. The process also involves high-pressure pressing, which increases operational complexity. Furthermore, the relationship between the properties of vaterite (e.g., particle size, morphology, and specific surface area), the processing parameters for cold sintering (e.g., pressure and temperature), and the properties and performance of CaCO₃ ceramics (strength, porosity, phase composition, etc.) requires further exploration. A standardized carbonation process that consistently produces vaterite with the best properties for CaCO₃ ceramics should be established.

The transition from laboratory-scale experiments to pilot-scale operations presents additional challenges, particularly in terms of reactor dimensions, mass transfer, and heat transfer, which often result in lower yields and reduced product quality. Moreover, many processes currently lack the capability for continuous operation, further hindering industrial scalability. Therefore, there is an immediate need for improved reactor design to address these limitations.

While the high-value products of these processes hold promise, market development remains an issue. Additionally, current studies often assume the use of low-carbon energy to achieve the carbon-negative potential of mineral carbonation. However, the availability of low-carbon energy varies significantly across regions. In the medium term, a comprehensive life cycle analysis of CO₂ emissions is essential to verify the true net carbon reduction of these processes. Moreover, the economic viability of these high-value products needs to be supported by accurate cost-benefit analyses that quantify both their market value and the financial implications of scaling up.

Finally, a significant hurdle in advancing the field is the

current reliance on a qualitative definition of "high-value" for carbonation products. In the literature, this term is often used without reference to explicit performance benchmarks (e.g., purity, particle size, reactivity) or clear market criteria (e.g., price comparable to commercial alternatives). This lack of standardization creates ambiguity, hinders fair comparison between different technologies, and ultimately obstructs market confidence and commercial adoption. Therefore, a critical long-term objective is to establish standardized product classifications. Defining quantitative thresholds for key properties is essential to move beyond qualitative claims, provide clear targets for research and development, and facilitate the creation of a transparent and reliable market for carbonation products derived from industrial waste.

6. Conclusions

Research has been developing the co-production of high-value products with mineral carbonation of industrial solid wastes to improve the economics and applicability. The main wastes that have the potential for high-value extraction are concrete waste, blast furnace slag, steel slag, coal fly ash, and municipal solid waste incineration fly ash. Zeolites and nanosized mesoporous silica gel are the two most popular products. Al-based products (alumina, aluminium alum) can be sourced from mineral carbonation of BFS and FA. However, their production often involves complicated processing and high energy input. Improving the reaction efficiency, reducing the energy consumption, and conducting a comprehensive life cycle and CO₂ emission analysis should be focused on in future research to maximise the potential of mineral carbonation in developing circular, sustainable, and low-carbon value-added material resources.

Author contribution statement

Tiejun Ding: Writing – original draft, Investigation, Formal analysis, Methodology, Conceptualization. **Jionghuang He:** Writing – review & editing, Methodology, Conceptualization. **Yong Zheng:** Writing – review & editing, Formal analysis. **Long Jiang:** Writing – review & editing. **Zhenjiang Gu:** Writing – review & editing. **Zihan Ma:** Writing – review & editing, Methodology.

Declaration of competing interest

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

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