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A Review of Direct Ex-situ Carbon Dioxide Mineralization Technology Focusing on Techno-economics

Zixu Wang

College of Chemistry and Chemical Engineering, Xiamen University, Xiamen, China
37420232204687@stu.xmu.edu.cn

Abstract. Direct ex-situ CO_2 mineralization converts CO_2 into stable carbonates by reacting with Ca/Mg-rich feedstocks in engineered reactors, offering permanent storage with low leakage risk. This review evaluates feedstocks (natural silicates and industrial alkaline wastes) and compares their sequestration capacity, kinetics, and techno-economic factors. We summarize key process intensification methods, including mechanical/thermal pretreatment and recyclable solvents that enhance CO_2 dissolution and enable cyclic operation. Mechanistic evidence indicates that silicate dissolution is commonly rate-controlling, and conversion is controlled by coupled effects of particle size, temperature, CO_2 partial pressure, ionic strength, and mixing. Finally, TEA frameworks and engineering cases are discussed to clarify the deployment opportunities in construction materials, etc. Challenges lie in accelerating kinetics under mild conditions, controlling product quality, and reducing site-dependent cost uncertainty.

Keywords: Carbon Dioxide, Direct Ex-situ Mineralization, Carbon Sequestration, Techno-economic Analysis, Industrial Alkaline Wastes

1. Introduction

Carbon dioxide (CO_2) emissions from fossil-fuel combustion remain a dominant driver of global climate change. Meeting the Paris Agreement temperature goals [1] requires deep decarbonization supported by large-scale carbon capture, utilization, and storage (CCUS), where durable storage ultimately determines whether net removal can be achieved. Conventional geological storage typically injects CO_2 into depleted oil and gas reservoirs or deep saline aquifers [2,3]. However, long-term monitoring requirements and concerns over leakage can limit public acceptance and deployment [4]. Carbon mineralization has therefore attracted growing interest as it converts CO_2 into thermodynamically stable carbonates, offering inherently permanent sequestration [5].

CO_2 mineralization can be broadly classified as passive mineralization, in-situ mineralization, and ex-situ mineralization. Passive mineralization relies on natural weathering and is kinetically slow under ambient conditions [6]. In-situ mineralization injects CO_2-rich fluids underground to react with host minerals but is constrained by site geology and limited process controllability. Ex-situ mineralization, in contrast, carries out controlled carbonation in engineered reactors using Ca- and Mg-rich feedstocks, including industrial alkaline wastes, enabling flexible siting and tunable operating conditions (Fig. 1) [7-9].

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This paper focuses on direct ex-situ mineralization. Compared with indirect routes that require strong acids/alkalis for metal extraction and subsequent reagent recovery, direct routes proceed within a single reaction environment, reducing process complexity and improving integration potential in selected industrial scenarios. Despite these advantages, high energy demand and feedstock-related costs remain key barriers to large-scale implementation [10,11]. This review synthesizes progress in feedstocks and pretreatment, solvent systems, reaction kinetics, and techno-economic analysis (TEA) with engineering applications, and discusses remaining challenges and future directions toward improved techno-economic performance of direct ex-situ mineralization.

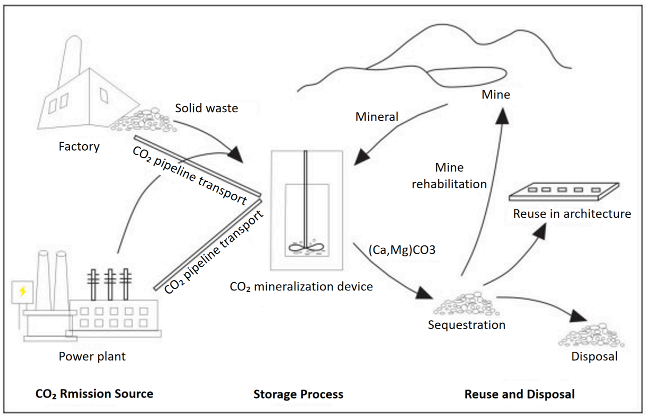

Figure 1. Schematic diagram of the ex-situ mineralization process

2. Feedstocks

The key to large-scale application of ex-situ mineralization lies in identifying feedstocks that are widely available, low in cost, and possess appropriate reactivity. These materials provide essential active components such as calcium and magnesium required for mineralization, as well as alkaline substances to regulate the reaction environment. Based on their origin, feedstocks can be divided into two categories: natural mineral resources and industrial solid wastes. Among them, industrial solid wastes are considered the most promising feedstock sources for ex-situ mineralization due to their accessibility and inherent chemical potential, offering significant environmental synergies.

2.1. Natural mineral ores

Natural minerals, although more frequently discussed for in-situ mineralization, also serve as relevant references for ex-situ routes, especially because similar mineral phases may exist in industrial wastes. Olivine, $(Mg,Fe)_2SiO_4$, is Mg-rich and exhibits high mineralization potential with relatively fast kinetics, and has therefore been extensively studied in ex-situ settings [12]. Serpentine, $Mg_3(Si_2O_5)(OH)_4$, is also abundant and Mg-rich [6], but its practical reactivity is often limited by its crystal chemistry; thermal activation is commonly required to obtain more reactive phases, which can substantially increase the upfront energy and cost burden [13]. Other candidates such as wollastonite ($CaSiO_3$) are typically used as model materials due to resource constraints, and are less favorable for large-scale deployment.

2.2. Industrial solid wastes

Given that techno-economic outcomes are highly site-dependent and strongly influenced by feedstock cost, industrial alkaline wastes are widely regarded as priority candidates for direct ex-situ

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mineralization. Global industrial solid waste generation has been estimated at 7–17 billion tonnes per year, corresponding to a theoretical carbon sequestration potential of 190–332 million tonnes of carbon annually [14]. Representative Ca/Mg-bearing wastes include steel slag, fly ash, cement-related wastes, waste gypsum, and mine tailings. These materials are often rich in reactive Ca phases and can be sourced at low cost while simultaneously addressing waste management challenges.

2.2.1. Steel slag

Steel slag is generated at approximately 10-15% of crude steel output. In China, where annual crude steel production exceeds 1 billion tonnes, steel slag generation reaches ~160 million tonnes per year [15]. Steel slag typically contains substantial reactive Ca-bearing phases (e.g., free CaO) and Mg-bearing components, which underpin its relatively high carbonation reactivity [17]. However, untreated steel slag used in concrete can suffer from volumetric instability, motivating stabilization strategies such as controlled carbonation (Fig. 2) [16]. Carbonation can improve the mechanical performance of slag-containing concrete and offers a pathway for simultaneous CO_2 sequestration and waste valorization [16]. Reported values suggest that 1 tonne of steel slag can mineralize up to ~250 kg CO_2 under suitable conditions [16], and the economic performance can be favorable depending on local costs and policy incentives [18].

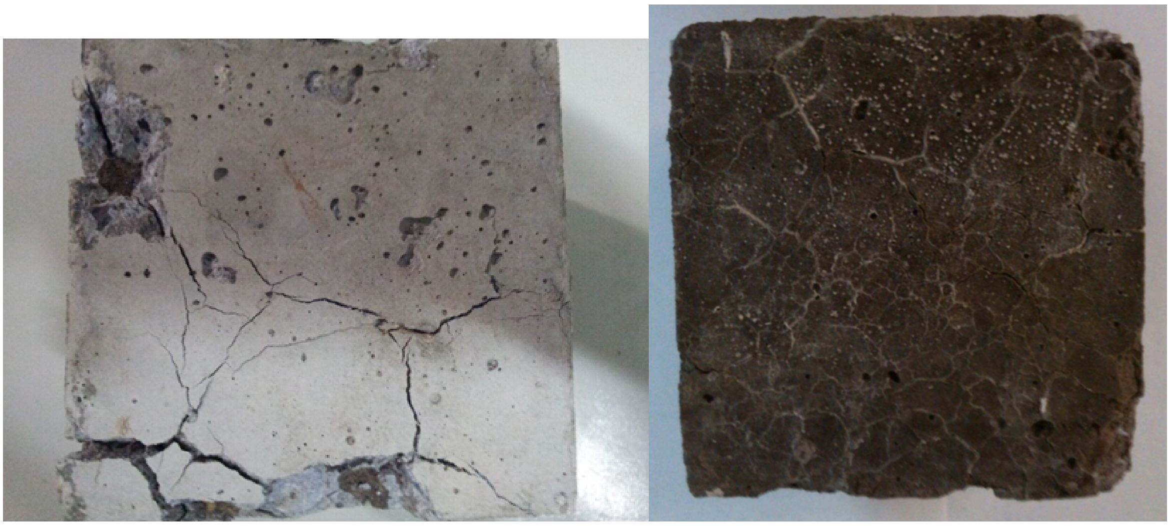

Figure 2. (A) condition of concrete containing steel slag aggregate after long-term simulation by autoclaving. (b) Condition of concrete containing steel slag aggregate after 360 days. Cracking occurs in both (a) and (b), leading to loss of rigidity

2.2.2. Fly ash

Fly ash is a major residue from coal combustion and remains one of the largest industrial waste streams. China produces over 600 million tonnes annually [19]. Fly ash typically consists of SiO_2, Al_2O_3, CaO, and Fe_2O_3, and is commonly classified as low-calcium (Class F) or high-calcium (Class C). Class C fly ash contains higher reactive Ca phases (including free CaO and cement-like minerals such as C_3A and C_2S) and therefore generally exhibits higher mineralization reactivity [20]. Under mild conditions, the theoretical CO_2 uptake is often limited (reported as ~5–80 kg CO_2 per tonne), but can be enhanced by process intensification (e.g., activators or elevated temperature/pressure) [21]. Surface precipitation of $CaCO_3$ during carbonation has been observed by SEM (Fig. 3) [22]. Overall, compared with steel slag, fly ash typically shows lower Ca availability and slower

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conversion; mechanical/chemical activation may be required to improve kinetics, which can increase cost. Consequently, its techno-economic performance depends strongly on process design and the value of carbonate products [22].

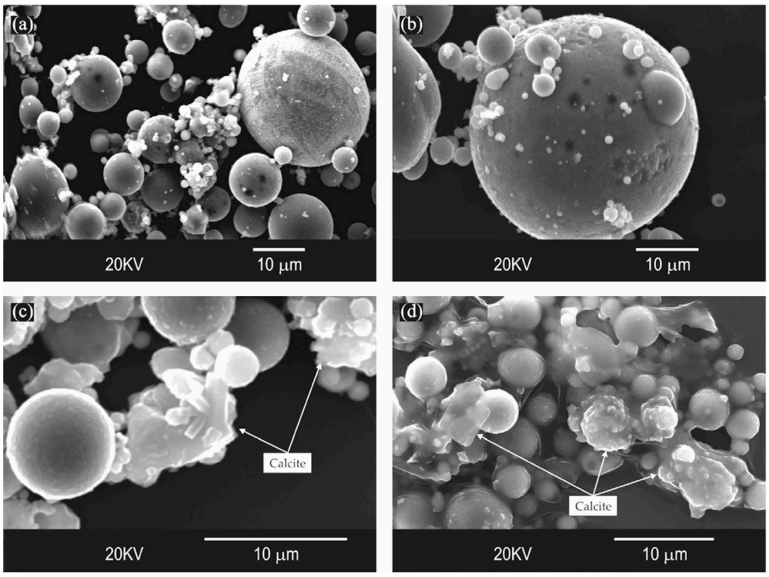

Figure 3. (A) and (b) SEM images of fly ash before reaction. (c) and (d) Calcite particles precipitated on fly ash surfaces during CO_2 sequestration experiments

2.2.3. Cement waste

Cement-related wastes mainly include waste concrete powder from demolition and cement kiln dust. Waste concrete powder contains carbonation-reactive phases such as $Ca(OH)_2$, C–S–H, and unhydrated cement minerals (C_2S and C_3S), while kiln dust is often enriched with reactive CaO and calcium silicates [23]. Reported CO_2 uptake for waste concrete powder can reach ~100 kg CO_2 per tonne, reflecting its substantial mineralization potential [24]. Owing to its porous structure and high surface area, carbonation can proceed rapidly, which is advantageous for continuous processing [24]. From a techno-economic perspective, partial cost recovery may be achieved via recycled aggregates, carbonate products, and carbon-credit revenues; nevertheless, profitability remains site- and boundary-condition dependent. When combined with carbon trading mechanisms, unit abatement costs as low as ~220 CNY tCO_2^{-1} have been reported for cement waste mineralization [25].

2.2.4. Summary

Table 1 compares representative industrial wastes in terms of CO_2 uptake, carbonation rate, and unit abatement cost. Industrial wastes are often co-located with point sources, offering practical advantages for large-scale deployment [8]. Available analyses suggest that steel slag contributes the largest share of direct emission reduction (~43.5%), whereas cement waste can dominate indirect emission reduction (~55.7%) when accounting for substitution effects and avoided emissions in conventional material production [26]. Overall, industrial solid wastes represent promising feedstocks for direct ex-situ mineralization, but techno-economic outcomes remain highly sensitive to feedstock logistics, pretreatment intensity, reactor performance, and product valorization.

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Table 1. Comparison of CO_2 sequestration capacity, carbonation reaction rate, and unit emission reduction cost for steel slag, fly ash, cement waste, waste gypsum, and mine tailings

Solid Waste	CO_2Sequestration Capacity (kgCO_2/ton)	Carbonation Reaction Rate	Techno-economics
Steel Slag	100-250	Fast	Profitable
Fly Ash	5-80	Slow	Hard to make profit
Cement Waste	≈100	Very fast	Cost≈220 yuan/tonCO_2
Gypsum Waste	120-160	Fast	-
Mine Tailings	-	Slow	Hard to make profit

3. Feedstock pretreatment

Pretreatment aims to accelerate direct ex-situ mineralization by increasing accessible reactive surface area and weakening mineral crystallinity, thereby promoting silicate dissolution and subsequent carbonation. Mechanical activation is the most widely used approach, typically including crushing, screening, and ball milling. Li et al. [27] reported that the carbonation conversion of olivine increased from 6% to 82% when the grinding time was extended from 5 to 30 min. Kleiv et al. [28] further showed a 2.3-fold increase in Mg^{2+} dissolution rate after mechanical activation, accompanied by a shift in apparent kinetics from surface-reaction control toward diffusion control. These improvements are commonly attributed to enlarged surface area as well as lattice distortion and partial amorphization induced by milling, which together lower the effective activation barrier for dissolution. Representative particle-size evolution for olivine and mine waste rock under different grinding durations is summarized in Fig. 4 [29]. Notably, the response to mechanical activation is strongly feedstock-dependent. For example, serpentine may exhibit limited reactivity enhancement even under increased grinding energy input [30], suggesting that pretreatment intensity should be selected based on mineralogy and the targeted conversion.

Energy demand is a primary constraint for scale-up. O'Connor et al. [31] estimated that initial crushing requires ~2 kWh t^{-1}, while grinding to 75 μm requires an additional ~11 kWh t^{-1}, and further size reduction to 38 μm can add ~70 kWh t^{-1} [32]. Consistently, Ariffin et al. [33] reported an energy consumption range of 16–52 kWh t^{-1} for producing ground calcium carbonate from marble depending on the final size distribution. These results highlight that aggressive comminution can rapidly dominate operating cost, and pretreatment design should therefore balance kinetic benefits against energy penalties within a techno-economic boundary.

Thermal activation is another common strategy, particularly for hydroxyl-bearing minerals with intrinsically low reactivity, such as serpentine. Heating serpentine to ~600–650 °C induces dehydroxylation, disrupts the layered structure, and increases surface reactivity, thereby improving mineralization efficiency [34]. By contrast, prolonged soaking treatments for olivine have only shown marginal increases in specific surface area [35] and are less attractive for practical deployment. Overall, pretreatment should be treated as an integrated part of process design, where the optimal route depends on feedstock mineralogy, target conversion, and the allowable energy and cost envelope.

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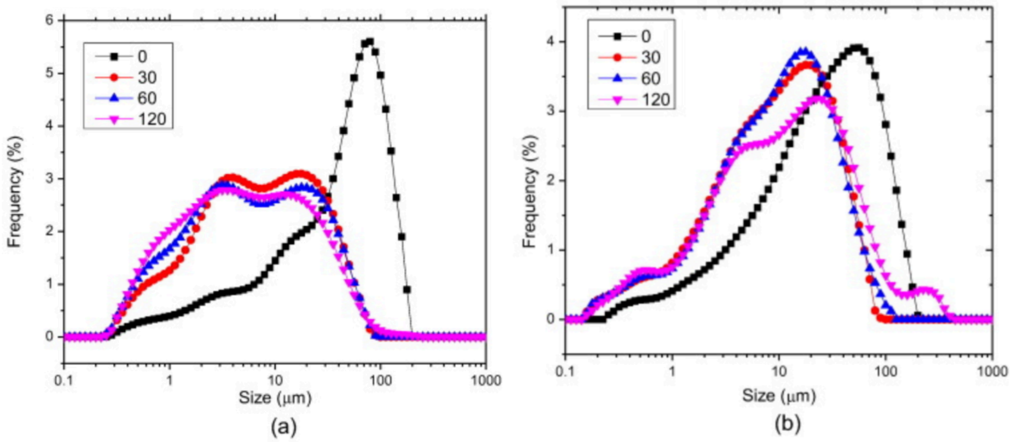

Figure 4. Particle size distribution of olivine and mine waste rock at different grinding times. (a) Initial olivine particle size is mainly 10–200 µm, shifting to 0.3–100 µm after 30, 60, and 120 min grinding. (b) Initial mine waste rock particle size is mainly 3–200 µm, shifting to 1–90 µm after 30, 60, and 120 min grinding

4. Solvents

In direct ex-situ mineralization, solvent selection is governed by two coupled requirements: sufficient CO_2 dissolution to sustain fast carbonation and robust recyclability to avoid prohibitive operating costs. Unlike indirect routes that rely on strong pH swings for regeneration, many direct ex-situ systems achieve in situ regeneration because alkaline mineral additives (e.g., CaO/MgO) shift the solution speciation, releasing free absorbent molecules while driving CO_3^{2-} formation and carbonate precipitation. Organic amines and amino-acid salts are therefore widely used. Monoethanolamine (MEA) has shown stable cyclic performance; Hu et al. [36] reported a CO_2 mineralization efficiency of 78.38% after 10 reuse cycles, and Thamsiriprideeporn et al. [37] also observed good reusability for 5 wt% MEA at 20–25 °C. Mixed amine formulations can further enhance equilibrium CO_2 loading; Feng et al. [38] reported 0.90 mol CO_2/mol amine for MEA + MDEA versus 0.60 mol CO_2/mol amine for 30 wt% MEA. Amino-acid salts such as sodium glycinate (NaGly) offer an alternative pathway via reversible complexation; Liu et al. [39] achieved maximum conversions of 87.50% (MgO) and 94.2% (CaO) with 1 M NaGly, indicating particular promise for Mg-rich feedstocks.

5. Kinetics

The overall kinetics of direct ex-situ mineralization typically involve three sequential steps: (i) CO_2 dissolution and speciation in the liquid phase, (ii) dissolution of Ca/Mg-bearing solids, and (iii) nucleation and precipitation of carbonate products [13]. Across many systems, solid dissolution is frequently identified as the rate-controlling step, especially once a product layer forms [40-42]. For high-purity olivine and serpentine, early-stage carbonation often follows surface-reaction control and gradually transitions to diffusion limitation through a carbonate (and/or silica-rich) layer [43,44]. Incongruent silicate dissolution can generate a silicon-rich amorphous layer that impedes cation transport until exfoliation exposes fresh reactive surfaces. Accordingly, apparent reaction rates are highly sensitive to particle size, temperature, CO_2 partial pressure (PCO_2), and ionic strength, as well as hydrodynamics (stirring) and solid–liquid ratio, which together regulate mass transfer and boundary-layer thickness [45,46]. Figure 5 visualizes the amorphous reaction layer on

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olivine during carbonation [47], and Figure 6 summarizes the dependence of mineralization efficiency on temperature, solid–liquid ratio, and stirring rate [48].

Beyond operating parameters, feedstock structure can dominate kinetics. The initial coordination environment and degree of amorphization of magnesium silicates have been shown to strongly influence mineralization rates [49]. For kinetic interpretation and scale-up, simplified frameworks such as shrinking-core, surface-coverage, Avrami-type, and mixed-control models are commonly adopted to fit conversion–time profiles, while recognizing that practical systems may shift between regimes as surface layers evolve and transport limitations emerge [50-54]. Therefore, it is recommended to report both conversion metrics and the inferred controlling regime under the tested conditions, rather than relying on a single universal correlation.

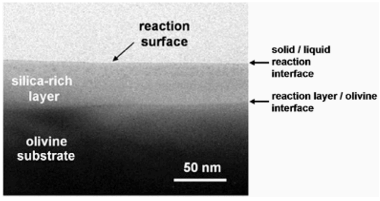

Figure 5. TEM cross-section image of the thin amorphous reaction layer that formed on the olivine single-crystal substrate early during unstirred carbonation

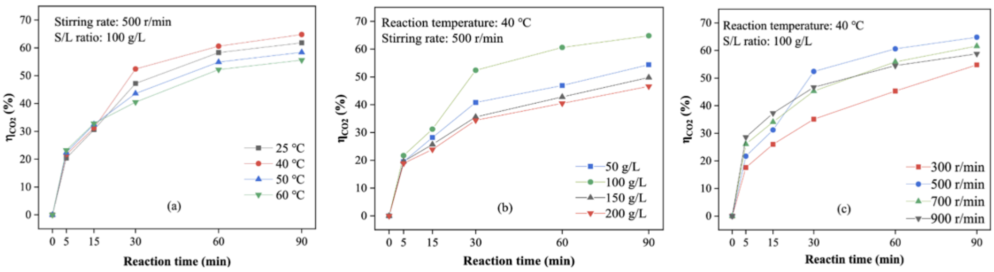

Figure 6. Carbon mineralization efficiency under different (a) temperatures, (b) solid–liquid ratios, and (c) stirring rates

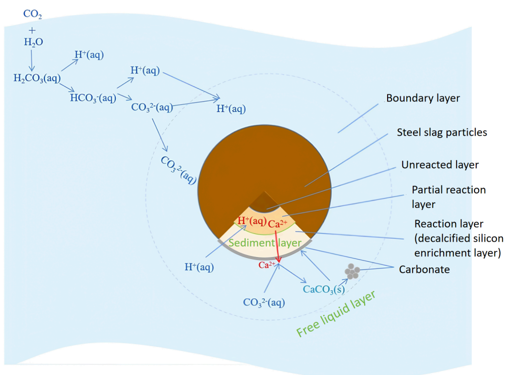

Figure 7. Schematic diagram of the direct carbon mineralization reaction based on the shrinking core model. This model idealizes the reacting solid particle as an unreacted core. As the reaction proceeds, reactants (e.g., CO_3^{2-}, H^+) diffuse through the precipitate layer formed on the exterior of the particle to reach the surface of the unreacted layer for reaction, causing the core radius to gradually shrink until complete conversion

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6. Techno-economics and engineering applications

Techno-economic assessment (TEA) is essential for evaluating whether direct ex-situ mineralization can be deployed beyond laboratory demonstrations. Reported conclusions vary widely because results are highly sensitive to system boundary and site conditions. For clarity and comparability, TEA should explicitly define: (i) the CO_2 source and delivery mode (captured CO_2 vs. flue gas; compression and transport distance), (ii) the feedstock supply chain (collection, pretreatment, transport, storage), (iii) reactor configuration and operating window (temperature, pressure, solid loading, residence time, solvent recycling), and (iv) the product pathway (disposal vs. utilization as construction materials). Within a consistent boundary, the net cost of mineralization can be expressed as a combination of capital expenditure (reactor and auxiliaries), operating expenditure (electricity/heat, solvent make-up, water, maintenance), and logistics costs, offset by potential revenues and avoided costs (e.g., reduced waste disposal, aggregate substitution, and carbon-credit income). Consequently, "profitability" should not be treated as a binary attribute; instead, unit abatement cost and sensitivity ranges are more informative for decision-making.

Across most engineering-relevant scenarios, pretreatment energy and feedstock logistics dominate operating cost, while solvent losses and reactor throughput largely determine scale-up feasibility. Aggressive comminution or thermal activation can accelerate kinetics, but the energy penalty can quickly erase economic gains. Similarly, using strong chemical additives may improve conversion but can shift the process toward an indirect route with higher complexity and reagent recovery burdens. Therefore, cost-effective designs typically prioritize moderate activation paired with process intensification that raises space–time yield, such as optimized solid–liquid ratios, improved mixing and mass transfer, and robust solvent recycling loops. From a design standpoint, high solids handling capability and stable long-term operation (without excessive scaling, clogging, or solvent degradation) are key prerequisites for industrialization

Direct ex-situ mineralization is most competitive when integrated with industries that already generate alkaline wastes or demand mineral products. Construction materials represent a major opportunity because carbonated products can serve as aggregates, supplementary cementitious materials, or binders, enabling co-benefits including improved volumetric stability (for slag-based materials), reduced alkalinity leaching, and partial replacement of carbon-intensive Portland cement. In such integrated pathways, the economics often hinge on local waste disposal policies, product qualification standards, and market acceptance, rather than reaction chemistry alone. In addition, co-location with point sources reduces transport costs and helps stabilize supply, whereas decentralized small-scale units can be viable when feedstock is abundant but CO_2 supply is fragmented. Overall, TEA consistently indicates that deployment feasibility is strongly site-dependent, and the most promising near-term route is industrial symbiosis: "waste-to-resource" mineralization coupled with construction material utilization under policy and carbon-market support.

7. Conclusion

Direct ex-situ carbon dioxide mineralization presents a promising technological pathway for achieving permanent carbon sequestration by converting CO_2 into stable carbonate minerals. The techno-economic viability of this approach is fundamentally dependent upon optimizing reaction kinetics, enhancing product value, and supportive industrial and policy ecosystems. Key barriers include slow kinetics under mild conditions, energy/logistics burdens, product quality qualification for construction use, and long-term solvent stability in recycling loops. Future efforts should prioritize low-energy activation, high-throughput multiphase reactor design, robust solvent recycling

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with impurity management, and deployment-oriented integration with local waste and construction markets. Policy support and credible carbon-credit accounting will be critical to reduce uncertainty and accelerate early deployment.

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