



Revisiting Basalt Reactivity for In Situ CO₂ Mineralization: A Condition-Dependent Reactivity Framework [†]

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Abstract

The identification of the most reactive basaltic phase for in situ CO₂ mineralization remains controversial because reported carbonation behaviour varies considerably across experimental studies. This perspective examines the carbonation characteristics of the principal reactive phases of basalt, including olivine, plagioclase, pyroxene, and basaltic glass, and proposes a Condition-Dependent Reactivity Framework (CDRF) to reconcile these inconsistencies. The framework demonstrates that apparent mineral reactivity is governed by the combined influence of thermo-hydro-geochemical environment, intrinsic mineral characteristics, surface accessibility, and temporal evolution, which collectively regulate reaction kinetics and subsequent carbonation behaviour. Comparative analysis further reveals that each reactive phase follows a distinct kinetic pathway controlled by different dominant constraints, indicating that no universally most reactive basaltic phase exists. Instead, mineral reactivity evolves dynamically with changing reaction conditions. The proposed CDRF provides a unified process-oriented framework for interpreting basalt carbonation and offers practical guidance for future experimental investigations and reservoir-scale evaluation of geological CO₂ mineralization.

Keywords: basalt; in situ mineralization; CO₂ sequestration; olivine; plagioclase

1. Introduction

Synopsis Statement: The proposed condition-dependent reactivity framework explains why no universally most reactive basaltic phase exists, guiding experimental design and improving geological CO₂ mineralization strategies.

The permanent geological storage of carbon dioxide (CO₂) through in situ mineralization in basaltic formations has emerged as one of the most promising strategies for achieving long-term and environmentally secure carbon sequestration [1,2]. Unlike conventional structural trapping, mineralization permanently converts injected CO₂ into thermodynamically stable carbonate minerals, thereby minimizing leakage risk while providing storage over geological timescales [3,4]. The abundance of divalent cation-bearing silicate minerals, coupled with widespread basaltic formations worldwide, has positioned basalt as a strategically important geological medium for accelerating mineral carbonation and supporting large-scale carbon storage [5].

The efficiency of this process is fundamentally governed by the dissolution of reactive silicate minerals, which releases Ca²⁺, Mg²⁺ and Fe²⁺ necessary for carbonate precipita-



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tion [6]. Consequently, identifying the minerals that contribute most effectively to cation release has become a central research focus in basalt carbonation. Among the major basaltic constituents, plagioclase feldspar and olivine have received particular attention because they are commonly regarded as the principal sources of calcium and magnesium required for carbonate formation [7,8]. However, despite decades of experimental and modelling studies, there remains no consensus regarding which mineral is intrinsically the most reactive during in situ CO₂ mineralization [9].

This apparent contradiction is evident throughout the literature. Plagioclase has long been recognized as a highly reactive silicate phase, and recent studies continue to investigate its dissolution and reaction behaviour across a range of geological environments, from magmatic systems to lower crustal deformation processes [10,11]. In contrast, studies performed under reservoir-relevant temperatures, elevated CO₂ pressures and saline fluids often report olivine as the dominant reactive phase because of its strong temperature dependence, rapid magnesium release and favourable carbonate-forming pathways [12–14]. Consequently, different studies identify different minerals as the dominant reactive phase depending on the experimental conditions investigated.

The continued search for a universally “most reactive” basaltic mineral may therefore represent the wrong scientific question. Mineral dissolution is controlled not only by intrinsic crystal chemistry but also by a complex interaction of temperature, pressure, CO₂ fugacity, fluid composition, reactive surface area, mineral composition, reaction affinity, secondary mineral precipitation and reaction timescale [15–20]. Consequently, mineral reactivity should be viewed as an emergent system response instead of an intrinsic mineral property. Comparisons between studies conducted under different thermo-hydro-geochemical conditions are therefore expected to produce different reactivity rankings without necessarily contradicting one another.

Despite this, current studies generally evaluate mineral reactivity within isolated experimental domains and often compare dissolution rates without a common framework for interpreting how environmental conditions govern these observations. As a result, the literature lacks a unified conceptual basis for explaining why apparently conflicting conclusions arise and how experimental findings can be translated into reservoir-scale CO₂ mineralization strategies.

To address this gap, this perspective proposes a Condition-Dependent Reactivity Framework (CDRF) for interpreting the apparent reactivity of the principal reactive phases of basalt during in situ CO₂ mineralization. The CDRF distinguishes four independent controls: thermo-hydro-geochemical (THG) environment, intrinsic mineral characteristics, surface accessibility, and temporal evolution which collectively govern reaction kinetics, reactive cation release, and surface evolution, ultimately determining apparent mineral reactivity and carbon mineralization efficiency. Evidence from surface-weathering, intermediate-temperature, and reservoir-condition studies is synthesized within this framework to explain why mineral rankings vary across geological settings. By reframing basalt reactivity as a condition-dependent phenomenon, the CDRF provides a unified conceptual basis for interpreting experimental observations, reconciling conflicting literature, and guiding future experimental design and geological CO₂ mineralization studies.

2. Development of Condition-Dependent Reactivity Framework (CDRF)

The dissolution of basaltic minerals is frequently interpreted through comparisons of experimentally measured reaction rates, often leading to attempts to identify a universally “most reactive” mineral. However, such comparisons are inherently limited because dissolution behaviour is governed by the coupled influence of mineralogical properties and the surrounding thermo-hydro-geochemical environment. Consequently, mineral reactivity

should not be deemed as a fixed intrinsic characteristic, but an emergent property resulting from multiple interacting controls that evolve throughout the mineralization process.

To address this limitation, we propose the CDRF (Figure 1), which provides a unified conceptual basis for interpreting basalt mineral reactivity during in situ CO₂ mineralization. The CDRF recognizes that apparent mineral reactivity emerges from the coupled interactions among the thermo-hydro-geochemical (THG) environment, intrinsic mineral characteristics (IMCs), and surface accessibility, while temporal evolution continuously modifies these controls throughout the reaction. Together, these controlling factors govern the coupled processes of dissolution kinetics, reactive cation release, and surface evolution, ultimately determining the apparent reactivity of basaltic minerals and the efficiency of CO₂ mineralization.

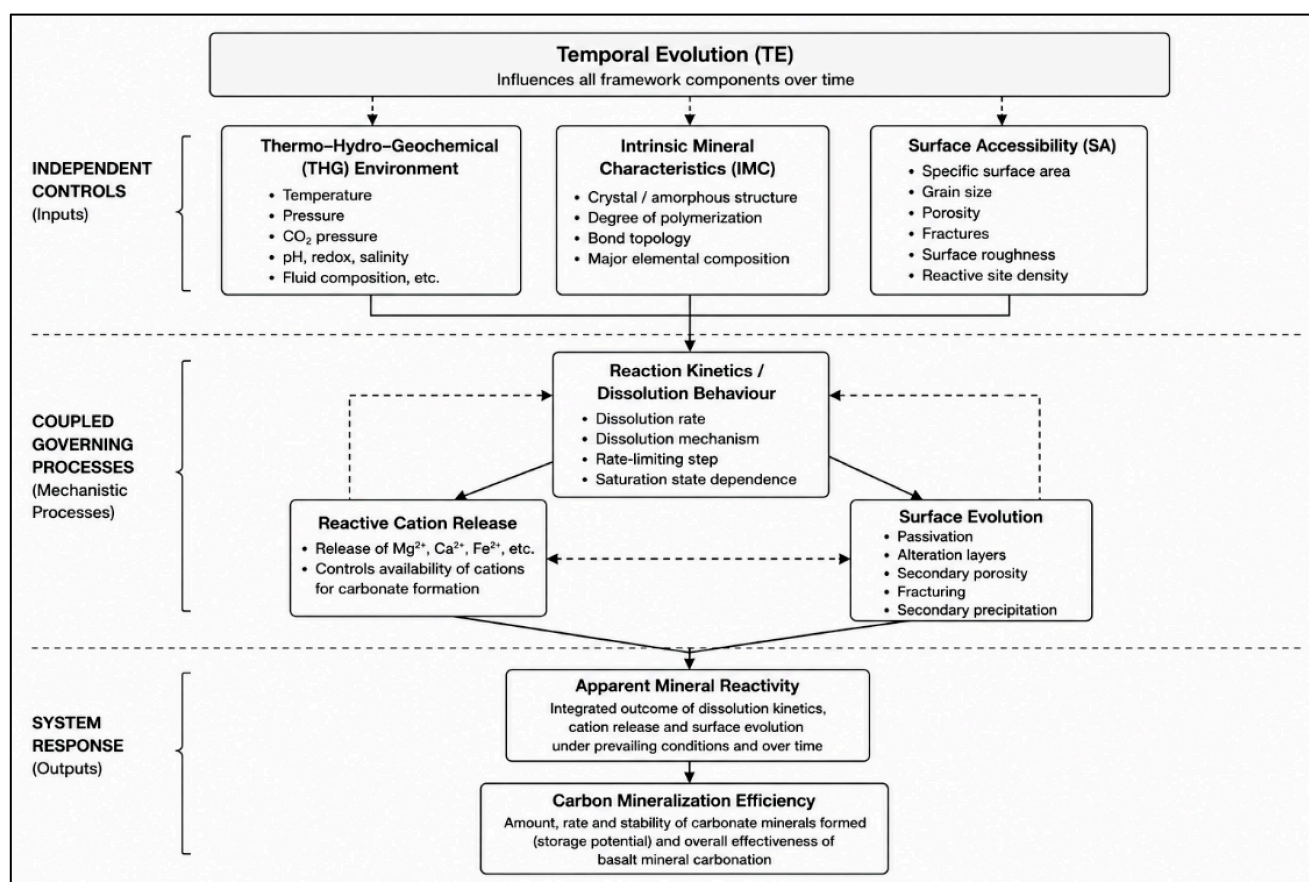


Figure 1. Condition-Dependent Reactivity Framework (CDRF).

The proposed framework provides the context required to interpret reactivity (reaction kinetics) consistently across different laboratory and reservoir conditions. As a result, apparently contradictory observations reported throughout the literature can be understood as expected outcomes arising from different operating domains instead of conflicting mineral behaviour. In the framework, apparent mineral reactivity represents the observable carbonation behaviour of a basaltic reactive phase under a given set of thermo-hydro-geochemical conditions. Unlike intrinsic reactivity, the apparent mineral reactivity emerges from the coupled influence of reaction kinetics, reactive cation release, and surface evolution throughout the mineralization process. Consequently, it is a dynamic, condition-dependent property that evolves with changing reaction conditions and cannot be represented by dissolution rate or carbonate yield alone.

2.1. Thermo-Hydro-Geochemical Environment

The thermo–hydro–geochemical (THG) environment defines the external boundary conditions governing mineral–fluid interactions and therefore represents the primary environmental control within the proposed framework. Temperature, pressure, CO₂ partial pressure (pCO₂), fluid composition, salinity, pH, water–rock ratio, and dissolved silica collectively regulate reaction thermodynamics, aqueous speciation, mineral saturation states, and the chemical driving force for dissolution and precipitation. Variations in these parameters alter reaction affinity and consequently influence both the rate and mechanism of mineral dissolution.

Among these variables, temperature generally exerts the strongest control by accelerating silicate dissolution and promoting carbonate nucleation. Elevated CO₂ pressure enhances carbonic acid formation and increases dissolved inorganic carbon availability, whereas pH regulates proton-promoted dissolution during the early stages of CO₂ injection. Salinity and dissolved silica further modify ion activities and silica solubility, influencing alteration-layer formation and long-term reaction behaviour. Consequently, mineral reactivity cannot be compared independently of the surrounding thermo–hydro–geochemical conditions because identical minerals may exhibit markedly different behaviours under different environmental regimes [3,21].

2.2. Intrinsic Mineral Characteristics (IMCs)

Intrinsic Mineral Characteristics (IMCs) describe the inherent structural and compositional attributes of a mineral that determine its fundamental susceptibility to dissolution before the influence of external environmental conditions. These characteristics include crystal or amorphous structure, degree of silicate polymerization, bond topology, elemental composition, and crystallographic defects, all of which govern the thermodynamic stability of the mineral and its resistance to fluid-mediated alteration. Unlike reaction kinetics, which represent the observable response of a mineral under specific experimental conditions, IMC reflects the intrinsic properties that remain unchanged unless the mineral itself undergoes structural modification.

Within basaltic systems, significant differences in IMC exist among the major mineral constituents. Olivine possesses a weakly polymerized nesosilicate structure that facilitates rapid hydrolysis and Mg release, whereas plagioclase exhibits a more polymerized framework aluminosilicate structure whose reactivity depends strongly on the anorthite–albite composition. Pyroxenes are characterized by single-chain inosilicate structures with intermediate structural stability, while basaltic glass lacks long-range crystallinity and therefore exists in a metastable, high-energy state that is inherently more susceptible to dissolution. Consequently, IMC establishes the intrinsic reactivity potential of each mineral, while the actual dissolution behaviour and apparent mineral reactivity emerge only after interaction with thermo-hydro-geochemical conditions, surface evolution, cation release, and reaction time [22].

2.3. Surface Accessibility and Texture

Mineral dissolution occurs exclusively at the mineral–fluid interface, making surface accessibility a critical control on reaction progression. The extent of reactive surface available for fluid interaction depends on grain size, fracture density, porosity, surface roughness, and the spatial distribution of reactive phases such as basaltic glass. These characteristics determine the density of reactive sites exposed to the fluid and therefore influence the efficiency of mass transfer during dissolution.

Surface accessibility is not static and evolves continuously throughout mineralization. Dissolution may enlarge fractures and generate secondary porosity, whereas silica-rich

alteration layers, secondary carbonates, and clay minerals progressively reduce the exposed reactive surface. Consequently, surface accessibility provides the essential link between intrinsic mineral characteristics and the subsequent governing processes that control apparent mineral reactivity [23–25].

2.4. Temporal Evolution of Reactivity

Mineral reactivity is inherently dynamic and evolves throughout the mineralization process. Initial dissolution rates measured during the early stages of reaction often differ substantially from long-term behaviour because mineral surfaces continuously change during water–rock interaction. As dissolution progresses, secondary silica-rich layers, clay minerals and carbonate precipitates may partially passivate mineral surfaces, reducing reactive surface area and slowing subsequent dissolution.

Consequently, short-term laboratory experiments frequently capture only the initial kinetic response, whereas long-term reservoir behaviour reflects the cumulative effects of passivation, changing saturation states, evolving fluid chemistry and progressive depletion of readily accessible reactive surfaces. The temporal evolution of these processes explains why minerals exhibiting rapid initial dissolution do not necessarily maintain the highest long-term contribution to CO₂ mineralization [26,27].

2.5. Governing Processes of Apparent Mineral Reactivity

The independent controlling factors described above do not directly determine apparent mineral reactivity. Instead, they interact through three coupled governing processes: reaction kinetics and dissolution behaviour, reactive cation release, and surface evolution. These processes are strongly interdependent and continuously influence one another throughout mineralization. For example, dissolution kinetics regulate the rate of cation release, while simultaneously modifying mineral surfaces through passivation, fracture generation, or secondary mineral precipitation. These surface modifications subsequently alter fluid accessibility and reaction rates, creating dynamic feedbacks that evolve with time.

Reactive cation release governs the supply of carbonate-forming ions such as Ca²⁺, Mg²⁺, and Fe²⁺, whereas surface evolution determines the availability of reactive sites for continued mineral dissolution. Together with dissolution kinetics, these coupled governing processes integrate the effects of thermo–hydro–geochemical conditions, intrinsic mineral characteristics, and surface accessibility into the observable behaviour of each mineral. Their combined interaction ultimately defines the apparent mineral reactivity and, consequently, the efficiency of in situ CO₂ mineralization [28,29].

3. Application of the Condition-Dependent Reactivity Framework to Basaltic Minerals

The CDRF provides a systematic basis for interpreting the behaviour of individual basaltic minerals under varying in situ CO₂ mineralization conditions. The framework recognizes that each mineral responds differently to changes in the thermo-hydro-geochemical environment, intrinsic dissolution kinetics, surface characteristics, elemental composition, and reaction timescale. Consequently, the apparent reactivity of a mineral cannot be evaluated independently of the conditions under which carbonation occurs.

Basalt is a mineralogically heterogeneous rock composed of several silicate phases that differ substantially in crystal structure, chemical composition, dissolution behaviour, and cation availability [30]. These differences ultimately control the type, rate, and extent of carbonate mineral formation during CO₂ mineralization. Table 1 summarizes the principal carbonation-relevant reactive groups commonly present in basalt, together with their

representative compositions, dominant cations released during dissolution, and simplified overall carbonation pathways. Table 1 establishes the mineralogical context for the subsequent discussion, where each mineral group is critically evaluated using the five components of the proposed Condition-Dependent Reactivity Framework.

Table 1. Major basaltic reactive phases and representative overall carbonation pathways during in situ CO₂ mineralization.

Basaltic Reactive Phases	Representative Mineral(s)	Representative Formula	Principal Cations Released	Representative Net Carbonation Pathway *	[†] Primary Carbonation Product(s)
Olivine	Forsterite–Fayalite	(Mg,Fe) ₂ SiO ₄	Mg ²⁺ , Fe ²⁺	Mg ₂ SiO ₄ + 2CO ₂ → 2MgCO ₃ + SiO ₂	Magnesite; siderite for Fe-rich olivine
Plagioclase feldspar	Albite–Anorthite	(Na,Ca)(Al,Si) ₄ O ₈	Ca ²⁺ , Na ⁺	CaAl ₂ Si ₂ O ₈ + CO ₂ + 2H ₂ O → CaCO ₃ + Al ₂ Si ₂ O ₅ (OH) ₄	Calcite from Ca-rich plagioclase
[‡] Pyroxene	Diopside, augite, enstatite	(Ca,Mg,Fe)Si ₂ O ₆	Ca ²⁺ , Mg ²⁺ , Fe ²⁺	CaMgSi ₂ O ₆ + 2CO ₂ → CaCO ₃ + MgCO ₃ + 2SiO ₂	Calcite, magnesite, siderite depending on composition
Basaltic glass (mineraloid)		Variable amorphous aluminosilicate	Ca ²⁺ , Mg ²⁺ , Fe ²⁺	Composition-dependent dissolution followed by carbonate precipitation	Mixed Ca–Mg–Fe carbonates

* Representative overall carbonation pathways are simplified net reactions intended to illustrate dominant mineral carbonation behaviour. Actual in situ mineralization proceeds through coupled dissolution–precipitation reactions and depends on temperature, pressure, pCO₂, fluid chemistry, reaction affinity and secondary mineral formation.

[†] Carbonation is primarily associated with Ca-rich plagioclase (anorthitic compositions). Na-rich plagioclase (albite) contributes mainly through alkalinity generation and indirect geochemical effects. [‡] Pyroxene comprises several mineral species with variable Ca–Mg–Fe compositions. The reaction shown is schematic and represents the overall release of divalent cations that subsequently precipitate as carbonate minerals.

Basaltic glass differs fundamentally from olivine, plagioclase, and pyroxene because it is an amorphous volcanic phase instead of a crystalline mineral. It is included within the CDRF because it commonly represents a major reactive constituent of fresh basalt and frequently dominates the earliest stages of CO₂ mineralization (if present).

Building upon the mineralogical overview presented in Table 1, the following sections evaluate each major basaltic mineral individually through the lens of the CDRF. The discussion considers how the input framework components collectively influence mineral behaviour under different carbonation environments. This approach enables a consistent interpretation of apparently conflicting observations reported across laboratory experiments, field demonstrations, and reactive transport simulations.

3.1. Olivine

Olivine is widely regarded as one of the most reactive basaltic minerals for in situ CO₂ mineralization because of its magnesium-rich composition and relatively simple nesosilicate crystal structure [31]. Nevertheless, within the proposed CDRF, olivine reactivity should be interpreted as the outcome of coupled thermo-hydro-geochemical controls, intrinsic dissolution kinetics, surface evolution, cation availability and reaction time. This framework provides a mechanistic explanation for the apparent inconsistency among previous studies, where olivine dominates carbonation under many reservoir conditions but may exhibit substantially lower apparent reactivity under low-temperature weathering environments.

Experimental evidence consistently demonstrates that the thermo-hydro-geochemical environment exerts the strongest external control on olivine dissolution. Hänchen et al. systematically investigated San Carlos olivine between 90 and 150 °C, 15–180 bar CO₂, and pH 2–12.5, reporting that dissolution remained largely stoichiometric and decreased with increasing pH under acidic to neutral conditions (pH 2–8.5). Regression of the complete dataset yielded an activation energy of 52.9 ± 6.9 kJ mol^{−1} and a proton reaction order of 0.46 ± 0.03, confirming that proton-promoted dissolution dominates olivine weathering

over a wide range of geological storage conditions [32]. Interestingly, under CO₂ atmospheres the dissolution behaviour diverged from CO₂-free systems above pH $\approx$ 5, where dissolution decreased more rapidly, indicating that dissolved CO₂ modifies surface reaction mechanisms instead of simply accelerating mineral dissolution.

The importance of environmental controls was further demonstrated by Wang and Giammar, who evaluated forsterite dissolution under conditions representative of geological carbon storage (25–100 °C, 10–100 bar CO₂, 0–50 g L^{−1} NaCl). Initial dissolution rates varied from $10^{-12} \times 7$ to $10^{-10} \times 3$ mol cm^{−2} s^{−1}, representing more than two orders of magnitude solely through changes in reaction conditions. Salinity further modified dissolution behaviour, with 50 g L^{−1} NaCl increasing the initial dissolution rate by approximately 27-fold at 100 °C and 100 bar CO₂, most likely by suppressing or modifying silica-rich surface layer development. However, increasing CO₂ pressure did not universally enhance dissolution. Under some high-temperature conditions, the expected kinetic enhancement was partially offset by rapid surface alteration, highlighting that apparent mineral reactivity emerges from competing kinetic and geochemical processes rather than any single environmental variable.

Although olivine possesses favourable intrinsic dissolution kinetics due to its isolated SiO₄ tetrahedral (nesosilicate) structure, the literature consistently shows that intrinsic kinetics alone cannot explain long-term carbonation behaviour. Instead, surface accessibility evolves continuously during reaction. Johnson et al. investigated olivine carbonation at 60 °C and 100 bar CO₂, obtaining an initial dissolution rate of 9.50×10^{-11} mol cm^{−2} s^{−1}, which declined to 1.69×10^{-12} mol cm^{−2} s^{−1} after prolonged reaction [33]. The dissolution rate decreased by approximately two orders of magnitude within four days, despite the system remaining far from equilibrium. XPS and SEM analyses showed that a Si-rich surface layer formed within two days, progressively passivating reactive sites and reducing effective surface accessibility. Nevertheless, carbonate formation continued, with up to 7 mol% of the original olivine converted into Mg-carbonate after 94 days, demonstrating that surface passivation slows but does not completely inhibit long-term mineralization.

The CDRF further recognizes that dissolution and carbonation are not synonymous processes. Giammar et al. showed that forsterite dissolution increased with both temperature and CO₂ pressure under experiments conducted at 30 and 95 °C and 1 and 100 bar CO₂. Although Mg and Si were released nearly stoichiometrically, dissolved silica concentrations eventually became controlled by amorphous silica solubility. More importantly, solutions reached supersaturation with respect to magnesite while no magnesite precipitation occurred for nearly four weeks in dissolution experiments initiated with deionized water, indicating that carbonate precipitation was limited primarily by nucleation kinetics [33,34]. Subsequent precipitation experiments demonstrated that magnesite nucleation required a critical saturation index (SI) between 0.25 and 1.14 at 95 °C and 100 bar CO₂, while pre-existing magnesite seeds significantly accelerated precipitation. These findings illustrate that the availability of Mg²⁺ alone does not guarantee immediate CO₂ mineralization and that nucleation kinetics constitute an additional control on apparent reactivity.

Collectively, these studies demonstrate that olivine should not simply be regarded as the “most reactive” basaltic mineral. Instead, its apparent reactivity emerges from the dynamic interaction between favourable intrinsic kinetics, environmentally controlled dissolution, evolving surface passivation, magnesium availability and carbonate nucleation. Under high-temperature, CO₂-rich reservoir conditions where fresh reactive surfaces are maintained and carbonate nucleation is favourable, olivine functions as an exceptionally efficient magnesium source for mineral trapping. Conversely, under less favourable thermo-hydro-geochemical conditions or where silica-rich alteration layers develop rapidly, its apparent reactivity decreases substantially.

3.2. Plagioclase Feldspar

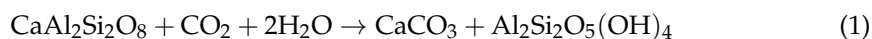
Plagioclase feldspar is the dominant calcium-bearing silicate in most basaltic rocks and therefore constitutes one of the principal mineral sources for long-term carbonate mineralization [35]. Unlike olivine, whose carbonation potential is primarily governed by rapid Mg^{2+} release, plagioclase contributes predominantly through sustained Ca^{2+} supply for calcite precipitation. However, within the proposed CDRF, plagioclase reactivity cannot be interpreted solely from its intrinsic dissolution kinetics. Instead, its apparent carbonation behaviour results from the combined influence of mineral composition, thermo-hydro-geochemical conditions, evolving surface morphology, fluid transport, and coupled secondary mineral precipitation. This explains why previous studies have reported markedly different carbonation efficiencies despite investigating the same mineral group.

The intrinsic reactivity of plagioclase is strongly controlled by its anorthite (Ca)-to-albite (Na) composition. Laboratory investigations by Casey et al. demonstrated that dissolution rates increase systematically with increasing anorthite content, with Ca-rich plagioclase dissolving approximately 2–3 orders of magnitude faster than albite under acidic conditions due to the weaker Ca–O bonds and greater susceptibility to proton-promoted hydrolysis [36]. Similarly, Gudbrandsson et al. investigated plagioclase compositions ranging from An_2 to An_{89} over pH 2–11 and observed a characteristic U-shaped pH dependence, with dissolution increasing under both strongly acidic and alkaline conditions while reaching minimum rates near neutral pH [37]. These studies demonstrate that mineral composition and fluid acidity jointly determine intrinsic dissolution behaviour, indicating that the term “plagioclase” alone provides little predictive value unless its chemical composition and surrounding geochemical environment are simultaneously considered.

While intrinsic kinetics initiate dissolution, the thermo-hydro-geochemical environment ultimately determines whether dissolution translates into effective CO_2 mineralization. Munz et al. investigated carbonation of anorthite-rich plagioclase (An_{67} – An_{73}) using both flow-through and batch reactors under 100–250 °C and 20–120 bar CO_2 [38]. Their experiments demonstrated that carbonation is fundamentally a coupled dissolution–precipitation process. In flow-through systems operated at 250 °C, between 11 and 30% of the plagioclase dissolved within 72–168 h, with substantially greater dissolution occurring at 120 bar than at 40 bar. Surprisingly, although higher pressure enhanced mineral dissolution, a smaller proportion of the released Ca^{2+} was ultimately trapped as calcite, indicating that enhanced cation release does not necessarily translate into greater mineral trapping efficiency. Instead, carbonation efficiency depended on the balance between dissolution kinetics, fluid transport, precipitation rates and reaction-front evolution.

Perhaps the most distinctive characteristic of plagioclase within the CDRF is the continuous evolution of surface accessibility during carbonation. Unlike olivine, where silica-rich passivation layers progressively suppress dissolution, Munz et al. observed that plagioclase dissolution initially generated secondary porosity between the shrinking feldspar grains and the surrounding Al-hydroxide reaction products. Petrographic observations revealed that proto-Al hydroxide was the first secondary phase to precipitate directly on dissolving feldspar surfaces, followed by calcite precipitation within the primary pore space and subsequent crystallization of boehmite after extensive plagioclase dissolution. The development of a migrating reaction front further demonstrated that dissolution, precipitation and pore evolution occurred simultaneously not sequentially. Consequently, surface accessibility is not static but continuously modified by competing processes that both enhance and restrict fluid access throughout carbonation.

From the perspective of cation availability, plagioclase represents the principal source of Ca^{2+} within basalt and therefore directly governs calcite formation through the overall reaction shown in Equation (1).



However, the experiments clearly demonstrate that carbonate precipitation is intrinsically coupled with the formation of Al-hydroxides, boehmite and, under stagnant conditions, clay minerals. Batch experiments produced plagioclase conversion ranging from below the analytical detection limit to 91% within only 24–72 h, with temperature and crystallinity emerging as the dominant controls, whereas increasing pressure and CaCl_2 addition exerted comparatively little influence on overall conversion. These observations indicate that mineral trapping is governed not only by Ca^{2+} release but also by the evolution of secondary reaction products that continuously modify both pore structure and reactive surface area.

Overall, the CDRF demonstrates that plagioclase should not be considered simply as a slower-reacting alternative to olivine. Instead, its apparent reactivity emerges from the interaction between mineral composition, thermo-hydro-geochemical conditions, evolving surface accessibility, calcium availability, and temporal changes in dissolution–precipitation coupling. Whereas olivine typically dominates the early release of Mg^{2+} under favourable reservoir conditions, plagioclase provides a sustained source of Ca^{2+} and increasingly controls long-term calcite formation through progressive reaction-front migration and continuous restructuring of the pore network. This behaviour strongly reinforces the central premise of the CDRF that mineral reactivity is an emergent, condition-dependent property.

3.3. Pyroxene

Pyroxenes constitute the second major group of basalt-forming silicates after plagioclase and encompass a wide compositional range from Mg-rich end members such as enstatite and diopside to Fe-rich varieties including augite and hedenbergite [39]. Consequently, their contribution to in situ CO_2 mineralization cannot be represented by a single dissolution rate or carbonation efficiency. Within the proposed CDRF, pyroxene reactivity emerges from the interaction between intrinsic crystal chemistry, thermo-hydro-geochemical conditions, evolving surface morphology, cation release, and progressive microstructural modification during reaction. Unlike olivine, where long-term carbonation is frequently constrained by silica-rich passivation layers, or plagioclase, where dissolution is coupled with secondary aluminosilicate precipitation, pyroxenes exhibit a distinct carbonation mechanism in which reaction-induced fracturing continuously modifies reactive surface accessibility.

The intrinsic dissolution behaviour of pyroxene has been extensively investigated using diopside as a representative clinopyroxene. Dixit and Carroll measured steady-state diopside dissolution in flow-through reactors at 125–175 °C and pH 7.5, demonstrating that dissolution remained essentially stoichiometric over the entire experimental range. Reaction rates continuously declined by approximately two orders of magnitude as equilibrium was approached, indicating that pyroxene reactivity progressively decreases with evolving solution chemistry. Their ion-exchange model, based on the formation of a Si-rich, Mg-deficient surface precursor, yielded an apparent activation energy of 332 kJ mol^{−1} with an Mg–H exchange coefficient of 1.39, emphasizing that dissolution is strongly influenced by surface reaction mechanisms and not solely by bulk mineral composition [40]. These observations demonstrate that intrinsic kinetics alone cannot define pyroxene reactivity, which instead varies continuously with the surrounding thermo-hydro-geochemical environment and solution saturation state.

Although dissolution initiates mineral carbonation, the subsequent evolution of reactive surfaces ultimately governs the long-term carbonation potential of pyroxenes. Monasterio-Guillot et al. investigated aqueous carbonation of single-crystal augite and diopside under hydrothermal conditions at 150 °C for 7, 14, and 28 days using bicarbonate- and carbonate-bearing solutions. Their results demonstrated a marked compositional dependence, with augite achieving approximately 38 wt% conversion, whereas diopside reached only about 15 wt% conversion under comparable conditions [41]. The enhanced reactivity of augite was attributed to its higher Fe and Al contents, which promoted greater cation release and facilitated the precipitation of secondary carbonate and silicate phases. Importantly, the principal carbonation products were low-Mg calcite and amorphous silica, while augite additionally produced Na-phillipsite and magnesium silicate hydrate (MSH) owing to its more complex chemistry. These observations demonstrate that mineral composition governs not only dissolution kinetics but also the identity of secondary reaction products, which subsequently influence the overall carbonation pathway.

Perhaps the most significant finding distinguishing pyroxenes from other basaltic minerals is the role of reaction-driven fracturing in controlling surface accessibility. The precipitation of secondary silicate phases within dissolution pits and microfractures generated crystallization stresses capable of propagating new fractures through the parent crystal. In augite, nucleation of Na-phillipsite within confined dissolution features generated crystallization pressures approaching 100 MPa, sufficient to exceed the tensile strength of the host mineral and induce extensive microfracturing. Diopside exhibited a similar but considerably weaker response, where localized precipitation of magnesium silicate hydrate produced only limited reaction-induced fracturing. The resulting fracture networks continuously exposed fresh reactive surfaces and created additional fluid pathways, establishing a positive feedback mechanism whereby carbonation itself enhanced subsequent dissolution. This behaviour contrasts sharply with olivine, where silica-rich alteration layers progressively reduce surface accessibility, and with plagioclase, where dissolution is primarily coupled to reaction-front migration and secondary porosity development.

From the perspective of cation availability, pyroxenes provide both Ca^{2+} and Mg^{2+} for carbonate formation depending on mineral composition. Representative carbonation of diopside may be expressed as Equation (2).



Although natural systems commonly deviate from this simplified pathway because Ca and Mg are partitioned among calcite, magnesium silicate hydrates, zeolites, and other secondary phases. Consequently, the amount of dissolved cations does not directly translate into carbonate precipitation. Instead, carbonation efficiency depends on the competition between carbonate nucleation, secondary silicate formation, and evolving pore-fluid chemistry. The progressive decrease in dissolution rates toward equilibrium observed by Dixit and Carroll, together with the reaction-enhanced permeability documented by Monasterio-Guillot et al., demonstrates that pyroxene carbonation is governed by a dynamic balance between kinetic slowdown and microstructural renewal.

Overall, pyroxene provides perhaps the clearest evidence supporting the proposed CDRF. Its apparent reactivity is not controlled by a single intrinsic property but emerges from the coupled influence of mineral composition, thermo-hydro-geochemical conditions, saturation state, reaction-induced fracturing, secondary mineral precipitation, and temporal evolution of reactive surfaces. Under conditions where dissolution promotes fracture generation and continuous exposure of fresh mineral surfaces, pyroxenes can sustain prolonged carbonation despite the thermodynamic tendency for dissolution rates to decrease toward equilibrium.

3.4. Basaltic Glass (Mineraloid)

Basaltic glass represents the most thermodynamically unstable constituent of fresh basalt and consequently exhibits the highest initial dissolution potential among basaltic phases [42]. Unlike crystalline minerals, basaltic glass lacks long-range atomic order, resulting in a metastable amorphous network that is considerably more susceptible to hydrolysis and water penetration. Within the proposed CDRF, however, basaltic glass should not be deemed as intrinsically superior for CO₂ mineralization. Instead, its apparent reactivity is governed by the combined influence of thermo-hydro-geochemical conditions, metastable dissolution kinetics, evolving alteration layers, multi-cation release, and progressive changes in surface chemistry. Consequently, basaltic glass represents an ideal example demonstrating that even highly reactive amorphous materials exhibit condition-dependent behaviour.

The intrinsic dissolution behaviour of basaltic glass has been comprehensively quantified by Gislason and Oelkers, who measured far-from-equilibrium dissolution kinetics over pH 2–11, temperatures between 6 and 150 °C, and open-system flow conditions. Their experiments demonstrated the characteristic U-shaped pH dependence observed for many silicates, with dissolution rates decreasing from acidic conditions toward circumneutral pH before increasing again under alkaline conditions. Regression of experimental data yielded a pH-independent activation energy of 25.5 kJ mol^{−1}, substantially lower than values commonly reported for crystalline silicates, reflecting the lower energetic barrier associated with breaking the disordered glass network. The authors further showed that dissolution kinetics could be described over an exceptionally broad range of environmental conditions (6–300 °C and pH 1–11) using a single kinetic expression, highlighting the dominant influence of temperature and proton activity on far-from-equilibrium glass dissolution [43]. These findings confirm that the absence of long-range crystallographic order provides basaltic glass with a significant kinetic advantage during the initial stages of water–rock interaction.

Nevertheless, the CDRF emphasizes that favourable intrinsic kinetics alone cannot sustain long-term carbonation. Recent work by Bas-Lorillot et al. demonstrated that basaltic glass dissolution under circumneutral to alkaline conditions (pH 6–10) and 30–90 °C is strongly influenced by the evolving thermo-hydro-geochemical environment. Although increasing pH accelerated dissolution under alkaline conditions, the dissolution rate decreased exponentially with increasing dissolved silica concentration, indicating that silica accumulation exerts a stronger kinetic limitation than predicted by classical transition-state models [44]. Instead, the authors proposed a kinetic framework based on classical nucleation theory, suggesting that aqueous silica fundamentally modifies the dissolution mechanism. Equally important, dissolved oxygen promoted oxidation of Fe(II) released from the glass, leading to the development of a Fe(III)-Si-rich alteration layer, while silica-rich solutions favoured formation of an amorphous silica-rich surface layer (ASSL). These observations demonstrate that dissolved oxygen and silica concentration constitute critical thermo-hydro-geochemical variables controlling basaltic glass reactivity in addition to temperature and pH.

The importance of temporal evolution was further demonstrated by Kanelis et al., who investigated basaltic glass dissolution under strongly alkaline conditions (pH ≈ 12) for 70 days at 22 and 50 °C. Although element release during the initial stages agreed well with previously reported flow-through dissolution rates, dissolution slowed markedly after approximately 2–3 weeks, despite geochemical calculations indicating that the solutions remained undersaturated with respect to basaltic glass. Surface analyses attributed this reduction not to thermodynamic equilibrium but to precipitation of brucite, minor calcite, and Al- and Fe-hydroxides, which progressively passivated the glass particles and

restricted further cation release [44]. The authors therefore concluded that efficient CO₂ mineralization using basaltic glass requires continuous exposure of fresh reactive surfaces to overcome secondary phase passivation. This finding reinforces the CDRF concept that apparent mineral reactivity evolves continuously with reaction time and cannot be inferred solely from initial dissolution kinetics.

From the perspective of cation availability, basaltic glass differs fundamentally from all crystalline basaltic minerals because it simultaneously supplies Ca²⁺, Mg²⁺ and Fe²⁺ during dissolution. This multi-cation release promotes the formation of diverse carbonate assemblages while also increasing solution alkalinity, thereby creating favourable conditions for subsequent mineral trapping. However, the rapid depletion of the most reactive amorphous material together with the progressive development of silica-rich alteration layers and secondary precipitates limits its long-term contribution relative to crystalline minerals. Consequently, basaltic glass functions primarily as an early-stage accelerator of mineralization, initiating rapid cation release before progressively transferring control of long-term carbonation to the more slowly dissolving crystalline constituents.

Overall, basaltic glass provides compelling support for the proposed CDRF. Its exceptionally high initial reactivity originates from its metastable amorphous structure, yet its apparent carbonation performance progressively declines as alteration layers develop, dissolved silica accumulates, secondary precipitates form, and reactive glass is consumed as shown in Table 2. Therefore, even the most reactive basaltic phase behaves as a condition-dependent material, whose carbonation efficiency emerges from the interaction between environmental conditions, intrinsic kinetics, evolving surface chemistry, cation release, and reaction time.

Table 2. Synthesis of basaltic reactive phases using the CDRF.

CDRF Parameter	Olivine [32,34,45]	Plagioclase [36–38,46]	Pyroxene [40,41,47]	Basaltic Glass (Amorphous Reactive Phase) [43,44,48]
THG environment	Studied across 25–150 °C, 10–180 bar CO ₂ , pH 2–12.5; salinity up to 50 g/L NaCl can strongly enhance dissolution.	Carbonation tested at 100–250 °C, 20–120 bar CO ₂ using An-rich plagioclase An67–An73.	Diopside kinetics tested at 125–175 °C, pH 7.5; carbonation of augite/diopside tested at 150 °C for 7–28 d.	Dissolution tested over 6–150 °C, pH 2–11; recent tests at 30–90 °C, pH 6–10 show strong SiO ₂ (aq)/O ₂ control.
Intrinsic Mineral Characteristics (IMCs)	Nesosilicate with isolated SiO ₄ tetrahedra and Mg-rich composition. Weakly polymerized structure promotes proton attack and rapid Mg release.	Reactivity depends strongly on An–Ab composition, with Ca-rich (anorthitic) plagioclase being intrinsically more reactive than Na-rich varieties.	Single-chain inosilicate with stronger polymerization than olivine. Crystal chemistry (diopside vs. augite) controls structural stability and dissolution susceptibility.	Amorphous, metastable aluminosilicate network lacking long-range crystallinity, resulting in high free energy and the greatest intrinsic susceptibility to dissolution.
Surface accessibility and texture	Si-rich surface layers/passivation reduce long-term reactive surface area; layer formation observed within days.	Al-hydroxide forms first; secondary porosity develops around shrinking feldspar, while calcite fills primary pores.	Reaction-driven fracturing creates new surface area; Na-phillipsite crystallization pressure can reach ~100 MPa in augite.	ASSL and Fe(III)-Si-rich alteration layers progressively reduce effective surface accessibility.
Cation availability and stoichiometry	Dominant Mg ²⁺ supplier for magnesite; Fe-rich olivine may contribute Fe ²⁺ for siderite.	Dominant Ca ²⁺ supplier for calcite; albite-rich compositions contribute indirectly via alkalinity.	Ca ²⁺ , Mg ²⁺ , Fe ²⁺ depending on composition	Simultaneously releases Ca ²⁺ , Mg ²⁺ , Fe ²⁺ during dissolution
Intrinsic dissolution kinetics	Rapid proton-promoted dissolution; initial forsterite rates 10 ^{−12.7} –10 ^{−10.3} mol cm ^{−2} s ^{−1} under GCS-relevant conditions.	Strongly composition-dependent; Ca-rich/anorthitic plagioclase dissolves much faster than Na-rich/albitic compositions.	Stoichiometric diopside dissolution; rates decrease by ~2 orders of magnitude as equilibrium is approached.	Metastable amorphous structure gives high initial reactivity; classic activation energy 25.5 kJ mol ^{−1} for far-from-equilibrium dissolution.
Temporal evolution of reactivity	High initial Mg release followed by slowdown; up to 7 mol% olivine converted to Mg-carbonate after 94 d.	Flow-through 11–30% dissolution in 72–168 h; batch conversion ranged from below detection to 91% in 24–72 h.	Carbonation conversion: augite ~38 wt%, diopside ~15 wt%; fracturing may sustain reaction over time.	Rapid early dissolution, followed by alteration-layer control and secondary phase passivation; alkaline batch tests show slowdown after 2–3 weeks.

4. Mechanistic Insights from the Condition-Dependent Reactivity Framework

The CDRF provides a mechanistic perspective for interpreting basalt carbonation beyond conventional comparisons based solely on dissolution rates or carbonate yields. While previous studies have predominantly ranked basaltic phases according to their apparent reactivity, the present framework demonstrates that each reactive phase follows a distinct kinetic pathway controlled by different rate-limiting mechanisms. As illustrated in Figure 2, reaction kinetics represent the central governing process linking intrinsic mineral behaviour with the subsequent evolution of reactive surfaces, cation release, and eventually apparent mineral reactivity. Consequently, the CDRF shifts the interpretation of basalt carbonation from a static comparison of minerals to a dynamic understanding of the processes controlling their evolution during CO₂ mineralization.

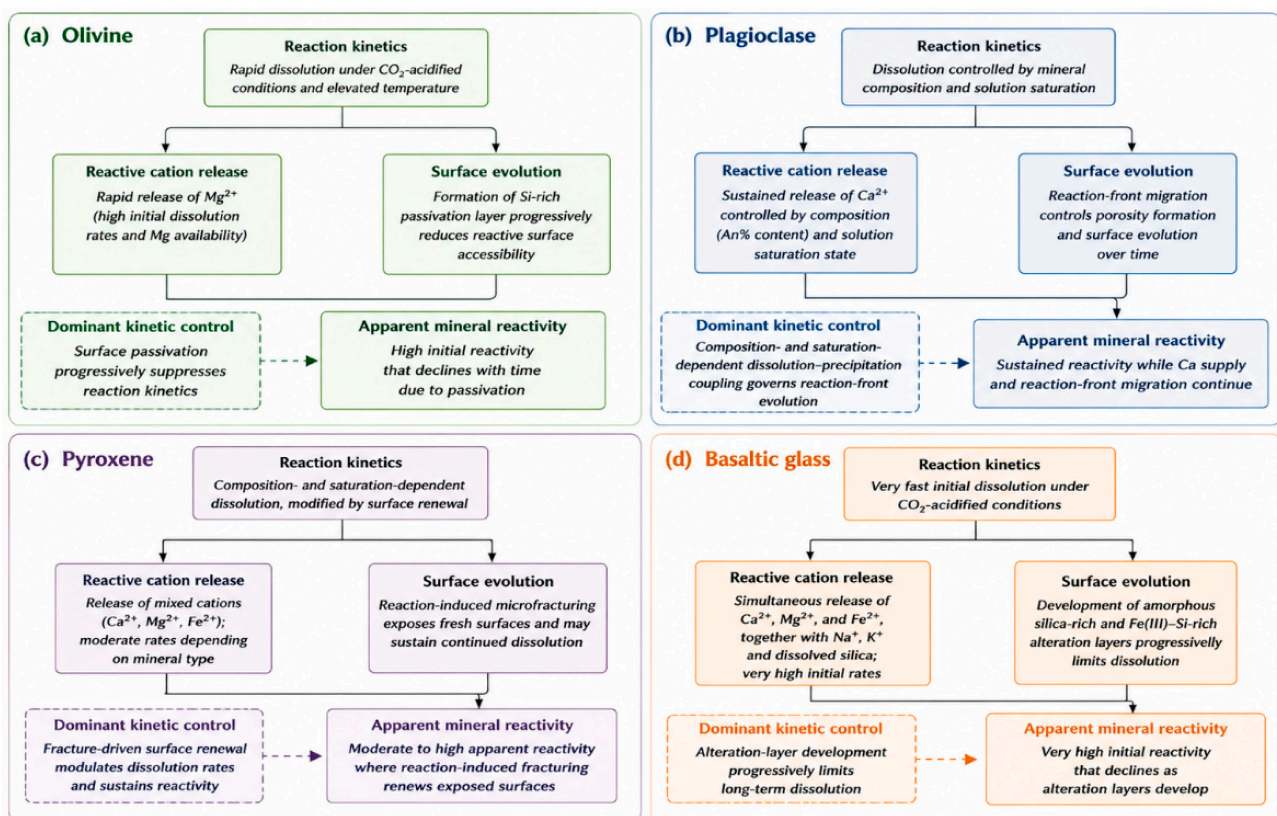


Figure 2. Mineral-specific kinetic controls governing the apparent reactivity of the principal reactive phases of basalt within the Condition-Dependent Reactivity Framework (CDRF).

4.1. Mineral-Specific Kinetic Constraints Governing Apparent Reactivity

Figure 2 demonstrates that although all basaltic reactive phases undergo dissolution upon exposure to CO₂-rich fluids, the mechanisms controlling the persistence of reaction kinetics differ fundamentally among them. Olivine exhibits rapid initial dissolution owing to its weakly polymerized nesosilicate structure and efficient Mg²⁺ release. However, this behaviour is progressively constrained by the formation of silica-rich passivation layers, which reduce reactive surface accessibility and suppress long-term dissolution. Consequently, the apparent reactivity of olivine is eventually governed not by its high intrinsic dissolution susceptibility but by the rate at which surface passivation develops.

Plagioclase follows a markedly different pathway. Its long-term reactivity is governed primarily by mineral composition and reaction-front evolution. The dissolution behaviour of anorthite-rich plagioclase promotes sustained Ca release, while the simultaneous de-

development of secondary porosity, Al-rich alteration products, and calcite precipitation continuously modifies transport pathways and reactive interfaces. These coupled processes explain why plagioclase frequently exhibits lower initial dissolution rates than olivine but may maintain significant carbonation potential over extended reaction periods.

Pyroxene demonstrates another distinct kinetic pathway in which reaction-induced fracturing becomes the dominant controlling mechanism. Unlike the other reactive phases, dissolution generates new fractures and microcracks that expose fresh reactive surfaces, partially compensating for the decline in dissolution rates expected as the system approaches equilibrium. Although pyroxene generally exhibits slower intrinsic dissolution kinetics than olivine and basaltic glass, reaction-induced surface renewal may sustain its apparent reactivity under favourable geological conditions.

Basaltic glass represents the most reactive amorphous constituent of fresh basalt and typically exhibits the highest initial dissolution susceptibility because of its metastable structure and lack of long-range crystallinity. Nevertheless, this initial advantage is progressively diminished by the formation of amorphous silica-rich surface layers (ASSLs) and Fe-rich alteration layers that increasingly restrict mass transfer and reactive surface accessibility. Therefore, despite its exceptionally rapid initial dissolution, the long-term behaviour of basaltic glass is finally constrained by alteration-layer development.

Collectively, these observations demonstrate that each reactive phase is governed by a unique dominant kinetic constraint, highlighting that apparent mineral reactivity cannot be generalized using a single dissolution-based ranking. Instead, the controlling mechanism evolves according to the interaction between dissolution behaviour, surface evolution, and reactive cation release.

4.2. Initial Dissolution Does Not Necessarily Predict Long-Term Carbonation Performance

One of the principal insights emerging from the CDRF is that rapid initial dissolution does not necessarily translate into superior long-term CO₂ mineralization performance. Conventional assessments frequently assume that minerals exhibiting the highest dissolution rates are also the most favourable carbonation candidates. However, the mechanistic pathways summarized in Figure 2 demonstrate that reaction kinetics continuously evolve as mineral surfaces, fluid chemistry, and secondary phases change during carbonation.

For example, both olivine and basaltic glass display rapid initial dissolution but subsequently experience substantial kinetic slowdown due to progressive surface passivation and alteration-layer development. Conversely, plagioclase may exhibit comparatively slower initial dissolution while sustaining prolonged Ca release through reaction-front migration and secondary porosity development. Similarly, pyroxene maintains reactive surface exposure through reaction-induced fracturing, partially mitigating the kinetic slowdown commonly associated with near-equilibrium conditions. These contrasting behaviours illustrate that apparent mineral reactivity is a dynamic property controlled by the evolution of multiple coupled processes.

The CDRF therefore provides a more comprehensive interpretation of basalt carbonation by recognizing that reaction kinetics represent only one stage within a continuously evolving mineralization pathway. The efficiency of CO₂ sequestration depends equally on the persistence of reactive surface area, the availability of carbonate-forming cations, and the evolution of secondary mineral phases throughout the reaction.

4.3. Insights from CarbFix

Insights from the CarbFix field demonstration further support the condition-dependent philosophy underpinning the proposed CDRF. CarbFix demonstrated that rapid and extensive mineral carbonation can be achieved under relatively low-temperature basaltic

reservoir conditions through optimization of the overall thermo-hydro-geochemical environment, including dissolved CO₂ injection, fluid–rock interaction, and groundwater chemistry. The observed high mineralization efficiencies therefore reinforce the view that reservoir-scale carbonation is governed by the coupled evolution of reaction kinetics, reactive cation release, and surface processes under site-specific conditions, rather than by the intrinsic reactivity of an individual basaltic constituent alone. Consequently, the CDRF provides a conceptual framework for interpreting field observations such as CarbFix by emphasizing process-driven reactivity instead of universal mineral ranking [49,50].

4.4. Engineering Implications and Future Basalt Carbonation Research

The mechanistic interpretation provided by the CDRF has important implications for both laboratory investigations and reservoir-scale CO₂ mineralization studies. As a practical example, the CDRF can guide both laboratory experimental design and reservoir evaluation by identifying the dominant controls expected under different geological settings. For shallow, low-temperature basalt formations, experimental programs should prioritize mineral composition, pH evolution, and reactive surface accessibility because dissolution kinetics are expected to dominate carbonation behaviour. Conversely, for deep, high-temperature reservoirs, greater emphasis should be placed on CO₂ pressure, fluid salinity, secondary mineral precipitation, and temporal evolution, as these factors increasingly govern long-term mineral trapping efficiency. The framework enables researchers to tailor experimental conditions, interpret conflicting observations consistently, and identify the controlling processes most relevant to specific geological environments. This condition-dependent approach may also support reservoir screening and reactive transport model parameterization by linking site-specific geological conditions to the dominant mechanisms controlling mineral carbonation.

Beyond experimental investigations, the framework may also provide a mechanistic basis for emerging predictive approaches, including reactive transport simulations and data-driven models, by helping identify the dominant reaction controls that should be represented under different geological conditions [6]. As data-driven approaches for CO₂ mineralization continue to emerge, the CDRF may also provide a physically interpretable framework for feature selection, model development, and interpretation of machine-learning predictions by linking model behaviour to the governing mineralogical and geochemical processes [51].

Future experimental studies should therefore adopt integrated characterization strategies capable of simultaneously monitoring dissolution kinetics, evolving reactive surface area, alteration-layer formation, reactive cation release, and carbonate precipitation under representative thermo-hydro-geochemical conditions. Such datasets would facilitate quantitative implementation of the CDRF and enable direct comparison of the principal reactive phases of basalt under standardized experimental conditions.

5. Conclusions

The following conclusions can be drawn from this perspective:

- i. The long-standing question of identifying a universally “most reactive” basaltic mineral is fundamentally inappropriate because mineral reactivity is condition-dependent. Apparent differences reported across previous studies primarily arise from variations in thermo-hydro-geochemical conditions, mineral characteristics, surface evolution, and reaction timescales.
- ii. The proposed CDRF provides a unified conceptual basis for interpreting basalt carbonation by integrating the key controls governing apparent mineral reactivity. The framework demonstrates that thermo-hydro-geochemical conditions, intrinsic mineral

- characteristics, surface accessibility, and temporal evolution collectively regulate the governing processes of reaction kinetics, reactive cation release, and surface evolution.
- iii. The four principal reactive phases of basalt exhibit distinct kinetic pathways and dominant reaction constraints during CO₂ mineralization. Olivine is primarily constrained by silica-rich passivation, plagioclase by mineral composition and reaction-front evolution, pyroxene by reaction-induced fracturing, and basaltic glass by alteration-layer development, highlighting that different mechanisms govern apparent reactivity across basaltic phases.
 - iv. Initial dissolution rates alone are insufficient for evaluating long-term carbonation performance. Sustained CO₂ mineralization depends on the persistence of reaction kinetics, the continuous availability of reactive cations, and the evolution of reactive surfaces throughout the mineralization process, demonstrating that apparent mineral reactivity is a dynamic, condition dependent property.
 - v. The CDRF shows that pyroxene should not be considered an unreactive phase. Although it dissolves more slowly than other basaltic phases, reaction-induced fracturing can expose fresh reactive surfaces and help sustain carbonation under suitable reservoir conditions.
 - vi. The CDRF provides a practical framework for future experimental design and reservoir-scale evaluation of in situ CO₂ mineralization. Future studies should adopt integrated approaches that simultaneously evaluate reaction kinetics, reactive cation release, surface evolution, and their temporal evolution under representative thermo-hydro-geochemical conditions.

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