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Wettability-aware phase-field micro-continuum modeling of two-phase carbonate acidizing with residual CO₂



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ABSTRACT

Following pre-CO₂ fracturing of carbonate reservoirs, residual CO₂ remaining in and remobilized from the wellbore-fracture system may introduce multiphase-flow and interfacial effects that are often neglected in conventional acidizing models. In this study, we develop a pore-scale micro-continuum framework that couples the Darcy-Brinkman-Stokes equations with a wettability-aware phase-field formulation. A key feature of the model is an implicit wetting scheme that allows the contact angle to evolve consistently with the receding fluid–solid interface during mineral dissolution, thereby addressing the moving contact-line problem. The simulations show that the CO₂ phase fundamentally reshapes acid migration and dissolution patterns through two competing mechanisms: flow diversion, whereby CO₂ clusters redirect acid into non-preferential pores, and bubble shielding, whereby adhered CO₂ shields reactive mineral surfaces. As a result, CO₂-bearing systems exhibit compact dissolution patterns and fluctuating permeability evolution driven by the competition between capillary trapping and pore enlargement. Surface wettability is further identified as a primary control factor: strongly water-wet conditions facilitate CO₂ mobilization, whereas wettability reversal promotes inlet blockage. In addition, parametric analysis reveals a nonlinear trade-off between acid concentration and injection rate. Specifically, increasing acid concentration under high-velocity injection aggravates live-acid bypass and reduces acid-utilization efficiency. These results provide mechanistic insight into gas-liquid-rock interactions and offer a physically grounded basis for optimizing acidizing strategies in carbonate reservoirs after pre-CO₂ fracturing.

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1. Introduction

Carbonate reservoirs represent globally significant hydrocarbon resources. Because of their low permeability, hydraulic fracturing is widely employed to enhance fluid flow and improve production performance (Soulaine et al., 2019; Carrillo et al., 2020; Zhang et al., 2023; Yan et al., 2025; Zhao et al., 2025a). Among these techniques, pre-pad CO₂ fracturing has gained increasing

attention owing to its high flowback efficiency and reduced formation damage (Zang et al., 2024; Yang et al., 2025a; Zhao et al., 2025b). Subsequent acidizing can further improve reservoir permeability by dissolving carbonate minerals and enlarging conductive flow paths (Zhang et al., 2022; Li et al., 2024; Liu et al., 2025). However, after pre-CO₂ fracturing, CO₂-bearing conditions may persist within fractures and pore spaces (Dudun and Feng, 2024; Zhang et al., 2024a). The presence of the CO₂ phase introduces complex physicochemical effects on acid–rock interactions. CO₂ bubbles adhering to mineral surfaces can shield reactive sites and suppress local dissolution, while the non-wetting CO₂ phase can alter local flow paths and acid distribution, thereby limiting wormhole propagation and promoting more

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compact dissolution patterns than those observed under single-phase conditions (Soulaine et al., 2018; You and Lee, 2021a; Zhang et al., 2024b). Understanding these pore-scale mechanisms is therefore essential for optimizing acidizing design in carbonate gas reservoirs after pre-CO₂ fracturing.

To investigate acid-carbonate rock interactions, researchers have developed both macroscale and pore-scale models (Liu et al., 2017; Jia et al., 2021; Xu et al., 2021; Asghari and Jahanbakhshi, 2025). Macroscale models capture dissolution front propagation at the reservoir scale but fail to resolve the fundamental pore-scale physicochemical processes, such as dynamic pore structure evolution and multiphase interactions. By contrast, pore-scale models explicitly solve fluid flow, solute transport, and chemical reaction kinetics in realistic pore geometries, offering deeper mechanistic insight into acidizing dynamics.

Various pore-scale approaches have been developed for reactive dissolution, including the lattice Boltzmann method (LBM), arbitrary Lagrangian-Eulerian (ALE) formulations, phase-field methods, and micro-continuum models based on DBS equations (Gao et al., 2017; Soulaine et al., 2017; Molins et al., 2021; Liu et al., 2022a; Zhang et al., 2024b; Wang et al., 2025a). These methods differ substantially in their treatment of moving interfaces, wettability, and topological evolution. LBM is particularly advantageous for complex pore geometries and multiphase interfacial dynamics, because fluid interfaces can be handled naturally and multiple sub-processes can be coupled within a unified numerical framework, but faces stability challenges at high Damköhler numbers and high-density ratios (Gao et al., 2017; Yang et al., 2021; Cai et al., 2025). While ALE formulations provide high accuracy for moving boundaries in single-phase flows, they entail prohibitive computational costs and mesh entanglement issues when handling topological changes (e.g., grain disappearance or coalescence) in 3D pore networks (Soulaine et al., 2018; Etancelin et al., 2020; Jyoti and Haese, 2021; Li et al., 2023). Phase-field methods are attractive because they can naturally describe diffuse interfaces and topological transitions, and are therefore well suited for dynamic tracking of evolving fluid-solid interfaces during dissolution; however, when used alone in porous reactive systems, they still require consistent coupling with pore-scale flow and evolving porous resistance (Li et al., 2023).

More recently, the micro-continuum approach has emerged as a powerful pore-scale modeling framework by combining DBS equations with volume-averaging theory to describe flow and transport in evolving porous media (Soulaine et al., 2018; You and Lee, 2021a). This approach avoids explicit interface tracking while retaining pore-scale fidelity and the ability to capture flow, transport, and structural evolution in complex porous media (Soulaine et al., 2019). Existing studies have demonstrated that the micro-continuum framework can be extended to multiphase reactive flow and mineral dissolution, including interfacial tension effects and moving contact lines at mineral surfaces (Soulaine et al., 2018, 2021; Soulaine, 2024).

Nevertheless, a critical methodological bottleneck persists in DBS-based multiphase reactive dissolution simulations: the stable, consistent characterization of wettability as fluid-solid boundaries continuously evolve during mineral dissolution. Conventional wettability enforcement schemes rely predominantly on explicit wall-normal corrections, which can reliably impose prescribed contact angle constraints under fixed solid boundary conditions; however, these methods exhibit notable limitations in preserving interfacial continuity, resolving moving contact lines, and capturing reactive front propagation when solid boundaries advance continuously via dissolution (Liu et al., 2022b; Liu, 2024). For conventional CFD, ALE, and other explicit-interface formulations, contact angles are typically prescribed via explicit boundary

conditions at the fluid-solid interface, which require repeated reconstruction and updating as the solid skeleton evolves—this makes it extremely challenging to maintain a physically consistent representation of wettability across dynamic gas-liquid-solid contact regions. Furthermore, several existing micro-continuum studies employ simplified or homogenized descriptions of gas-liquid interfaces, which severely limit the ability to resolve discontinuous gas morphology, dynamic bubble redistribution, and wettability-governed shielding effects within dissolving pore spaces (Maes et al., 2022; Yang et al., 2022; Zhang et al., 2024a). Against this background, the present modeling strategy combines the continuum advantages of the DBS framework with a phase-field formulation for explicit interfacial description. This provides a robust, fit-for-purpose framework to capture interfacial tension, dynamic gas-cluster evolution, and wettability-dependent shielding during carbonate dissolution, while entirely avoiding the need for repeated remeshing or reassignment of wetting conditions on evolving solid surfaces.

In this work, we develop a pore-scale micro-continuum framework that explicitly couples DBS flow with a phase-field formulation. To address the limitations of explicit boundary-based wetting treatments in dissolving porous media, we implement an implicit wetting scheme embedded within the phase-field framework. This treatment allows the contact angle to evolve naturally with the moving fluid-solid boundary, ensuring a more accurate and consistent representation of wettability at dynamic gas-liquid-solid interfaces. In this way, wettability is incorporated into the interfacial free-energy description rather than being imposed solely through repeated explicit boundary reconstruction. Using this framework, we investigate the mechanistic role of CO₂ in carbonate acidizing, specifically the bubble shielding effect and flow diversion, with emphasis on its blocking-reopening behavior and impact on reactive flow. The results provide fundamental insights into how CO₂ modifies pore-scale reactive transport and dissolution patterns, and offer physically grounded guidance for optimizing acid concentration, injection strategy, and gas management in carbonate gas reservoirs.

2. Physicochemical processes

This study investigates carbonate acidizing under a CO₂-bearing condition that remains after pre-CO₂ fracturing. Here, the term residual CO₂ is used in an engineering sense to denote CO₂ that remains in the wellbore-fracture-near-wellbore system and can be intermittently remobilized during subsequent acid injection, rather than a strictly immobile trapped phase. At the pore-scale model level, this behavior is represented by periodic CO₂ supply at the inlet, which serves as an idealized boundary condition for episodic entry and migration of residual CO₂ into the local porous domain. The model therefore focuses on the transport and interfacial consequences of a post-fracturing CO₂-bearing environment, as illustrated in Fig. 1.

Based on this engineering context and the available literature, the following assumptions and modeling treatments are adopted:

- (1) CO₂-bearing inflow representing remobilized residual CO₂. The injected system is modeled as an immiscible CO₂-liquid two-phase flow. The CO₂ phase is introduced through periodic inlet pulses, which idealize the intermittent remobilization and local entry of residual CO₂ from the wellbore/fracture system during acidizing. Accordingly, the local flow regime and gas saturation in the pore-scale domain are governed by the imposed CO₂-entry schedule rather than by a prescribed static trapped-gas distribution.

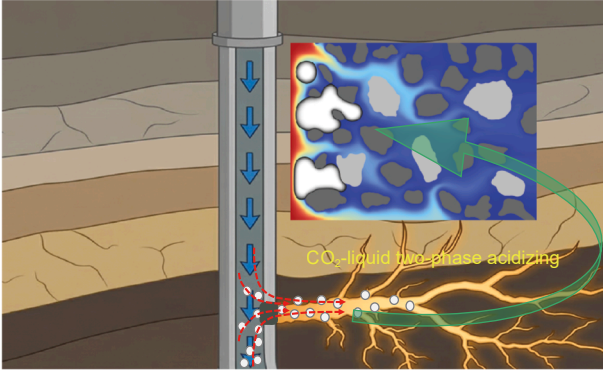


Fig. 1. Schematic of gas-liquid two-phase acidizing in tight carbonate rock after hydraulic fracturing.

- (2) Evolving pore geometry. The pore geometry, porosity, permeability, and flow characteristics of the entire medium evolve due to chemical reactions occurring at the solid boundaries, with mineral dissolution primarily driven by reactions at the calcite surface.
- (3) Liquid-phase reaction dominance. Carbonate minerals on the rock surface within the acidizing zone primarily interact with the acid liquid phase. Accordingly, fluid flow, mass transfer, and heterogeneous reactions occur within the acid phase, while the CO_2 phase influences the process by altering the flow field and interfacial coverage.
- (4) Treatment of reaction-generated CO_2 . Previous studies have shown that the CO_2 produced by the calcite-acid reaction dissolves rapidly into the liquid phase or reaches local phase equilibrium over a sufficiently short timescale (Yan et al., 2019; Alarji et al., 2022; Khojastehmehr et al., 2025). Reaction-generated CO_2 is not explicitly resolved in the present model. This simplification is adopted to isolate the hydrodynamic and interfacial effects associated with externally supplied or remobilized residual CO_2 during post-fracturing acidizing. Under the short pore-scale times and injection-controlled conditions considered here, the imposed CO_2 phase is treated as the dominant contributor to gas-liquid morphology and flow redistribution, whereas the additional contribution from reaction-generated CO_2 is neglected in the governing equations. The present model should therefore be interpreted as a focused description of CO_2 -bearing transport and wettability effects, rather than a complete compositional model of all CO_2 sources.

3. Numerical setup and discretization

3.1. Development of the pore-scale two-phase acid-rock reaction model

Under post-fracturing CO_2 -bearing conditions, carbonate acidizing is modeled as an immiscible CO_2 -liquid reactive system. In the present model, the CO_2 phase is introduced through periodic inlet injection rather than as a pre-existing trapped phase in the pore space. The model therefore focuses on the transport, interfacial, and reactive effects induced by the CO_2 phase during two-phase acidizing, while CO_2 generated by the acid-rock reaction is not considered.

The pore geometry is represented by a scalar porosity field, ε , which denotes the local pore-volume fraction at the pore scale.

Unlike bulk porosity defined at the core or reservoir scale, ε varies continuously between 0 and 1 and serves as a micro-continuum indicator distinguishing fluid and solid regions. The porosity and solid fraction satisfy:

$$\varepsilon + \varepsilon_s = 1 \quad (1)$$

Symbols: ε is the local porosity; ε_s is the solid fraction. Here, $\varepsilon = 1$ indicates free flow, $0 < \varepsilon < 1$ denotes mixed zones with partial solid obstruction, and $\varepsilon \rightarrow 0$ corresponds to solid regions with negligible flow. To avoid numerical singularities, a very small porosity ($\varepsilon \approx 10^{-6}$) is assigned to fully solid cells. This scalar formulation is advantageous for two-phase dissolution simulations because it enables smooth tracking of evolving interfaces without dynamic remeshing.

Within each cell, liquid and gas occupy distinct fractions, such that:

$$\alpha_w + \alpha_g = 1 \quad (2)$$

where, α_w and α_g are defined as intrinsic phase fractions within the local pore space, such that $\alpha_w + \alpha_g = 1$ in fluid-accessible regions. The corresponding cell-averaged phase holdups over the whole computational volume are therefore $\varepsilon\alpha_l$ and $\varepsilon\alpha_g$, respectively. Combined with Eq. (1), the local volume closure for each grid cell can be written as $\varepsilon_s + \varepsilon\alpha_w + \varepsilon\alpha_g = 1$. In the phase-field representation, the pore-space phase fractions are related to the order parameter ω through $\alpha_w = (1 - \omega)/2$ and $\alpha_g = (1 + \omega)/2$, so that $\omega = -1$ corresponds to the liquid phase and $\omega = 1$ to the CO_2 phase. Inside the solid skeleton, ω is retained only as a diffuse-field variable, while its evolution is suppressed by the immersed-boundary switching treatment introduced later in Eqs. (25)–(29).

Because the liquid and gas fractions evolve continuously during two-phase transport and mineral dissolution, mass conservation is formulated at the phase level rather than by directly imposing a divergence-free mixture condition. Let $\varepsilon_{fw} = \varepsilon\alpha_w$ and $\varepsilon_{fg} = \varepsilon\alpha_g$ denote the liquid- and gas-phase volumetric fractions in the total computational volume, respectively, with $\varepsilon_{fw} + \varepsilon_{fg} = \varepsilon$. Under the present isothermal immiscible-displacement assumption, without explicit interphase mass transfer and without explicit treatment of reaction-generated CO_2 , the phase mass-balance equations are written as:

$$\begin{aligned} \frac{\partial \varepsilon_{fw} \rho_w}{\partial t} + \nabla \cdot (\varepsilon_{fw} \rho_w \bar{u}) &= m_s \\ \frac{\partial \varepsilon_{fg} \rho_g}{\partial t} + \nabla \cdot (\varepsilon_{fg} \rho_g \bar{u}) &= 0 \end{aligned} \quad (3)$$

where m_s is the volumetric source term associated with fluid-solid interfacial reaction in the liquid-phase mass balance. Summation of the two phase-balance equations gives the mixture continuity equation:

$$\frac{\partial \varepsilon \rho}{\partial t} + \nabla \cdot (\varepsilon \rho \bar{u}) = m_s \quad (4)$$

where ρ is the mixture density and $\bar{u}$ is the mass-averaged mixture velocity.

In the present one-fluid micro-continuum framework, impermeability in the solid region is not imposed through a separate sharp-wall boundary condition, but is recovered continuously through the porosity field ε together with the Darcy-resistance term in the governing momentum equation as $\varepsilon \rightarrow 0$. The velocity field is obtained from the DBS formulation, which incorporates porosity-dependent inertial effects, viscous stress, capillary forcing, and Darcy drag (Wang et al., 2024; Geng et al., 2025; Xu et al., 2025; Yang et al., 2025b):

$$\frac{\rho}{\varepsilon} \left(\frac{\partial \bar{u}}{\partial t} + \frac{\bar{u}}{\varepsilon} \cdot \nabla \bar{u} \right) = -\nabla \bar{p} + \frac{\mu}{\varepsilon} \nabla^2 \bar{u} + f_{st} - \mu k^{-1} \bar{u} \quad (5)$$

where, ρ is the mixture density, kg/m³; ε is the local porosity; m/s; $\bar{u}$ is the mass-averaged mixture velocity, m/s; $\bar{p}$ is the averaged pressure, Pa; μ is the effective viscosity, Pa·s; k is the permeability tensor, m²; f_{st} is the capillary force, N/m³; t is time, s. The effective density and viscosity appearing in Eq. (5) are evaluated from the local phase distribution by smooth phase-field interpolation:

$$\begin{aligned} \rho(\omega) &= \alpha_w \rho_w + \alpha_g \rho_g = \frac{(1-\omega)\rho_w + (1+\omega)\rho_g}{2} \\ \mu(\omega) &= \alpha_w \mu_w + \alpha_g \mu_g = \frac{(1-\omega)\mu_w + (1+\omega)\mu_g}{2} \end{aligned} \quad (6)$$

This interpolation ensures continuous property variation across the diffuse CO₂-liquid interface and is fully consistent with the phase-field description of the two-phase system.

The gas-liquid interface is resolved using the phase-field method (von Wolff and Pop, 2022; Li et al., 2023). The order parameter ω evolves according to:

$$\varepsilon \frac{\partial \omega}{\partial t} + \nabla(\omega \bar{u}) = \nabla \cdot (M \nabla G(\omega)) \quad (7)$$

$$G(\omega) = \frac{\lambda}{\delta^2} F(\omega) - \lambda \Delta \omega = \frac{\lambda}{\delta^2} (\omega(\omega^2 - 1) - \delta^2 \nabla^2 \omega) \quad (8)$$

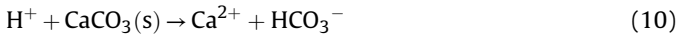
Here, the mobility M controls the diffusive relaxation timescale in the Cahn-Hilliard equation and is commonly expressed as $M = \zeta \delta^2$, where δ is the capillary thickness and ζ is a mobility tuning parameter (Gao et al., 2023a). The order parameter ω varies between -1 and 1 and describes the transition from the wetting phase to the non-wetting phase.

The corresponding capillary force in the momentum equation is (Yue et al., 2006; Gao et al., 2023b):

$$f_{st} = G(\omega) \nabla \omega \quad (9)$$

where, f_{st} denotes the interfacial force, N/m³; $G(\omega)$ is the chemical potential.

During acidizing, mineral dissolution modifies the pore structure. The primary reaction is the irreversible calcite dissolution under HCl injection (Molins et al., 2021):



Since H^+ is the key reactive species, its transport is governed by a mass balance equation (You and Lee, 2021b):

$$\frac{\partial(\varepsilon \bar{c}_f)}{\partial t} + \nabla \cdot (\bar{u} \cdot \bar{c}_f) = \nabla \cdot (\varepsilon D_k \nabla \bar{c}_f) - R(c) \quad (11)$$

where, ε is the porosity; $\bar{c}_f$ is the H^+ concentration, mol/m³; D_k is the effective diffusion coefficient of H^+ in the fluid phase, m²/s; $R(c)$ is the reaction rate, mol/(m³·s).

The reaction rate is expressed as (Zhang et al., 2024c):

$$R(c) = a_v k_H \bar{c}_f \quad (12)$$

where, a_v is the effective surface area per unit volume within the grid cell, m²/m³; k_H is the reaction rate constant, m/s.

Mineral dissolution simultaneously alters porosity, permeability, and surface area. The governing relations are (Soulaine et al., 2018; You and Lee, 2021c):

$$\frac{\partial \varepsilon \rho_s}{\partial t} = -m_s = -\beta R(c) \quad (13)$$

where, ρ_s denotes solid density, kg/m³; β is the stoichiometric coefficient, kg/mol.

$$k^{-1} = k_0^{-1} \frac{(1-\varepsilon)^2}{\varepsilon^3} \quad (14)$$

where, k_0 is the reference permeability, m².

$$\begin{aligned} a_v &= \|\nabla \varepsilon\| \psi \\ \psi &= 4\varepsilon(1-\varepsilon) \end{aligned} \quad (15)$$

where, ψ is the diffuse-interface function which is used to enforce the localization of the reactive boundary solution at the liquid-solid interface.

Together, Eqs. (1)–(15) define a closed reactive-transport framework in which flow drives species transport, interfacial dynamics govern CO₂-liquid redistribution, dissolution updates the pore structure, and the evolving structure feeds back into transport and reaction. This framework captures the essential features of CO₂-bearing two-phase acidizing, including pore enlargement, wormhole initiation, and the dynamic interplay among flow, transport, reaction, and evolving pore geometry.

3.2. Phase-field model incorporating interfacial tension and wettability conditions

Conventional micro-continuum models often homogenize pore-scale geometry and therefore fail to represent the effects of interfacial tension and wettability on two-phase reactive transport explicitly. To overcome this limitation, the present model employs a phase-field formulation with immersed boundary treatment, in which wettability effects are introduced directly into the free energy functional. This treatment allows the diffuse interface to recover realistic wetting behavior without explicitly prescribing contact-angle boundary conditions on sharp solid walls.

A contact angle θ forms at the interface between the two-phase fluids (w, g) and the solid surface γ (as shown in Fig. 2). At equilibrium, the net force on any element of the triple line vanishes. The relationship between the fluid-solid contact angle and interfacial tensions is described by Young's equation along the directions perpendicular to the triple line for the w_g , w_γ , and g_γ interfaces:

$$\sigma_{wg} \cos \theta = \sigma_{g\gamma} - \sigma_{w\gamma} \quad (16)$$

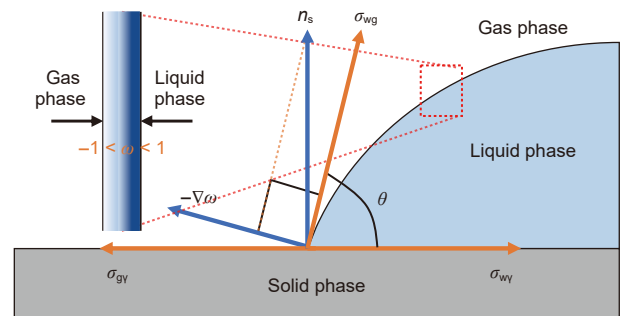


Fig. 2. Schematic diagram of contact angle at the triple junction.

where, σ_{wg} is the liquid-gas interfacial tension coefficient, N/m; σ_{wy} is the liquid-solid interfacial tension coefficient, N/m; σ_{gy} is the gas-solid interfacial tension coefficient, N/m.

Multicomponent immiscible fluid models often employ the multi-component Cahn-Hilliard equation for numerical simulation. A common approach fixes one component within the domain containing solids, treating the region where the order parameter corresponds to this fixed component as a solid obstacle or arbitrary domain. Thus, solving for the remaining fluid components describes the flow state and interfacial contact relationships within the domain. Leveraging this concept, ε_s is used as the fixed component to interpret the contact relationship at the fluid-solid interface in this model. If the contours of the order parameters in the diffuse interface remain parallel, the interface normal vector $\mathbf{n}$ can be expressed using the gradient of the introduced fixed variable $\mathbf{n}_s$ (the normal to the solid surface).

$$\mathbf{n}_s = \nabla \varepsilon_s / |\nabla \varepsilon_s| \quad (17)$$

At the contact line, the contact angle can be geometrically computed from ε_s as:

$$\tan\left(\frac{\pi}{2} - \theta\right) = \frac{-\mathbf{n}_s \cdot \nabla \omega}{|\nabla \omega - (\mathbf{n}_s \cdot \nabla \omega) \mathbf{n}_s|} \quad (18)$$

Eq. (18) gives the geometric relation for contact-angle evaluation and remains accurate when the diffuse interface is sufficiently resolved and close to local equilibrium near the solid surface. By substituting Eq. (17) into Eq. (18), the formula can also be rewritten as (Gao et al., 2021):

$$\mathbf{n}_s \cdot \nabla \omega = \frac{\nabla \varepsilon_s \cdot \nabla \omega}{|\nabla \varepsilon_s|} = |\nabla \omega| \cos \theta \quad (19)$$

The equilibrium condition of the diffuse interface gives $F(\omega) = \delta^2 |\nabla \omega|^2 / 2$ in the near-wall region, which can be rearranged to obtain the explicit expression of the order parameter gradient modulus:

$$|\nabla \omega| = \frac{\sqrt{2F(\omega)}}{\delta} = -\frac{(\omega^2 - 1)}{\sqrt{2}\delta} \quad (20)$$

Here, $F(\omega)$ denotes the dimensionless symmetric double-well free energy density function. Substituting Eq. (20) back into Eq. (19) and rearranging leads to:

$$\nabla \varepsilon_s \cdot \nabla \omega + \frac{(\omega^2 - 1)|\nabla \varepsilon_s| \cos \theta}{\sqrt{2}\delta} = 0 \quad (21)$$

Eq. (21) is termed the surface energy formula for calculating microscale contact angles. It is equivalent to calculating the contact angle from a given ω distribution near the contact line. This formulation helps better reconcile the energy structure described by Young's equation Eq. (16) and effectively avoids numerical computation of $|\nabla \omega|$. Since the right-hand side of Eq. (21) is zero, it can be treated as a "zero contribution term" and added back into the chemical potential $G(\omega)$:

$$\begin{aligned} G(\omega) &= \frac{\lambda}{\delta^2} F(\omega) - \lambda \Delta \omega + \delta^2 \nabla \varepsilon_s \cdot \nabla \omega + \frac{\delta(\omega^2 - 1)|\nabla \varepsilon_s| \cos \theta}{\sqrt{2}} \\ &= \frac{\lambda}{\delta^2} \left(F(\omega) + \frac{\delta}{\sqrt{2}} (\omega^2 - 1) |\nabla \varepsilon_s| \cos \theta - \delta^2 \nabla \cdot ((1 - \varepsilon_s) \nabla \omega) \right. \\ &\quad \left. - \delta^2 \varepsilon_s \Delta \omega \right) \end{aligned} \quad (22)$$

where, the term $\Delta \omega$ is rewritten using Young's equation $\nabla \cdot ((1 - \varepsilon_s) \nabla \omega) + \nabla \varepsilon_s \cdot \nabla \omega + \varepsilon_s \Delta \omega$ and substituted into the polynomial

for decomposition. In the free-flow region, ε_s is zero, Eq. (22) reduces to the original chemical potential form. For the diffuse interface, as it narrows with mesh refinement, the term $\delta^2 \varepsilon_s \Delta \omega$ in Eq. (22) does not contribute significantly to mass conservation or wettability (i.e., it does not explicitly contain θ). As ε_s tends to zero, the last term in Eq. (22) also vanishes. Therefore, for computational convenience, $\delta^2 \varepsilon_s \Delta \omega$ is neglected, and the expression is revised to:

$$G(\omega) = \frac{\lambda}{\delta^2} \left(F(\omega) + \alpha(\omega) + \frac{\delta}{\sqrt{2}} (\omega^2 - 1) |\nabla \varepsilon_s| \cos \theta - \delta^2 \nabla \cdot ((1 - \varepsilon_s) \nabla \omega) \right) \quad (23)$$

Equation (23) imposes the wetting condition implicitly through the governing equations rather than through an explicit wall boundary condition. An additional Lagrange multiplier $\alpha(\omega)$ is introduced in Eq. (23) to ensure the model recovers its original conservative properties, expressed as:

$$\alpha(\omega) = \int_{\Omega} F(\omega) d\mathbf{x} / \left(\delta^2 \int_{\Omega} d\mathbf{x} \right) \quad (24)$$

Simultaneously, to suppress the evolution of ω within the solid region, the Cahn-Hilliard phase-field equation is rewritten as:

$$\varepsilon \frac{\partial \omega}{\partial t} + \nabla(\omega \bar{\mathbf{u}}) = \nabla \cdot ((1 - \varepsilon_s) M \nabla G(\omega)) \quad (25)$$

$$\begin{aligned} G(\omega) &= \frac{\lambda}{\delta^2} \left(F(\omega) + \alpha(\omega) + \frac{\delta}{\sqrt{2}} (\omega^2 - 1) |\nabla \varepsilon_s| \cos \theta \right. \\ &\quad \left. - \delta^2 \nabla \cdot ((1 - \varepsilon_s) \nabla \omega) \right) = \frac{\lambda}{\delta^2} \left(\omega (\omega^2 - 1) + \alpha(\omega) \right. \\ &\quad \left. + \frac{\delta}{\sqrt{2}} (\omega^2 - 1) |\nabla \varepsilon_s| \cos \theta - \delta^2 \nabla \cdot ((1 - \varepsilon_s) \nabla \omega) \right) \end{aligned} \quad (26)$$

where, $F(\omega)$ is a function (e.g., Heaviside) that is 1 in the fluid domain and 0 in the solid domain, ensuring ε only evolves where fluid is present and ε_s is fixed initially.

Considering the effect of the immersed boundary and inspired by the special property mentioned earlier, multiply the above $F(\omega)$ term by $(1 - \varepsilon_s)$ and revise it further as:

$$\varepsilon \frac{\partial \omega}{\partial t} + \nabla(\omega \bar{\mathbf{u}}) = \nabla \cdot ((1 - \varepsilon_s) M \nabla G(\omega)) \quad (27)$$

$$G(\omega) = P_a(\omega) + P_b(\omega, \varepsilon_s) + \alpha(\omega) \quad (28)$$

$$P_b(\omega, \varepsilon_s) = \delta \left(\omega^2 - 1 \right) |\nabla \varepsilon_s| \cos \theta / \sqrt{2} - \delta^2 \nabla \cdot ((1 - \varepsilon_s) \nabla \omega) \quad (29)$$

3.3. Model implementation and solution

In the present pore-scale simulations, the imposed periodic CO₂ entry should therefore be interpreted as an idealized representation of remobilized residual CO₂ migration under post-fracturing acidizing conditions. The coupled formulation was implemented according to the functional grouping of Eqs. (1)–(29). Eqs. (1)–(4) define the porosity field, solid fraction, and phase-volume constraints. Eq. (5) governs the Darcy-Brinkman-Stokes flow field. Eqs. (7)–(9) describe phase-field evolution and capillary effects. Eqs. (10)–(15) represent reactive transport, including calcite dissolution, hydrogen-ion transport, porosity evolution, permeability update, and effective surface-area decay. Eqs. (16)–(21) define the contact-angle condition and its diffuse-interface reformulation, while Eqs. (22)–(29) introduce wettability corrections into the chemical potential and phase-field equations.

As shown in Fig. 3, the simulation starts from a heterogeneous micrometer-scale porous geometry generated from the porosity-related definitions. The domain is discretized with sufficient resolution to resolve the diffuse gas-liquid interface and evolving solid boundary. Initial conditions are specified for the flow field, solute concentration, porosity-dependent properties, and phase distribution. At each time step, the flow field is updated by Eq. (5), the gas-liquid interface and wettability response are updated by Eqs. (7)–(9) and Eqs. (22)–(29), and reactive transport together with pore-structure evolution is solved by Eqs. (10)–(15). The updated porosity and phase distribution are then transferred to the subsequent time step.

The model was implemented in COMSOL Multiphysics using the finite element method. The computational domain was discretized with unstructured triangular elements. A segregated solution scheme was adopted, and the resulting algebraic system was solved using the direct sparse solver PARDISO. Time integration employed a variable-order Backward Differentiation Formula (BDF) scheme with a maximum order of 2. The initial and maximum allowable time steps were automatically adjusted to improve convergence robustness and to avoid numerical singularities. For two-phase simulations, the phase field was initialized prior to transient computation to define the initial gas-liquid interface morphology. The physical properties of the two fluid phases and the rock matrix are listed in Table 1.

3.4. Model validation

In conventional interface-tracking methods, the contact angle is typically imposed explicitly as a geometric boundary condition on sharp solid walls. In the present micro-continuum DBS framework, however, the solid skeleton is represented by a continuous volume-fraction field rather than a sharp boundary, which makes explicit wall-based wetting conditions inapplicable.

Table 1

Physical property settings for two-phase displacement simulation (Deng et al., 2016; Jin et al., 2021; Li and Jin, 2019; Rezaei et al., 2021).

Symbol	Value	Unit	Description
ρ_w	1010	kg/m ³	Liquid density
μ_w	0.656×10^{-3}	Pa·s	Liquid viscosity
ρ_{nw}	650	kg/m ³	CO ₂ density
μ_{nw}	0.471×10^{-3}	Pa·s	CO ₂ viscosity
ρ_s	2200	kg/m ³	Solid density
D_k	4.5×10^{-9}	m ² /s	Diffusion coefficient
k_0	1×10^{-15}	m ²	Initial permeability
β	1.37	kg/mol	Stoichiometric coefficient
θ	$\pi/3$	rad	Contact angle
σ	0.07	N/m	Surface tension coefficient
u	1×10^{-4}	m/s	Injection velocity
c_f	2000	mol/m ³	Inlet acid concentration
k_H	8.9×10^{-4}	m/s	Reaction rate constant

To address this issue, a modified phase-field formulation is introduced, in which wettability is incorporated implicitly through a solid-fluid interaction term in the free-energy functional. The contact angle therefore evolves consistently with the moving fluid–solid interface.

To assess the reliability of the proposed model, a hierarchical validation strategy is adopted. First, the implicit wettability treatment is verified using static droplet benchmarks. Second, numerical accuracy is evaluated against a high-fidelity ALE benchmark. Finally, physical consistency is examined by comparison with experimental observations.

3.4.1. Validation of implicit wettability control

To verify that the presented implicit formulation can accurately describe two-phase wettability behavior, a benchmark validation case is established to compare the consistency of the traditional phase-field method (Explicit BC) and the modified implicit phase-field method.

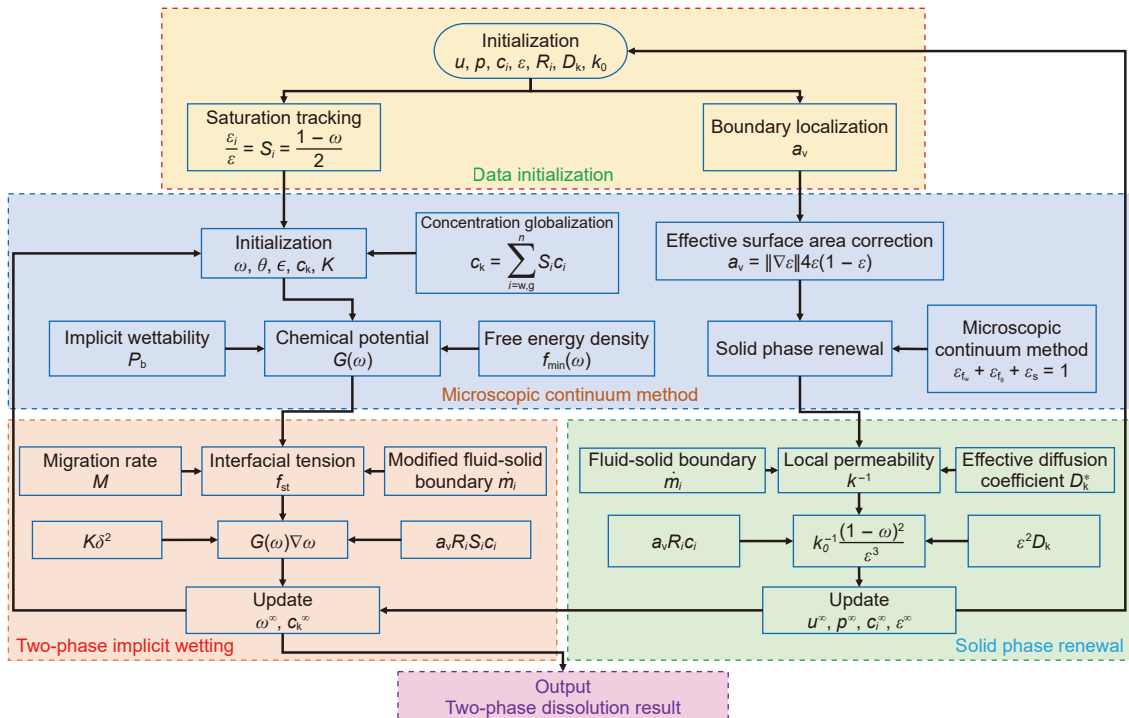


Fig. 3. Multi-physics coupling framework for two-phase reactive transport and rock dissolution.

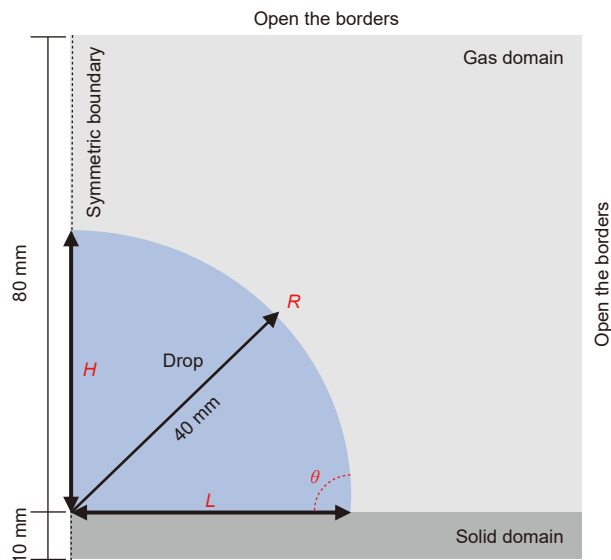


Fig. 4. Static droplet spreading model and triangular solid structure channel model containing a bubble.

Fig. 4 shows the computational geometry and the initial droplet configuration. In this benchmark, the spreading of a static droplet on a flat surface was simulated under gravity-free conditions. The droplet evolves toward an equilibrium shape governed by interfacial tension and the prescribed wall contact angle. Five contact angles, 30° , 60° , 90° , 120° , and 150° , were simulated to equilibrium. For a droplet of radius R , the equilibrium spreading height H and spreading radius L can be obtained analytically as functions of R and θ :

$$H = R \left(\frac{1 - \cos \theta}{1 + 0.5 \cos \theta} \right)^{1/3} \quad (30)$$

$$L = \frac{\sin \theta}{1 - \cos \theta} R \left(\frac{1 - \cos \theta}{1 + 0.5 \cos \theta} \right)^{1/3} \quad (31)$$

The numerical results of both methods were compared with the analytical solution. As shown in Figs. 5–7, both formulations reproduce the droplet spreading behavior under different wettability conditions, while the modified implicit model shows closer agreement with the analytical solution. The relative errors of the equilibrium spreading height and spreading radius are 1.61% and 1.82%, respectively, compared with 5.35% and 5.24% for the conventional formulation. These results confirm that the proposed implicit treatment can accurately impose wettability in the diffuse-interface framework.

3.4.2. Quantitative numerical validation with ALE benchmark

Although the reactive mass-transport solver for single-phase flow has been validated in our previous work (Fig. 8), additional verification is required for the present coupling among multiphase flow, interfacial dynamics, and mineral dissolution. For this purpose, the Arbitrary Lagrangian–Eulerian (ALE) method is adopted as a high-accuracy numerical benchmark.

A benchmark problem was constructed for the flow and dissolution of acidic fluid with continuously injected bubbles in a simple capillary medium, as shown in Fig. 9. A 2 mol/L acid solution was injected from the inlet at $u_0 = 0.001$ m/s, together with periodically injected non-reactive bubbles of 2 mm diameter at 1 s intervals. The interfacial tension was set to 0.07 N/m, and two contact angles, 60° and 120° , were tested.

Fig. 10 shows that the proposed model and the ALE benchmark produce highly consistent predictions for bubble position, morphology, wall-contact behavior, and the associated concentration distribution. Further quantitative comparison in Fig. 11 shows that the calculated solid-volume loss from the two methods is nearly identical, with an average relative error below 2%. These results demonstrate that the proposed model achieves numerical

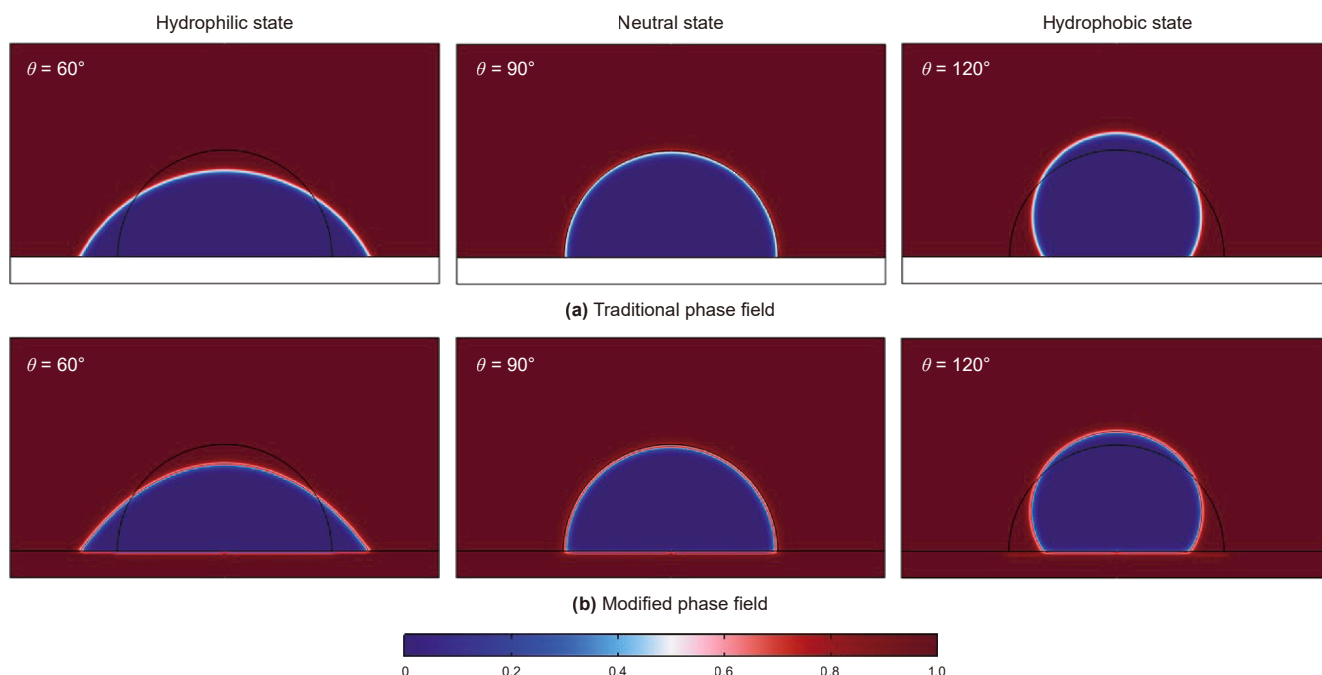


Fig. 5. Droplet-spreading contours at different contact angles.

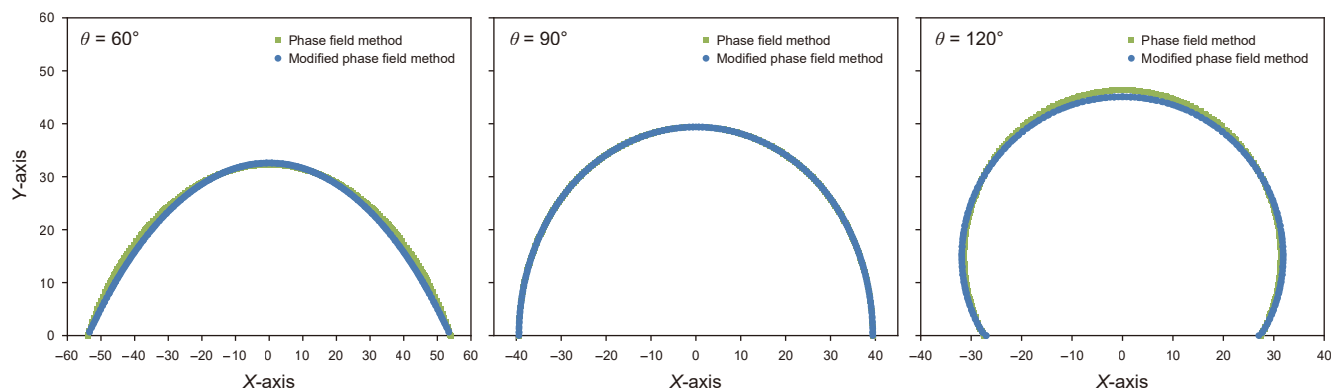


Fig. 6. Comparison of numerical results for droplet spreading under different contact angles.

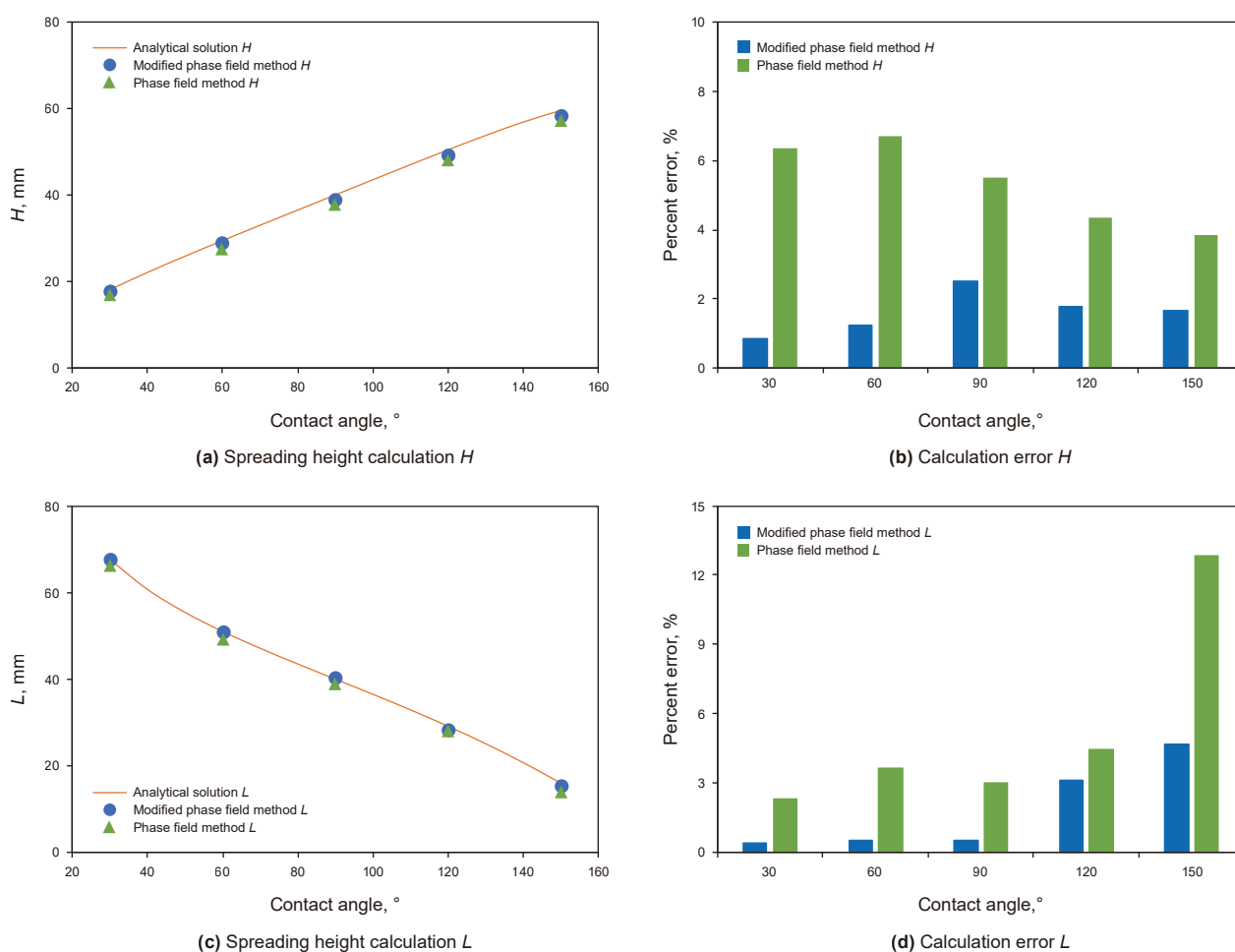


Fig. 7. Comparison of numerical and analytical solutions for the spreading height H and spreading radius L .

accuracy comparable to that of the ALE method for short-term moving-boundary reactive-flow problems.

3.4.3. Physical consistency validation with experiments

To further evaluate the physical consistency of the proposed model, the simulation results were compared with the two-phase acid-etching experiment (Zhou et al., 2025), as shown in Fig. 12(a).

In the experiment, CO_2 bubbles adhered to the mineral surface and locally inhibited acid-rock contact. The simulation reproduced

the same qualitative behavior, including bubble attachment to the solid surface, local shielding of the reactive interface, and coalescence of adjacent bubbles during transport. In addition, using a contact angle of 60° , consistent with the water-wet character of the experimental mineral surface, produced a three-phase contact morphology in good agreement with the experimental observations.

Overall, the comparison indicates that the proposed model captures the main pore-scale phenomena observed in the

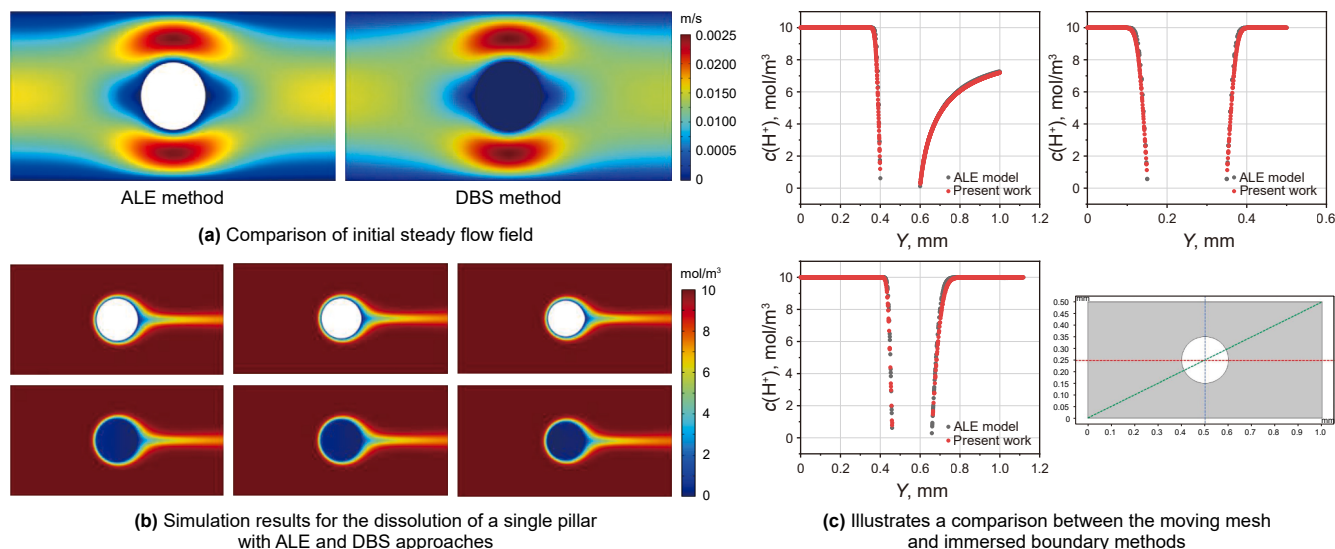


Fig. 8. Validation of the DBS method against the classical ALE method for moving-boundary dissolution (Wang et al., 2025b).

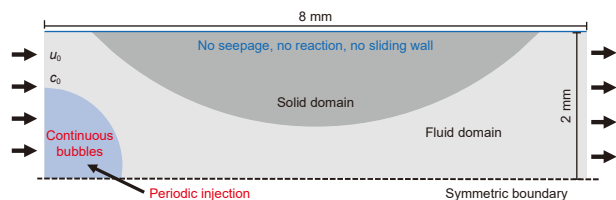


Fig. 9. Schematic of the dissolution model involving continuous bubble flow in a capillary medium.

experiment and is physically reliable for describing residual- CO_2 -bearing reactive transport during carbonate acidizing.

Taken together, the benchmark, numerical, and experimental validations demonstrate that the proposed micro-continuum model with implicit wettability treatment provides a numerically robust and physically reliable framework for pore-scale studies of coupled multiphase flow, solute transport, and mineral dissolution.

4. Results and discussion

This study examines CO_2 -bearing two-phase acidizing at the pore scale. A two-dimensional porous-medium model with

alternating CO_2 -liquid injection boundaries was established to capture the coupled evolution of two-phase transport, interfacial dynamics, and mineral dissolution under post-fracturing conditions. The model boundary conditions are shown in Fig. 13.

The overall computational domain is $6.4 \text{ mm} \times 5.0 \text{ mm}$, and is discretized using 45,811 free triangular elements, with a minimum element size of 0.05 mm. Black pixels represent reactive minerals (calcite), while light gray pixels represent non-reactive minerals (non-calcite). The inlet features three segments (each $150 \text{ }\mu\text{m}$ wide) designated as gas injection boundaries. The injection follows a periodic pattern dictated by the flow regime, ensuring each gas injection forms bubbles of consistent diameter. Acid at varying concentrations is injected from the left side at a constant velocity $u_0 = 0.001 \text{ m/s}$. The outlet pressure p_0 is set to 0. Interfacial tension is set to $\sigma = 0.07 \text{ N/m}$, and the mineral contact angle θ is 60° . The fundamental physical properties are detailed in Table 1.

4.1. Influence of the CO_2 phase on acidizing

To isolate the role of the CO_2 phase, a gas-bearing CO_2 -acid system was compared with single-phase acid injection. In these simulations, the injection velocity was 0.1 mm/s , the temperature was 343 K , and the inlet acid concentration was 2.0 mol/L . To characterize the transport and reaction regime, we computed the

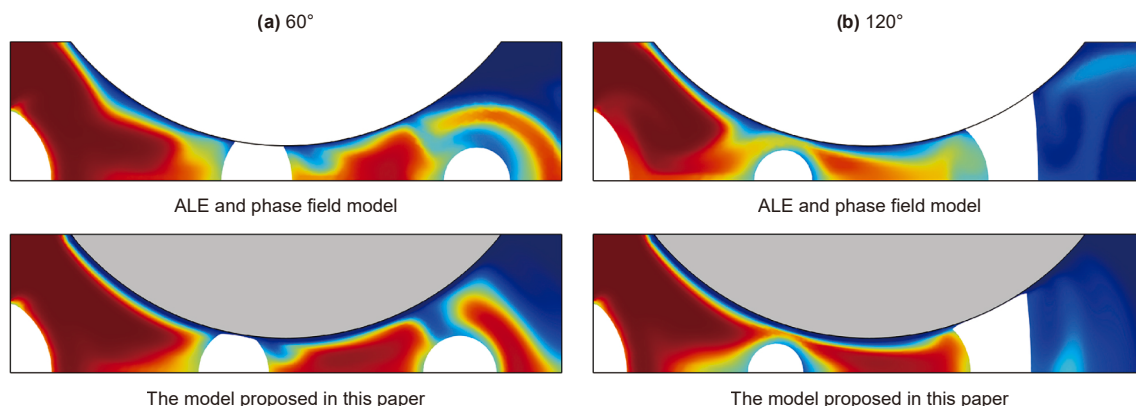


Fig. 10. Comparison of two-phase dissolution simulation results between the DBS + PFM model and the ALE benchmark.

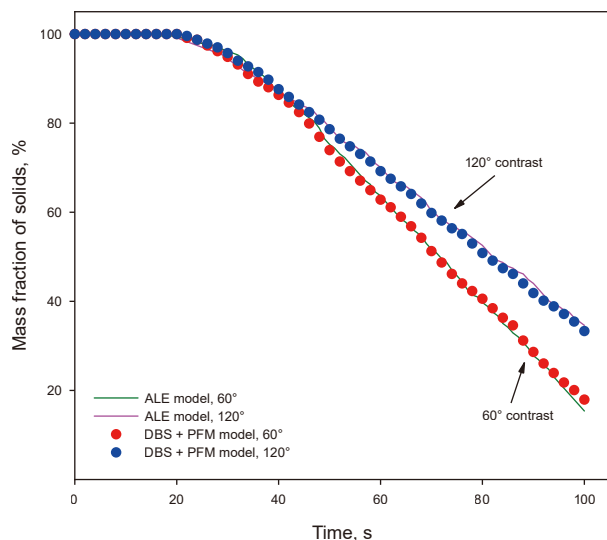


Fig. 11. Comparison of solid mass fraction loss between the two methods under different contact angle conditions.

Péclet number (Pe) and Damköhler number (Da). Under the present conditions, both Pe and Da exceed unity, indicating that convective transport dominates over diffusion, and that the reaction proceeds under convection-controlled conditions.

Fig. 14 shows that the presence of the CO_2 phase substantially alters dissolution dynamics. In the single-phase case, H^+ transport follows a relatively stable preferential pathway, producing localized dissolution and progressive channel deepening. In the two-phase system, by contrast, CO_2 continuously perturbs the

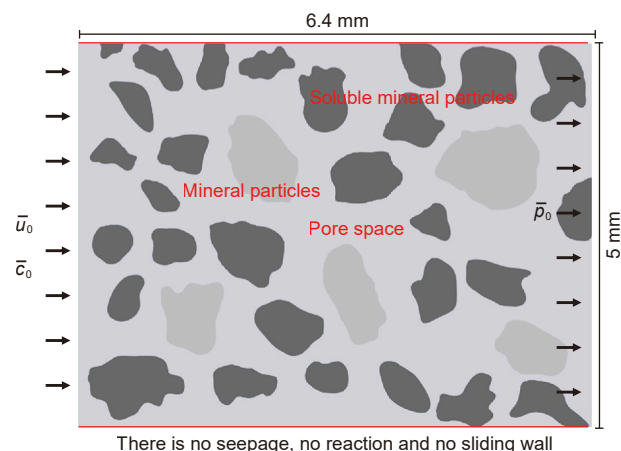


Fig. 13. The 2D idealized porous structure used in the simulation. Black and gray regions represent reactive mineral (calcite) and non-reactive minerals, respectively.

velocity field and redistributes the acid. CO_2 preferentially accumulates in dominant flow channels: part of the phase is transported downstream by convection, whereas the remainder adheres to solid surfaces under capillary forces. The trapped CO_2 gradually coalesces into larger clusters that obstruct major pathways and redirect the liquid into neighboring pores. As a result, the two-phase system develops a markedly different dissolution pattern. A cyclic sequence of blockage, detachment, and re-entry is observed, producing a more heterogeneous dissolution morphology than that in the single-phase case.

The spatial distribution of dissolution along the flow direction is compared in Fig. 15. During the early stage, up to approximately

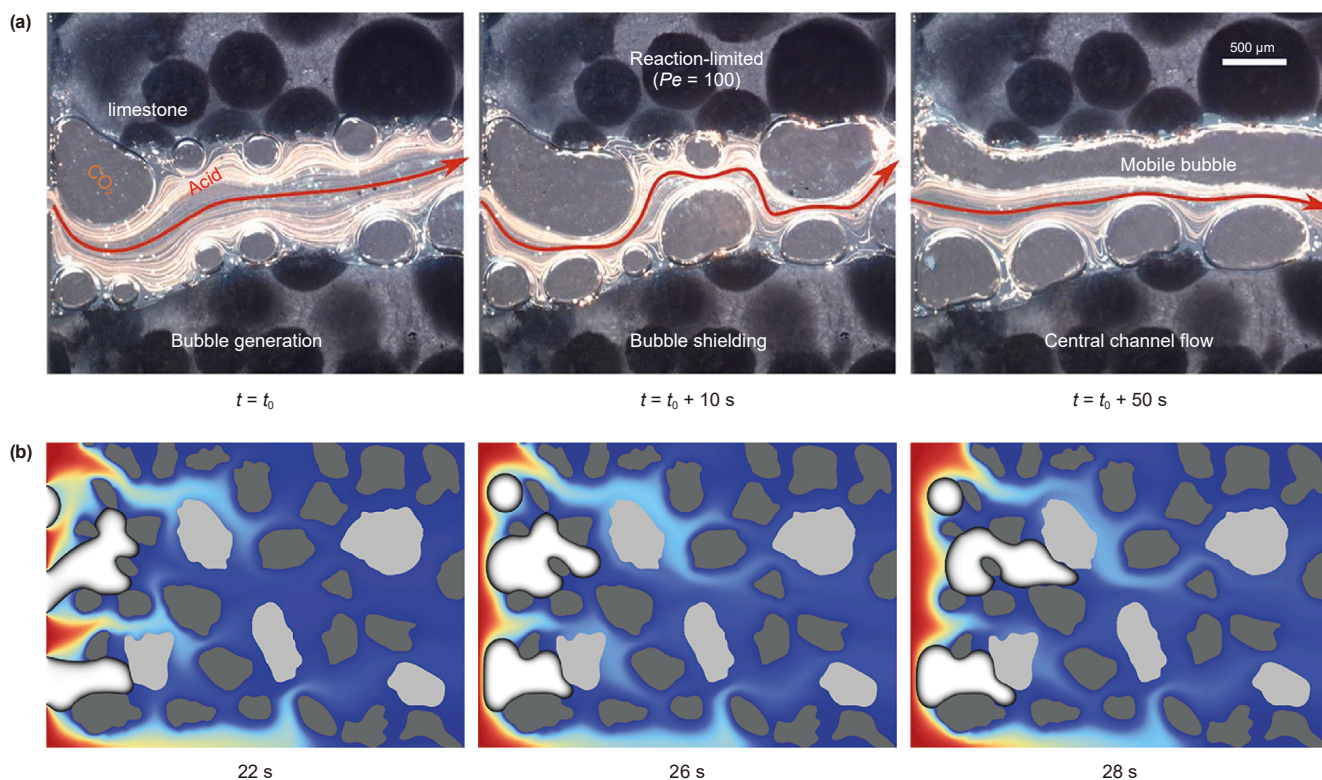


Fig. 12. Comparison between (a) the experimental two-phase acid-etching process reported by Zhou et al. (2025) and (b) the corresponding simulation results, showing bubble attachment, shielding of the reactive interface, and bubble coalescence. Reprinted with permission from Zhou et al. (2025).

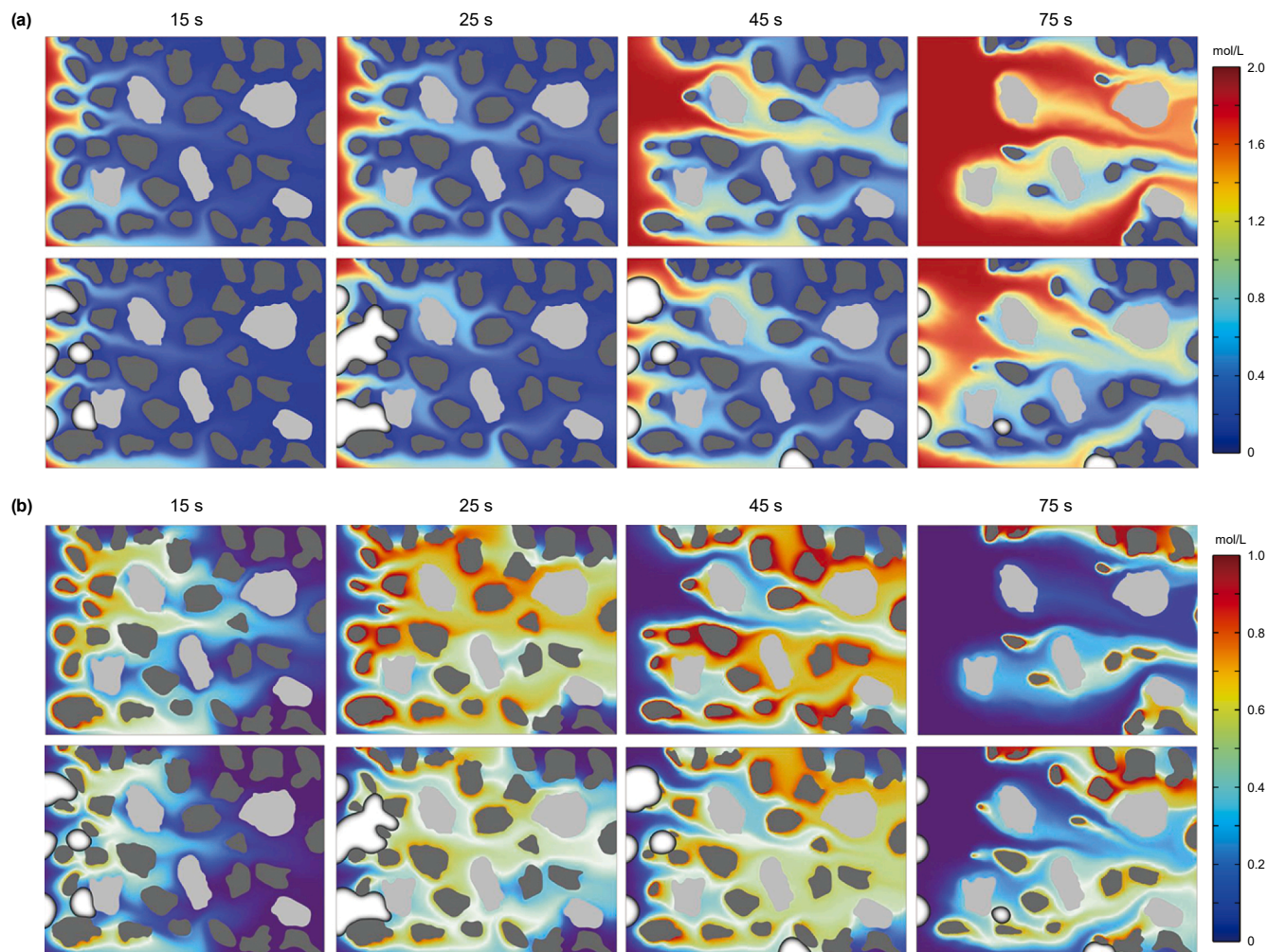


Fig. 14. Temporal evolution of ion concentrations during the acidizing process: (a) H^+ concentration and (b) Ca^{2+} concentration. The upper and lower panels correspond to single-phase and two-phase (CO_2 and acid solution) dissolutions, respectively.

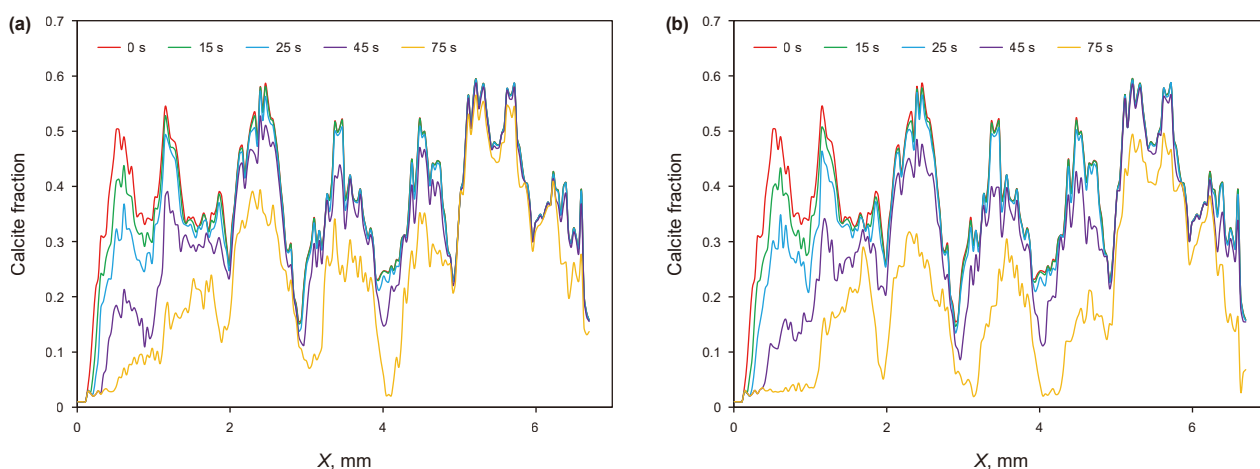


Fig. 15. Evolution of solid fraction along the flow direction (X-axis) at different time points: (a) gas-liquid two-phase and (b) single-phase.

15 s, both systems show similar mineral removal near the inlet. As the process continues, the difference becomes increasingly pronounced. In the single-phase case, the calcite fraction decreases more uniformly along the flow direction, indicating sustained penetration of the acid into deeper channels. In the two-phase

case, dissolution becomes progressively confined to the inlet region. Beyond approximately 25 s, much less mineral is removed downstream in the presence of CO_2 , demonstrating the inhibiting effect of the CO_2 phase on acid transport. CO_2 accumulation and cluster formation block the dominant pathways, redirect the acid,

and retard the advance of the dissolution front. Consequently, the two-phase system evolves more slowly overall and exhibits much stronger spatial heterogeneity. At later times, from 45 to 75 s, the single-phase system shows substantial mineral removal throughout the domain, whereas the two-phase system retains a significantly higher calcite fraction downstream. These results indicate that the CO₂ phase reduces effective acid penetration and shifts dissolution toward near-inlet regions.

Further insight into the transient response is provided by Fig. 16. During the early stage, from 0 to 5 s, the two-phase system exhibits a higher instantaneous dissolution rate than the single-phase system. This initial enhancement results from CO₂-induced flow diversion and transverse redistribution of fresh acid. In single-phase flow, the acid preferentially follows high-permeability pathways, leaving stagnant regions weakly accessed. Once the non-wetting CO₂ phase is introduced, bubbles occupy the centers of larger pores and force the liquid into narrower throats and previously bypassed regions. This redistribution enlarges the effective reactive contact area and temporarily enhances mineral accessibility. As a result, the two-phase system can

dissolve regions that remain unreachable in the single-phase case during the same interval.

After approximately 5 s, however, CO₂ blockage becomes dominant. Persistent CO₂ clusters obstruct the flow paths, reduce effective acid supply, and cause the two-phase dissolution rate to fall below that of the single-phase case. CO₂ phase therefore plays a dual role: it can transiently enhance dissolution by redistributing the acid, but over longer times it suppresses the overall reaction through blockage and shielding.

The effect of CO₂ phase on transport capacity is reflected in Fig. 17. In the two-phase case, adhesion and detachment of CO₂ clusters generate pronounced permeability fluctuations. During the early dissolution stage, particularly before about 35 s, CO₂ accumulation near the inlet substantially reduces liquid-flow capacity. The normalized permeability decreases to about 0.7, mainly because of the Jamin effect, whereby trapped CO₂ clusters impose strong capillary resistance in pore throats. As dissolution proceeds, permeability exhibits a distinctly fluctuating evolution, reflecting the competition between CO₂ blockage and pore enlargement.

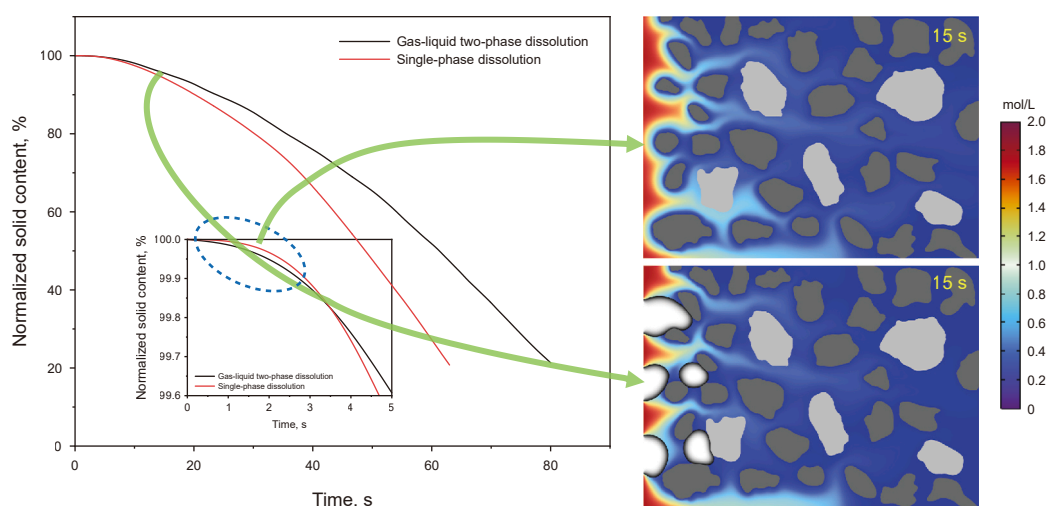


Fig. 16. Left: temporal evolution of normalized solid content in single-phase vs. two-phase dissolution modes. Right: reaction patterns at specific time points for single-phase (top) and two-phase (bottom) dissolution modes.

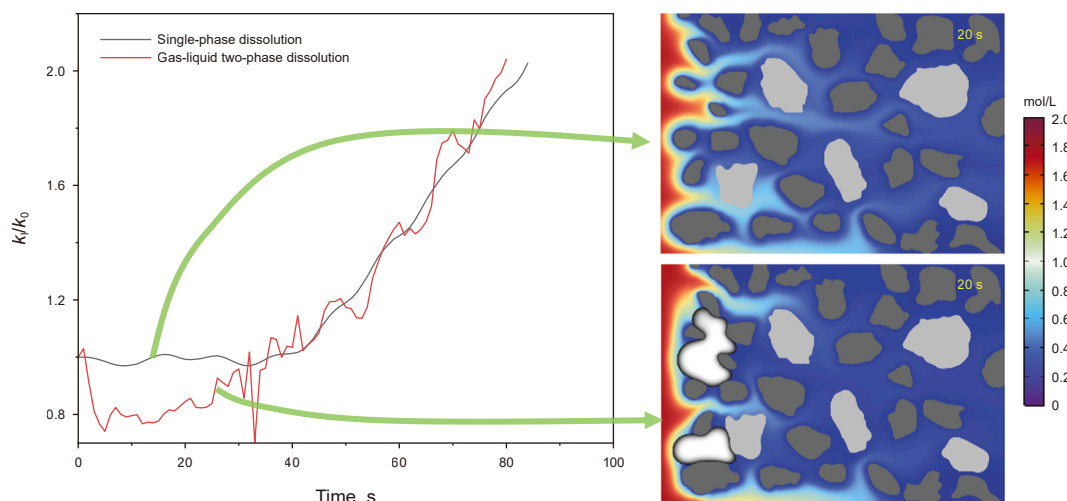


Fig. 17. Left: time evolution of the liquid-phase permeability normalized by its initial value for single- and two-phase dissolution modes. Right: representative reaction patterns at selected times for single-phase (top) and two-phase (bottom) dissolution.

According to the Young-Laplace relation, the capillary entry pressure required to trap a bubble is inversely proportional to throat radius. As dissolution widens pore throats, the capillary holding force weakens, enabling trapped CO_2 clusters to mobilize and be flushed out. At the same time, dissolution channels widen and reconnect, allowing blocked pathways to reopen. By approximately 80 s, the normalized liquid permeability rebounds to about 2.0, indicating that newly created pore space eventually outweighs the transient blocking effect of CO_2 . The period of minimum permeability coincides with the interval in which the two-phase dissolution rate falls below that of the single-phase case in Fig. 16, confirming that CO_2 -induced permeability loss directly suppresses reaction progress. Overall, the two-phase system exhibits a strongly nonlinear coupling between transport and reaction, in which CO_2 redistribution modifies the flow and modulates the accessibility of reactive surfaces over time.

4.2. Effect of surface wettability

In the acidizing process, the dissolution of mineral grains is fundamentally constrained by the wetting behavior of the liquid phase on the rock surface. To investigate this mechanism, we established simulation models with three distinct contact angles ($\theta = 30^\circ, 60^\circ, 120^\circ$), while keeping the temperature at 343 K, the injection velocity at 0.1 mm/s, and the inlet acid concentration at 2.0 mol/L.

Fig. 18 shows that wettability exerts strong control on two-phase transport and dissolution. Varying the contact angle from 30° to 120° spans the transition from a strongly water-wet regime to a wettability-reversed, gas-wet regime. Under strongly water-wet conditions, the mineral surface favors the acid phase, and the non-wetting CO_2 phase passes through pore throats more readily. This promotes continuous delivery of fresh acid to the

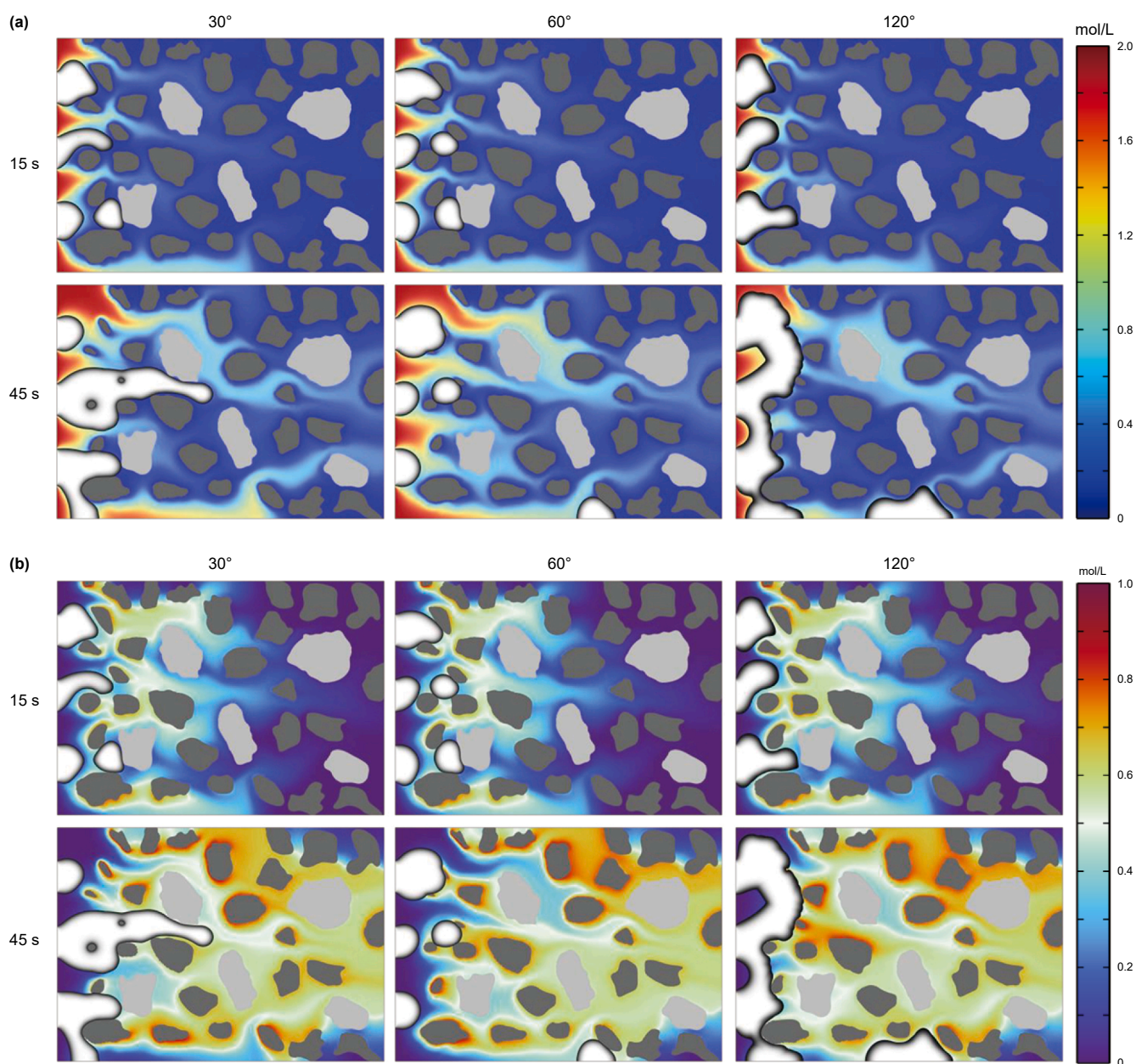


Fig. 18. Temporal evolution of ion concentrations during the acidizing process: (a) H^+ concentration and (b) Ca^{2+} concentration.

reactive interface. At 120°, by contrast, the CO₂ phase adheres strongly to the mineral surface, accumulates near the inlet, and forms an extended shielding layer that suppresses acid–rock contact and retards dissolution.

The quantitative impact of wettability is further detailed in Fig. 19. Results indicate that a strongly water-wet condition ($\theta = 30^\circ$) effectively mitigates the interference of the CO₂ phase. Under this condition, the acid can penetrate deeper into the formation (away from the inlet) more rapidly, leading to an accelerated rate of solid dissolution and a faster increase in porosity. Conversely, as the system shifts towards gas-wet conditions, the reaction is retarded.

4.3. Effect of acid concentration

The effect of acid strength was examined using five hydrochloric-acid concentrations: 1.5, 2.0, 3.0, 4.0, and 5.0 mol/L. In all cases, the injection velocity was kept at 0.1 mm/s and the temperature at 343 K, so that the only variable was the acid concentration.

Fig. 20 presents the temporal evolution of key parameters (solid calcite fraction, fluid–solid reaction front length, porosity, and liquid permeability) for the different acid concentrations. As

expected, higher acid concentrations provide a greater supply of H⁺ ions, which led to faster mineral dissolution. Increasing acid concentration also altered the evolution of the fluid–solid reaction-front length, because the more reactive fluid was able to dissolve minerals quickly and thus advance the dissolution front further in the same timeframe. All cases showed a monotonic increase in overall porosity as the acid reacted with calcite. The permeability curves for all concentrations exhibited some fluctuations due to CO₂ blocking events (similar to those described earlier in the two-phase system). However, with higher acid concentrations these fluctuations tended to be somewhat less severe over time, because the faster reaction can enlarge pore spaces more quickly and help release trapped gas. Indeed, as the solid phase was consumed and CO₂ bubbles detached from the dissolving surfaces, the amplitude of the permeability oscillations gradually diminished for each case. In summary, using a stronger acid accelerated the dissolution kinetics and shortened the fluid–solid reaction-front length, while eventually alleviating some of the gas-induced flow disruptions by more rapidly creating flow pathways.

The effect of acid concentration on dissolution rate is further quantified in Fig. 21. As the acid concentration increases from 1.5 to 5.0 mol/L, the average dissolution rate increases in both the single-

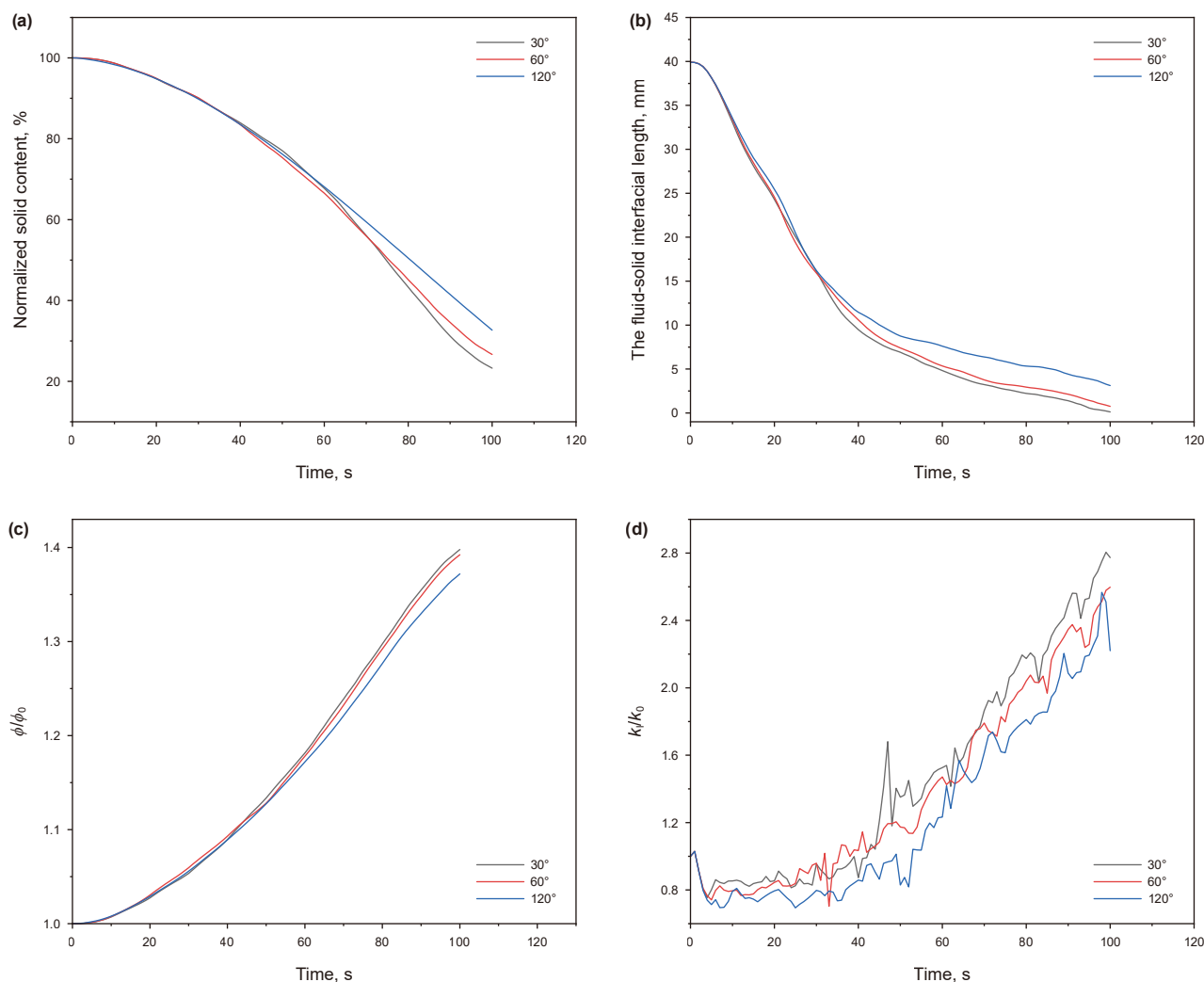


Fig. 19. Temporal evolution of: (a) normalized solid content, (b) fluid–solid reaction length, defined as the total length of the reactive liquid–solid interface satisfying, (c) porosity change, (d) liquid permeability change.

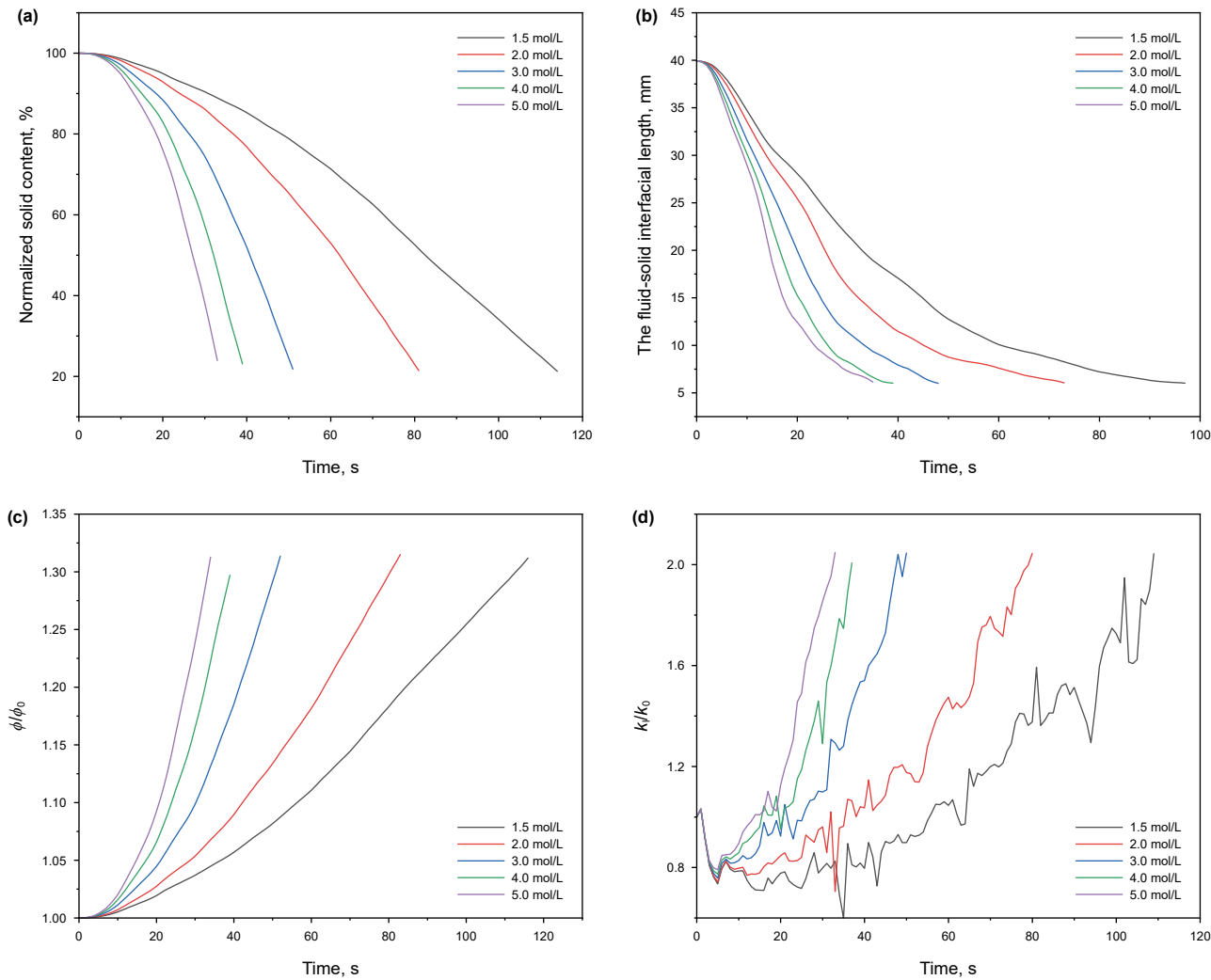


Fig. 20. Temporal evolution of: (a) normalized solid content, (b) fluid-solid reaction length, (c) porosity change, (d) liquid permeability change.

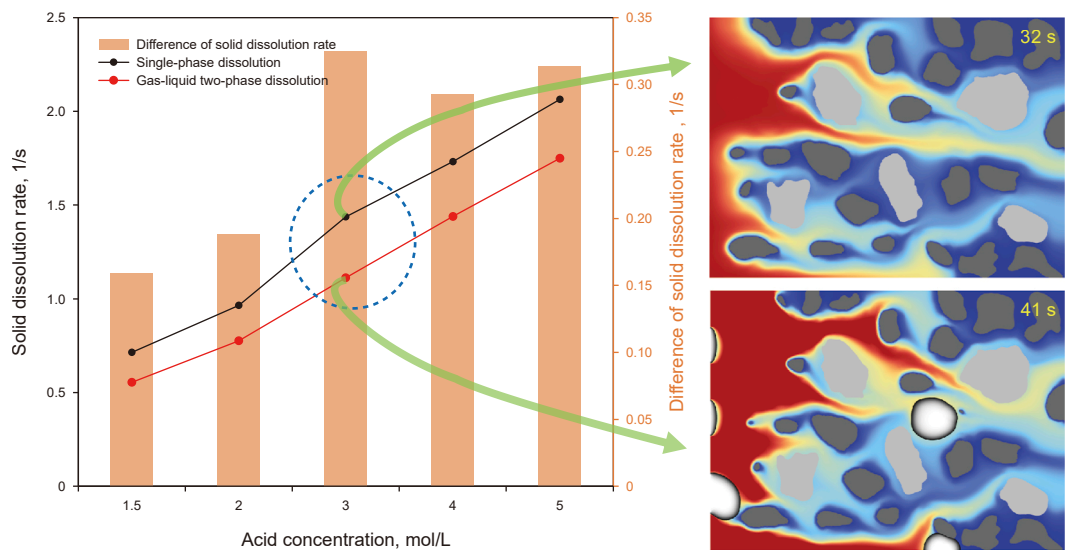


Fig. 21. Left: temporal evolution of solid volume fraction in single-phase vs. two-phase modes for different acid concentrations. Right: reaction patterns at specific times for single-phase (top) and two-phase (bottom) dissolution modes.

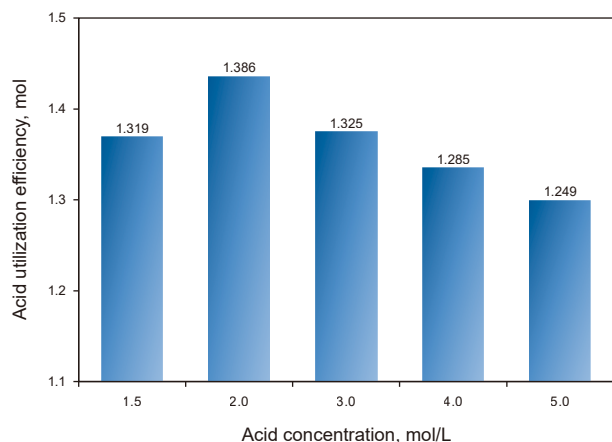


Fig. 22. Acid utilization efficiency vs. acid concentration.

phase and two-phase systems. This reflects the greater proton supply available for the acid–rock reaction. However, the difference between the two systems is not constant. The largest disparity occurs at an intermediate concentration, around 3.0 mol/L. At low acid concentrations, CO₂ phase strongly suppresses the

two-phase reaction because limited H⁺ supply is compounded by blockage and shielding. As the concentration increases, the relative penalty imposed by the CO₂ phase becomes smaller, indicating that stronger acid can partially offset the mass-transfer limitation caused by CO₂ blockage. At 5.0 mol/L, the dissolution rates of the two systems become more similar, suggesting that sufficiently strong acid improves acid–rock contact efficiency even in the presence of CO₂. These results imply the existence of a concentration threshold above which the negative impact of the CO₂ phase on reaction rate is substantially reduced.

A higher acid concentration does not necessarily imply more efficient acid use. Fig. 22 shows the acid-utilization efficiency, defined as the ratio of the relative solid consumption to the moles of injected acid, with a unit of mol⁻¹. The efficiency is non-monotonic with respect to concentration. It increases initially, reaches a maximum at approximately 2.0 mol/L, and then declines at higher concentrations. This trend indicates that increasing acid strength improves utilization only up to an optimum level. Beyond that point, the process becomes increasingly limited by available reactive surface area and residence time rather than by acid supply alone. Consequently, a larger fraction of the injected acid remains unreacted and exits the domain as live acid. In practical terms, excessively concentrated acid provides diminishing returns while increasing operational cost, corrosion risk, and safety burden. An

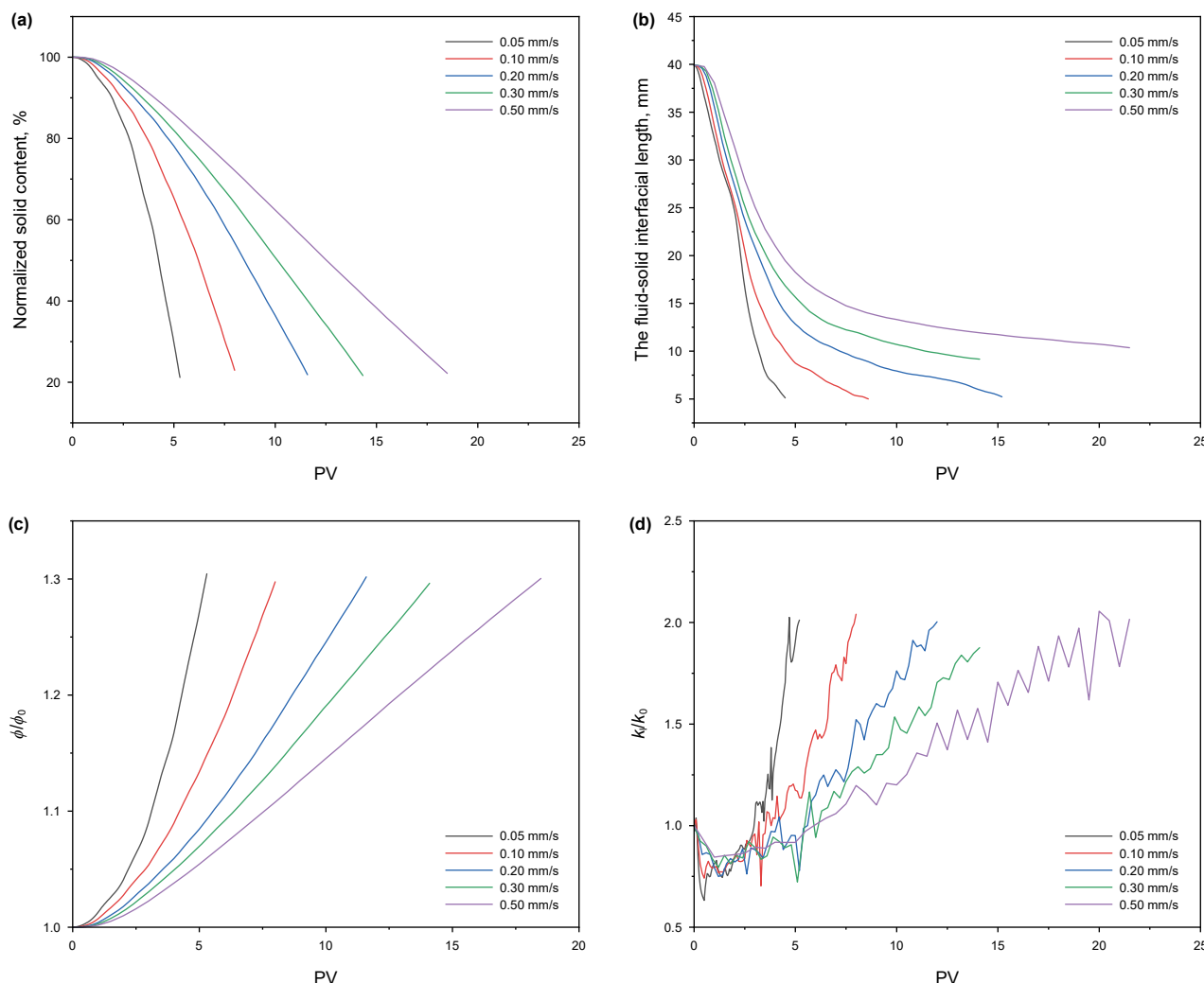


Fig. 23. Injection volume evolution of: (a) normalized solid content, (b) fluid-solid reaction length, (c) porosity change, (d) liquid permeability change.

intermediate concentration is therefore more favorable for balancing dissolution performance and chemical efficiency.

4.4. Injection rate

The influence of injection rate was investigated under two-phase conditions using five inlet velocities: 0.05, 0.1, 0.2, 0.3, and 0.5 mm/s. The acid concentration was fixed at 2.0 mol/L and the temperature at 343 K.

Fig. 23 summarizes the response of solid fraction, fluid–solid reaction-front length, porosity, and liquid permeability to changes in injection velocity. As the injection rate increases, acid is delivered into the porous medium more rapidly and reaches deeper regions over the same physical time. Consequently, higher inlet velocities produce faster initial dissolution and earlier porosity increase. The permeability response, however, remains complex because CO₂ migration and blockage introduce strong transient fluctuations. At higher injection rates, these fluctuations are more pronounced during the early stage, since rapid flow can abruptly mobilize and redistribute CO₂ clusters, producing intermittent surges and drops in flow capacity. As dissolution proceeds and the pore space enlarges, the amplitude of these oscillations decreases in all cases.

Although increasing the injection rate accelerates acid delivery, it does not translate linearly into higher efficiency in the presence of CO₂. Fig. 24 compares the average dissolution rates of the two-phase and single-phase systems at different injection rates. The absolute dissolution rate increases in both systems with increasing velocity because fresh acid is supplied more rapidly. However, the gap between the two-phase and single-phase rates also becomes larger at higher injection velocities. In other words, the detrimental effect of the CO₂ phase becomes more pronounced as the flow rate increases. This behavior shows that faster injection does not eliminate CO₂ interference; instead, it amplifies the residence-time limitation. At high velocities, the acid traverses the pore space more rapidly, leaving less time for interfacial mass transfer and surface reaction. CO₂ clusters remain in the flow paths and continue to divert or shield the acid, while the shortened contact time magnifies the impact of those barriers. Increasing injection rate alone is therefore not an effective strategy for overcoming CO₂ interference during acidizing.

The diminishing returns of high injection speeds are clearly shown in Fig. 25, which plots acid-utilization efficiency as a function of injection velocity. This metric reflects the effectiveness of acid use in promoting solid dissolution and differs from the conventional wormholing efficiency, which is typically evaluated by the pore volumes to breakthrough (PV_{bt}) and primarily describes penetration behavior.

As shown in Fig. 25, the efficiency is highest at the lowest tested velocity, 0.05 mm/s, and decreases steadily as the injection rate increases. This inverse relationship reflects the importance of residence time. At low flow rates, the acid remains in the pore space long enough to react more completely with calcite even in the presence of CO₂-induced barriers. At high flow rates, by contrast, the acid is swept through the domain too rapidly. The shortened residence time, combined with shielding by CO₂ clusters, prevents the acid from utilizing the available reactive surface effectively. As a result, a larger fraction of the injected acid exits the system unreacted. These results indicate a clear trade-off in CO₂-bearing systems: high pump rates may accelerate delivery, but they significantly reduce acid-utilization efficiency.

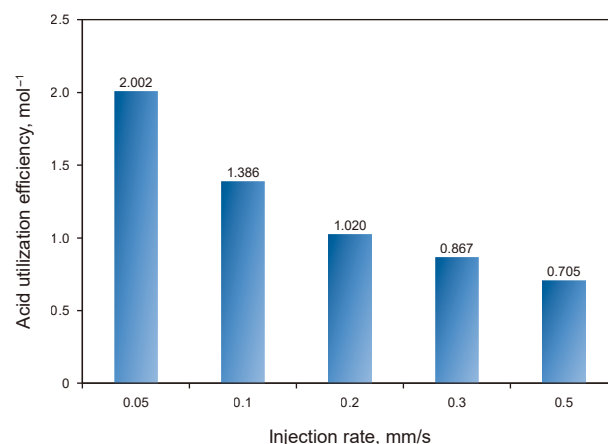


Fig. 25. Acid utilization efficiency versus injection velocity.

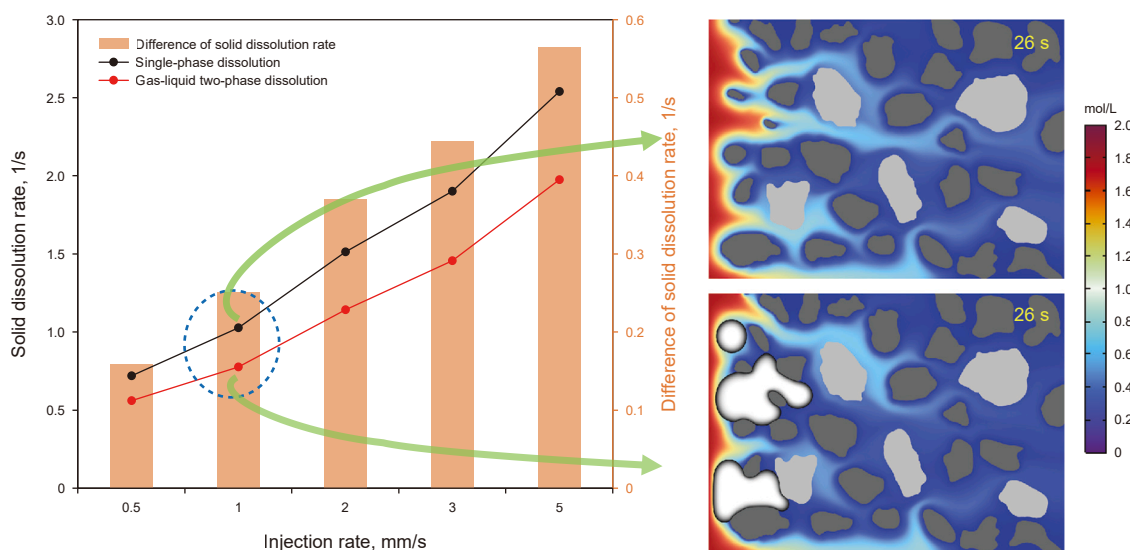


Fig. 24. Left: temporal evolution of solid volume fraction in single-phase vs. two-phase modes for different injection velocities. Right: reaction patterns at specific times for single-phase (top) and two-phase (bottom) dissolution modes.

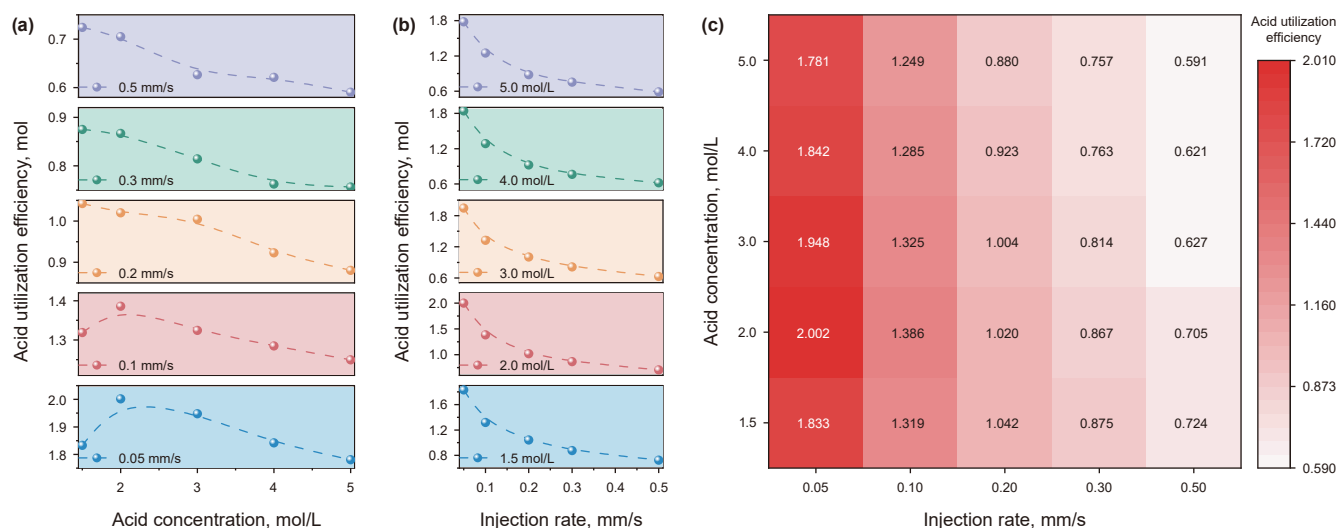


Fig. 26. Coupled analysis of acid utilization efficiency. (a) Variation of efficiency with acid concentration under different injection rates; (b) Variation of efficiency with injection rate under different acid concentrations; (c) Heatmap of acid utilization efficiency under coupled conditions.

4.5. The coupled effect of acid concentration and injection rate

Within the explored pore-scale parameter space, we conducted a comprehensive parametric study to investigate the coupling effects between acid concentration and injection rate. As detailed in the simulation setup, a matrix of 25 scenarios was established, covering five acid concentrations (1.5, 2.0, 3.0, 4.0, and 5.0 mol/L) and five injection velocities (0.05, 0.1, 0.2, 0.3, and 0.5 mm/s), with the system temperature maintained at 343 K.

The primary objective is to determine how the optimal acid concentration shifts in response to varying injection rates. The computed acid utilization efficiency for all scenarios is summarized and visualized in Fig. 26.

As illustrated in the heatmap (Fig. 26(c)), acid-utilization efficiency depends nonlinearly on both parameters. Consistent with the trends identified in Section 4.4, increasing injection velocity causes a systematic decline in efficiency across all concentration levels. For example, at 2.0 mol/L, the efficiency decreases from about 2.00 at 0.05 mm/s to about 0.70 at 0.5 mm/s, confirming that high flow rates reduce the residence time required for effective acid–rock reaction.

More importantly, the optimum acid concentration is not fixed but shifts with injection rate. At low injection rates, 0.05–0.1 mm/s, the highest efficiency occurs at a moderate concentration of approximately 2.0 mol/L, where residence time is sufficient for effective acid consumption. At higher injection rates, 0.3–0.5 mm/s, the efficiency becomes less sensitive to concentration overall but tends to decrease as concentration increases. For example, at 0.5 mm/s, increasing the concentration from 1.5 to 5.0 mol/L lowers the efficiency from about 0.72 to about 0.59. These results indicate that under high-velocity conditions, higher acid concentrations aggravate live-acid bypass because a larger fraction of the injected acid is swept out of the domain before it can be fully spent.

5. Conclusions

A pore-scale micro-continuum model coupling Darcy-Brinkman-Stokes flow with a wettability-corrected phase-field formulation was developed for gas–liquid two-phase acidizing in carbonate rocks. The proposed framework captures the coupled evolution of flow, interfacial dynamics, and reactive dissolution, and shows good agreement with benchmark tests. The main conclusions are as follows.

- (1) The CO₂ phase modifies dissolution through two competing mechanisms, namely flow diversion and bubble shielding. CO₂ clusters occupying pore centers redirect acid into secondary flow paths, whereas CO₂ adhering to mineral surfaces shields the reactive interface and suppresses local dissolution.
- (2) Surface wettability exerts strong control on CO₂ mobility and acid accessibility. Under strongly water-wet conditions ($\theta = 30^\circ$), CO₂ is more readily mobilized and shielding is weakened, allowing deeper acid penetration. In contrast, under gas-wet conditions ($\theta = 120^\circ$), CO₂ adhesion and inlet blockage become more severe, leading to a marked retardation of the dissolution process.
- (3) Unlike the monotonic permeability increase observed in single-phase acidizing, permeability under two-phase conditions exhibits pronounced temporal fluctuations. This behavior reflects the competition between capillary trapping, represented by the Jamin effect, and dissolution-induced pore enlargement. Permeability recovery occurs only after throat widening reduces the capillary holding force sufficiently to mobilize trapped CO₂ clusters.
- (4) Acid concentration and injection rate jointly control acid-utilization efficiency. The optimum acid concentration is not fixed, but shifts with flow conditions. At low injection rates, a high-efficiency operating window occurs at a moderate acid concentration of about 2.0 mol/L, where reaction capacity and residence time are balanced. At high injection rates, increasing acid concentration produces diminishing returns because live-acid bypass becomes more severe. Increasing acid strength alone therefore cannot compensate for residence-time limitations under high-velocity two-phase conditions.

The present two-dimensional idealization underestimates pore connectivity relative to real three-dimensional carbonate rocks and may therefore overestimate CO₂-blockage effects because out-of-plane bypass pathways are absent. Nevertheless, the two core mechanisms identified here, namely flow diversion and bubble shielding, arise from local interfacial physics and are expected to remain relevant in three-dimensional systems. Future work will extend the present framework to three-dimensional digital rock geometries to quantify these topological effects more rigorously.

CRediT authorship contribution statement

Qi-Gui Wang: Writing – original draft, Methodology. **Cheng-Yong Li:** Writing – review & editing, Supervision. **Yin Feng:** Visualization, Methodology. **Run-Hua Zhu:** Visualization, Validation, Methodology. **Ya-Qi Li:** Visualization, Software, Resources. **Yi Liu:** Data curation. **Dan-Ni Tang:** Data curation. **Yang Zeng:** Data curation.

Data availability

The data that support the findings of this study are available within the article and from the corresponding author upon reasonable request.

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Conflict of 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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