Numerical Modeling of Electromigration Effects for Subsurface Geological Carbon Sequestration*

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Abstract. Geological Carbon Storage (GCS) is a critical technology for mitigating anthropogenic CO_2 emissions, but accurate reactive transport modeling (RTM) of subsurface ionic species remains challenging due to neglected electromigration effects. This paper focuses on the numerical modeling of Electrical Double Layer (EDL) effects, a key electromigration phenomenon, and its implications for GCS processes. Building on the theoretical framework of Poisson-Nernst-Planck (PNP) equations, we develop a reactive transport modeling workflow that integrates EDL effects with chemical kinetics and diffusive transport. The characteristic time scales of carbonate reactions and electromigration are analyzed to justify the selection of kinetic models for reaction terms. Two core GCS scenarios are investigated: CO_2 dissolution in nanoscale water films attached to solid surfaces and CO_2 diffusive leakage through caprock/well cement nanopores. Numerical results demonstrate that EDL effects are length-scale dependent: negligible at the micrometer scale but dominant at the nanometer scale, significantly altering ionic concentration distributions, pH conditions, and reaction kinetics. For CO_2 leakage through nanopores, EDL-induced electrostatic forces modify ionic fluxes, facilitating positive ion transport and repelling negative ions, which lead to lower pH and increase corrosion risk. High salinity conditions mitigate but do not eliminate EDL impacts, highlighting the necessity of incorporating electromigration effects in GCS RTM. The proposed modeling framework advances the accuracy of subsurface transport simulations, providing critical insights for GCS project design and risk assessment.

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1 Introduction

1.1 Background

Geological Carbon Storage (GCS) has emerged as a cornerstone technology in the global transition toward a sustainable energy landscape, proving pivotal for achieving ambitious net-zero emissions targets. By capturing and sequestering anthropogenic CO_2 within deep subsurface formations (specifically saline aquifers, depleted hydrocarbon reservoirs, and shale caprocks), GCS offers a viable pathway to mitigate the greenhouse effect [1,2]. However, the long-term viability, safety, and containment security of these storage systems depend fundamentally on the high-fidelity modeling of complex, coupled multi-physical and geochemical processes. These include the intricate dynamics of CO_2 dissolution into formation brines, the advective-diffusive transport of ionic species, complex mineral precipitation/dissolution cycles, and the evolving nature of fluid-rock interactions over decadal to millennial timescales [3,4].

Reactive Transport Modeling (RTM) serves as the primary computational framework for simulating these evolutionary processes. Despite its utility, conventional RTM approaches frequently rely on simplified transport formulations that neglect electromigration effects, specifically the electrostatic interactions that regulate and redistribute ionic species within charged porous media [5,6]. By failing to account for the electrical potential gradients that emerge during transport, traditional models may significantly misrepresent the chemical evolution of the storage reservoir.

In the context of GCS, two fundamental electromigration phenomena are of critical importance: **Coulomb effects**, which result from transient charge imbalances caused by differential ionic diffusivities, and **Electrical Double Layer (EDL) effects**, which arise from the intrinsic surface charge of mineral grains within the porous matrix [7,8]. This research focuses specifically on the EDL effects that manifest at the solid-fluid interface. These phenomena occur when charged mineral surfaces (common in clay-rich caprocks or wellbore cement) attract counterions and repel co-ions, establishing a structured electrochemical region characterized by the **Debye length** (λ_D) where electrostatic forces dominate over random thermal diffusion [9,10]. In subsurface sequestration environments, these effects are particularly pronounced in nanoporous media, such as low-permeability caprocks and primary wellbore seals. In these materials, the Debye length (typically ranging from 1 to 100 nm depending on ionic strength) becomes comparable to the characteristic pore throat size. This leads to a substantial overlap between the EDL and the pore cross-section, effectively transforming the pore space into an electrostatic filter that governs all mass transport [11,12].

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The omission of EDL effects in GCS-focused Reactive Transport Models can result in profound inaccuracies regarding predicted ionic fluxes, localized pH distributions, and the resulting reaction kinetics, all of which are critical metrics for assessing CO_2 trapping efficiency and the catastrophic risk of leakage [13,14]. For instance, EDL-induced alterations in the local concentration of H^+ ions can significantly accelerate the rate of mineral dissolution or carbonation-induced corrosion in both caprocks and wellbore cement. Such geochemical degradation compromises the mechanical integrity of the storage complex, potentially creating preferential flow paths for buoyant CO_2 [15,16].

Despite their demonstrated significance to storage safety, EDL effects are seldom incorporated into comprehensive, large-scale GCS simulations. This research gap is primarily attributed to two formidable hurdles:

- The absence of a robust, unified mathematical framework capable of seamlessly integrating microscopic EDL physics with macroscopic reactive transport equations.
- The extreme numerical complexity and computational cost associated with solving the highly non-linear, coupled **Poisson-Nernst-Planck (PNP) equations** that rigorously describe EDL-induced ionic transport within the multi-component chemical systems characteristic of the deep subsurface [17,18].

This paper seeks to address these challenges by developing a simplified yet physically consistent approach to bridge the gap between interface electrochemistry and reservoir-scale carbon sequestration modeling.

1.2 Literature Review

Historically, foundational investigations into Electrical Double Layer (EDL) effects relied predominantly on equilibrium-based formulations, most notably the Poisson-Boltzmann (PB) equation. This classical approach provides a rigorous mathematical description of how ionic density distributions are structured in the immediate vicinity of charged mineral surfaces, assuming a state of thermodynamic equilibrium [19,20]. While the PB equation has been instrumental in interfacial science, its applicability to Geological Carbon Storage (GCS) is severely constrained by its core assumptions. It is typically limited to low-salinity environments, whereas GCS sites often involve high-salinity brines, and it fundamentally neglects the dynamic flux of ions through the pore space. This inherent absence of a transport component renders the PB framework unsuitable for capturing the non-equilibrium nature of sequestration scenarios, such as the transient dissolution of CO_2 into formation water or the long-term diffusive leakage of acidified fluids through caprock seals [21,22].

To overcome these limitations, the Poisson-Nernst-Planck (PNP) equations have emerged as a more robust and generalized mathematical framework. By explicitly coupling the Nernst-Planck equation, which governs the advective-diffusive-electromigrative flux of individual ionic species, with the Poisson equation, which calculates the self-consistent electric potential, the PNP system can

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simulate complex, time-dependent EDL dynamics [23,24]. However, the integration of PNP models into reservoir-scale simulations is hampered by the intense non-linear coupling between the local electrostatic potential and the concentration fields of multiple ionic species. These mathematical dependencies pose formidable numerical challenges, frequently leading to instability or prohibitive computational costs when applied to the multi-component electrolyte solutions and complex geometries characteristic of deep subsurface formations.

Recent breakthroughs in the numerical treatment of PNP systems have shifted toward the development of high-order energy-stable schemes. These sophisticated algorithms are specifically engineered to satisfy essential physical laws, including the strict preservation of mass conservation, the guaranteed positivity of ionic concentrations, and the maintenance of thermodynamic consistency across all temporal steps [25,26]. A prominent example of this progress is the work of Qiao et al. [18], who introduced a linear, unconditionally energy-stable scheme [27] based on a unique energy factorization approach. Their method allows for the efficient simulation of EDL effects in charged porous media without the restrictive time-step limitations common in traditional explicit solvers. Despite the elegance of these numerical advancements, the practical synthesis of such energy-stable schemes with established reactive transport models (RTMs) remains largely underexplored, particularly for the high-pressure, chemically reactive environments found in CO_2 storage sites.

Within the specific domain of GCS, a growing but still limited body of literature has begun to quantify the impact of EDL physics on subsurface transport processes. For instance, Yang et al. [28] conducted a rigorous analysis of EDL-mediated ion diffusion within wellbore cement pastes, demonstrating that electrostatic forces significantly alter the flux of chloride ions and thereby heighten the risk of steel casing corrosion. Similarly, Tournassat and Steefel [29] established a landmark benchmark for EDL modeling in clay-rich porous media, underscoring the vital necessity of fine-scale spatial discretization to capture the extreme electrostatic heterogeneity inherent in clay matrices. Despite these incremental advances, a glaring gap persists in the field. A comprehensive reactive transport framework that fully integrates microscopic EDL effects, non-linear chemical kinetics such as mineral carbonation, and multi-species diffusive transport for GCS-specific scenarios is still currently lacking in the scientific community. Such a tool is necessary for predicting CO_2 behavior in ultra-thin water films and simulating the localized mineral transformations that occur during nanopore-scale leakage events.

1.3 Research Objectives and Contributions

This paper primarily aims to establish a robust numerical modeling framework that integrates Electrical Double Layer (EDL) effects into reactive transport simulations, specifically to quantify their influence on fundamental Geological Carbon Storage (GCS) processes. To achieve this, we present a unified reactive transport workflow that seamlessly couples the Poisson-Nernst-Planck (PNP) equations, governing EDL-induced ionic transport, with advanced kinetic models

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for carbonate reactions, all validated through a rigorous characteristic time scale analysis. Utilizing this framework, the study provides a detailed numerical investigation into how EDL phenomena affect CO_2 dissolution within nanoscale water films and its diffusive leakage through the nanoporous structures of caprocks and wellbore cement. Furthermore, the research quantifies the critical dependence of these effects on spatial length scalescontrasting nanometer and micrometer environmentsas well as varying salinity levels to determine their impact on ionic concentrations, pH distributions, and overall reaction kinetics. Ultimately, these findings offer essential guidance for GCS modelers, identifying the specific conditions and methodologies required to incorporate EDL effects into larger-scale simulations to enhance predictive accuracy and long-term risk assessment.

1.4 Paper Organization

This paper is organized as follows: Section 2 presents the theoretical framework, including EDL fundamentals, PNP equations for ionic transport, and kinetic models for carbonate reactions. Section 3 details the numerical modeling workflow, including characteristic time scale analysis and spatial/temporal discretization. Section 4 presents numerical results for CO_2 dissolution in water films and diffusive leakage through nanopores, with analysis of EDL effects on transport and reactions. Section 5 discusses the implications of EDL modeling for GCS design and risk assessment. Section 6 provides conclusions and future research directions.

2 Theoretical Framework

2.1 Electrical Double Layer (EDL) Fundamentals

The EDL is a charge structure forming at the solid-fluid interface in porous media, consisting of two layers: (1) a compact layer (Stern layer) of adsorbed counterions directly adjacent to the mineral surface, and (2) a diffuse layer of mobile counterions and co-ions extending into the bulk solution [30,31]. The EDL is characterized by the Debye length (λ_D), the distance from the surface where electrostatic forces decay to 1/e of their maximum value:

$$\lambda_D = \sqrt{\frac{\epsilon R T}{2F^2 I}},$$

where ϵ is the permittivity of the aqueous medium (F/m), R is the universal gas constant (J/(mol·K)), T is absolute temperature (K), F is Faradays constant (C/mol), and I is ionic strength (mol/m^3). For typical subsurface brines (I = 0.01 - 1 mol/L), λ_D ranges from 1 - 10 nm, making EDL effects significant in pores smaller than 100 nm [32,33].

The zeta potential (ζ) (the electric potential at the shear plane between the compact and diffuse layers) governs EDL strength. For common mineral surfaces (e.g., clay, cement), ζ ranges from -50 to -25 mV (negatively charged), leading to attraction of positive ions (e.g., H^+, Na^+) and repulsion of negative ions (e.g., HCO_3^-, CO_3^{2-}) [34,35].

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2.2 Poisson-Nernst-Planck (PNP) Equations for Ionic Transport

The PNP equations describe the dynamic transport of charged ionic species under the influence of diffusion and electromigration. For a system of N ionic species, the Nernst-Planck equation (mass conservation for ionic transport) is [24,25]:

$$\frac{\partial c_i}{\partial t} = \nabla \cdot \left(D_i \nabla c_i + \frac{z_i D_i F}{RT} c_i \nabla \psi \right) + r_i.$$

In this formulation, c_i represents the concentration of the i-th species (mol/m^3), D_i denotes its effective diffusion coefficient (m^2/s), z_i is the dimensionless valence, ψ signifies the electric potential (V), and r_i accounts for the reaction source or sink term (mol/(m^3·s))).

The effective diffusion coefficient D_i accounts for porosity (ϕ) and tortuosity (τ_i) of porous media:

$$D_i = \frac{\phi}{\tau_i} D_{i,s},$$

where $D_{i,s}$ is the self-diffusion coefficient of the i-th species in aqueous solution (m^2/s).

The Poisson equation (coupling electric potential to charge density) is:

$$\nabla \cdot (\epsilon \nabla \psi) = -F \sum_{i=1}^{N} z_i c_i,$$

where the right-hand side represents the net charge density from ionic species.

Reactive Transport for Carbonate Systems CO_2 dissolution in aqueous brines initiates a cascade of carbonate reactions that generate charged ionic species, governing geochemical feedbacks in GCS [26,27,28]. The key reactions are: (1) CO_2 dissolution: $CO_2(aq) + H_2O \rightleftharpoons H_2CO_3$. (2) Carbonic acid dissociation: $H_2CO_3 \rightleftharpoons H^+ + HCO_3^-$. (3) Bicarbonate dissociation: $HCO_3^- \rightleftharpoons H^+ + CO_3^{2-}$. (4) Water dissociation: $H_2O \rightleftharpoons H^+ + OH^-$.

Reaction kinetics are described using a rate law for each forward/reverse reaction [32]:

$$r_i = k_{+n} \prod_j [A_j] - k_{-n} \prod_k [B_k],$$

where k_{+n} and k_{-n} are forward and reverse rate constants, and $[A_j]$ and $[B_k]$ are reactant and product concentrations. The equilibrium constant $K_n = k_{+n}/k_{-n}$ is temperature-dependent, with values for carbonate reactions at 25°C and 1 atm listed in Table 1.

For neutral species (e.g., $CO_2(aq)$), transport is governed by the diffusion-reaction equation (neglecting convection in low-permeability media):

$$\frac{\partial c_{CO_2}}{\partial t} = D_{CO_2} \nabla^2 c_{CO_2} + r_{CO_2},$$

where r_{CO_2} is the net rate of CO_2 dissolution.

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Table 1: Rate constants for carbonate system at 25°C and 1 atm

Rate Constant	Magnitude
k_1 (CO_2 dissolution)	6×10^{-2} s^{-1}
k_{-1} (reverse)	2×10^{1} s^{-1}
k_2 (H_2CO_3 dissociation)	1×10^{7} s^{-1}
k_{-2} (reverse)	5×10^{10} M^{-1}s^{-1}
k_3 (HCO_2^- dissociation)	3×10^{0} s^{-1}
k_{-3} (reverse)	5×10^{10} M^{-1}s^{-1}

3 Numerical Modeling Workflow

Characteristic Time Scale Analysis The choice of reaction model (kinetic vs. equilibrium) depends on the characteristic time scale of reactions (t_{rxn}) relative to transport (t_{trans}). For carbonate reactions, t_{rxn} is the time to reach 99% equilibrium:

$$t_{rxn} = \frac{-\ln(0.01)}{k_{+n} + k_{-n}[B_k]}.$$

For CO_2 dissolution and dissociation reactions, t_{rxn} ranges from 10^{-6} to 10^{-4} seconds (microsecond scale), while transport time scales in nanopores ($t_{trans} = L^2/D$) range from 10^{-3} to 10^{0} seconds (millisecond to second scale) for $L = 10 - 1000$ nm and $D = 10^{-9}$ m^2/s [31]. Since $t_{\text{rxn}} \ll t_{\text{trans}}$, kinetic models are required to capture transient reaction dynamics [29].

Coupled Modeling Workflow The reactive transport system is solved using a sequential non-iterative approach (SNIA), integrating transport equations, PNP equations, and chemical kinetics [33]. The workflow is illustrated in Figure 1:

1. **Input Parameters:** Physical properties (porosity, tortuosity), length scale, electrical boundary conditions (zeta potential), initial concentrations, and reaction rate constants.
2. **Neutral Species Transport:** Solve the diffusion-reaction equation for the concentration of CO_2(aq).
3. **Chemical Kinetics:** Calculate reaction rates and update ionic concentrations.
4. **Charged Species Transport:** Solve PNP equations to update ionic concentrations and electric potential, accounting for EDL effects.
5. **Convergence Check:** Repeat steps 2 - 4 until steady-state or final time.

Spatial and Temporal Discretization The PNP equations and diffusion-reaction equations are discretized using Cell-Centered Finite Difference (CCFD) method, suitable for structured grids in porous media [34]. For temporal discretization, a linear semi-implicit scheme is used for PNP equations to ensure unconditional energy stability and concentration positivity [18]:

$$\frac{c_i^{n+1} - c_i^n}{\Delta t} = \nabla \cdot \left(D_i c_i^n \nabla \mu_i^{n+1} \right),$$

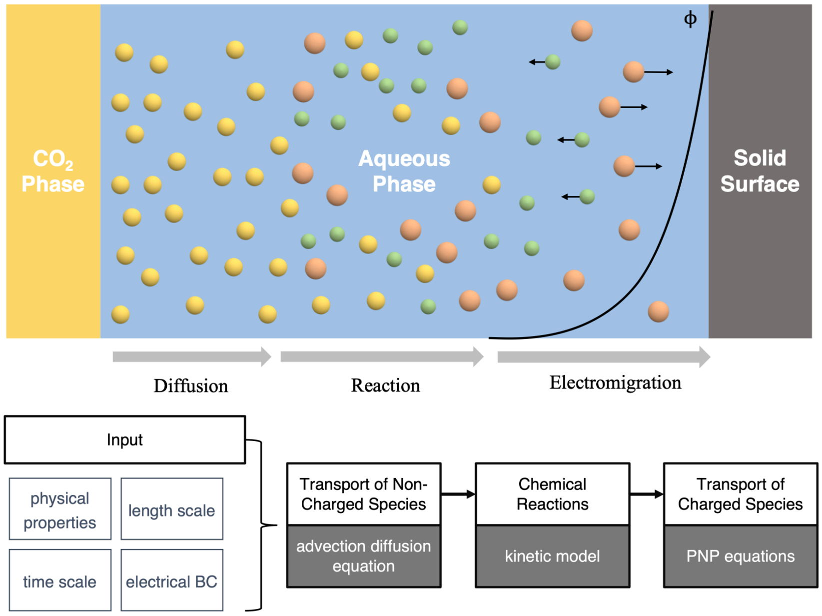

Fig. 1: Physical processes and the simulation workflow flow chart coupling transport, electromigration, and chemical reactions

$$\mu_i^{n+1} = \log c_i^n + \frac{c_i^{n+1}}{c_i^n} - 1 + z_i \psi^{n+1},$$

where μ_i is the chemical potential, and $n/n+1$ denote current/future time steps. The Poisson equation is discretized using a fully implicit scheme to maintain coupling with ionic concentrations.

The boundary conditions are implemented using Dirichlet constraints to maintain a constant $CO_2(aq)$ concentration at the inlet and a fixed zeta potential at the solid surfaces. Optionally, complementary Neumann conditions are applied to enforce no-flux constraints for concentration at solid boundaries and to represent insulated electric potential at the inlet and outlet.

The fully discrete CCFD scheme for PNP equations is derived using discrete gradient (δ) and average (A) operators (notations and details see [18]). For a 2D grid with cell size h, the discrete Nernst-Planck equation is:

$$\frac{c_i^{n+1} - c_i^n}{\mathit{\Delta} t} = \delta_x^u \left(D_i A_x^c[c_i^n] \delta_x^c[\mu_i^{n+1}] \right) + \delta_y^v \left(D_i A_y^c[c_i^n] \delta_y^c[\mu_i^{n+1}] \right),$$

where δ_x^c and δ_y^c are spatial gradient operators, and A_x^c and A_y^c are arithmetic average operators.

4 Numerical Results and Analysis

We investigate EDL effects on two GCS-relevant scenarios: (1) CO_2 dissolution in nanoscale water films, and (2) CO_2 diffusive leakage through caprock/well cement nanopores. Simulation parameters are summarized in Table 2.

Table 2: Simulation parameters

Parameter	Value
Temperature (T)	25°C
Permittivity (ϵ)	7.08×10^{-10} F/m
Zeta potential (ζ)	-50 mV
Porosity (ϕ)	0.3
Tortuosity (τ_i)	1.5
CO_2 inlet concentration	0.11 M
Salinity (NaCl)	01 M
Pore length (leakage scenario)	1 mm
Pore width (leakage scenario)	10 nm
Water film thickness	100 nm - 100 μm

Scenario 1: CO_2 Dissolution in Water Films Water films attached to mineral surfaces in GCS reservoirs range from nanometers to micrometers in thickness, with EDL effects varying by length scale [35]. We simulate CO_2 dissolution in a 1D water film with thicknesses of 100 nm (nanoscale) and 100 μm (micrometer scale), with a CO_2 inlet at $x = 0$ and a solid surface (zeta potential = -50 mV) at $x = L$.

Micrometer Scale (100 μm) At the micrometer scale, the Debye length ($\lambda_D \approx 10$ nm) is much smaller than the film thickness, so EDL effects are confined to a thin region near the solid surface (Figure 2a). Ionic concentrations (H^+, HCO_3^-, CO_3^{2-}) are nearly identical to simulations without EDL, with only minor deviations within 100 nm of the surface. The total amount of reacted CO_2 is similar to the no-EDL case (Figure 2b), indicating that EDL effects are negligible for micrometer-scale water films.

Nanoscale (100 nm) At the nanoscale, the Debye length is comparable to the film thickness, leading to full EDL overlap across the pore (Figure 3a). The negatively charged surface attracts H^+ (positive counterions) and repels HCO_3^-/CO_3^{2-} (negative co-ions), altering concentration distributions throughout the film. H^+ concentration is enhanced near the surface, while HCO_3^-/CO_3^{2-} concentrations are reduced. This leads to a 30% reduction in total reacted CO_2 compared to the no-EDL case (Figure 3b), as EDL-induced electrostatic forces inhibit carbonate reactions by reducing reactant (HCO_3^-) availability.

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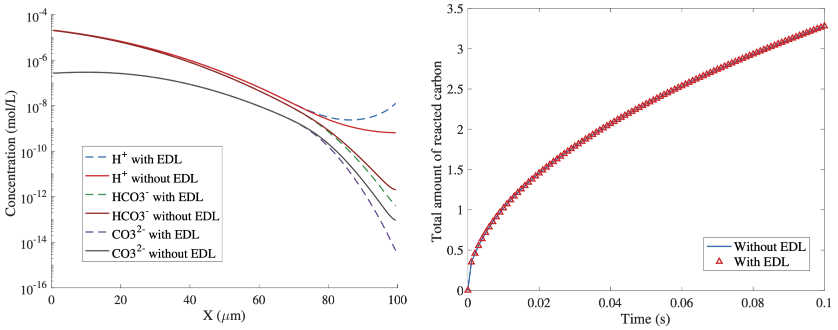
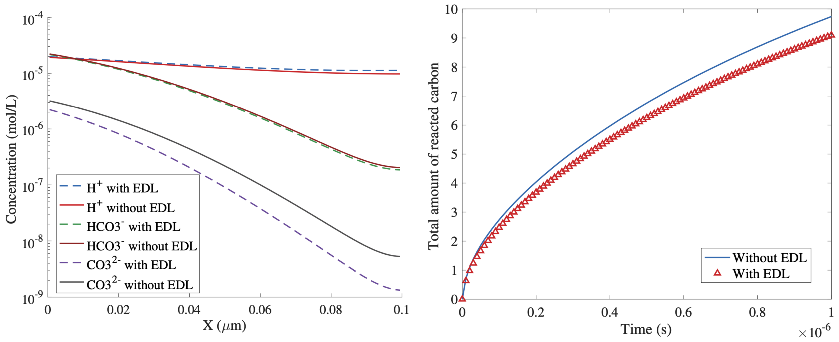


(a) Ionic concentration profiles (b) Total reacted CO_2

Fig. 2: EDL effects on CO_2 dissolution in a 100 micrometer water film

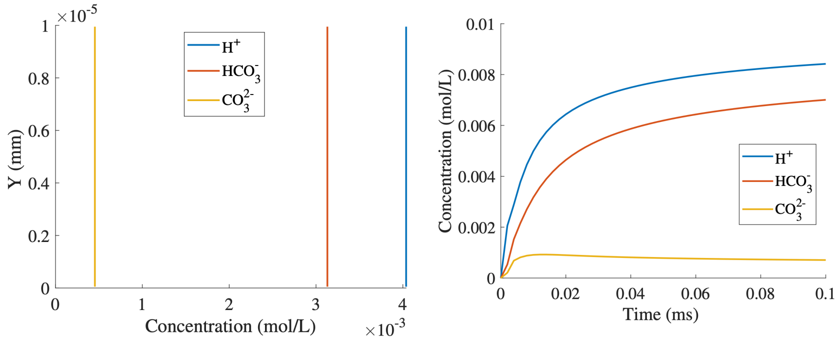


(a) Ionic concentration profiles (b) Total reacted CO_2

Fig. 3: EDL effects on CO_2 dissolution in a 100 nm water film

Scenario 2: CO_2 Diffusive Leakage Through Nanopores Caprocks and well cement contain nanopores (10 - 100 nm) where diffusive transport dominates, making EDL effects critical for assessing CO_2 leakage risk [36]. We simulate 2D CO_2 leakage through a nanopore (width = 10 nm, length = 1 mm) with charged walls (zeta potential = -50 mV), inlet CO_2 concentration = 0.1 M, and variable salinity (0 - 1 M NaCl).

Low Salinity (0 M NaCl) Without background salinity, EDL effects strongly modify ionic transport (Figure 4a). H^+ (positive) is concentrated near the negatively charged walls, while HCO_3^- and CO_3^{2-} (negative) are depleted in the EDL region. The average H^+ concentration across the pore cross-section is 2x higher than the no-EDL case (Figure 4b), leading to a lower pH (≈4.5 vs. ≈5.5 without EDL) and increased corrosion risk for cement/caprock minerals.



(a) Concentration profile at $x = 10\mu$m

(b) Average concentration over time

Fig. 4: EDL effects on CO_2 leakage (0 M NaCl)

High Salinity (1 M NaCl) Background salinity increases ionic strength (I), reducing the Debye length ($\lambda_D \approx 1$ nm) and mitigating EDL overlap (Figure 5a). Na^+ and Cl^- from NaCl dominate the EDL, reducing EDL-induced alterations to carbonate ion concentrations. However, H^+ concentration is still 1.3x higher than the no-EDL case (Figure 5b), indicating that EDL effects persist even in high-salinity brines. This is critical for GCS in saline aquifers, where conventional models may underestimate corrosion risk by neglecting residual EDL effects.

Key Findings: The primary findings of this research demonstrate that Electrical Double Layer (EDL) effects exhibit a profound dependence on length scales, as they remain largely negligible at the micrometer level but become dominant at the nanometer scale, which underscores the vital necessity of pore-scale modeling within nanoporous media. Furthermore, these effects manifest significant ionic specificity by preferentially facilitating the transport of positive ions while repelling negative ions, a process that leads to localized pH reductions and a heightened risk of corrosion. The study also reveals a salinity mitigation effect where high saline concentrations serve to reduce but not entirely eliminate EDL influences, as background ions like Na^+ and Cl^- provide only a partial screening of the solid surface charge. Finally, it was observed that EDL-induced decreases in HCO_3^- concentrations act to inhibit carbonate reactions, thereby substantially reducing CO_2 trapping efficiency within nanoscale water films.

5 Implications for Geological Carbon Storage

The numerical results have critical implications for GCS design and risk assessment:

1. Caprock/Wellbore Integrity: EDL-induced pH reductions in nanopores accelerate mineral dissolution and cement corrosion, increasing leakage risk. GCS models should incorporate EDL effects to accurately predict wellbore longevity.

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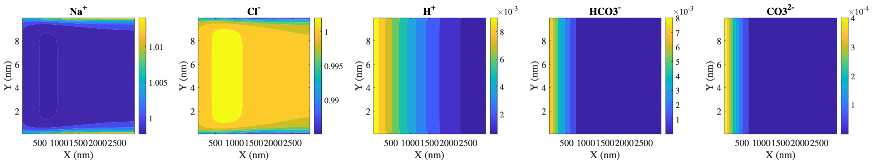
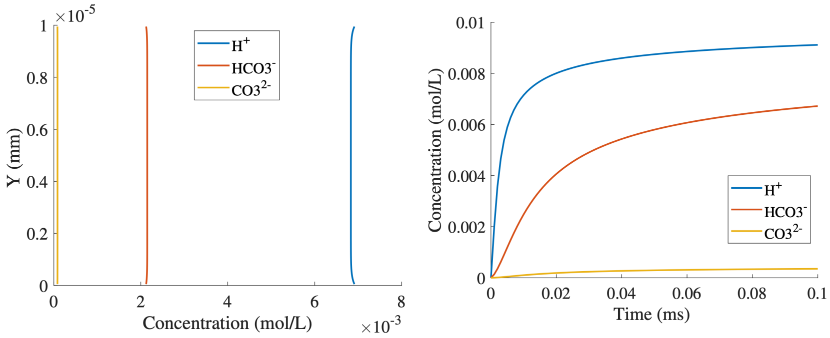


(a) Snapshots of the concentration profile



(b) Concentration profile at $x = 10\mu\text{m}$ (c) Average concentration over time

Fig. 5: EDL effects on CO_2 leakage (1 M NaCl)

2. CO_2 Trapping Efficiency: EDL effects inhibit carbonate reactions in nanoscale water films, reducing mineral trapping efficiency. This is particularly relevant for tight reservoirs with high nanoporosity.

3. Modeling Guidance: EDL effects should be included in GCS RTM for pore sizes < 100 nm and low-permeability formations (caprocks, shale). For larger pores (> 1 μm), EDL effects can be safely neglected.

4. Salinity Considerations: While high salinity mitigates EDL effects, residual impacts persist, requiring integration into saline aquifer models.

6 Conclusions and Future Work

This paper established a numerical modeling framework designed to integrate Electrical Double Layer (EDL) effects into reactive transport simulations for Geological Carbon Storage (GCS), with a specific emphasis on CO_2 dissolution within water films and its diffusive leakage through nanoporous structures. The study demonstrated that EDL effects are fundamentally dependent on length scales, exerting a dominant influence at the nanometer scale while remaining largely negligible at the micrometer scale. These electrostatic forces significantly alter ionic concentrations, local pH distributions, and reaction kinetics. By coupling Poisson-Nernst-Planck (PNP) equations with advanced kinetic models, the proposed framework offers a robust computational tool for simulating complex EDL phenomena in GCS-relevant scenarios.

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In term of novelty, this work introduced a pioneering numerical modeling framework that integrates Electrical Double Layer (EDL) effects into reactive transport simulations for Geological Carbon Storage (GCS), filling a critical gap in traditional models that often overlook electromigration. By coupling Poisson-Nernst-Planck (PNP) equations with chemical kinetics, the study revealed that electrostatic forces become a dominant driver of ionic transport and reaction kinetics at the nanometer scale, particularly in water films and caprock nanopores. The impact of this research is profound: it demonstrated that EDL effects can significantly alter local pH and accelerate corrosion risks during CO_2 leakagephenomena that are length-scale dependent and persist even under high-salinity conditions. Ultimately, this framework provides a more rigorous computational tool for project design, offering essential insights into the long-term safety and trapping efficiency of subsurface carbon sequestration.

Looking forward, future research efforts will prioritize upscaling these pore-scale EDL effects to field-scale GCS models while incorporating the complexities of permeability and porosity heterogeneity. The framework will also be extended to account for mineral precipitation/dissolution processes and the chemical behavior of multi-component brines containing ions such as Ca^{2+} and Mg^{2+}. Furthermore, we aim to validate these numerical findings against experimental data regarding EDL-induced ionic transport in nanoporous media and develop high-order numerical schemes for PNP equations to enhance both temporal accuracy and overall computational efficiency. Ultimately, by incorporating EDL effects into GCS Reactive Transport Modeling, this research significantly advances the fidelity of subsurface transport simulations and provides critical support for the design of secure and efficient carbon sequestration projects.

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References

1. Metz, Bert, Ogunlade Davidson, H. C. De Coninck, Manuela Loos, and Leo Meyer. IPCC special report on carbon dioxide capture and storage. Cambridge: Cambridge University Press, 2005.

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2. Tariq, Zeeshan, Ertugrul Umut Yildirim, Manojkumar Gudala, Bicheng Yan, Shuyu Sun, and Hussein Hoteit. “Spatialtemporal prediction of minerals dissolution and precipitation using deep learning techniques: An implication to Geological Carbon Sequestration.” Fuel 341 (2023): 127677.
3. Steefel, Carl I., Donald J. DePaolo, and Peter C. Lichtner. “Reactive transport modeling: An essential tool and a new research approach for the Earth sciences.” Earth and Planetary science letters 240, no. 3-4 (2005): 539-558.
4. Gaus, Irina. “Role and impact of CO2rock interactions during CO2 storage in sedimentary rocks.” International journal of greenhouse gas control 4, no. 1 (2010): 73-89.
5. Steefel, Carl I., C. Anthony J. Appelo, Balveer Arora, Diederik Jacques, Thomas Kalbacher, Olaf Kolditz, Vincent Lagneau et al. “Reactive transport codes for subsurface environmental simulation.” Computational Geosciences 19, no. 3 (2015): 445-478.
6. Birkholzer, Jens T., Curtis M. Oldenburg, and Quanlin Zhou. “CO2 migration and pressure evolution in deep saline aquifers.” International Journal of Greenhouse Gas Control 40 (2015): 203-220.
7. Sprocati, Riccardo, and Massimo Rolle. “On the interplay between electromigration and electroosmosis during electrokinetic transport in heterogeneous porous media.” Water research 213 (2022): 118161.
8. Leroy, P., and A. Revil. “A triple-layer model of the surface electrochemical properties of clay minerals.” Journal of Colloid and interface Science 270, no. 2 (2004): 371-380.
9. Israelachvili, Jacob N. Intermolecular and surface forces. Academic press, 2011.
10. Hunter, Robert J. Zeta potential in colloid science: principles and applications. Vol. 2. Academic press, 2013.
11. Tournassat, Christophe, and Carl I. Steefel. “Ionic transport in nano-porous clays with consideration of electrostatic effects.” Reviews in Mineralogy and Geochemistry 80, no. 1 (2015): 287-329.
12. Railsback, L. Bruce. “A geochemical view of weathering and the origin of sedimentary rocks and natural waters.” Journal of Geological Education 41, no. 5 (1993): 404-411.
13. Kutchko, Barbara G., Brian R. Strazisar, David A. Dzombak, Gregory V. Lowry, and Niels Thaulow. “Degradation of well cement by CO_2 under geologic sequestration conditions.” Environmental science and technology 41, no. 13 (2007): 4787-4792.
14. Bourg, Ian C., and Carl I. Steefel. “Molecular dynamics simulations of water structure and diffusion in silica nanopores.” The Journal of Physical Chemistry C 116, no. 21 (2012): 11556-11564.
15. Choi, Yoon-Seok, David Young, Srdjan Nei, and Linda GS Gray. “Wellbore integrity and corrosion of carbon steel in CO_2 geologic storage environments: A literature review.” International Journal of Greenhouse Gas Control 16 (2013): S70-S77.
16. Shukla, Richa, Pathegama Ranjith, Asadul Haque, and Xavier Choi. “A review of studies on CO_2 sequestration and caprock integrity.” Fuel 89, no. 10 (2010): 2651-2664.
17. Squires, Todd M., and Martin Z. Bazant. “Induced-charge electro-osmosis.” Journal of Fluid Mechanics 509 (2004): 217-252.
18. Qiao, Tian, Zhonghua Qiao, Shuyu Sun, and Shenggao Zhou. “An unconditionally energy stable linear scheme for PoissonNernstPlanck equations.” Journal of Computational and Applied Mathematics 443 (2024): 115759.

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DOI: 10.1007/978-3-032-29918-5_10

19. Bohinc, Klemen, Guilherme Volpe Bossa, Sergei Gavryushov, and Sylvio May. “Poisson-Boltzmann model of electrolytes containing uniformly charged spherical nanoparticles.” The Journal of Chemical Physics 145, no. 23 (2016).
20. Chapman, David Leonard. “LI. A contribution to the theory of electrocapillarity. ” The London, Edinburgh, and Dublin philosophical magazine and journal of science 25, no. 148 (1913): 475-481.
21. Kim, Kangjoo, Natarajan Rajmohan, Hyun-Jung Kim, Seok-Hwi Kim, Gab-Soo Hwang, Seong-Taek Yun, Baohua Gu, Min Joe Cho, and Sang-Ho Lee. “Evaluation of geochemical processes affecting groundwater chemistry based on mass balance approach: a case study in Namwon, Korea.” Geochemical Journal 39, no. 4 (2005): 357-369.
22. Bonto, Maria, Ali Akbar Eftekhari, and Hamid Nick. “Reactive transport modeling in carbonate reservoirs and assessment of the adsorption of oil polar components on the rock surface.” In 11th Annual Meeting Interpore 2019 Valencia, pp. 80-80. Universitat Politecnica De Valencia, 2019.
23. Squires, Todd M., and Martin Z. Bazant. “Induced-charge electro-osmosis.” Journal of Fluid Mechanics 509 (2004): 217-252.
24. Meng, Da, Bin Zheng, Guang Lin, and Maria L. Sushko. “Numerical solution of 3D Poisson-Nernst-Planck equations coupled with classical density functional theory for modeling ion and electron transport in a confined environment.” Communications in Computational Physics 16, no. 5 (2014): 1298-1322.
25. Shen, Jie, Jie Xu, and Jiang Yang. “A new class of efficient and robust energy stable schemes for gradient flows.” SIAM Review 61, no. 3 (2019): 474-506.
26. Liu, Chun, Cheng Wang, Steven M. Wise, Xingye Yue, and Shenggao Zhou. “A second order accurate, positivity preserving numerical method for the PoissonNernstPlanck system and its convergence analysis.” Journal of scientific computing 97, no. 1 (2023): 23.
27. Kou, Jisheng, Shuyu Sun, and Xiuhua Wang. “Linearly decoupled energy-stable numerical methods for multicomponent two-phase compressible flow.” SIAM Journal on Numerical Analysis 56, no. 6 (2018): 3219-3248.
28. Li, Xinxin, and Yi Xu. “Microstructure-based modeling for water permeability of hydrating cement paste.” Journal of Advanced Concrete Technology 17, no. 7 (2019): 405-418.
29. Tournassat, Christophe, and Carl I. Steefel. “Ionic transport in nano-porous clays with consideration of electrostatic effects.” Reviews in Mineralogy and Geochemistry 80, no. 1 (2015): 287-329.
30. Stern, Otto. “Zur theorie der elektrolytischen doppelschicht.” Zeitschrift für Elektrochemie und angewandte physikalische Chemie 30, no. 2122 (1924): 508-516.
31. Bourg, Ian C., and Carl I. Steefel. “Molecular dynamics simulations of water structure and diffusion in silica nanopores.” The Journal of Physical Chemistry C 116, no. 21 (2012): 11556-11564.
32. Steefel, Carl I., and Kate Maher. “Fluid-rock interaction: A reactive transport approach.” Reviews in mineralogy and geochemistry 70, no. 1 (2009): 485-532.
33. Steefel, Carl I., and K. T. B. McQuarrie. “Approaches to modeling of reactive transport in porous media.” Reviews in mineralogy (1996): 83-130.
34. Peaceman, Donald W. Fundamentals of numerical reservoir simulation. Vol. 6. Elsevier, 2000.
35. Tokunaga, Tetsu K. “Physicochemical controls on adsorbed water film thickness in unsaturated geological media.” Water Resources Research 47, no. 8, W08514. (2011).

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