

Document Version

Final published version

Licence

CC BY

Citation (APA)

Lu, J., Novikov, A., Zhao, M., Yan, L., Farajzadeh, R., & Voskov, D. (2026). Augmented operator-based linearization for modeling of history-dependent behavior with applications to CO2 sequestration. *Journal of Computational Physics*, 566, Article 115287. <https://doi.org/10.1016/j.jcp.2026.115287>

Important note

To cite this publication, please use the final published version (if applicable).
Please check the document version above.

Copyright

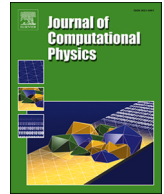
In case the licence states "Dutch Copyright Act (Article 25fa)", this publication was made available Green Open Access via the TU Delft Institutional Repository pursuant to Dutch Copyright Act (Article 25fa, the Taverne amendment). This provision does not affect copyright ownership.
Unless copyright is transferred by contract or statute, it remains with the copyright holder.

Sharing and reuse

Other than for strictly personal use, it is not permitted to download, forward or distribute the text or part of it, without the consent of the author(s) and/or copyright holder(s), unless the work is under an open content license such as Creative Commons.

Takedown policy

Please contact us and provide details if you believe this document breaches copyrights.
We will remove access to the work immediately and investigate your claim.



Augmented operator-based linearization for modeling of history-dependent behavior with applications to CO₂ sequestration

Jianxin Lu ^a, Aleksei Novikov ^a, Mengjie Zhao ^a, Lifei Yan ^a, Rouhi Farajzadeh ^a, Denis Voskov ^{a,b,*}

^a Department of Geoscience and Engineering, Delft University of Technology, Stevinweg 1, Delft, 2628 CN, the Netherlands

^b Department of Energy Science and Engineering, Stanford University, 367 Panama Street, Stanford, CA, 94305, USA

ARTICLE INFO

Keywords:

Operator-based linearization (OBL)

History-dependent behavior

Hysteresis

CO₂ sequestration

ABSTRACT

Accurate simulation of CO₂ sequestration in deep saline aquifers requires consistent treatment of history-dependent processes. Operator-Based Linearization (OBL) provides an efficient and robust framework for compositional flow simulation. However, it was originally designed for reversible physical processes, since the operator evaluation depends solely on the instantaneous thermodynamic state. In this work, we extend the OBL approach by introducing history-dependent variables into the operator parameter space while keeping them outside the Newton unknown vector, thereby augmenting OBL with history dependency while preserving the dimension of the Jacobian matrix. To apply the augmented OBL framework to hysteresis modeling, a new hysteresis algorithm is proposed, in which the maximum gas saturation is used as the history coordinate; the flow equations are solved fully implicitly with respect to the primary variables, while this history variable is held fixed during Newton iterations and updated locally after timestep convergence to capture drainage–imbibition transitions and CO₂ dissolution feedback. The augmented OBL approach is validated against the academic DARSim simulator and the commercial CMG simulator, demonstrating close agreement with conventional hysteresis modeling approaches. A sensitivity analysis of the operator parameterization shows that the resolution of the parameter space governs both accuracy and computational cost within the OBL framework. Numerical experiments in one-dimensional homogeneous and two-dimensional heterogeneous models further demonstrate that the proposed approach provides a practical and accurate means of incorporating history-dependent physics into reservoir-scale compositional simulations of CO₂ sequestration.

1. Introduction

Carbon capture and storage (CCS) is a key option to reduce CO₂ emissions from large point sources [1]. A central element of CCS is geological CO₂ storage (GCS), in which CO₂ is injected into deep subsurface formations [2,3]. Among the available options, deep saline aquifers are widely distributed and offer large storage potential [4,5]. After injection, CO₂ displaces brine, and is trapped by different mechanisms: structural, residual, dissolution and mineral trapping [6–8].

Many storage scenarios exhibit history-dependent effects, notably hysteresis in relative permeability [9] and capillary pressure [10], where the current response depends on the flow history. That can lead to strong influences on residual trapping and thus

* Corresponding author.

E-mail address: d.v.voskov@tudelft.nl (D. Voskov).

<https://doi.org/10.1016/j.jcp.2026.115287>

Received 14 December 2025; Received in revised form 11 June 2026; Accepted 5 August 2026

Available online 8 August 2026

0021-9991/© 2026 The Author(s). Published by Elsevier Inc. This is an open access article under the CC BY license (<http://creativecommons.org/licenses/by/4.0/>).

interplays with other trapping mechanisms [11]. Such history dependency is particularly important during shut-ins, pressure management, and cyclic injection (e.g., water-alternating-gas cycles) [12,13]. To represent the hysteretic process of a two-phase system, several mathematical models have been developed, including Land equation [14], Killough model [15] and Carlson model [16]. The Killough model makes more accurate predictions for gas/water systems [17]. Building on such mathematical models, the hysteretic effect on CO₂ storage is investigated in reservoir simulation [18–20].

A common challenge in a predictive simulation of flow and transport in such complex systems is a coupling of highly nonlinear properties with a fully implicit method of approximation, which showed its efficiency in the accurate modeling of CO₂ sequestration processes [21–23]. The nonlinear discrete system must be linearized at each time step. This is commonly achieved with Newton-type methods, which require the assembly of the residual and Jacobian for the coupled system of equations. Depending on the chosen formulation, all thermodynamic and transport properties and their derivatives need to be evaluated and stored with respect to the nonlinear unknowns which is an expensive part in compositional simulation [24]. To reduce expensive phase behavior computations during the course of a simulation, a tie-line-based parametrization method was proposed [25], which recast multicomponent multiphase equilibrium calculation in a parameterized compositional space. Based on this approach, adaptive parametrization approach was developed, casting the governing equations in tie-simplex parameter space [26,27].

Inspired by these methods, an operator-based linearization (OBL) approach was proposed to address high nonlinearity issues by abstracting property evaluations into state-dependent operators tabulated over a parameter space [28]. During simulation, operators and their derivatives are obtained using interpolation, which minimizes the expensive multiphase flash and property evaluations. The balance between approximation accuracy and nonlinear solver performance can be controlled through the resolution of the parameter-space hypervolume [29]. Within the OBL framework, adaptive parameterization is used to refine operators dynamically during simulation [30], translating expensive physics-based calculations into generic interpolation over supporting points that store operator values [31]. This provides an efficient and flexible means for assembling the Jacobian and residual.

Previous extensions of OBL have incorporated additional physical coordinates, such as temperature in thermal formulations or mechanical variables in coupled flow–geomechanics formulations. In such cases, the added coordinates are physical state variables that participate both in the operator parameterization and in the residual/Jacobian assembly of the coupled nonlinear system. The history coordinate considered here has a different role. It represents a local memory of the flow path rather than an additional conserved or mechanical unknown. Therefore, it is used to augment the operator parameter space, but it is not added to the Newton unknown vector. During a nonlinear solve, the history coordinate is kept fixed and only updated after timestep convergence. This distinction allows irreversible, path-dependent physics to be represented within OBL while preserving the original Jacobian dimension; the additional cost is mainly associated with the enlarged operator space and the generation of additional operator points.

To incorporate hysteretic behavior in simulation, history-dependent variables must be included in the mathematical formulation and numerical implementation [15,32]. However, in conventional OBL, the operator parameter space is coordinated only by the instantaneous thermodynamic state (primary unknowns such as pressure, composition, and temperature). This construction implicitly assumes that all physical processes are reversible in the same thermodynamic state, and thus limits the applicability of standard OBL to history-dependent process such as hysteresis.

In this study, we address this limitation by introducing a history-dependent state coordinate to the parameter space. The resulting augmented OBL framework makes operator evaluation both state- and history-dependent. The simulation history is embedded directly into the parameterization process, while consistent derivatives for the fully implicit Jacobian are preserved without ad hoc switching. History-dependent processes are handled by introducing specific history variables for the relevant physics and storing them locally for each grid cell. As a first application, we employ the augmented OBL framework to model hysteresis in capillary pressure and relative permeability for CO₂ storage in saline aquifers. We propose a new hysteresis algorithm based on the augmented OBL framework, in which the flow equations are solved fully implicitly with respect to the primary variables, while the maximum gas saturation is treated as a locally stored history coordinate, held fixed during Newton iterations, and updated after timestep convergence. Additionally, in the new hysteresis algorithm, we incorporate feedback from CO₂ dissolution into the hysteresis modeling. Taken together, these developments enable large-scale CO₂ storage simulations to represent key hysteretic trapping mechanisms within a history-consistent OBL framework whose flow solve remains fully implicit with respect to the primary variables.

The developed methods are implemented in the open-source Delft Advanced Research Terra Simulator (open-DARTS) [33]. DARTS is a scalable parallel simulator that has been successfully applied to hydrocarbon production [34], geothermal energy [31,35,36] and CO₂ sequestration [37–39]. Recently, it was extended to support fully coupled geomechanics based on finite-volume discretization [40,41]. Building on these developments, this work adds history-dependent process modeling to DARTS, extending its capabilities from reversible to hysteretic CCS processes and strengthening its role as a platform for realistic CO₂ storage scenario assessment.

The remainder of this paper is organized as follows. In Section 2, we present the governing equations, the OBL framework, and its extension to history-dependent processes, followed by the coupling to the implicit hysteresis algorithm. In Section 3, we validate the augmented OBL implementation in DARTS against the conventional hysteresis model in the academic Delft Advanced Reservoir Simulator (DARSim) and commercial CMG simulator. Besides, we perform a sensitivity analysis of the parameter-space resolution and illustrate the impact of the OBL extension on operator generation. In Section 4, we assess the performance and applicability of the proposed simulation approach through representative test cases, and Section 5 summarizes the main findings.

2. Methodology

In this section, we present the governing equations for a general isothermal compositional system and summarize the Operator-Based Linearization (OBL) methodology and its extension for history-dependent process modeling.

2.1. Governing equations

The governing equations in compositional simulation describe the conservation of mass in a system with n_p phases and n_c components:

$$\frac{\partial}{\partial t} \left(\phi \sum_{j=1}^{n_p} x_{cj} \rho_j S_j \right) + \nabla \cdot \sum_{j=1}^{n_p} (x_{cj} \rho_j \mathbf{v}_j + \mathbf{J}_{cj}) + \sum_{j=1}^{n_p} x_{cj} \rho_j \tilde{q}_j = 0, \quad c = 1, \dots, n_c. \quad (1)$$

where

- ϕ – porosity.
- x_{cj} – mole fraction of component c in phase j .
- ρ_j – molar density of phase j .
- S_j – saturation of phase j .
- $\mathbf{v}_j$ – Darcy velocity of phase j .
- $\tilde{q}_j$ – volumetric source/sink term for phase j .

The rock is assumed compressible and represented by the change of porosity through:

$$\phi = \phi_0 (1 + c_r(p - p_{\text{ref}})) \quad (2)$$

where ϕ_0 is the initial porosity, c_r is rock compressibility and p_{ref} is reference pressure.

Phase velocities are given by Darcy's law:

$$\mathbf{v}_j = -\mathbf{K} \frac{k_{rj}}{\mu_j} (\nabla p_j - \rho_j M_j g \nabla z), \quad j = 1, \dots, n_p, \quad (3)$$

where

- $\mathbf{K}$ – permeability tensor,
- k_{rj} – relative phase permeability,
- μ_j – phase viscosity,
- p_j – pressure in phase j ,
- M_j – phase molar weight,
- z – depth, positive downwards.

The diffusive flux of component c in phase j follows Fick's law:

$$\mathbf{J}_{cj} = -\phi S_j D_{cj} \rho_j \nabla x_{cj}, \quad c = 1, \dots, n_c, \quad j = 1, \dots, n_p, \quad (4)$$

where D_{cj} is scalar molecular diffusion coefficient of component c in phase j . The phase pressures p_j are driven by the capillary pressure p_c , which depends on the saturation:

$$p_n - p_w = p_c(S_w), \quad (5)$$

where n and w denote the non-wetting and wetting phases, respectively.

We adopt the overall composition formulation, assuming thermodynamic equilibrium at each nonlinear iteration of Eq. (1). The multiphase flash ensures that, for a given overall composition z_c , the equilibrium phase compositions x_{cj} and phase fractions v_j are uniquely determined. Solving the flash procedure yields all phase properties and their derivatives required to evaluate the Jacobian for Eq. (1).

2.2. Operator-based linearization approach

Operator-Based Linearization (OBL) approach is used in linearization stage, including Jacobian and residuals assembly. The efficiency of OBL stems from the localization of state-dependent operators, i.e. functions of single-cell unknowns ω , called a state. In this section, we briefly describe the application of the OBL approach to the general isothermal-compositional formulation.

We discretize the fully coupled system in space and time using the finite volume method with the first-order backward Euler time integration, a single-point upstream weighting, and the two-point flux approximation. Under the OBL strategy, the mass balance of the c th component in Eq. (1) is rewritten in the following discretized operator residual form

$$V \phi_0 [\alpha_c(\omega) - \alpha_c(\omega_n)] - \Delta t \sum_{l \in L(i)} \sum_{j=1}^{n_p} \left[\Gamma^l \beta_{cj}^l(\omega^u) \Delta \psi_j^l + \Gamma_d^l \phi_0 \gamma_j^l(\omega) \Delta \chi_{cj}^l \right] + \Delta t \theta_c(\omega) = 0. \quad (6)$$

where V is the control volume, $\omega = p, z_c$ is the physical state(unknowns) at the new timestep, ω_n is the physical state(unknowns) at the previous timestep, ω^u is the physical state of upstream block, Γ^l is the Darcy flux transmissibility, Γ_d^l is geometrical part of the transmissibility, and $L(i)$ is a set of interfaces for the gridblock. Here, $\Delta \chi_{cj}^l = \chi_{cj}(\omega^a) - \chi_{cj}(\omega^b)$ denotes the diffusive-operator gradient across interface l between neighboring cells a and b . When molecular diffusion is disabled, $D_{cj} = 0$ and the diffusive term vanishes.

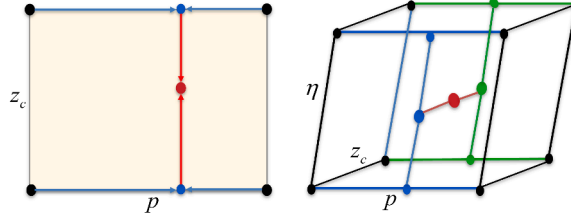


Fig. 1. Multilinear operator interpolation in parameter space before (left) and after (right) adding a history-dependent coordinate.

In this study, diffusive transport is disabled in the one-dimensional validation, resolution-sensitivity, and hysteresis examples, and is activated only in the SPE 11b example.

Here we defined the following state-dependent operators,

$$\alpha_c(\omega) = (1 + c_r(p - p_{\text{ref}})) \sum_{j=1}^{n_p} x_{cj} \rho_j S_j, \quad c = 1, \dots, n_c; \quad (7)$$

$$\beta_{cj}(\omega) = x_{cj} \rho_j k_{rj} / \mu_j, \quad c = 1, \dots, n_c, \quad j = 1, \dots, n_p; \quad (8)$$

$$\gamma_j(\omega) = (1 + c_r(p - p_{\text{ref}})) \rho_j S_j, \quad j = 1, \dots, n_p; \quad (9)$$

$$\chi_{cj}(\omega) = D_{cj} x_{cj}, \quad c = 1, \dots, n_c, \quad j = 1, \dots, n_p; \quad (10)$$

$$\theta_c(\omega) = \sum_{j=1}^{n_p} \rho_j x_{cj} q_j(\omega), \quad c = 1, \dots, n_c. \quad (11)$$

The phase-potential-upwinding (PPU) strategy for OBL parametrization is applied to model the gravity and capillary effect. The potential difference of phase j on the interface l between block a and b can be written as

$$\Delta\psi_j^l = p^a + p_j^c(\omega^a) - p^b - p_j^c(\omega^b) + \frac{\rho_j(\omega^a)M_j(\omega^a) + \rho_j(\omega^b)M_j(\omega^b)}{2} g(z^b - z^a) \quad (12)$$

where p_j^c is capillary pressure of phase j .

In the OBL framework, the state-dependent operators are tabulated over a parameter space (spanned by the chosen primary variables) and adaptively evaluated by interpolation during the Newton process. The space-dependent operators are calculated in the pre-processing stage and remain fixed during the simulation. This separation enables consistent residual and Jacobian assembly from tabulated responses while retaining the robustness of a fully implicit scheme.

2.3. History-dependent physics extension in OBL and its application

In conventional OBL, the operator parameter space is spanned by the instantaneous thermodynamic state (primary variables), which implies reversible physics. To represent history-dependent behavior, we augment the operator space with a history coordinate η (Fig. 1). The extended space is

$$\hat{\omega} = \{\omega, \eta\},$$

and all physics-based operators are tabulated and interpolated in this enlarged space. After extension, the original state ω is translated to $\hat{\omega}$ and all operators become history-aware. The history variable η is not added to the nonlinear unknowns of the Newton solve. Therefore it is updated outside the linearization, and the Jacobian size is unchanged.

In the extended OBL framework, the set of operators is interpolated as

$$\{\alpha_c, \beta_{cj}, \rho_j, p_j^c, \gamma_j, \chi_{cj}\}(\hat{\omega}) = f(\hat{\omega}), \quad (13)$$

where f denotes the interpolant that reconstructs operator values between supporting points. For a regular grid in the extended state space, we define

$$\hat{\omega} = \bar{p} \times \bar{z}_1 \times \dots \times \bar{z}_{n_c-1} \times \bar{\eta} \quad (\times T \text{ if thermal}), \quad (14)$$

with axis point sets

$$\bar{p} = \{p_1, \dots, p_{N_p}\}, \quad \bar{z}_i = \{z_{i,1}, \dots, z_{i,N_{z_i}}\}, \quad i = 1, \dots, n_c - 1, \quad \bar{\eta} = \{\eta_1, \dots, \eta_{N_h}\}, \quad (15)$$

where $\bar{p}$, $\bar{z}_i$ denote conventional state-space axes coordinates, while $\bar{\eta}$ represents coordinates of the additional history axis. The distribution of points along the axes in Eq. (15) can be dictated by specific model properties, e.g. to achieve higher resolution of phase envelope. By default, they are uniformly distributed for simplicity. Generally, the history coordinate η is not limited to a single dimension.

For local-memory processes, any model that can be written as an operator response $f(\omega, \eta)$, with η denoting internal history variables, can be incorporated by adding these variables to the operator parameter space. The model-specific part is the definition

and update rule of η . If a hysteresis or other history-dependent model requires several memory variables, the history vector can be written as $\eta = (h_1, h_2, h_3, \dots)$. The operator-space dimension then becomes $d = d_\omega + d_\eta$. For a full tensor-product table with N_k supporting points along axis k , the storage for N_{op} operators scales as $O(N_{\text{op}} \prod_{k=1}^d N_k)$, and the same scaling applies to a priori operator generation. In the adaptive OBL strategy used here, however, operators are generated on demand only in the regions visited by the simulation. If M operator points are generated, the practical storage scales as $O(N_{\text{op}} M)$, plus interpolation metadata, and the generation cost scales with M and the cost of the underlying property evaluator. Additional history coordinates therefore increase storage and operator-generation cost, but the Newton unknown vector and Jacobian dimension remain unchanged.

Different strategies can be employed to evaluate operator values between the vertices of the regular grid defined in Eq. (14), such as linear barycentric or multilinear interpolations [42]. Here we outline the procedure for the multilinear interpolation. Although this interpolation is computed recursively across dimensions, the resulting expression for any scalar operator O at an arbitrary state $\hat{\omega}$ can be written in a compact form as:

$$I_h[O](\hat{\omega}) = \sum_{k \in \mathcal{N}(\hat{\omega})} w_k(\hat{\omega}) O(\hat{\omega}^k), \quad \sum_k w_k(\hat{\omega}) = 1, \quad w_k \geq 0, \quad (16)$$

where $\mathcal{N}(\hat{\omega})$ denotes the set of vertices of the hypercube containing $\hat{\omega}$, and $w_k(\hat{\omega})$ are the corresponding interpolation weights. For a state outside the tabulated augmented range, the closest boundary hypercube is used and the same multilinear stencil is continued as a linear extrapolation along the out-of-range coordinate(s); this treatment applies to both thermodynamic and history axes. The derivatives of the interpolant with respect to the instantaneous and historical components of the state $\hat{\omega} = \{\omega, \eta\}$ are obtained analytically for given interpolant shape:

$$\frac{\partial}{\partial \omega} I_h[O](\hat{\omega}) = \sum_{k \in \mathcal{N}(\hat{\omega})} \frac{\partial w_k}{\partial \omega}(\hat{\omega}) O(\hat{\omega}^k), \quad \frac{\partial}{\partial \eta} I_h[O](\hat{\omega}) = \sum_{k \in \mathcal{N}(\hat{\omega})} \frac{\partial w_k}{\partial \eta}(\hat{\omega}) O(\hat{\omega}^k). \quad (17)$$

Let $d = d_\omega + d_\eta$ be the number of variables in the extended space $\hat{\omega}$. For isothermal compositional problems, $d_\omega = n_c$ (pressure plus $n_c - 1$ overall compositions), while d_η corresponds to the number of history variables included, which is equal to one for the hysteretic examples considered in this work.

During a nonlinear iteration, η is treated as a frozen parameter, so the chain rule gives

$$\frac{\partial r_c}{\partial \omega} = \left(\frac{\partial r_c}{\partial \omega} \right) \left(\frac{\partial \omega}{\partial \omega} \right)_\eta. \quad (18)$$

Expanding $\partial r_c / \partial \omega$ from (6) yields

$$\frac{\partial r_c}{\partial \omega} = V \phi_0 \frac{\partial \alpha_c}{\partial \omega} - \Delta t \sum_{l \in L(i)} \sum_{j=1}^{n_p} \left(\Gamma^l \left(\frac{\partial \beta_{cj}^l}{\partial \omega} \Delta \psi_j^l + \beta_{cj}^l \frac{\partial \Delta \psi_j^l}{\partial \omega} \right) + \Gamma_d^l \phi_0 \left(\frac{\partial \gamma_j^l}{\partial \omega} \Delta \chi_{cj}^l + \frac{\partial \Delta \chi_{cj}^l}{\partial \omega} \gamma_j^l \right) \right), \quad (19)$$

where the derivatives are obtained from the interpolation in (17). Because η is excluded, the Jacobian dimension is identical to that of the reversible OBL system.

The history coordinate extends the operator parameter space but is not added to the nonlinear unknown vector. During Newton iterations, it is held fixed, so the residual is linearized consistently with respect to the primary variables at a fixed history state. This treatment preserves the standard Newton structure when the history variable is not an independent conserved unknown, the history-dependent operators are continuous and bounded for a fixed history state, and the history update is local, bounded, and applied after timestep convergence. Thus, the flow problem remains fully implicit with respect to the primary variables, while the history variable is updated locally after convergence.

2.4. Application of extended OBL in hysteresis modeling

First, we need to identify the history-dependent state variable corresponding to the hysteresis process. For two-phase CO_2 -brine flow, we use the maximum gas saturation, $S_{g,\text{max}}$, as the history-dependent coordinate, so that $\eta = \{S_{g,\text{max}}\}$. During hysteresis modeling, the operators are queried and interpolated in the expanded space $\hat{\omega} = \{\omega, S_{g,\text{max}}\}$. This choice is specific to the Killough hysteresis model [15] used in this work. In this model, the scanning curves are anchored by the reversal saturation, represented here by the maximum historical gas saturation, and the residual gas saturation is computed from the maximum historical non-wetting saturation through Land's relation. Therefore, the current gas saturation S_g together with the stored $S_{g,\text{max}}$ provides the information needed to select the primary-drainage or scanning branch and to evaluate the hysteretic k_r and p_c operators.

Here, we propose a new hysteresis algorithm as an application of the augmented OBL framework. The algorithm uses $S_{g,\text{max}}$ as the history coordinate: it is held fixed during Newton iterations for operator evaluation and updated locally after timestep convergence. Conventional hysteresis implementations may express saturation functions as $k_r, p_c = f(S, h, b)$, where h denotes history variables and b is an explicit drainage/imbibition branch label. Such branch switching is often treated carefully or delayed to the end of a timestep to improve nonlinear robustness in fully implicit reservoir simulators [11,43,44]. In the present Killough implementation, no separate branch label is stored as an additional history variable. Instead, the effective branch indicator is inferred locally during operator evaluation by comparing the current Newton saturation iterate S_g^{n+1} with the fixed history coordinate $S_{g,\text{max}}^n$. Thus, the transition decision follows the current primary-variable iterate, while the memory variable itself is updated after timestep convergence. This avoids using a separate drainage/imbibition label as a stored memory variable, unlike implementations that track such a label explicitly or infer it from previous time levels. Here, we adopted the logic as follows: (i) using $S_{g,\text{max}}$ as the sole history-dependent

input to k_r and p_c ; and (ii) holding $S_{g,\max}$ fixed during Newton iterations and updating it locally after timestep convergence using the converged saturation S_g^n . The evaluation of k_r and p_c at time $n+1$ is summarized in Algorithm (1).

Algorithm 1 Relative permeability and capillary pressure calculation with hysteresis effect.

```

Input:  $S_g^{n+1}$ ,  $S_{g,\max}^n$ 
1: if  $S_g^{n+1} \geq S_{g,\max}^n$  then
  /* Hysteresis calculation in primary drainage curve */
2:    $k_{rg}^{n+1} \leftarrow k_{rg,d}(S_g^{n+1}, S_{g,\max}^n)$ 
3:    $p_c^{n+1} \leftarrow p_{c,d}(S_g^{n+1}, S_{g,\max}^n)$ 
4: else
  /* Hysteresis calculation in scanning curve */
5:    $k_{rg}^{n+1} \leftarrow k_{rg,i}(S_g^{n+1}, S_{g,\max}^n)$ 
6:    $p_c^{n+1} \leftarrow p_{c,i}(S_g^{n+1}, S_{g,\max}^n)$ 
7: end if

```

After convergence of the local nonlinear iteration at time level n , as shown in Algorithm (2), $S_{g,\max}^n$ required in the next timestep $n+1$ is updated according to the S_g^n after the iteration converged. Since dissolution can reduce the local residual gas saturation S_{gr} ,

Algorithm 2 $S_{g,\max}$ updating after iteration converged in a single cell.

```

1: if  $S_g^n > S_{g,\max}^{n-1}$  then
  /* Update local  $S_{g,\max}$  on primary drainage curve */
2:    $S_{g,\max}^n = S_g^n$ 
3: else
  /* Update local  $S_{g,\max}$  on scanning curves */
4:    $S_{gr}^n = S_{g,\max}^{n-1} / (1 + C_{Land} * S_{g,\max}^{n-1})$ 
5:   if  $S_g^n < S_{gr}^n$  then
    /* Update  $S_{g,\max}$  when  $\text{CO}_2$  dissolved */
6:      $S_{g,\max}^n = S_g^n / (1 - C_{Land} * S_g^n)$ 
7:   else
8:      $S_{g,\max}^n = S_{g,\max}^{n-1}$ 
9:   end if
10: end if

```

the update rules above regenerate the scanning curves when dissolution drives S_g^n below S_{gr}^n . In effect, as shown in Algorithm (2), this hysteresis algorithm self-consistently updates both S_{gr} (via the Land relation) and the implied $S_{g,\max}$ used in the Killough scanning, thereby capturing dissolution impacts on hysteresis. Therefore, $S_{g,\max}$ is monotone non-decreasing only when dissolution feedback is inactive. With dissolution feedback, it should be interpreted as an effective history coordinate rather than a strict maximum saturation; it may decrease when dissolution reduces S_g below the residual saturation implied by the previous history state.

This dissolution–hysteresis coupling should be interpreted as a physically motivated constitutive closure within the Killough model. Dissolution changes the phase partitioning and reduces the local gas saturation through the compositional mass-conservation solve; the update of $S_{g,\max}$ does not create or remove CO_2 mass, but only updates the history parameter used in subsequent evaluations of k_r and p_c . Therefore, local and global component mass conservation are controlled by the conservative flow equations and phase-equilibrium calculation, while the history update modifies only the saturation-function branch selected after convergence. The inverse Land update is applied only when $S_g < S_{gr}$. Under this condition, $C_{Land} S_g < 1$, so the reconstructed effective $S_{g,\max} = S_g / (1 - C_{Land} S_g)$ remains well defined and lower than the previous history state. The value of C_{Land} controls the strength of residual trapping and therefore the magnitude of dissolution feedback.

Since the wettability of the aquifer is the key consideration in hysteresis modeling [3], in this study, we assume a strongly water-wet rock for all the cases. Consequently, hysteresis is most pronounced in the non-wetting gas phase and has only a minor effect on the wetting phase [13,45]. Although predictions are sensitive to rock type through the required parameter settings (e.g., endpoints, Corey exponents, and C_{Land}), a systematic analysis of rock-type dependence is beyond the scope of this work. All information and parameter settings needed to reproduce the Killough hysteresis implementation in this study are provided in Appendix A. To illustrate this model, we perform a single-cell test with cyclic drainage and imbibition processes. Fig. 2 shows the resulting loops for relative permeability and capillary pressure.

3. Validation and sensitivity analysis

In this section, we validate the extended OBL framework for hysteresis modeling against the open-source academic simulator DARSim and the commercial simulator CMG, both of which have been widely employed for studies involving hysteresis [11,19,46]. In

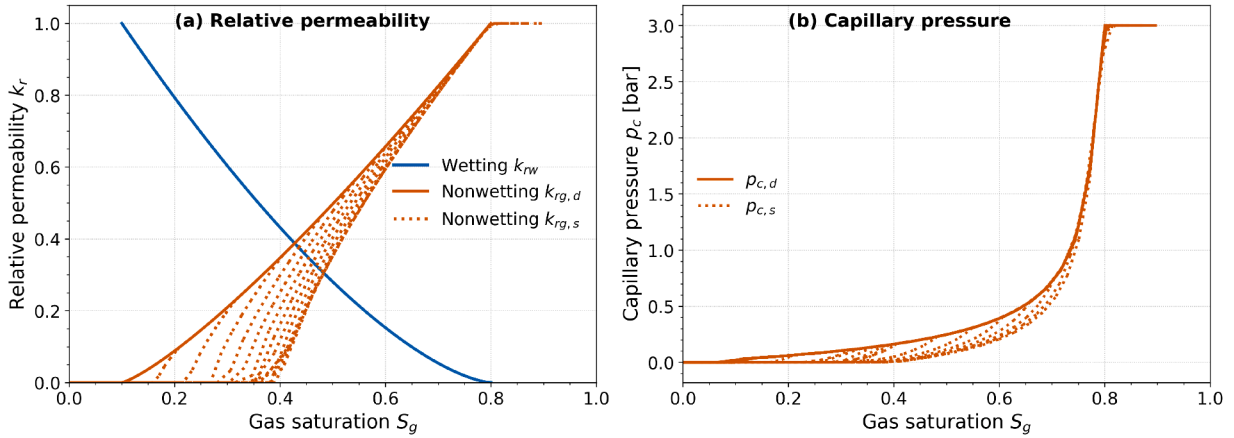


Fig. 2. Single-cell hysteresis test: (a) relative permeability and (b) capillary pressure under cyclic drainage-imbibition.

Table 1

Physical parameters and simulation settings for the 1D benchmark case.

Parameter	Value	Unit
Domain length	100	m
dx, dy, dz	1, 1, 1	m
Porosity	0.20	–
Absolute permeability	40	mD
Initial pressure	250	bar
Bottom-hole pressure (producer)	250	bar
Temperature	338.15	K
Gas-phase density	800	kg/m ³
Aqueous-phase density	1000	kg/m ³
Gas-phase viscosity	0.064	cP
Aqueous-phase viscosity	0.47	cP
Injection rate (CO ₂ /H ₂ O)	6.4 / 1.728	kg/day
Total simulation time	1000	days
Total injection time (CO ₂ + H ₂ O)	600	days

addition, we present a sensitivity study to quantify errors and performance impacts induced by the parameter-space parametrization, and assess the numerical behavior of the extended OBL framework relative to the conventional (reversible) OBL formulation.

3.1. Validation against conventional simulators

To validate hysteresis modeling with the extended OBL framework in DARTS, we consider a horizontal 1D column, no gravity cyclic injection case designed to make hysteresis effects more pronounced, and simulate it with DARTS, DARSim, and CMG under harmonized physics. To minimize discrepancies arising from differences in thermodynamic and property correlations between simulators, we use constant K -values in the flash calculations and keep phase properties (densities and viscosities) constant. Table 1 summarizes the physical parameters and simulation settings. The boundary conditions are a specified mass-rate control at the inlet and a fixed production pressure of 250 bar at the outlet. To enhance hysteretic behavior, we impose a three-stage injection schedule: CO₂ injection from day 0 to day 400, a shut-in period (no injection) from day 400 to day 800, followed by water injection from day 800 to day 1000.

Fig. 3 compares gas saturation results between all three simulators at three characteristic times: 400, 800, and 1000 days, corresponding to the end of CO₂ injection, the end of shut-in (start of water injection), and the end of the simulation, respectively. In this 1D model, hysteresis arises from capillary-driven backflow of water during the shut-in period, and becomes more pronounced after the subsequent water injection stage, as is evident from the profiles at 1000 days. Across all stages, the extended OBL implementation in DARTS shows excellent agreement with DARSim and CMG in gas saturation profile. This 1D case demonstrates that the extended OBL formulation successfully reproduces hysteresis behavior consistent with the results generated by conventional simulation techniques.

3.2. Sensitivity analysis of parameter space

With spatial and temporal discretizations fixed, the dominant numerical error arises from the accuracy of the nonlinear physics representation, which in OBL is controlled by the parameter-space resolution (the discretization used for α_c , β_c , and related operators). We therefore perform a performance and sensitivity study of the OBL framework over several operator resolutions, based on the

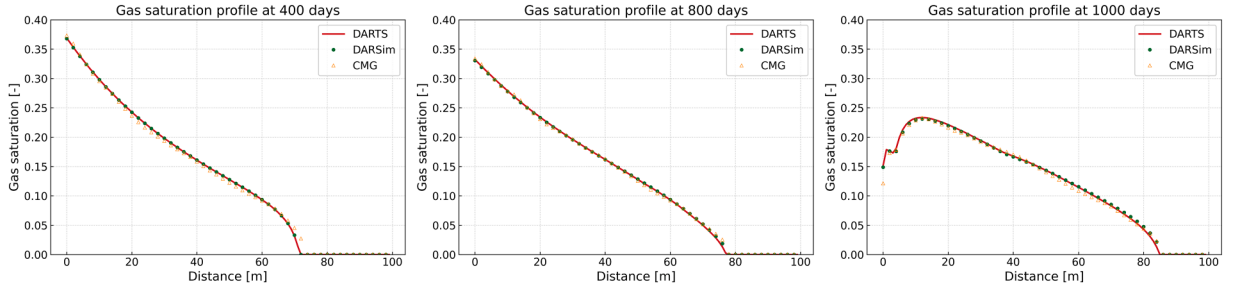


Fig. 3. Gas saturation profile in three timesteps: comparison between DARTS (extended OBL), DARSim and CMG for hysteresis modeling at three characteristic times.

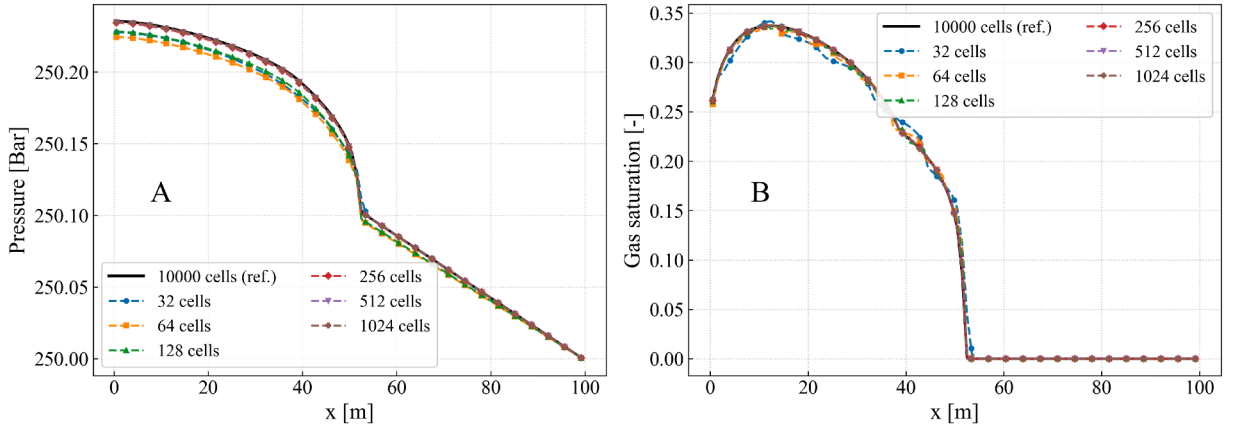


Fig. 4. Simulation results at 1000 days for different operator resolutions: (A) pressure, (B) gas saturation.

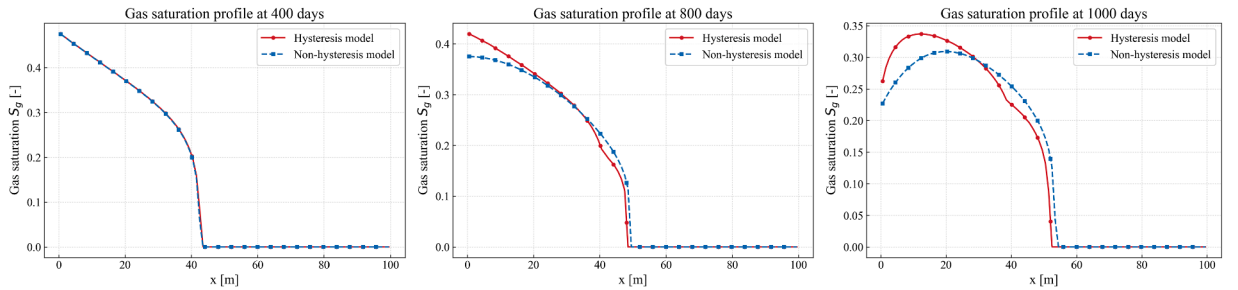


Fig. 5. Gas saturation profiles at different times: comparison between non-hysteretic and hysteretic simulations.

1D reservoir model described in Section 3.1. Compared with the simplified benchmark presented in Section 3.1, we adopt here a thermodynamically rigorous brine- CO_2 model for equilibrium and property calculations, as introduced in [39,47], to provide a more realistic test case.

We measure error using the accumulated root-squared error (ARSE), taking the highest-resolution results (10^4 points in the operator space) as reference. For gas saturation at a given time,

$$\text{ARSE}(n) = \sqrt{\sum_{i=1}^N [\text{sat}V(i, n) - \text{sat}V(i, 10^4)]^2}, \quad (20)$$

where n is the operator-space resolution, N the number of grid cells, and i the cell index.

Fig. 4 compares the pressure, CO_2 molar fraction, and gas saturation at the end of the simulation (1000 days) for different operator resolutions. Low resolutions exhibit noticeably poorer agreement with the reference. Table 2 summarizes performance and numerical error in the parameter-space sensitivity analysis. As expected, increasing the resolution reduces errors in all variables, while the number of nonlinear iterations per simulation remains in a comparable range. The reference case (10^4 -points) provides the most accurate solution but is also the most expensive. The 1024-point case offers a favorable compromise between accuracy and

Table 2

1D CO₂ injection: performance and accuracy versus operator resolution. Iters. = total Newton iterations; E_p , E_z , and E_s are ARSEs(%) in pressure, overall composition, and saturation, respectively, against the reference case.

Resolution	Iters.	E_p (%)	E_z (%)	E_s (%)	CPU (s)
32	1122	6.39	4.21	8.07	0.51
64	1186	8.67	1.97	4.00	0.45
128	1228	7.93	1.30	3.12	0.71
256	1363	0.97	0.44	1.11	1.53
512	1419	0.53	0.21	0.53	2.85
1024	1455	0.26	0.08	0.21	3.80
Reference	1494	–	–	–	41.83

Note: CPU timings were obtained on AMD EPYC 7742 CPUs using one Python process, no MPI, one thread, and SuperLU solver.

Table 3

Simulation performance of hysteretic and non-hysteretic runs.

Model	Iterations	Generated points	CPU (s)
Hysteretic	1455	12,571	3.80
Non-hysteretic	1154	1910	1.17

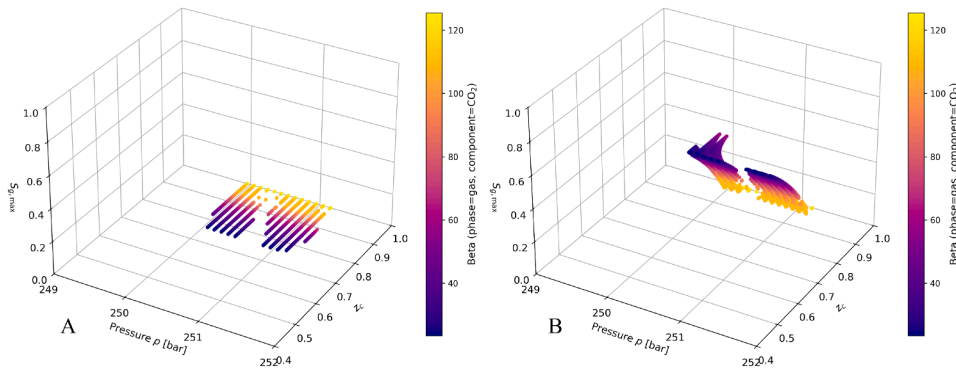


Fig. 6. Operator points generated in parameter space for (A) non-hysteretic and (B) hysteretic simulations, colored by the operator value β_c . Axis ranges are adjusted to the actual domain explored during the simulation.

computational cost, reducing saturation errors to below 0.3% while being more than an order of magnitude cheaper than the reference case.

In addition, non-hysteretic simulations are carried out alongside hysteretic simulations to highlight the impact of the extended OBL framework. Fig. 5 compares gas saturation profiles in the 1D model between hysteretic and non-hysteretic runs at the same three timesteps. Fig. 6 shows the distribution of operator points in the parameter space before (non-hysteretic) and after (hysteretic) OBL extension. As summarized in Table 3, hysteresis modeling increases the number of nonlinear iterations and generates more operator points in this one-dimensional test, as expected from the added history-dependent coordinate and scanning-curve dynamics. This also explains the significant increase in simulation time, as point generation dominates the computational cost in these simplified one-dimensional models.

4. Numerical results

Building on the methodology in Section 2, we apply the extended OBL framework coupled with the new hysteresis algorithm to model realistic problems relevant to CO₂ sequestration in saline aquifers. Different reservoir models (a 1D homogeneous reservoir and a 2D heterogeneous reservoir) and injection strategies are considered to test the new framework's capability to model history-dependent (hysteretic) processes. In particular, we utilize continuous CO₂ injection and cyclic injection with water-alternating-gas (WAG) sequences, which can enhance hysteresis effects.

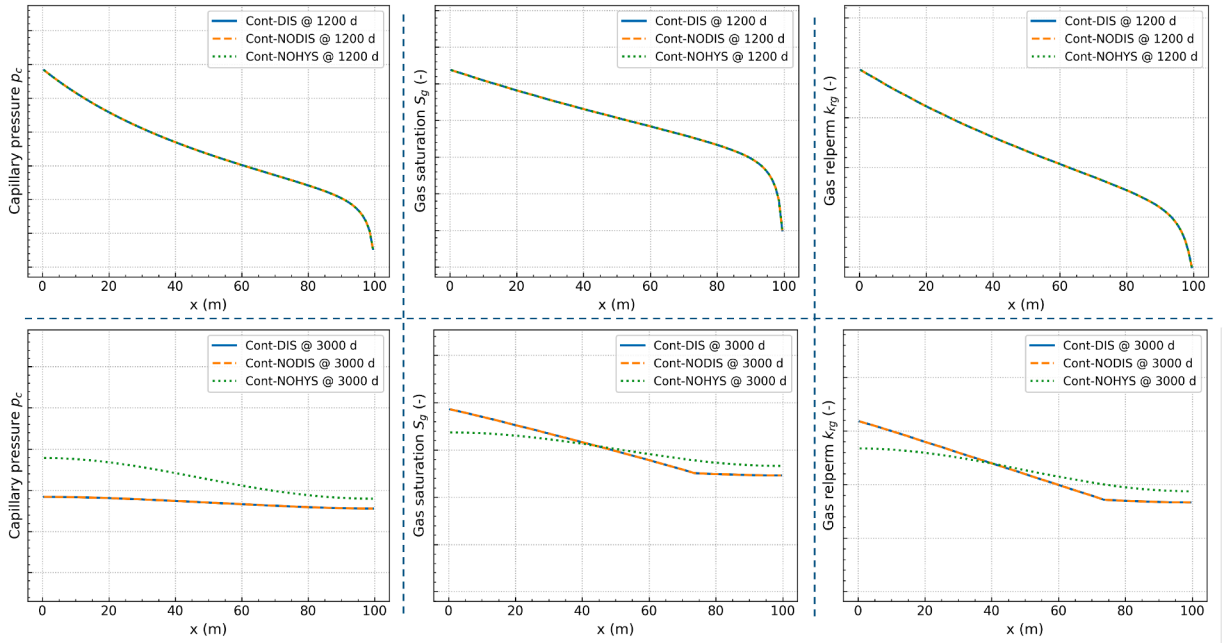


Fig. 7. Profiles of capillary pressure, gas saturation, and gas relative permeability at 1200 days (end of injection) and 3000 days (end of simulation) for the continuous-injection 1D case.

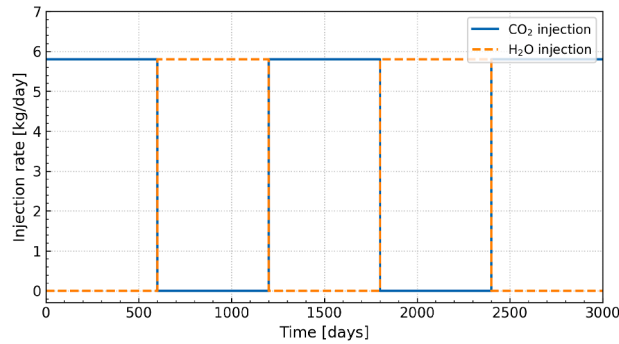


Fig. 8. Cyclic injection schedule (WAG) in the 1D model.

4.1. One-dimensional homogeneous reservoir

The main objective of this example is to illustrate the impact of dissolution feedback on hysteresis. We use the same 1D reservoir setup described in Section 3.1 but with higher resolution ($N_b = 1000$ gridblocks), together with the same fluid-property models as those in Section 3.2. For the continuous-injection case, CO_2 is injected at a constant mass rate of 5.8 kg/day for 1200 days, while the production pressure is fixed at 250 bar. Fig. 7 shows profiles of capillary pressure, gas saturation, and gas relative permeability. At 1200 days (end of injection), all models show close agreement. By 3000 days, the hysteretic runs (*Cont-DIS* and *Cont-NODIS*) deviate from the non-hysteretic baseline (*Cont-NOHYS*), while the two hysteretic variants remain in close agreement with each other. This indicates that dissolution feedback in *Cont-DIS* has minimal effect on hysteresis under continuous injection.

For the cyclic injection case, the injection schedule follows the pattern shown in Fig. 8.

Fig. 9 shows the corresponding profiles. The two hysteretic variants—with dissolution feedback (*Cyclic-DIS*) and without it (*Cyclic-NODIS*)—match at 1200 days (end of the WAG sequence) but diverge by 3000 days (post-injection). *Cyclic-NODIS* exhibits higher p_c , S_g , and k_{rg} than *Cyclic-DIS*, with the largest differences near the injector.

To diagnose the mechanisms, we monitor the first grid cell near the injector (Fig. 10). In this case, *Cyclic-DIS* and *Cyclic-NODIS* begin to diverge after the end of the first WAG cycle (vertical dashed line). With dissolution feedback, the history variable $S_{g,\max}$ (panel C) is updated according to Algorithm 2, thereby generating a new scanning curve for k_r and p_c (panels B and A). Without feedback, *Cyclic-NODIS* continues to use the scanning curve anchored at the start of the WAG phase, leading to higher p_c , S_g , and k_{rg} at later times.

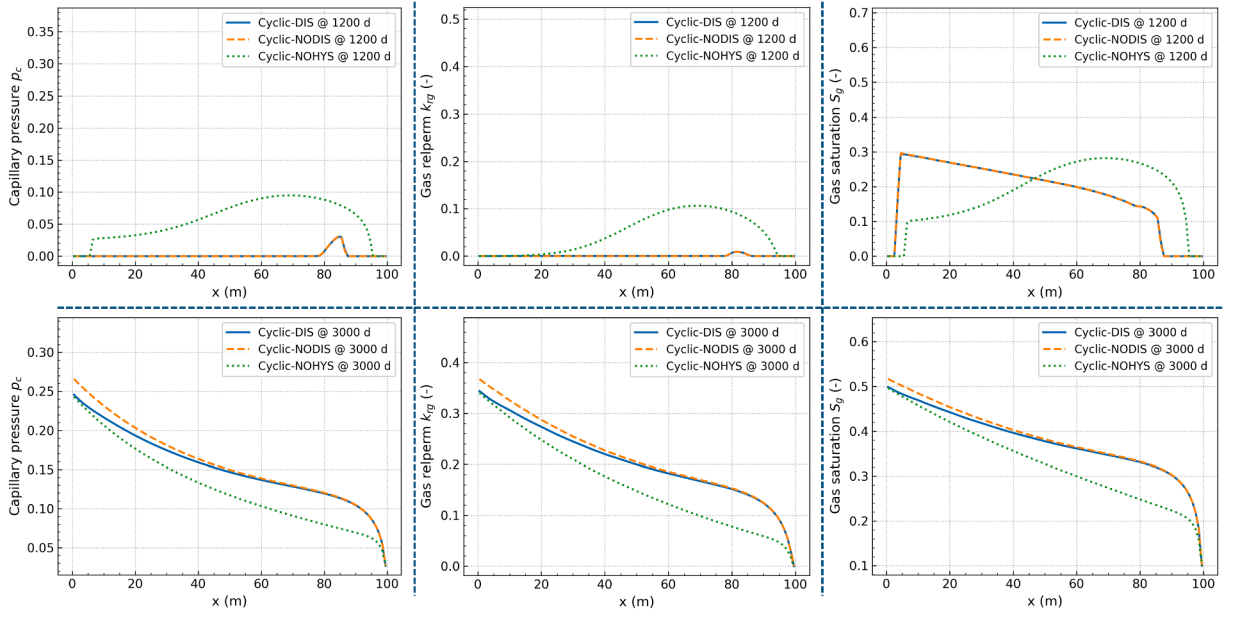


Fig. 9. Profiles of capillary pressure, gas saturation, and gas relative permeability at 1200 days (end of the WAG sequence) and 3000 days (end of simulation) for the cyclic-injection 1D case.

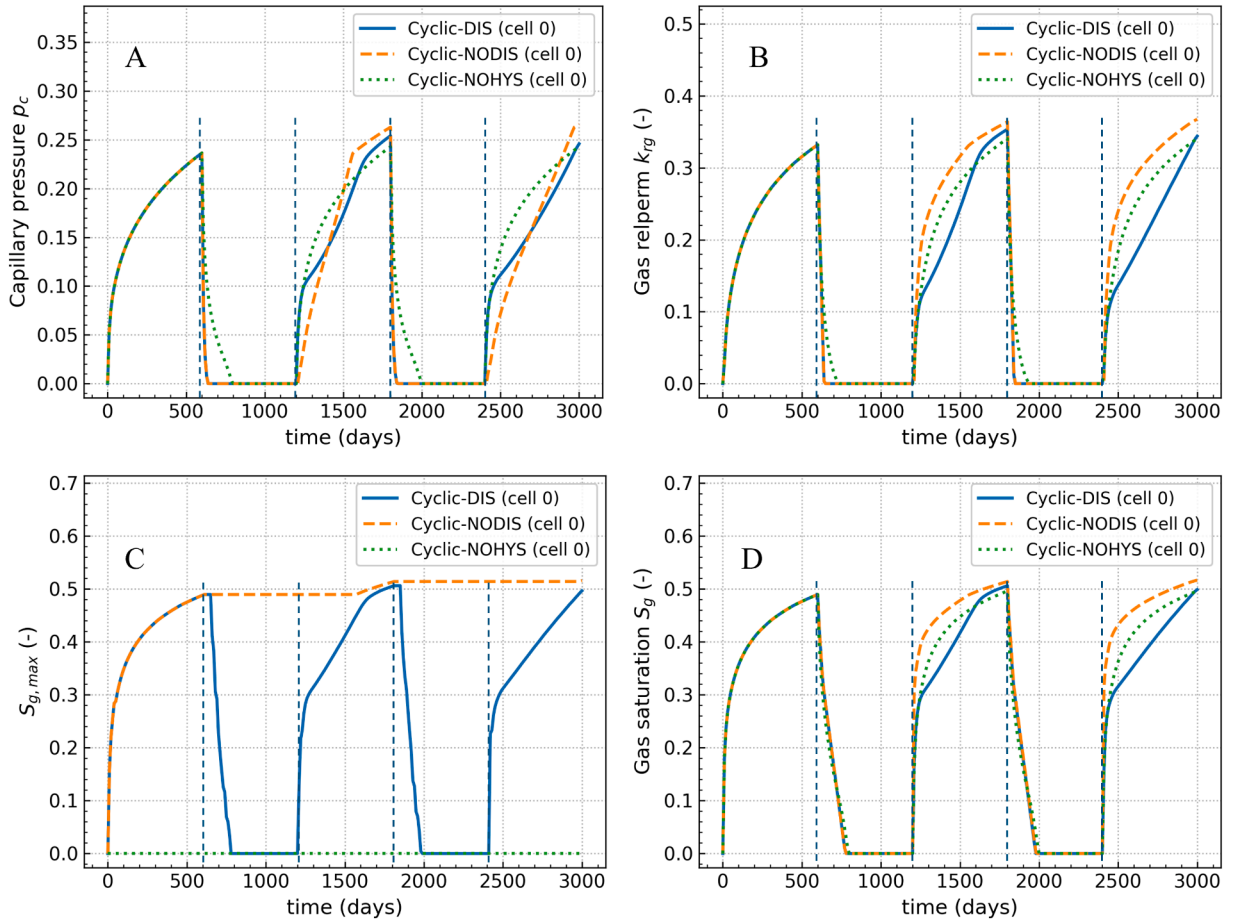


Fig. 10. Time evolution in the first grid cell for the cyclic 1D case: (A) capillary pressure; (B) gas relative permeability; (C) $S_{g,max}$; (D) gas saturation. Vertical dashed lines indicate cycles of WAG.

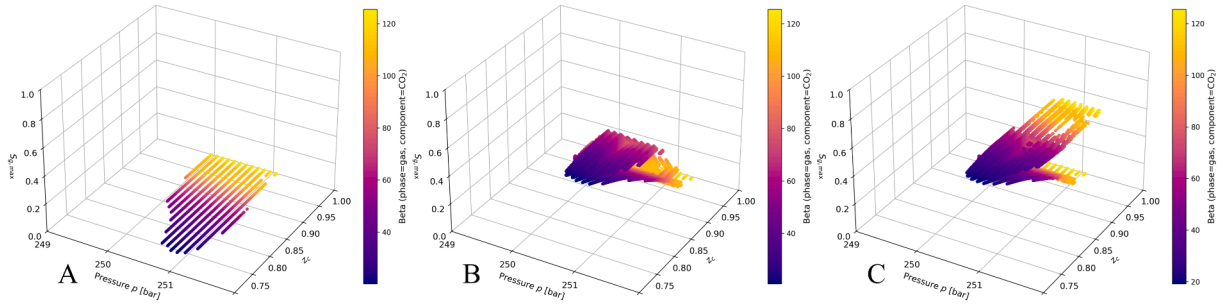


Fig. 11. Operator points generated in parameter space in (A) the non-hysteretic case and hysteretic cases (B) with and (C) without dissolution feedback, colored by the operator value β_c .

Table 4

Numerical performance for the 1D example.

Model	Iters.	Generated points	CPU (s)
Dis-hysteresis	4 086	95 653	35.3
Nodis-hysteresis	4 057	111 023	40.0
Non-hysteresis	3 220	4 940	3.2

Table 5

Facies properties of the SPE 11b model.

Facies no.	Permeability k (mD)	Porosity ϕ	S_{wc}	S_{gr}
1	0.101	0.10	0.32	0.10
2	101	0.20	0.14	0.10
3	202	0.20	0.12	0.10
4	507	0.20	0.12	0.10
5	1013	0.20	0.12	0.10
6	2027	0.25	0.12	0.10
7	0	0.00	0.10	0.10

We further compare the generation of operator points among these cases. As shown in Fig. 11, compared with non-hysteretic simulations, hysteretic simulations generate additional operator points along the history coordinate $S_{g,max}$. The range and distribution of operator points in the hysteretic cases with and without dissolution feedback also differ. As summarized in Table 4, the number of operator points in hysteretic simulations is roughly twenty times larger than in the non-hysteretic simulation, which substantially increases the simulation complexity. This also affects the overall simulation time, since in this simple one-dimensional example—similar to the case in the previous section—the majority of the computational effort is devoted to property evaluations. Moreover, fewer operator points are generated in the hysteretic case with dissolution feedback (*Dis-hysteresis*) than in the case without dissolution feedback (*Nodis-hysteresis*).

4.2. Two-dimensional heterogeneous reservoir

We now examine a heterogeneous two-dimensional aquifer, which is a more realistic representation for CO₂ storage. The model is based on the SPE11b benchmark, representative of conditions on the Norwegian Continental Shelf [48,49]. The reservoir structure and well configuration are shown in Fig. 12; facies properties are listed in Table 5.

The original SPE 11b setup includes thermal effects. Here, however, we use an isothermal variant to isolate the impact of the proposed history-dependent OBL extension on flow, hysteresis, residual trapping, and dissolution, and to reduce computational cost. This simplification may affect CO₂ properties, solubility, and plume migration quantitatively. The domain is discretized on an 840 × 120 grid and is initially water-saturated. We consider two injection strategies: (i) continuous injection—two injectors at a rate of 3024 kg/day for 50 years; and (ii) cyclic injection with the schedule shown in Fig. 13. The total simulated time is 1000 years. Fluid-property calculations follow the same choices as in Section 3.2, and we now also include diffusive transport. For hysteresis examples, we employ the hysteresis model with dissolution feedback. The operator parametrization used here follows the resolution choices previously assessed for the SPE11 benchmark with open-DARTS [47].

Figs. 14 and 15 show gas saturation and dissolved CO₂ at three representative times: end of injection (50 years for continuous injection, 60 years for cyclic injection), early post-injection (100 years), and late post-injection (1000 years). In both strategies, near-well regions exhibit clear differences between the hysteretic and non-hysteretic models by 100 years. Buoyant CO₂ rises locally and induces imbibition; in the hysteretic model this leads to gradual evolution of residual gas saturation via scanning curves, whereas the non-hysteretic model maintains fixed residual values. Other field-scale patterns are broadly similar in these snapshots.

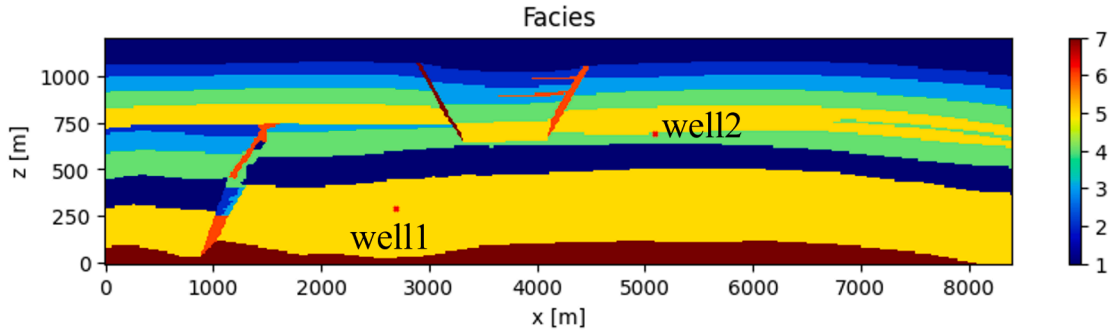


Fig. 12. Reservoir structure and well placement in the SPE 11b model.

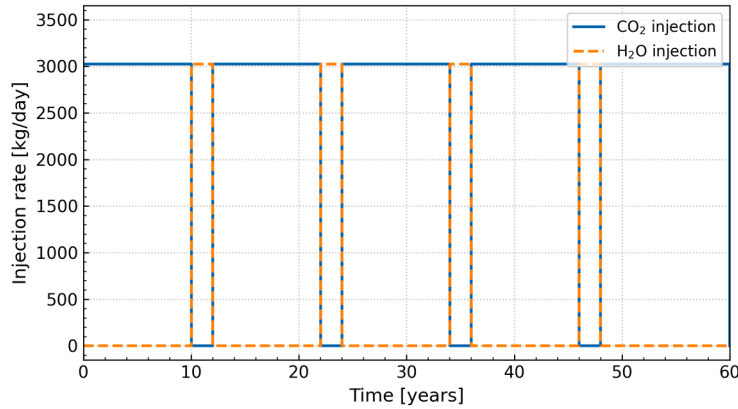


Fig. 13. Cyclic injection schedule in the SPE 11b model.

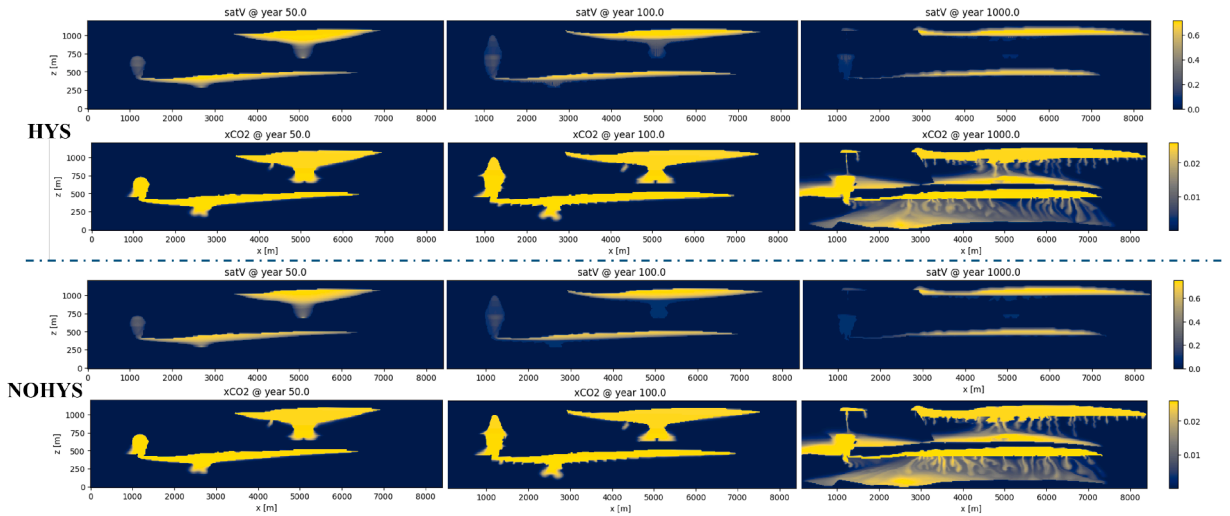


Fig. 14. Continuous injection in the SPE 11b model: comparison of hysteretic (HYS) and non-hysteretic (NOHYS) simulations at three representative times.

To quantify differences that are not obvious from snapshots, we partition injected CO_2 into mobile ($S_g > S_{gr}$), immobile (residually trapped, $S_g \leq S_{gr}$), and dissolved fractions (liquid-phase CO_2 molar fraction), and compute sweep efficiency. Fig. 16 summarizes the temporal evolution. The hysteretic model yields a higher immobile CO_2 fraction than the non-hysteretic model after injection shut-in. As post-injection flow proceeds, this gap narrows because dissolution acts first in immobile gas regions, gradually reducing the immobile fraction. The dissolved fraction increases in both cases but remains lower with hysteresis overall, reflecting earlier immobilization that limits subsequent dissolution from the mobile phase.

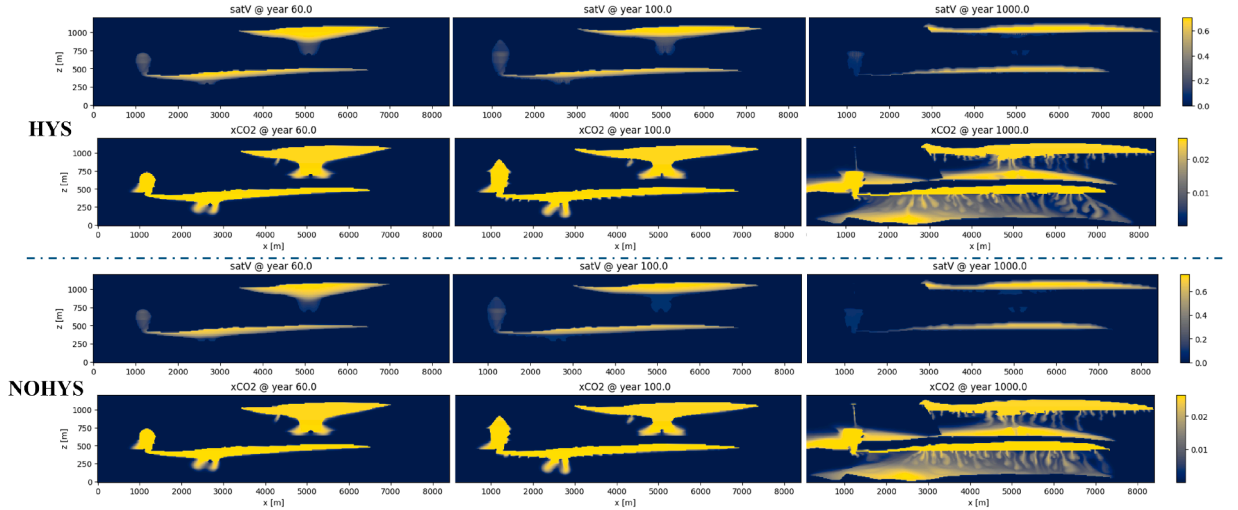


Fig. 15. Cyclic injection in the SPE 11b model: comparison of hysteretic (HYS) and non-hysteretic (NOHYS) simulations at three representative times.

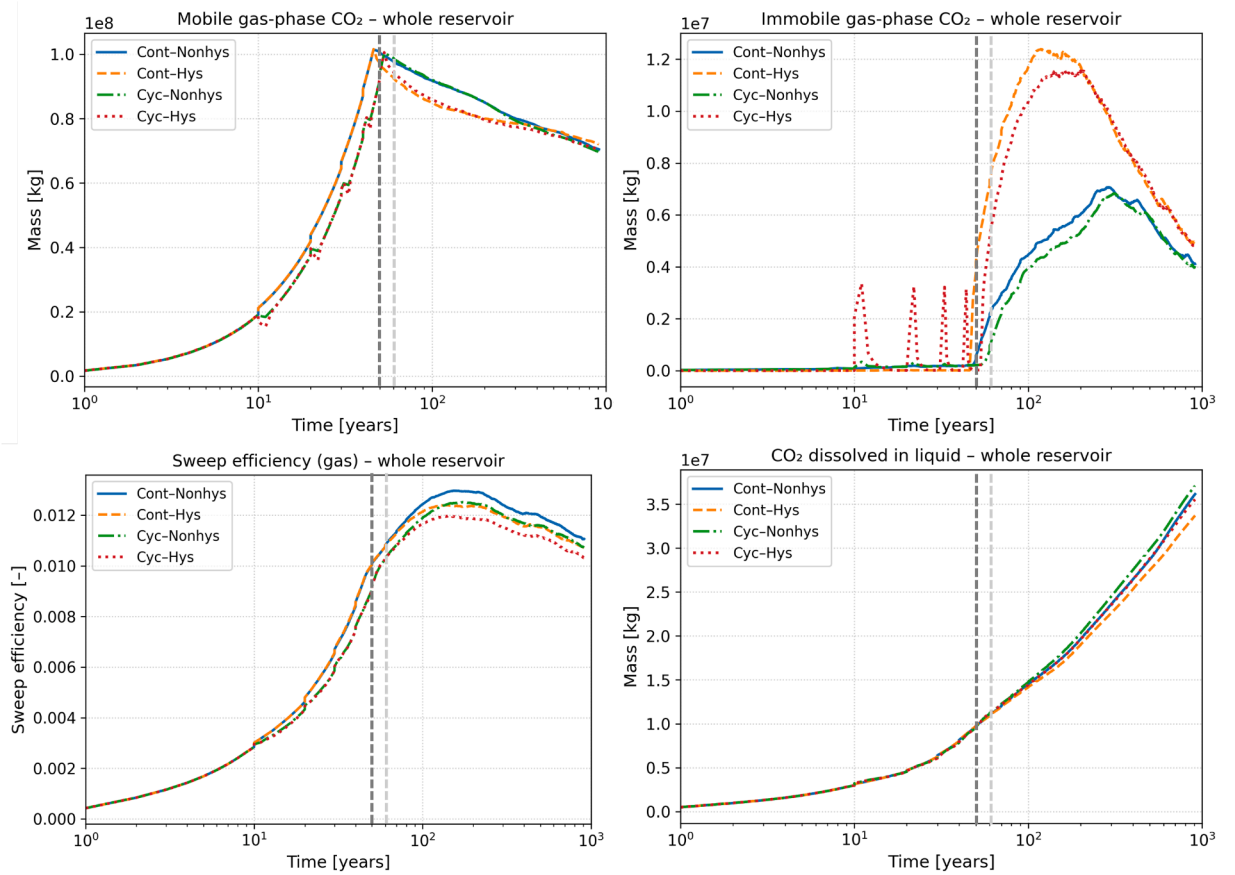


Fig. 16. Temporal dynamics of CO₂ trapping and sweep efficiency in the SPE 11b model. The vertical dashed line indicates the time at which injection stops.

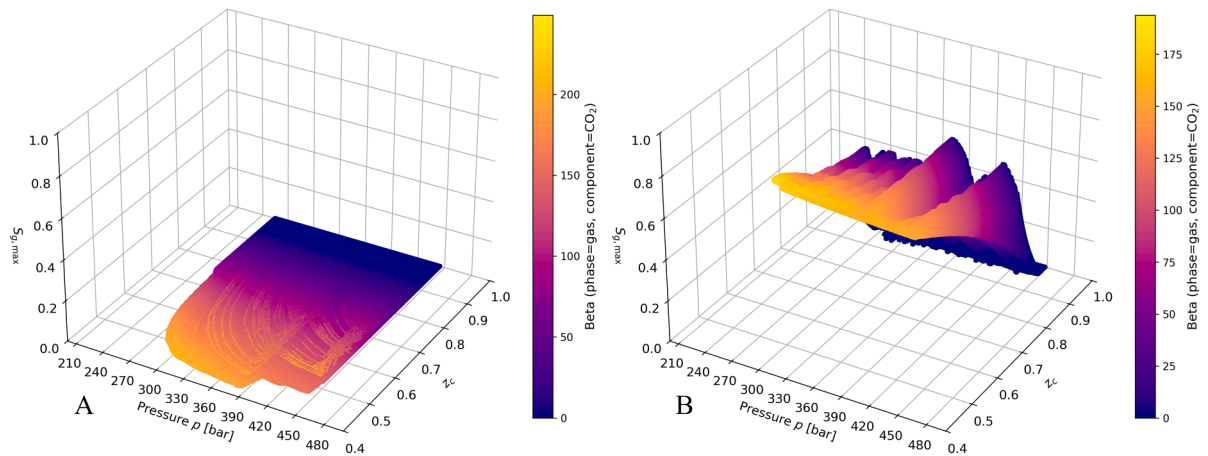


Fig. 17. Operator points generated in parameter space in (A) non-hysteretic and (B) hysteretic simulations for SPE 11b, colored by the operator value β_c .

Table 6
Numerical performance for the SPE11b example.

Model	Iters.	Generated points	CPU (s)
Hysteresis	63 430	339 730	8 995
Non-hysteresis	63 852	147 910	7 322

In this configuration, total trapping is only weakly sensitive to whether injection is continuous or cyclic. However, WAG tends to increase the immobile fraction during the water injection stage, while a following gas injection can partly undo that gain. Conversely, if WAG were scheduled as the final injection stage, the immobile CO_2 fraction at shut-in would increase. Sweep efficiency is reduced by cyclic injection and, in the post-injection period, is also lower for the hysteretic model than for the non-hysteretic model.

Finally, we compare the generation of operator points between hysteretic and non-hysteretic simulations for the SPE 11b model. As shown in Fig. 17 and Table 6, the hysteretic simulation generates substantially more operator points due to the additional history coordinate $S_{g,\max}$, compared with the non-hysteretic case. This increases the computational cost, although the relative overhead is noticeably smaller than that observed for one-dimensional models before. Notably, the hysteretic SPE11b case requires slightly fewer nonlinear iterations than the non-hysteretic case, while generating more operator points and requiring more CPU time. This indicates that the convergence impact of the fixed-history treatment is problem-dependent, and that the main additional cost in this case is associated with operator generation rather than a systematic increase in Newton iterations. Moreover, the range and distribution of operator points differ between the two simulations, reflecting the additional history dependence introduced by hysteresis.

5. Conclusion and discussion

We have proposed and validated an augmented operator-based linearization (OBL) framework that extends the conventional, reversible OBL formulation to incorporate modeling of history-dependent processes. The extension is achieved by embedding a history-dependent coordinate, represented in this study by the maximum gas saturation, into the operator parameter space. This enables operators to depend on both the instantaneous state and the flow history, while the nonlinear system retains the original dimension. Unlike thermal or coupled mechanical extensions, where the added coordinates are solved as part of the coupled nonlinear state, the history coordinate introduced here acts as a locally stored memory variable that augments operator evaluation without enlarging the Newton system. Within this framework, we have constructed a new hysteresis algorithm in which $S_{g,\max}$ is used as the history coordinate and updated locally after timestep convergence, while the flow problem is solved fully implicitly with respect to the primary variables. This update ensures consistent treatment of drainage–imbibition transitions. By coupling the hysteresis model with dissolution-driven updates of residual gas saturation, the approach also accounts for solubility trapping effects.

Validation against academic and commercial reservoir simulators confirms the consistency of the augmented OBL framework. Sensitivity analysis shows that the operator-space resolution critically controls both accuracy and computational cost. Numerical experiments in 1D and 2D reservoir models, including the SPE 11b benchmark, demonstrate the ability of the extended framework to capture the hysteresis phenomenon. Compared with non-hysteretic models, the new approach predicts enhanced residual trapping and distinct post-injection dissolution dynamics, at the cost of increased nonlinearity in the governing physics, which corresponds to additional operator points generated along the history coordinate.

The scope of this extension is limited to history-dependent constitutive responses that can be represented by a finite set of local internal variables. If an apparent history effect is already described by physical state variables included in the model, such as mineral amounts or reaction-progress variables in a reactive-transport formulation, it can be handled within the standard OBL for-

mulation without introducing a separate history coordinate. Processes suited to the augmented framework include, for example, adsorption/desorption hysteresis, and wettability-alteration hysteresis represented by cumulative exposure or irreversible alteration indices. Processes requiring complete path storage, nonlocal memory, spatially coupled history, or additional governing equations require additional internal variables or model-specific coupling strategies and cannot be captured by a single local history coordinate alone.

A remaining limitation is that additional history dimensions increase the number of generated operator points, memory use, and operator-generation cost. This cost becomes more important for high-dimensional history descriptions, such as models involving multiple hysteresis variables, history-dependent geomechanical closures, or reactive-transport closures with local memory variables. Feasible mitigation strategies include adaptive/on-demand operator generation, selective refinement of frequently visited or high-error regions, pruning or cache management of unused points, compression of tabulated operator values, and parallel operator evaluation. Ongoing high-dimensional OBL interpolator developments based on multi-index keys can further reduce practical limitations associated with legacy full-grid enumeration and user-defined axis bounds.

Overall, the proposed methodology extends the applicability of OBL to history-dependent physics in porous-media flow. This work establishes a consistent and effective foundation for incorporating hysteresis and related history-dependent processes into large-scale reservoir simulations of CO₂ storage, and provides a generalization path for subsurface processes whose history dependence can be represented by finite-dimensional local memory variables.

CRedit authorship contribution statement

Jianxin Lu: Writing – original draft, Visualization, Validation, Software, Methodology, Conceptualization; **Aleksei Novikov:** Writing – review & editing, Software, Methodology, Conceptualization; **Mengjie Zhao:** Writing – review & editing, Validation; **Lifei Yan:** Writing – review & editing, Validation; **Rouhi Farajzadeh:** Writing – review & editing, Supervision, Methodology; **Denis Voskov:** Writing – review & editing, Supervision, Methodology, Funding acquisition, Conceptualization.

Declaration of competing interest

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

Acknowledgment

JL acknowledges the [China Scholarship Council](#) for supporting his PhD work at TU Delft. AN thanks to TotalEnergies for funding through FC-MAELSTROM Project.

Data and code availability

The implementation of this work is made available to the public via the open-source Delft Advanced Research Terra Simulator ([open-DARTS](#)).

Data availability

No data was used for the research described in the article.

Appendix A. Killough hysteresis model

The hysteresis formulation used in this work follows the Killough model [15], which generates scanning curves for capillary pressure and relative permeability based on saturation history.

A.1. Capillary-pressure hysteresis

During decreasing wetting-phase saturation (drainage), capillary pressure follows the primary drainage curve $p_{c,d}(S_w)$; during increasing saturation (imbibition), it follows the primary imbibition curve $p_{c,i}(S_w)$. When a reversal occurs, a scanning curve is constructed. For a drainage-to-imbibition reversal at saturation S_w^{Hyst} , the scanning relation is

$$p_c(S_w) = p_{c,i}(S_w) + F(S_w) [p_{c,d}(S_w) - p_{c,i}(S_w)], \quad (21)$$

with interpolation function

$$F(S_w) = \frac{S_w^{\text{Hyst}} - S_w}{S_w^{\text{Hyst}} - S_w + \epsilon}, \quad \epsilon \in [0.05, 0.10], \quad (22)$$

where ϵ is a small regularization parameter, typically chosen in the range $[0.05, 0.10]$ [15]; S_w^{Hyst} is the saturation at which the reversal occurred; for a two-phase water–gas system with $S_w + S_g = 1$, we set

$$S_w^{\text{Hyst}} = 1 - S_{g,\max}. \quad (23)$$

A new reversal event defines a new anchor point and a new scanning curve, ensuring continuity of p_c across reversals.

A.2. Relative-permeability hysteresis

Relative-permeability hysteresis is modeled by accounting for non-wetting-phase trapping using Land's relation [14]. The trapped non-wetting saturation S_{Nr} is

$$S_{Nr} = \frac{S_N^{\text{Hyst}}}{1 + C_{\text{Land}} S_N^{\text{Hyst}}}, \quad (24)$$

where S_N^{Hyst} is the maximum historical non-wetting saturation and C_{Land} is an empirical constant.

For the non-wetting phase, the imbibition relative permeability between S_{Nr} and S_N is interpolated as

$$k_{rN}^{\text{imb}}(S_{Nr}) = k_{rN}^{\text{dr}}(S_N^{\text{Hyst}}) \left(\frac{S_N - S_{Nr}}{S_N^{\text{Hyst}} - S_{Nr}} \right)^A, \quad (25)$$

where k_{rN}^{dr} is the drainage relative permeability, A is a curvature (exponent) parameter, and S_N^{Hyst} is the saturation at reversal. In this work, with gas as the non-wetting phase, we use $S_N \equiv S_g$, $S_{Nr} \equiv S_{gr}$, and $S_N^{\text{Hyst}} \equiv S_{g,\max}$. Thus $k_{rN}^{\text{imb}} = 0$ at S_{gr} , and $k_{rN}^{\text{imb}} = k_{rN}^{\text{dr}}$ at $S_{g,\max}$.

A.3. Corey primary curves used in the Killough hysteresis model

Primary drainage curves for relative permeability are constructed using the Corey model [50,51]. These primaries seed the Killough scanning rules (Appendix A) to produce history-dependent imbibition/scanning curves.

Let S_w and S_g denote water and gas saturations, with connate (irreducible) endpoints S_{wc} and S_{gc} . Define the effective (normalized) saturations

$$S_w^* = \frac{S_w - S_{wc}}{1 - S_{wc} - S_{gc}}, \quad S_g^* = \frac{S_g - S_{gc}}{1 - S_{wc} - S_{gc}}, \quad S_w^*, S_g^* \in [0, 1]. \quad (26)$$

Endpoint relative permeabilities at $S_w^* = 1$ and $S_g^* = 1$ are k_{rw}^e and k_{rg}^e , respectively.

For primary *drainage*, the Corey forms are

$$k_{rw,d}(S_w) = k_{rw}^e (S_w^*)^{n_w}, \quad (27)$$

$$k_{rg,d}(S_g) = k_{rg}^e (S_g^*)^{n_g}, \quad (28)$$

where n_w and n_g are Corey exponents controlling curvature. Typical engineering ranges are $n_w \in [1, 5]$ and $n_g \in [1, 4]$, tuned to match rock/wettability and endpoints; the Brooks–Corey family further links these curves to capillary behavior [51].

Notes. (i) The Corey primaries $(k_{rw,d}, k_{rg,d})$ define the drainage envelopes used by the Killough scanning rules; (ii) imbibition/scanning curves for k_r are not Corey functions but are generated via Killough/Land; (iii) capillary-pressure primaries $p_{c,d}(S_w)$ and $p_{c,i}(S_w)$ are supplied from lookup tables; scanning follows Eq. (21).

A.4. Hysteresis setting in this study

In our simulations:

- For gas relative permeability (k_{rg}), the primary drainage curve $k_{rg,d}(S_g)$ was generated using a Corey model (or, where stated, from tabulated input); imbibition/scanning curves were computed using Eq. (25).
- For capillary pressure, primary drainage and imbibition curves were provided as tabulated input; scanning curves were generated using Eq. (21).

Thus, hysteresis in both p_c and k_r is consistently represented within the augmented OBL framework, with $S_{g,\max}$ serving as the history variable.

The augmented OBL framework is not restricted to the Killough hysteresis model used in this study. The role of the framework is to include the history variables required by a chosen hysteresis law into the operator parameter space, so that path-dependent saturation functions can be evaluated consistently during the simulation. In the present CO₂–brine examples, we assume strongly water-wet conditions and use the Killough/Land formulation because hysteresis is mainly associated with the non-wetting gas phase, and the maximum historical gas saturation $S_{g,\max}$ provides the natural memory variable for constructing the scanning curves and residual trapping relation. This choice also gives a clear first application in which the difference between hysteretic and non-hysteretic

Table A.1

Hysteresis/relperm parameter settings used in this paper. Section numbers refer to the main text.

Section	Relative permeability parameters							A	Tabulated input
	n_w	n_g	S_{gr}^{max}	k_{rw}^e	k_{rg}^e	S_{wc}	S_{gc}		
2.4	1.5	1.25	0.40	1.0	1.0	0.20	0.10	0.75	–
3.1	–	–	0.40	1.0	0.8	0.20	0.00	–	[11]
3.2	1.5	1.5	0.40	1.0	1.0	0.20	0.10	0.80	–
4.1	1.5	1.5	0.40	1.0	1.0	0.20	0.10	0.80	–
4.2	1.5	1.5	0.30	1.0	1.0	facies-dependent		0.80	–

behavior can be isolated. Other hysteresis models, such as Carlson- or Land-type formulations, can be treated in the same augmented-parameter-space framework by replacing the saturation-function evaluator and adding the history coordinates required by that model. For mixed-wet or oil-wet systems, or for three-phase water–oil–gas flow, additional memory variables may be needed to track reversal states or trapping histories for different phase pairs; these variables would be added as additional history dimensions in the operator space.

Parameter sources. Parameter choices for the Killough/Land hysteresis (e.g., S_{gr}^{max} , A, Corey exponents, endpoint saturations) follow [11,45,52,53]. Table A.1 lists the values used in this paper. For capillary pressure, the tabulated primaries are taken from [11] where indicated.

References

- [1] M. Bui, C.S. Adjiman, A. Bardow, E.J. Anthony, A. Boston, S. Brown, P.S. Fennell, S. Fuss, A. Galindo, L.A. Hackett, J.P. Hallett, H.J. Herzog, G. Jackson, J. Kemper, S. Krevor, G.C. Maitland, M. Matuszewski, I.S. Metcalfe, C. Petit, G. Puxty, J. Reimer, D.M. Reiner, E.S. Rubin, S.A. Scott, N. Shah, B. Smit, J.P.M. Trusler, P. Webley, J. Wilcox, N.M. Dowell, Carbon capture and storage (CCS): the way forward, *Energy Environ. Sci.* 11 (5) (2018) 1062–1176. Publisher: The Royal Society of Chemistry, <https://doi.org/10.1039/C7EE02342A>
- [2] P.S. Ringrose, A.-K. Furre, S.M.V. Gilfillan, S. Krevor, M. Landrø, R. Leslie, T. Meckel, B. Nazarian, A. Zahid, Storage of carbon dioxide in saline aquifers: physicochemical processes, key constraints, and scale-up potential, *Annu. Rev. Chem. Biomol. Eng.* 12 (Volume 12, 2021) (2021) 471–494. Publisher: Annual Reviews, <https://doi.org/10.1146/annurev-chembioeng-093020-091447>
- [3] T. Ajayi, J.S. Gomes, A. Bera, A review of CO2 storage in geological formations emphasizing modeling, monitoring and capacity estimation approaches, *Pet. Sci.* 16 (5) (2019) 1028–1063. <https://doi.org/10.1007/s12182-019-0340-8>
- [4] A. Izadpanahi, M.J. Blunt, N. Kumar, M. Ali, C.C. Gaeta Tassinari, M.A. Sampaio, A review of carbon storage in saline aquifers: mechanisms, prerequisites, and key considerations, *Fuel* 369 (2024) 131744. <https://doi.org/10.1016/j.fuel.2024.131744>
- [5] A. Bashir, M. Ali, S. Patil, M.S. Aljawad, M. Mahmoud, D. Al-Shehri, H. Hoteit, M.S. Kamal, Comprehensive review of CO2 geological storage: exploring principles, mechanisms, and prospects, *Earth Sci. Rev.* 249 (2024) 104672. <https://doi.org/10.1016/j.earscirev.2023.104672>
- [6] O. Massarweh, A.S. Abushaikh, CO2 Sequestration in subsurface geological formations: a review of trapping mechanisms and monitoring techniques, *Earth Sci. Rev.* 253 (2024) 104793. <https://doi.org/10.1016/j.earscirev.2024.104793>
- [7] S. Krevor, M.J. Blunt, S.M. Benson, C.H. Pentland, C. Reynolds, A. Al-Menhali, B. Niu, Capillary trapping for geologic carbon dioxide storage—from pore scale physics to field scale implications, *Int. J. Greenh. Gas Control* 40 (2015) 221–237. <https://doi.org/10.1016/j.ijggc.2015.04.006>
- [8] Y. Li, P.G. Ranjith, M.S.A. Perera, Q. Yu, Residual water formation during the CO2 storage process in deep saline aquifers and factors influencing it: a review, *J. CO2 Util.* 20 (2017) 253–262. <https://doi.org/10.1016/j.jcou.2017.05.022>
- [9] C. Ruprecht, R. Pini, R. Falta, S. Benson, L. Murdoch, Hysteretic trapping and relative permeability of CO2 in sandstone at reservoir conditions, *Int. J. Greenh. Gas Control* 27 (2014) 15–27. <https://doi.org/10.1016/j.ijggc.2014.05.003>
- [10] R. Pini, S.M. Benson, Capillary pressure heterogeneity and hysteresis for the supercritical CO2/water system in a sandstone, *Adv. Water Resour.* 108 (2017) 277–292. <https://doi.org/10.1016/j.advwatres.2017.08.011>
- [11] Y. Wang, C. Vuik, H. Hajibeygi, Analysis of hydrodynamic trapping interactions during full-cycle injection and migration of CO2 in deep saline aquifers, *Adv. Water Resour.* 159 (2022) 104073. <https://doi.org/10.1016/j.advwatres.2021.104073>
- [12] Y. Ghomian, G.A. Pope, K. Sepehrnoori, Hysteresis and field-scale optimization of WAG injection for coupled CO2-EOR and sequestration, *OnePetro*, 2008. <https://doi.org/10.2118/110639-MS>
- [13] R. Sedaghatinasab, S. Kord, J. Moghadasi, A. Soleymanzadeh, Relative permeability hysteresis and capillary trapping during CO2 EOR and sequestration, *Int. J. Greenh. Gas Control* 106 (2021) 103262. <https://doi.org/10.1016/j.ijggc.2021.103262>
- [14] C.S. Land, Calculation of imbibition relative permeability for two- and three-phase flow from rock properties, *Soc. Pet. Eng. J.* 8 (02) (1968) 149–156. <https://doi.org/10.2118/1942-PA>
- [15] J.E. Killough, Reservoir simulation with history-dependent saturation functions, *Soc. Pet. Eng. J.* 16 (01) (1976) 37–48. <https://doi.org/10.2118/5106-PA>
- [16] F.M. Carlson, Simulation of relative permeability hysteresis to the nonwetting phase, *OnePetro*, 1981. <https://doi.org/10.2118/10157-MS>
- [17] S. Foroudi, A. Gharavi, M. Fatemi, Assessment of two-phase relative permeability hysteresis models for oil/water, gas/water and gas/oil systems in mixed-wet porous media, *Fuel* 309 (2022) 122150. <https://doi.org/10.1016/j.fuel.2021.122150>
- [18] R. Juanes, E.J. Spiteri, F.M. Orr Jr., M.J. Blunt, Impact of relative permeability hysteresis on geological CO2 storage, *Water Resour. Res.* 42 (12) (2006). <https://doi.org/10.1029/2005WR004806>
- [19] Y. Wang, Z. Zhang, C. Vuik, H. Hajibeygi, Simulation of CO2 storage using a parameterization method for essential trapping physics: fluidflow benchmark study, *Transp. Porous Media* 151 (5) (2024) 1053–1070. <https://doi.org/10.1007/s12422-023-01987-5>
- [20] R. Khoramian, M. Pourafshary, Peyman Riaz, The role of capillary hysteresis in enhancing CO2 trapping efficiency and storage stability, *Kazakhstan J. Oil Gas Ind.* 7 (1) (2025) 90–99. <https://doi.org/10.54859/kjogi108781>
- [21] M.T. Elenius, D.V. Voskov, H.A. Tchelepi, Interactions between gravity currents and convective dissolution, *Adv. Water Resour.* 83 (2015) 77–88. <https://doi.org/10.1016/j.advwatres.2015.05.006>
- [22] H. Ni, B. Li, N. Darraj, B. Ren, C. Harris, P.G. Krishnamurthy, I. Bukar, S. Berg, J. Snippe, P. Ringrose, T.A. Meckel, S. Krevor, S. Benson, The impact of capillary heterogeneity on CO2 flow and trapping across scales, *Earth Sci. Rev.* 270 (2025) 105257. <https://doi.org/10.1016/j.earscirev.2025.105257>
- [23] M. Zhao, M. Gerritsma, M. Al Kobaisi, H. Hajibeygi, Algebraic dynamic multilevel (ADM) method for CO2 storage in heterogeneous saline aquifers, *J. Comput. Phys.* 539 (2025) 114202. <https://doi.org/10.1016/j.jcp.2025.114202>

- [24] D.V. Voskov, H.A. Tchelepi, Comparison of nonlinear formulations for two-phase multi-component EoS based simulation, *J. Pet. Sci. Eng.* 82–83 (2012) 101–111. TLDR: The behavior of the Newton-based nonlinear solver as a function of timestep size for different variable sets and for different nonlinear updating strategies is analyzed., <https://doi.org/10.1016/j.petrol.2011.10.012>
- [25] D.V. Voskov, H.A. Tchelepi, Compositional space parameterization: theory and application for immiscible displacements, *SPE J.* 14 (03) (2009) 431–440. <https://doi.org/10.2118/106029-PA>
- [26] R. Zaydullin, D.V.V. Voskov, H.A.A. Tchelepi, Nonlinear formulation based on an equation-of-state free method for compositional flow simulation, *SPE J.* 18 (02) (2013) 264–273. <https://doi.org/10.2118/146989-PA>
- [27] R. Zaydullin, D.V. Voskov, H.A. Tchelepi, Formulation and solution of compositional displacements in tie-simplex space, in: *SPE Reservoir Simulation Symposium*, SPE, The Woodlands, Texas, USA, 2013, pp. SPE–163668–MS. <https://doi.org/10.2118/163668-MS>
- [28] D.V. Voskov, Operator-based linearization approach for modeling of multiphase multi-component flow in porous media, *J. Comput. Phys.* 337 (2017) 275–288. TLDR: A new approach for the linearization of governing equations that describe flow and transport in porous media is proposed, based on an approximate representation of the exact physics of the problem, which is similar to an approximate representations of space and time discretization performed in conventional simulation., <https://doi.org/10.1016/j.jcp.2017.02.041>
- [29] M. Khait, D. Voskov, Operator-based Linearization for Non-isothermal Multiphase Compositional Flow in Porous Media, 2016. <https://doi.org/10.3997/2214-4609.201601800>
- [30] M. Khait, D. Voskov, Adaptive parameterization for solving of thermal/compositional nonlinear flow and transport with buoyancy, *SPE J.* 23 (02) (2018) 522–534. <https://doi.org/10.2118/182685-PA>
- [31] M. Khait, D. Voskov, Operator-based linearization for efficient modeling of geothermal processes, *Geothermics* 74 (2018) 7–18. <https://doi.org/10.1016/j.geothermics.2018.01.012>
- [32] H.M. Nilsen, K.-A. Lie, O. Andersen, Fully-implicit simulation of vertical-equilibrium models with hysteresis and capillary fringe, *Comput. Geosci.* 20 (1) (2016) 49–67. <https://doi.org/10.1007/s10596-015-9547-y>
- [33] D. Voskov, I. Saifullin, A. Novikov, M. Wapperom, L. Orozco, G. Seabra, Y. Chen, M. Khait, L. Xiacong, X. Tian, S. de Hoop, A. Palha, Open delft advanced research terra simulator (open-DARTS), *J. Open Source Softw.* 9 (2024) 6737. <https://doi.org/10.21105/joss.06737>
- [34] X. Lyu, Simulation of foam in Enhanced Oil Recovery (EOR) and Carbon Capture and Storage (CCS) applications, Ph.D. thesis, Delft University of Technology, 2021. <https://doi.org/10.4233/UUID:127B6A6F-C947-4624-A72C-11F6539C41D7>
- [35] Y. Wang, D. Voskov, M. Khait, S. Saeid, D. Bruhn, Influential factors on the development of a low-enthalpy geothermal reservoir: a sensitivity study of a realistic field, *Renew. Energy* 179 (2021) 641–651. <https://doi.org/10.1016/j.renene.2021.07.017>
- [36] Y. Chen, D. Voskov, A. Daniilidis, Rigorous numerical methodology and heat recovery analysis for modeling of direct use geothermal systems, *Geoenergy Sci. Eng.* 247 (2025) 213661. <https://doi.org/10.1016/j.geoen.2025.213661>
- [37] X. Lyu, M. Khait, D. Voskov, Operator-based linearization approach for modeling of multiphase flow with buoyancy and capillarity, *SPE J.* 26 (04) (2021) 1858–1875. <https://doi.org/10.2118/205378-PA>
- [38] M. Wapperom, X. Lyu, D. Voskov, Accurate modeling of near-wellbore effects induced by supercritical CO₂ injection, in: *ECMOR 2022, European Association of Geoscientists & Engineers, The Hague, Netherlands / Online*, 2022, pp. 1–13. <https://doi.org/10.3997/2214-4609.202244092>
- [39] M. Wapperom, X. Tian, A. Novikov, D. Voskov, FluidFlow benchmark: lessons learned from the perspective of subsurface simulation, *Transp. Porous Media* 151 (5) (2024) 1033–1052. <https://doi.org/10.1007/s11242-023-01984-8>
- [40] A. Novikov, D. Voskov, M. Khait, H. Hajibeygi, J.D. Jansen, A scalable collocated finite volume scheme for simulation of induced fault slip, *J. Comput. Phys.* 469 (2022) 111598. <https://doi.org/10.1016/j.jcp.2022.111598>
- [41] A. Novikov, I. Saifullin, H. Hajibeygi, D. Voskov, A finite volume framework for the fully implicit thermal-hydro-mechanical-compositional modeling in subsurface applications, *J. Comput. Phys.* 538 (2025) 114152. <https://doi.org/10.1016/j.jcp.2025.114152>
- [42] A. Novikov, D.V. Voskov, Interpolation strategies for complex thermal-multiphase-reactive modeling with operator-based linearization, *OnePetro*, 2025. <https://doi.org/10.2118/223919-MS>
- [43] C. Doughty, Modeling geologic storage of carbon dioxide: comparison of non-hysteretic and hysteretic characteristic curves, *Energy Convers. Manag.* 48 (6) (2007) 1768–1781. <https://doi.org/10.1016/j.enconman.2007.01.022>
- [44] C. Doughty, User's Guide for Hysteretic Capillary Pressure and Relative Permeability Functions in TOUGH2, *Tech. Rep. LBNL-6533E, Lawrence Berkeley National Laboratory, Berkeley, California*, 2013. https://tough.lbl.gov/assets/docs/Hysteresis_Users_Guide.pdf
- [45] E.A. Al-Khdheawi, S. Vialle, A. Barifcani, M. Sarmadivaleh, S. Iglaue, Effect of wettability heterogeneity and reservoir temperature on CO₂ storage efficiency in deep saline aquifers, *Int. J. Greenh. Gas Control* 68 (2018) 216–229. <https://doi.org/10.1016/j.ijggc.2017.11.016>
- [46] M. Zhao, Y. Wang, M. Gerritsma, H. Hajibeygi, Efficient simulation of CO₂ migration dynamics in deep saline aquifers using a multi-task deep learning technique with consistency, *Adv. Water Resour.* 178 (2023) 104494. <https://doi.org/10.1016/j.advwatres.2023.104494>
- [47] G. Hadjisotiriou, J. Sass, M. Wapperom, A. Novikov, D.V. Voskov, SPE11: Convergence study and extension to realistic physics, *OnePetro*, 2025. <https://doi.org/10.2118/223922-MS>
- [48] J.M. Nordbotten, M.A. Ferno, B. Flemisch, A.R. Kovscek, K.-A. Lie, The 11th society of petroleum engineers comparative solution project: problem definition, *SPE J.* 29 (05) (2024) 2507–2524. <https://doi.org/10.2118/218015-PA>
- [49] J.M. Nordbotten, M.A. Ferno, B. Flemisch, A.R. Kovscek, K.-A. Lie, J.W. Both, O. Møyner, T.H. Sandve, E. Ahusborde, S. Bauer, Z. Chen, H. Class, C. Di, D. Ding, D. Element, E. Flauraud, J. Franc, F. Gasanzade, Y. Ghomian, M.A. Giddins, C. Green, B.R.B. Fernandes, G. Hadjisotiriou, G. Hammond, H. Huang, D. Kachuma, M. Kern, T. Koch, P. Krishnamurthy, K.O. Lye, D. Landa-Marbán, M. Nole, P. Orsini, N. Ruby, P. Salinas, M. Sayyafzadeh, J. Torben, A. Turner, D.V. Voskov, K. Wendel, A.A. Youssef, Benchmarking CO₂ storage simulations: results from the 11th society of petroleum engineers comparative solution project, *Int. J. Greenh. Gas Control* 148 (2025). <https://doi.org/10.1016/j.ijggc.2025.104519>
- [50] A.T. Corey, C.H. Rathjens, Effect of stratification on relative permeability, *J. Pet. Technol.* 8 (12) (1956) 69–71. <https://doi.org/10.2118/744-G>
- [51] R.H. Brooks, A.T. Corey, Hydraulic properties of porous media and their relation to drainage design, *Trans. ASAE* 7 (1) (1964) 26–28. <https://doi.org/10.13031/2013.40684>
- [52] E.A. Al-Khdheawi, S. Vialle, A. Barifcani, M. Sarmadivaleh, S. Iglaue, Influence of injection well configuration and rock wettability on CO₂ plume behaviour and CO₂ trapping capacity in heterogeneous reservoirs, *J. Nat. Gas Sci. Eng.* 43 (2017) 190–206. <https://doi.org/10.1016/j.jngse.2017.03.016>
- [53] K. Rasmusson, M. Rasmusson, Y. Tsang, S. Benson, F. Hingerl, F. Fagerlund, A. Niemi, Residual trapping of carbon dioxide during geological storage—insight gained through a pore-network modeling approach, *Int. J. Greenh. Gas Control* 74 (2018) 62–78. <https://doi.org/10.1016/j.ijggc.2018.04.021>