

## Impact of capillary pressure hysteresis and injection-withdrawal schemes on performance of underground hydrogen storage

Nazari, Farzaneh; Aghabozorgi Nafchi, Shokoufeh; Vahabzadeh Asbaghi, Ehsan; Farajzadeh, Rouhi; Niasar, Vahid J.

**DOI**

[10.1016/j.ijhydene.2023.09.136](https://doi.org/10.1016/j.ijhydene.2023.09.136)

**Publication date**

2023

**Document Version**

Final published version

**Published in**

International Journal of Hydrogen Energy

**Citation (APA)**

Nazari, F., Aghabozorgi Nafchi, S., Vahabzadeh Asbaghi, E., Farajzadeh, R., & Niasar, V. J. (2023). Impact of capillary pressure hysteresis and injection-withdrawal schemes on performance of underground hydrogen storage. *International Journal of Hydrogen Energy*, 50, 1263-1280. <https://doi.org/10.1016/j.ijhydene.2023.09.136>

**Important note**

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

**Copyright**

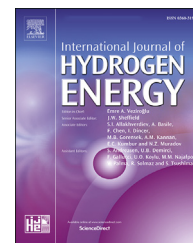
Other than for strictly personal use, it is not permitted to download, forward or distribute the text or part of it, without the consent of the author(s) and/or copyright holder(s), unless the work is under an open content license such as Creative Commons.

**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.

Available online at [www.sciencedirect.com](http://www.sciencedirect.com)

ScienceDirect

journal homepage: [www.elsevier.com/locate/he](http://www.elsevier.com/locate/he)

# Impact of capillary pressure hysteresis and injection-withdrawal schemes on performance of underground hydrogen storage

Farzaneh Nazari <sup>a</sup>, Shokoufeh Aghabozorgi Nafchi <sup>b</sup>,  
Ehsan Vahabzadeh Asbaghi <sup>a</sup>, Rouhi Farajzadeh <sup>c,d</sup>, Vahid J. Niasar <sup>a,\*</sup>

<sup>a</sup> Department of Chemical Engineering, The University of Manchester, Oxford Road, Manchester, M13 9PL, UK

<sup>b</sup> Wood Group PLC, Aberdeen, Scotland, United Kingdom

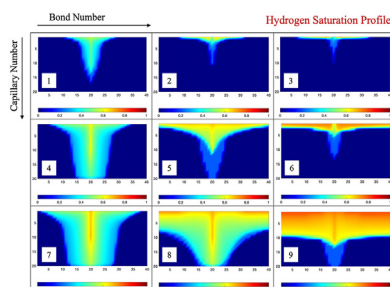
<sup>c</sup> Faculty of Civil Engineering and Geosciences, Delft University of Technology, P.O. Box 5048, Delft, GA 2600, the Netherlands

<sup>d</sup> Shell Global Solutions International B.V., The Hague, the Netherlands

## HIGHLIGHTS

- Buoyancy plays a significant role on UHS performance.
- A dimensionless number analysis to study the influence of injection/withdrawal schemes on UHS performance.
- Capillary pressure scanning curves did not significantly affect the hydrogen saturation distribution.
- Water upconing limits the UHS performance at high flow rates.
- Interplay between Bond and capillary numbers should be considered for UHS.

## GRAPHICAL ABSTRACT



## ARTICLE INFO

### Article history:

Received 7 June 2023

Received in revised form

11 July 2023

Accepted 12 September 2023

Available online 04 October 2023

### Keywords:

Capillary Pressure Hysteresis

Underground hydrogen Storage

## ABSTRACT

**Hypothesis:** Underground hydrogen storage in depleted hydrocarbon reservoirs and aquifers has been proposed as a potential long-term solution to storing intermittently produced renewable electricity, as the subsurface formations provide secure and large storage space. Various phenomena can lead to hydrogen loss in subsurface systems with the key cause being the trapping especially during the withdrawal cycle. Capillary trapping, in particular, is strongly related to the hysteresis phenomena observed in the capillary pressure/saturation and relative-permeability/saturation curves. This paper address two key points: (1) the sole impact of hysteresis in capillary pressure on hydrogen trapping during withdrawal cycles and (2) the dependency of optimal operational parameters (injection/withdrawal

\* Corresponding author.

E-mail address: [vahid.niasar@manchester.ac.uk](mailto:vahid.niasar@manchester.ac.uk) (V.J. Niasar).

<https://doi.org/10.1016/j.ijhydene.2023.09.136>

0360-3199/© 2023 The Author(s). Published by Elsevier Ltd on behalf of Hydrogen Energy Publications LLC. This is an open access article under the CC BY license (<http://creativecommons.org/licenses/by/4.0/>).

Scanning Curves  
Numerical Simulation

flow rate) and the reservoir characteristics, such as permeability, thickness and wettability of the porous medium, on the remaining hydrogen saturation.

**Model:** To study the capillary hysteresis during underground hydrogen storage, Killough [1] model was implemented in the MRST toolbox [2]. A comparative study was performed to quantify the impact of changes in capillary pressure behaviour by including and excluding the hysteresis and scanning curves. Additionally, this study investigates the impact of injection/withdrawal rates and the aquifer permeability for various capillary and Bond numbers in a homogeneous system.

**Findings:** It was found that although the hydrogen storage efficiency is not considerably impacted by the inclusion of the capillary-pressure scanning curves, the impact of capillary pressure on the well properties (withdrawal rate and pressure) can become significant. Higher injection and withdrawal rates does not necessarily lead to a better performance in terms of productivity. The productivity enhancement depends on the competition between gravitational, capillary and viscous forces. The observed water upconing at relatively high capillary numbers resulted in low hydrogen productivity, highlighting the importance of well design and placement.

© 2023 The Author(s). Published by Elsevier Ltd on behalf of Hydrogen Energy Publications LLC. This is an open access article under the CC BY license (<http://creativecommons.org/licenses/by/4.0/>).

## 1. Introduction

### 1.1. Underground hydrogen storage

The low-carbon energy industry requires a long-term solution to overcome the intermittent nature of the renewable energy sources and to provide year-round electricity. The cyclic intermittent nature of these resources leads to energy surplus during high energy production season and vice versa [3]. Moreover, heavy industries, such as steel mining, require high-temperature heat that cannot be easily supplied by electricity. The energy produced during a productive season can be stored for use during the shortage times. Additionally, this excess produced energy needs a durable and efficient storage medium able to hold it for different durations of time (days, months, or even years). Due to its desirable characteristics,  $H_2$  is a favorable medium to/from which the excess low-carbon electricity can be converted [4,5] through electrolyzers and fuel cells a [6,7], for instance Refs. [8–11], while it can accommodate the high-temperature requirements that heavy industries deal with. The successful implementation of hydrogen economy [5,7] requires proper storage spaces to hold the hydrogen itself. The produced  $H_2$  can be stored in surface or subsurface structures. Hydrogen storage in high-pressure gas cylinders, and cryogenic tanks [12–14] can be some example cases of surface hydrogen storage. However, surface energy storage may not be a long-term solution as surface structures only provide limited storage capacities. Underground Hydrogen Storage (UHS), on the other hand, can become a viable solution enabling industries to have access to continuous energy supply as they provide safe storage spaces compared to the limited spaces that the surface structures provide. These subsurface structures may include aquifers [15–17], depleted oil and gas reservoirs [18], and salt caverns [19,20]. Subsurface hydrogen storage, although an intriguing option, comes with complexities that need to be assessed

prior to the storage process.  $H_2$  storage in salt caverns, for instance, is restricted by the limited volume that these structures offer. Depleted oil and gas reservoirs also contain significant amounts of trapped/residual hydrocarbons that can react with  $H_2$  and also reveal relatively high levels of microbial activity that can lead to  $H_2$  loss [21]. Aquifers, on the other hand, maybe a suitable alternative as they can provide vast storage spaces, and demonstrate less chance of microbial/chemical reactions [16,22].

### 1.2. Hysteresis and trapping

There are various hydromechanical processes that may cause hydrogen loss in UHS. Examples include loss of purity of hydrogen due to mixing with the cushion gas [23,24] or other in-situ gases [25], lateral spreading of hydrogen due to strong buoyancy [26,27], and ultimately trapping of hydrogen due to structural trapping, dissolution trapping, mineral trapping, and residual trapping, similar to  $CO_2$  storage in aquifers [28]. It should also be noted that in  $CO_2$  storage, the main objective is maximising the trapping efficiency, whereas in UHS, the aim is to minimise the trapping to decrease hydrogen loss, and therefore, a formation suitable for  $CO_2$  storage might be unfit for UHS.

The residual trapping, in particular, is initiated by the wettability preference of the subsurface rocks. It is closely linked with the hysteresis phenomena, which shows the dependence of flow on its history. This path dependence can be studied in terms of contact angle, capillary pressure-saturation ( $P_c-S_w$ ) and relative permeability-saturation ( $k_r-S_w$ ) functions. Due to capillary trapping and hysteresis, the  $P_c-S_w$  and  $k_r-S_w$  functions will not be unique and will differ for drainage and imbibition processes [29–31]. This behaviour can be enhanced during the UHS because of its cyclic nature and also the strong water wetting conditions [32].

A complete hysteresis analysis can be done through scanning curves to consider the full hysteresis cycles of  $P_c-S_w$

and  $k_r$ - $S_w$ . Depending on the initial condition and two-phase flow process,  $P_c$ - $S_w$  and  $k_r$ - $S_w$  will follow one of the primary drainage, main drainage, primary imbibition, or imbibition curves (these are called bounding curves) until they reach a turning point due to the reversal in flow path. From that point, a new scanning curve would be formed, which is bounded by the main drainage-imbibition curves. The formation of the scanning curves from a previous one continues as long as the reversals occur. Different models have been suggested to generate the scanning curves from the bounding drainage/imbibition curves and characterize trapping. Avid reader is referred to literature for a detailed study of such models [1,32–41].

The  $\text{CO}_2$  capillary trapping has proven to happen significantly in underground storage sites, which is a favorable process in CCS as opposed to hydrogen capillary trapping in UHS [42,43]. Akbarabadi and Piri [42] observed high capillary-trapping efficiency, particularly for gaseous  $\text{CO}_2$  [42] in their experiments. Juanes et al. [31] employed the Killough [1] and Land [35] models to investigate the impact of relative-permeability hysteresis and the non-wetting phase trapping in the context of  $\text{CO}_2$  sequestration. However, they ignored the effect of capillary-pressure hysteresis on the flow properties [31,44]. Ide et al. [45] investigated the effect of dimensionless gravity number, aquifer inclination, and capillarity on the amount of trapped  $\text{CO}_2$ , and concluded that as the gravity number increases, the amount of the trapped gas decreases, although the aquifer inclination showed the opposite trend [45]. Gershenzon et al. [46] used Killough [1]  $k_r$ - $S_w$  model to study the  $\text{CO}_2$  trapping in terms of snap-off and capillary pinning using ECLIPSE-300 [46–49], however, they ignored the capillary-pressure hysteresis.

To investigate the impact of aquifer heterogeneity on the capillary trapping of  $\text{CO}_2$ , Yang et al. [50] employed an invasion percolation model combined with Land [35] and Spiteri et al. [37] trapping models. Wang et al. [30] investigated the solubility and residual trapping of  $\text{CO}_2$  during full-cycle injections, employing the Killough [1]  $P_c$ - $S_w$  and  $k_r$ - $S_w$  model to generate the first scanning curves; however, they did not consider all possible cycles and scanning curves due to hysteresis in the system. To examine the importance of the gas/liquid relative-permeability hysteresis on the UHS, Bo et al. [17] employed the experimentally-measured data of Boon and Hajibeygi [51] along with the GEM simulator [52]. Their results indicate that hysteresis effect in gas and liquid flows gain importance at differing injection/withdrawal rates. Moreover, they emphasized the importance of considering the measured  $k_r$ - $S_w$  hysteresis data for the  $\text{H}_2$ -water fluids, as it highly influences the  $\text{H}_2$  productivity estimation. In another study, Pan et al. [16] examined the impact of relative-permeability hysteresis, wettability, and injection/withdrawal scenarios during UHS, and compared the performance of  $\text{CO}_2$ -brine systems with  $\text{H}_2$ -brine systems. They showed that the inclusion of hysteresis in  $\text{H}_2$ -brine relative permeability lowers the  $\text{H}_2$  productivity in terms of withdrawal efficiency [16]. Lysy et al. [53] used the experimentally-measured  $\text{H}_2$ -brine  $k_r$  values [54] to investigate the impact of hysteresis in  $k_r$  data on  $\text{H}_2$  storage. The findings demonstrated that the ultimate recovery factor was overestimated by nonhysteretic  $k_r$  values.

### 1.3. The present study

There is limited information in the literature regarding the sole influence of capillary-pressure hysteresis on UHS performance as most studies only include the relative-permeability hysteresis. Clearly, in Darcy's law, relative permeability and its hysteresis directly control the two-phase flow dynamics and trapped saturation. As hysteresis calculations are computationally expensive and time consuming, here, we seek to explore the hysteresis in capillary pressure to determine if it presents a substantial impact on the flow characteristics and efficiency and to see whether we can exclude its effect to increase the calculation speed. Therefore, we will investigate the impact of hysteresis in  $P_c$ - $S_w$  curves and trapping during cyclic  $\text{H}_2$  injection and withdrawal by taking into account the full scanning behaviour in each individual grid block of the model during drainage and imbibition processes, while ignoring the hysteresis in relative permeability. Moreover, we investigate the impact of flow rates, on the UHS efficiency. In summary, this study provides the following key contributions by applying the Killough [1] model for hysteresis in capillary pressure and MRST toolbox [2,55].

- Investigating the impact of  $P_c$ - $S_w$  curves on  $\text{H}_2$  loss due to trapping and saturation distribution in aquifers.
- Performing a comparative study to quantify the impact of including and ignoring  $P_c$ - $S_w$  curves and their hysteresis on the amount of trapped  $\text{H}_2$  and saturation distribution after injection/withdrawal cycles,
- Probing the impact of dimensionless numbers, namely capillary, Bond, and buoyancy numbers (as the ratios of viscous, capillary and gravitational forces) on  $\text{H}_2$  injection/withdrawal cycles.
- Discussing the relevance of water upconing on  $\text{H}_2$  production efficiency.

The remainder of this paper first presents a summary of the methodology and tools used for generating the bounding/scanning  $P_c$ - $S_w$  curves and modeling the  $\text{H}_2$  injection/withdrawal cycles. Next, we will present the results regarding the impact of intermediate  $P_c$ - $S_w$  scanning curves as well as the influence of the viscous/buoyancy forces on the  $\text{H}_2$  storage efficiency. Finally, a discussion on the numerical instability for hysteresis modeling and the complexities will be provided.

## 2. Methodology

### 2.1. The Main Capillary Pressure - Saturation Curves

It has been shown in the literature that  $\text{H}_2$  and  $\text{N}_2$  exhibit similar (flow) characteristics in porous media, for example in terms of three-phase contact angle [56], because the three-phase systems containing these gases (gas-brine-rock) present similar van der Waals and electrostatic interactions. Therefore, the  $\text{N}_2$ -water  $P_c$ - $S_w$  data from Plug and Bruining [57] is used as the initial input for calculating  $\text{H}_2$ -water  $P_c$ - $S_w$  curve. The data is scaled using the Leverett J-function

$J(S_w) = \frac{P_c(S_w)\sqrt{K/q}}{\sigma \cos \theta}$  [58] to account for the impact of permeability.

Afterwards, the Brooks-Corey [59] formulation (equation (1)) is used to fit the scaled  $P_c$ - $S_w$  curves:

$$P_c(S_w) = P_e \times S_w^{*n} \quad (1)$$

where  $P_e$  is the entry capillary pressure,  $S_w^*$  is the scaled wetting phase saturation ( $S_w^* = \frac{S_w - S_{rw}}{1 - S_{rw} - S_{rnw}}$ ),  $S_{rw}$  and  $S_{rnw}$  are the residual saturations of the wetting (water) and non-wetting (hydrogen) phases, respectively. The relative-permeability values are also calculated using the Brooks-Corey equations [59], where  $k_{ra}^{max}$  denotes the endpoint relative permeability for each phase:

$$k_{ra}(S_a) = k_{ra}^{max} \times S_a^{*m} \quad (2)$$

## 2.2. The Scanning Capillary Pressure - Saturation Curves

The  $P_c$  along the scanning curve is computed using Equation (1), considering a secondary imbibition process starting from the main drainage curve (Equation (3)). In this equation,  $P_c^{Dr}$  and  $P_c^{Im}$  show the main drainage and imbibition curves,  $S_w^{Hyst}$  is the turning point wetting-phase saturation,  $S_w^{Max}$  denotes the maximum attainable wetting-phase saturation, and  $\epsilon$  is a curvature parameter [1] (See Fig. 1b for an example of scanning curves generated by this model).

$$P_c^{Sc}(S_w) = P_c^{Dr}(S_w) - F \times [P_c^{Dr}(S_w) - P_c^{Im}(S_w)]$$

$$F = \frac{\frac{1}{S_w - S_w^{Hyst} + \epsilon} - \frac{1}{\epsilon}}{\frac{1}{S_w^{Max} - S_w^{Hyst} + \epsilon} - \frac{1}{\epsilon}} \quad (3)$$

The secondary scanning  $P_c$ - $S_w$  curves starting from a prior scanning curve (other than the main curves) can also be calculated using the methodology outlined in Killough [1] and Guo [60]. The generation of the  $n$ -th scanning curves requires the turning point history of the grid blocks as well as their  $P_c$ - $S_w$  functions. For instance, to generate the  $n$ -th drainage scanning curve (initiated from an imbibition scanning curve), one can follow Equations (4)–(6) as outlined by Guo [60]. In these equations,  $S_{w1}$  and  $S_{w2}$  are imbibition-to-drainage and previous drainage-to-imbibition turn-around saturations, respectively, and  $P_c^{Sc}$  is the capillary pressure on the scanning curve. Similar formulations are also applied for the scanning imbibition-to-drainage processes and can be found in Guo [60] and Killough [1].

$$P_c^{Sc} = P_c^{Im}(S_w) + \left[ \frac{\frac{1}{\Delta S_w^{Hyst} + \epsilon} - \frac{1}{\epsilon}}{\frac{1}{\Delta S_w + \epsilon} - \frac{1}{\epsilon}} \right] [P_c^{Dr}(S_w) - P_c^{Im}(S_w)] + (\delta P_1 - \delta P_2) \quad (4)$$

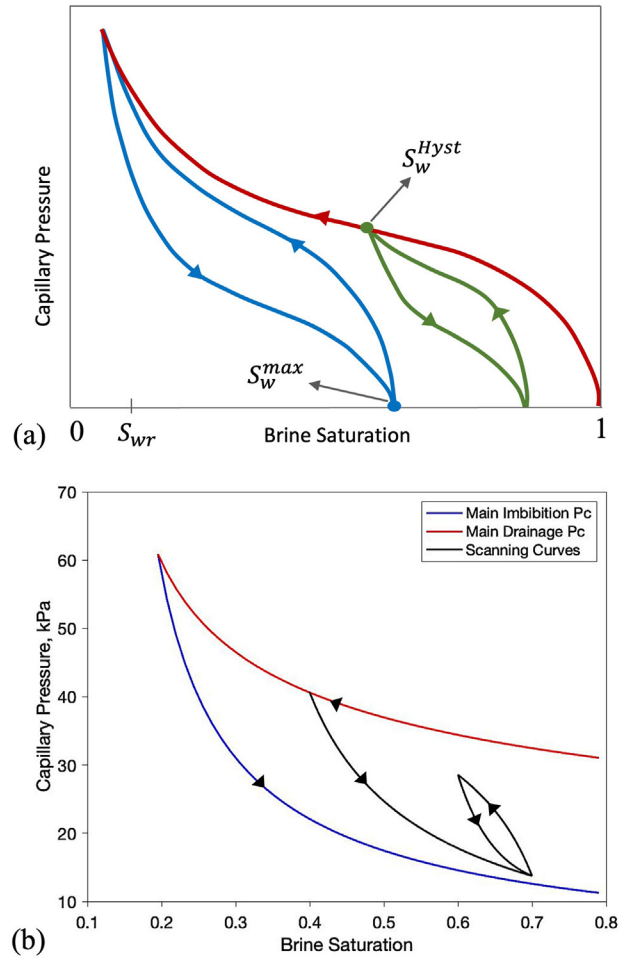
where:

$$\delta P_1 = [P_c^{Sc}(S_{w1}) - P_c^{Im}(S_{w1})] \frac{\Delta S_w^{Hyst}}{\Delta S_w} \quad (5)$$

$$\delta P_2 = [P_c^{Dr}(S_{w2}) - P_c^{Sc}(S_{w2})] \frac{\Delta S_w^{Hyst}}{\Delta S_w}$$

and:

$$\Delta S_w^{Hyst} = S_{w1} - S_w \quad , \quad \Delta S_w^{Hyst} = S_w - S_{w2} \quad \text{and} \quad \Delta S_w = S_{w1} - S_{w2} \quad (6)$$



**Fig. 1 – (a) Schematic of the capillary-pressure hysteresis and the scanning curves. The red, blue, and green curves illustrate the primary drainage, the main hysteresis loop, and a scanning loop generated at a different turning point saturation (reproduced from Joekar-Niasar et al. [29]).  $S_w^{Hyst}$  denotes the turning-point saturation of the non-wetting phase and  $S_w^{max}$  is the maximum attainable wetting phase saturation. (b) An example of the scanning  $P_c$ - $S_w$  curves generated using the Killough [1] model based on the main Brooks-Corey capillary-pressure model. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)**

Note that the application of any generated scanning curve from another scanning curve is only valid in the saturation range of the two consecutive turning points ( $S_{w2} < S_w < S_{w1}$ ). For saturations outside of this range, the scanning curve continues on the capillary-pressure curve generated at time-step  $t^{i-2}$ , where  $i$  is an index indicating the current timestep number. A diagram of the  $P_c$  scanning behaviour is shown in Fig. 1a, where depending on the saturation at the turning point, various scanning curves can be produced [29]. Fig. 1b also represents a real scanning curve analysis generated based on the Killough [1] model. It can be seen that depending on the saturation of the turning point, the scanning curves may/may not reach the main  $P_c$ - $S_w$  curves.



In a multi-phase system, capillary forces are only one factor influencing flow efficiency; other density/viscosity-driven forces also impose major impacts on flow behaviour and productivity. Viscous and buoyancy forces, in particular, gain tremendous importance as the  $H_2$ -brine system present large viscosity and density differences. Thus, it is vital to quantify impact of these forces, which will be done through a dimensionless number analysis by considering three numbers, namely capillary number ( $Ca$ ) [58], Bond Number ( $Bo$ ), and Buoyancy Number ( $Bu$ ) [61,62].  $Ca$  is defined as the ratio of viscous to capillary forces,  $Bo$  signifies the ratio of gravitational to capillary forces. Based on these two numbers,  $Bu$  number is proposed as the ratio of gravitational to viscous forces, which is less commonly used. The following definitions are used to calculate these numbers, respectively, where  $\mu_g$ ,  $q_g$ ,  $\sigma$ , and  $\varphi$  represent the gas viscosity, injection/withdrawal Darcy velocity, interfacial tension, and aquifer porosity, respectively.  $K_v$  and  $r$  also represent the aquifer permeability (in the vertical direction) and characteristic pore radius, in turn.

$$Ca = \frac{\mu_g q_g}{\sigma \varphi \cos \theta}, \quad Bo = \frac{\Delta \rho g h K_v}{\sigma r}, \quad \text{and} \quad Bu = \frac{\Delta \rho g h K_v \varphi \cos \theta}{\mu_g q_g r} \quad (7)$$

### 2.3. Model implementation

As previously demonstrated, a complete  $P_c$ - $S_w$  hysteresis study was performed by considering all the predictable scanning curves that the simulation grid blocks can follow. The main steps for conducting the  $P_c$ - $S_w$  hysteretic behaviour are presented in Fig. 2. To this end, the saturations at three

consecutive time steps are used to determine whether a grid block will continue to drain, imbibe, or will have a turning point. Note that the local grid imbibition/drainage cycles are independent of the main injection/withdrawal schemes, meaning some grid blocks can go through an imbibition process while the main operation is controlled by the injection of the non-wetting phase (drainage). If a grid block demonstrates a reversal in its saturation trend, the Killough [1] model will be used to generate a new  $P_c$ - $S_w$  function based on the turning point saturation value, otherwise, the  $P_c$  functions would stay the same as the previous timestep. This procedure will be repeated at every time step for all the grid blocks to determine the local grid processes and their corresponding  $P_c$ - $S_w$  functions.

We have implemented the mentioned  $P_c$ - $S_w$  hysteresis model in the MATLAB Reservoir Simulation Toolbox (MRST) [2,55], which is used for simulating the  $H_2$  injection/withdrawal. In particular, we used the object-oriented, automatic-differentiation (AD-OO) framework in MRST employing a fully-implicit scheme to calculate pressure and saturation simultaneously.

## 3. Results and discussion

### 3.1. Input parameters

The parameters outlined in Table 1 are used as the input to the aforementioned  $P_c$ - $S_w$  hysteresis model. It should be noted that the impact of fluid and rock compressibilities has been neglected in this study. To elaborate on the potential importance of hydrogen compressibility, we conducted two simulations using a single injection scenario assuming

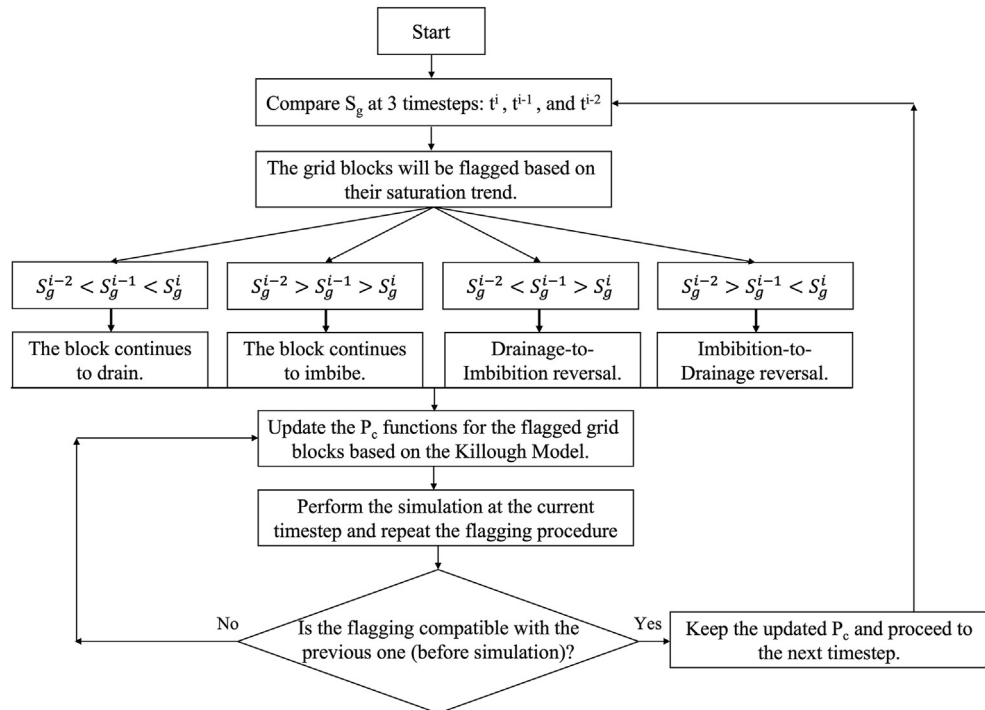


Fig. 2 – A diagram outlining the steps taken to simulate the injection and withdrawal of  $H_2$  using scanning analysis.

**Table 1 – A summary of basic parameters used in the model.**

| Parameter                                 | Value           |
|-------------------------------------------|-----------------|
| Permeability, mD                          | 250 [2]         |
| Porosity, (–)                             | 0.25 [2]        |
| Aquifer Depth, m                          | 1500            |
| Aquifer Thickness, m                      | 100             |
| Aquifer Lateral Size, m                   | 1000            |
| Brine Density, kg/m <sup>3</sup>          | 1000 [63]       |
| Brine Viscosity, cp                       | 0.80 [63]       |
| H <sub>2</sub> Density, kg/m <sup>3</sup> | 15 <sup>a</sup> |
| H <sub>2</sub> Viscosity, cp              | 0.01 [64]       |
| Interfacial Tension, mN/m                 | 72 [65,66]      |
| Reference Pressure, MPa                   | 15 [2]          |

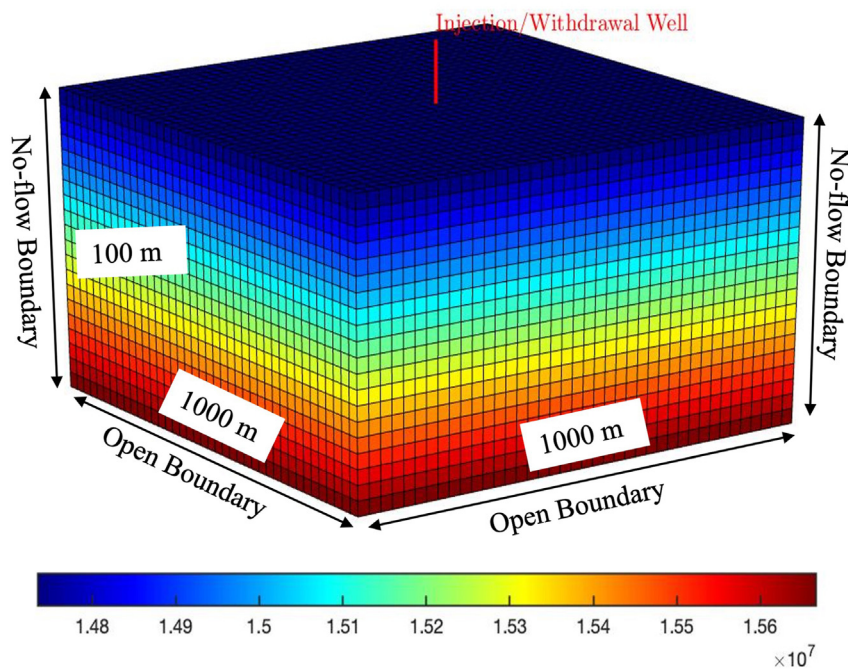
<sup>a</sup> Calculated based on the ideal gas law (Compressibility factor = 1).

**Table 2 – Petrophysical properties used in the model. The  $P_c$  data are derived by consecutive scaling [58] and Brooks-Corey [59,67] fitting of Nitrogen data presented in Plug and Bruining [57].**

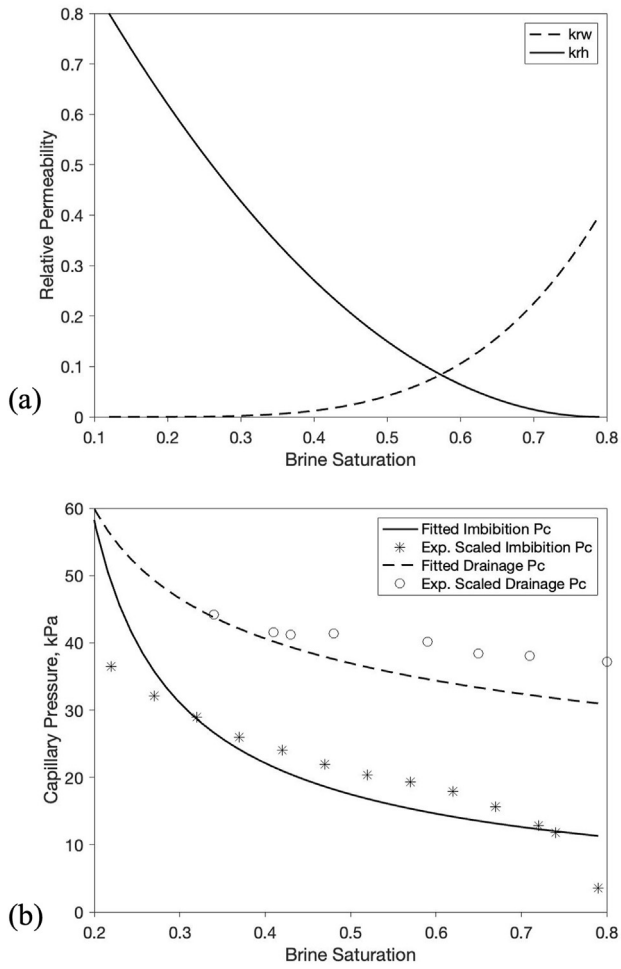
| Parameter                                        | Value    |
|--------------------------------------------------|----------|
| Corey Exponent for Water $k_r$ function          | 4        |
| End point $k_r$ value for Water                  | 0.40     |
| Corey Exponent for H <sub>2</sub> $k_r$ function | 2        |
| End point for H <sub>2</sub> $k_r$ value         | 0.80     |
| Corey Exponent for Drainage $P_c$ function       | 0.31     |
| Entry $P_c$ value for Drainage, kPa              | 31       |
| Corey Exponent for Imbibition $P_c$ function     | 0.77     |
| Entry $P_c$ value for Imbibition, kPa            | 11       |
| Irreducible Water Saturation                     | 0.12 [2] |
| Residual Gas Saturation                          | 0.21 [2] |

compressible and incompressible gases. Note that the compressible simulation has been done using the constant gas compressibility assumption. For comparison, the hydrogen saturation fronts were plotted as a function of aquifer depth and length as shown in Figure Appendix C.1. It can be seen that the two cases present small differences, especially near the well. However at very long injections when full segregation takes place, there is a considerable difference in fronts. Given the cyclic injection - withdrawal of hydrogen at much smaller PVs and to reduce further complexities in the model, we have neglected the impact of fluid compressibility in the simulations. Table 2 shows the petrophysical properties used in conjunction with the Brooks-Corey  $k_r$ - $S_w$  and  $P_c$ - $S_w$  formulations. As mentioned, we used N<sub>2</sub>-water  $P_c$ - $S_w$  data [57] and scaled it for the H<sub>2</sub>-water system. The entry capillary pressure and exponents demonstrate the Brooks-Corey [59] fitted values after scaling. The data presented by Plug and Bruining [57] is shown in Fig. 4 as well. The aquifer is also considered to be fully water wet ( $\cos \theta = 1$ ). Fig. 3 shows the simulation domain, initial conditions, and boundary conditions. We have considered a  $1000 \times 1000 \times 100$  m<sup>3</sup> homogeneous aquifer with its bottom border regarded as an open flow boundary, while the vertical boundaries are designated as no-flow boundaries. The injection/withdrawal well is perforated down to half of the aquifer thickness and is placed in the middle of X–Y plane. A sensitivity analysis was performed to check the impact of the grid resolution on the results, which is presented in Appendix A.

The H<sub>2</sub> injection/withdrawal schemes considered in this study include two consecutive cycles of injection-withdrawal in a fully water-saturated aquifer. Note that the injection/



**Fig. 3 – The simulation domain and initial pressure conditions of the aquifer in Pa. Hydrostatic initial pressure condition has been assumed, all boundary faces are closed (no flow), except the bottom boundary, which is open flow. The injection/withdrawal well is considered at the centre of the aquifer and has penetrated the aquifer to its half depth.**



**Fig. 4 – (a) The H<sub>2</sub> ( $k_{rh}$ - $S_w$ ) and water ( $k_{rw}$ - $S_w$ ) relative permeability curves used in this study. The  $k_r$ - $S_w$  values are calculated according to Brooks-Corey Equation [59]. The Corey exponents for H<sub>2</sub> and water are 2 and 4, respectively. The endpoint relative permeability values for water and H<sub>2</sub> are set equal 0.4 and 0.8, respectively. (b) The imbibition and drainage  $P_c$ - $S_w$  data used in this study, fitted by the Brooks-Corey equation [59]. The Corey exponents for imbibition and drainage are equal 0.77 and 0.31, respectively. The entry capillary pressure values are also set equal 11 and 31 kPa. To derive this data first the N<sub>2</sub> data of Plug and Bruining [57] were scaled using the Leverett J-Function [58], then the capillary pressure values was fitted to the Brooks-Corey equation.**

withdrawal rates are kept constant for each simulation case. The first injection phase continues for 6 months, followed by 3-month withdrawal-injection-withdrawal phases.

The remaining of this paper is devoted to how the hysteresis in  $P_c$ - $S_w$  curves affects the UHS. The impact of the flow rate and aquifer characteristics will be examined by running the simulations for various  $Ca$  and  $Bo$  numbers. The last

section will also include a discussion on the numerical complexities that may be encountered using the scanning curves.

### 3.2. Capillary pressure - saturation hysteresis

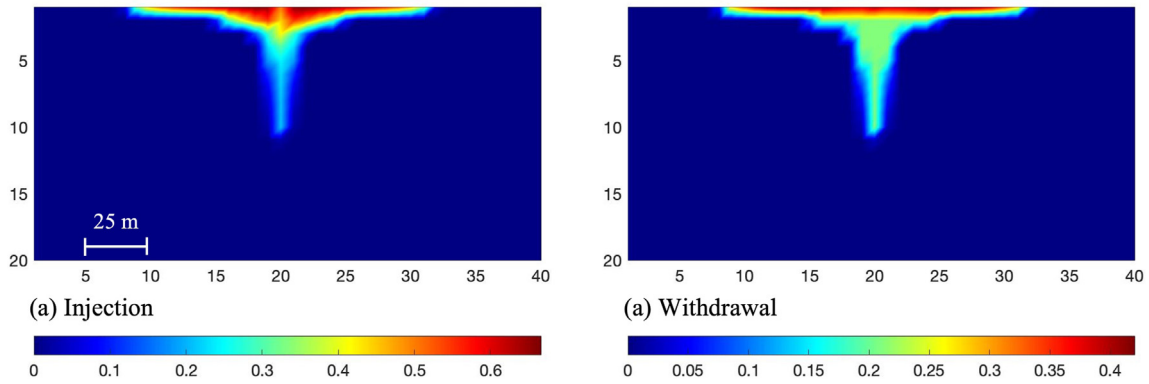
This first modeling case probes the effect of  $P_c$ - $S_w$  hysteresis on the efficiency of the H<sub>2</sub> injection/withdrawal cycles when considering a low capillary number ( $Ca = 2 \times 10^{-7}$ ) equivalent to 0.01 PV/year (684 m<sup>3</sup>/day) and  $Bo = 0.44$ . H<sub>2</sub> injection/withdrawal is regarded to happen in two cycles, with the first injection lasting six months, and the subsequent withdrawal, injection, withdrawal each for three months.

For comparison of the impact of  $P_c$ - $S_w$  hysteresis, the cyclic simulations were repeated for four cases: 1)  $P_c$ - $S_w$  hysteresis with full scanning behaviour, 2)  $P_c$ - $S_w$  hysteresis only with main loops (no scanning), 3) drainage  $P_c$ - $S_w$  curve with no hysteresis, and 4) zero capillary pressure. Fig. 5 shows the H<sub>2</sub> saturation profile at the end of the second injection and withdrawal when the  $P_c$ - $S_w$  scanning curves were included. Table 3 details the difference between the four scenario cases in terms of produced H<sub>2</sub> volume and remained H<sub>2</sub> saturation in the aquifer. It can be concluded that assumptions related to ignoring the hysteresis or ignoring the capillary pressure (Scenarios 2–4) slightly underestimate remaining H<sub>2</sub> saturation for this specific case. However, due to the competition between viscous, capillary, and gravitational forces, this underestimation may be pronounced under different conditions. Figure Appendix B.1 also presents the data regarding the H<sub>2</sub> operational flow rate and bottom-hole pressure as a function of time for these four cases. It can be seen that the  $P_c$  mainly influences the bottom-hole pressure and flow rate at late-time withdrawal, when the H<sub>2</sub> withdrawal rate is fallen as a result of water production. Moreover, during the second withdrawal stage, the bottom-hole pressure has fallen less noticeably for the scanning (first) scenario compared to second and third scenarios. Moreover, it can be deducted that the flow rate will be overestimated when neglecting the  $P_c$  effect. It can be concluded that although the impact of  $P_c$  scenarios is not significant in terms of saturation distribution and remaining saturation, their influence on the pressure field and well properties, especially at late-time, can become noticeable.

### 3.3. Dimensionless number analysis

The interaction of viscous, capillary, and gravitational forces determines how efficiently a UHS design operates. The relative significance of these forces can be quantified using the dimensionless capillary, Bond, and buoyancy numbers as defined in Equation (7). Capillary number, for instance, is directly connected to injection/withdrawal rates, which its optimization is vital for a successful and low-loss UHS operation. In this section, impact of variations of these dimensionless numbers on H<sub>2</sub> removal has been investigated. To determine how effective certain scenarios are, we set a withdrawal cut-off constraint equal to 10% gas-cut ( $Q_g/Q_{total} >$





**Fig. 5** –  $H_2$  saturation profile at the end of (a) second injection and (b) second withdrawal with including  $P_c$ - $S_w$  scanning curves (first case in Table 3) for  $Ca = 2 \times 10^{-7}$  and  $Bo = 0.44$ . Given the slow viscous force and intermediate Bond numbers, hydrogen saturation override is significant.

**Table 3** – A quantitative comparison between four cases with different  $P_c$  models. For these scenarios, no cut-off constraint is applied, and the second withdrawal time is set to three months. The injection/withdrawal rate is equal to 0.01 PV/year and  $Ca = 2 \times 10^{-7}$ . The error value is calculated considering the first scenario as the base case ( $|(S_{g,rem}^i - S_{g,rem}^1)/S_{g,rem}^1|$ ). The total injected pore volume is equal to 0.009.

| Case Number | Description                                               | Produced $H_2$ Pore Volume | Remaining $H_2$ Saturation | Remaining $H_2$ Relative Difference, % |
|-------------|-----------------------------------------------------------|----------------------------|----------------------------|----------------------------------------|
| 1           | Hysteresis in $P_c$ - $S_w$ curves - with scanning curves | $4.638 \times 10^{-3}$     | 0.276                      | —                                      |
| 2           | Hysteresis in $P_c$ - $S_w$ curves - bounding curves only | $4.622 \times 10^{-3}$     | 0.269                      | −2.460                                 |
| 3           | No hysteresis - only drainage $P_c$ - $S_w$               | $4.642 \times 10^{-3}$     | 0.268                      | −2.858                                 |
| 4           | No capillary pressure                                     | $4.778 \times 10^{-3}$     | 0.272                      | −1.483                                 |

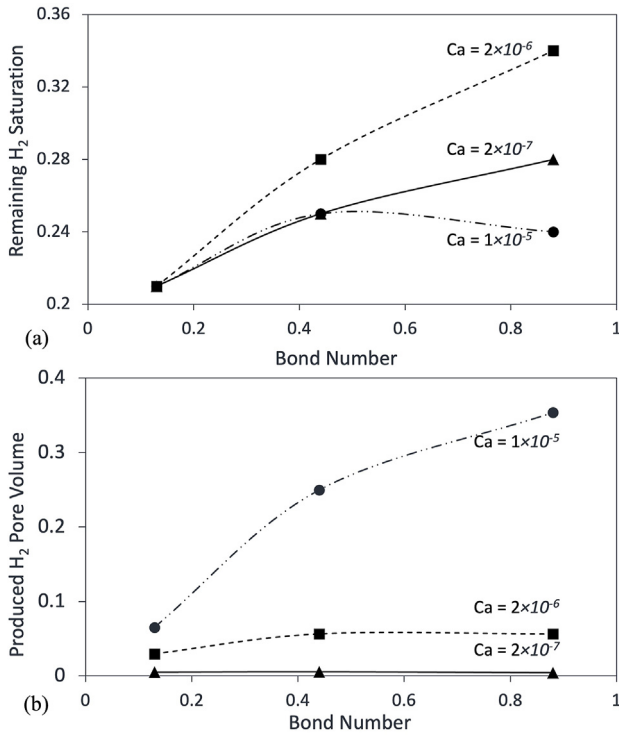
**Table 4** – A quantitative comparison between nine cases of  $H_2$  injection/withdrawals indicating the significance of Capillary, Bond, and Buoyancy numbers on the  $H_2$  productivity. The second withdrawal stops when the gas-cut reaches 10%. The remaining  $H_2$  saturations quantifies the average saturation of grid blocks containing  $S_g > 0.05$ . The  $H_2$  saturation profiles regarding these cases are shown in Figs. 7 and 8.

| Case Number | Injected PV, 1/year | Capillary Number, (–) | Bond Number, (–) | Buoyancy Number, (–) | Produced Pore Volume  | Remained $H_2$ Saturation | Withdrawal Time, Days |
|-------------|---------------------|-----------------------|------------------|----------------------|-----------------------|---------------------------|-----------------------|
| 1           | 0.01                | $2 \times 10^{-7}$    | 0.13             | $6.5 \times 10^5$    | $5.00 \times 10^{-3}$ | 0.21                      | 97.25                 |
| 2           | 0.01                | $2 \times 10^{-7}$    | 0.44             | $2.20 \times 10^6$   | $5.44 \times 10^{-3}$ | 0.25                      | 210.01                |
| 3           | 0.01                | $2 \times 10^{-7}$    | 0.88             | $4.40 \times 10^6$   | $4.40 \times 10^{-3}$ | 0.28                      | 289.91                |
| 4           | 0.13                | $2 \times 10^{-6}$    | 0.13             | $6.50 \times 10^4$   | $2.95 \times 10^{-2}$ | 0.21                      | 46.11                 |
| 5           | 0.13                | $2 \times 10^{-6}$    | 0.44             | $2.20 \times 10^5$   | $5.64 \times 10^{-2}$ | 0.28                      | 148.84                |
| 6           | 0.13                | $2 \times 10^{-6}$    | 0.88             | $4.40 \times 10^5$   | $5.64 \times 10^{-2}$ | 0.34                      | 208.19                |
| 7           | 0.63                | $1 \times 10^{-5}$    | 0.13             | $1.30 \times 10^4$   | $6.52 \times 10^{-2}$ | 0.21                      | 20.54                 |
| 8           | 0.63                | $1 \times 10^{-5}$    | 0.44             | $4.40 \times 10^4$   | $2.49 \times 10^{-1}$ | 0.25                      | 123.73                |
| 9           | 0.63                | $1 \times 10^{-5}$    | 0.88             | $8.80 \times 10^4$   | $3.53 \times 10^{-1}$ | 0.24                      | 166.18                |

0.1) during the second  $H_2$  withdrawal phase. For simulations,  $P_c$ - $S_w$  hysteresis without the scanning curves were considered (case 2 in Table 3), as inclusion of the scanning curves in MRST led to significantly higher computational cost. To elaborate, the variations in the capillary number and Buoyancy number can be achieved by changing the flow rates. Altering the aquifer permeability, which would directly affect the  $P_c$ - $S_w$  functions, is another way to examine the effect of Bond and Buoyancy numbers. In order to do this, three different permeability scenarios (50, 250, and 1000 mD) and three injection/withdrawal rates were considered, which resulted in

nine cases. The average pore radius to calculate the Bond numbers were taken from the study of Kashif et al. [68]. A summary of these cases is presented in Table 4 and Fig. 6 with the visualisation displayed in Figs. 7 and 8 showing Y-Z aquifer slices where the injection/withdrawal happens.

Considering the variations in capillary number, it can be seen that the withdrawal constraint is satisfied earlier when the capillary number rises (flow rate increases), and the  $H_2$  removal would be halted due to the high water withdrawal. As Pan et al. [16] also mentioned, larger withdrawal rates cause the reservoir pressure to drop more noticeably, therefore it is



**Fig. 6 – Impact of Bond number and capillary number on (a) remaining H<sub>2</sub>, (b) produced H<sub>2</sub> in pore volume. All plotted results are related to the end of second withdrawal for the nine cases presented in Table 4. The second withdrawal stops when the gas-cut reaches 10%. The remaining H<sub>2</sub> saturations quantifies the average saturation of grid blocks containing  $S_g > 0.05$ . The H<sub>2</sub> saturation profiles regarding these cases are shown in Figs. 7 and 8.**

critical to maintain the aquifer pressure by injecting a large initial H<sub>2</sub> volume or other cushion gases. Variation of the pressure and flow rate versus time for some cases is shown in Figure Appendix B.2. Higher H<sub>2</sub> removal rates can be attained when the aquifer pressure is maintained because viscous forces predominate and reduce H<sub>2</sub> lateral spreading [16]. However, caution must be taken while increasing the capillary number since at high flow rates, the magnitude of viscous forces increases and therefore, water upconing would be unavoidable and would stop the withdrawal of H<sub>2</sub>. A graphical representation of water upconing during the second H<sub>2</sub> withdrawal at a rate of 0.13 PV/year ( $Ca = 2 \times 10^{-6}$ ,  $Bo = 0.44$ ) is shown in Fig. 9. The graph shows the H<sub>2</sub> saturation at six different timesteps, from the second withdrawal up until the water upconing is fully formed. When upconing takes place, water bypasses H<sub>2</sub> and will breakthrough to the withdrawal well faster, trapping the H<sub>2</sub> in the aquifer.

The relative magnitude of gravitational forces compared to capillary forces can be investigated using the dimensionless

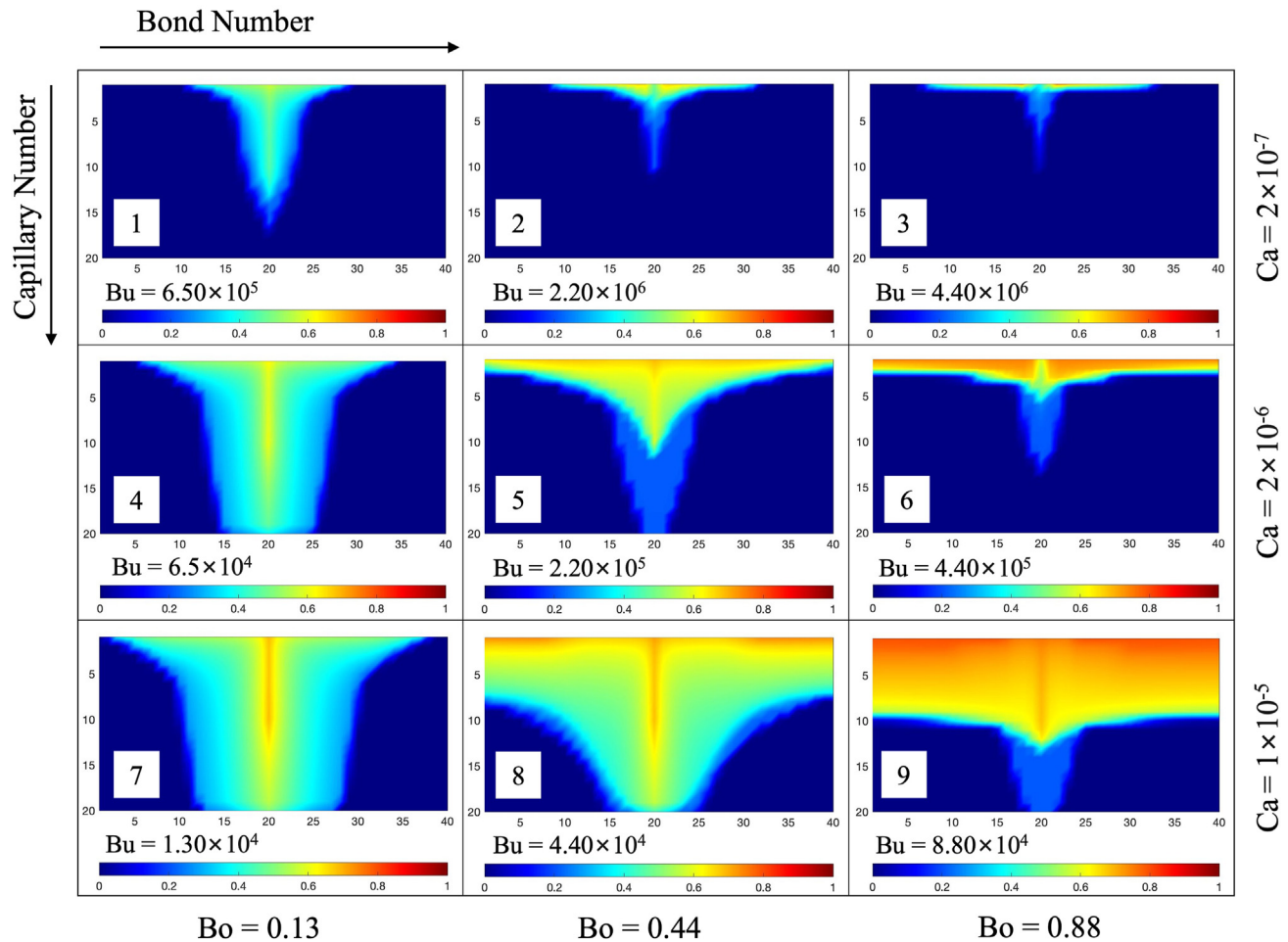
Bond number. In order to consider the effect of variations in aquifer permeability on capillary pressure curves, we have used the Leverett J-Fuction  $J(S_w) = \frac{P_c(S_w) \sqrt{K/\phi}}{\sigma \cos \theta}$  [58] to re-scale the  $P_c-S_w$  curves. At a constant capillary number, as shown in Table 4 and Figs. 6–8, when the Bond number drops (permeability reduces), the interfaces become more diffuse and H<sub>2</sub> finds it more difficult to travel upwards, which causes it to spread out more in the aquifer. As a result, at low Bond numbers, as contrasted to high Bond numbers (high permeability values), the cut-off requirement is satisfied significantly earlier. Although higher vertical aquifer permeability appears to be beneficial for H<sub>2</sub> removal, caution should be used when selecting a UHS site. Higher horizontal aquifer permeability results in faster lateral spreading of H<sub>2</sub>, which allows H<sub>2</sub> to escape through the aquifer's spill points. Another important implication is that although an increase in capillary number results in reduced withdrawal time, according to Fig. 6a, there is non-monotonic trend between remaining H<sub>2</sub> and Bond number at high capillary numbers. This means that at a certain range of Bond number values, it is even more beneficial to increase the operational flow rate as the trapped saturation would decrease. This also enhances the significant impact of aquifer permeability on the UHS design.

### 3.4. Discussion on numerical challenges

As previously noted, we have simulated the H<sub>2</sub> injections and withdrawals as well as the hysteresis behaviour using the MRST toolbox [2,55]. Killough [1] methodology was used to implement the  $P_c$  model in the MRST AD-OO solver to create the scanning curves. However, there are a few issues that needs to be discussed and clarified because they have a significant impact on the simulation operation for including scanning curves.

The complexities can be first discussed regarding the Killough [1] methodology. The turning point history of the grid blocks, the relative placement of the turning points to one another, and the gas saturation at those turning points are all important factors in achieving appropriate, bounded scanning curves from the model, especially when grid blocks go through subsequent drainage and imbibition phases and present a high number of turning points. Not-bounded scanning curves may be achieved depending on the occurrence of the stated factors, which needs to be handled. The new scanning curves can also only be generated between the saturation range of two subsequent turning points. The model must be able to manage saturation values outside of this range by re-calling the  $P_c-S_w$  curve at timestep  $t^{i-2}$  (either the scanning or main curve depending on the number of turning points).

On the other hand, the inclusion of the  $P_c-S_w$  scanning model presented a high level of complexity by significantly increasing the number of timesteps needed as well as the time required for solving each timestep. Additionally, these



**Fig. 7 – Visualisation of hydrogen plume at different capillary, Bond, and Buoyancy numbers at the end of the second injection. Each row presents 3 cases at a constant capillary number. The quantitative difference between these cases is also shown in Table 4.**

difficulties would increase, especially at larger flow rates, where timestep sizes under an hour were necessary. Another important issue was regarding the  $P_c$ - $S_w$  behaviour in the vicinity and below the gas residual saturation region. A large number of consecutive turning points may be produced by the numerical saturation oscillations in this region, ultimately leading to the scanning curves becoming unbounded. We considered the primary drainage curve for grid blocks starting to fill with gas until their saturation approaches the residual gas saturation to overcome these problems. Moreover, some grid blocks experience substantial gas saturation drops during the  $H_2$  withdrawal. As a result, their saturations approach residual gas, and they may also experience saturation oscillations near this area that can result in unbounded scanning curves. Transferring from the last properly-generated scanning curve to the main drainage curve appeared to be the simplest way to get around this issue.

#### 4. Shortcomings and future work

The main purpose of this study has been to investigate the impact of hysteresis in  $P_c$ - $S_w$  functions and injection/withdrawal rates on the efficiency of the UHS operations while ignoring the  $k_r$ - $S_w$  hysteresis. The authors are fully aware of the significance of  $k_r$  data while simulating the hysteresis. However, the primary goal of this study was to solely consider the impact of  $P_c$ - $S_w$  hysteresis as it is largely disregarded in the literature. This provided insights under what conditions complexities of  $P_c$ - $S_w$  hysteresis are worth investment for practical industrial scale. Clearly, heterogeneity of the reservoirs and wettability distribution (in depleted reservoirs) are major factors that need to be addressed in future studies. Moreover, higher injection/withdrawal rates (capillary number) result in different  $k_r$  curves, and including the dynamic  $k_r$  effect can be investigated in future studies.

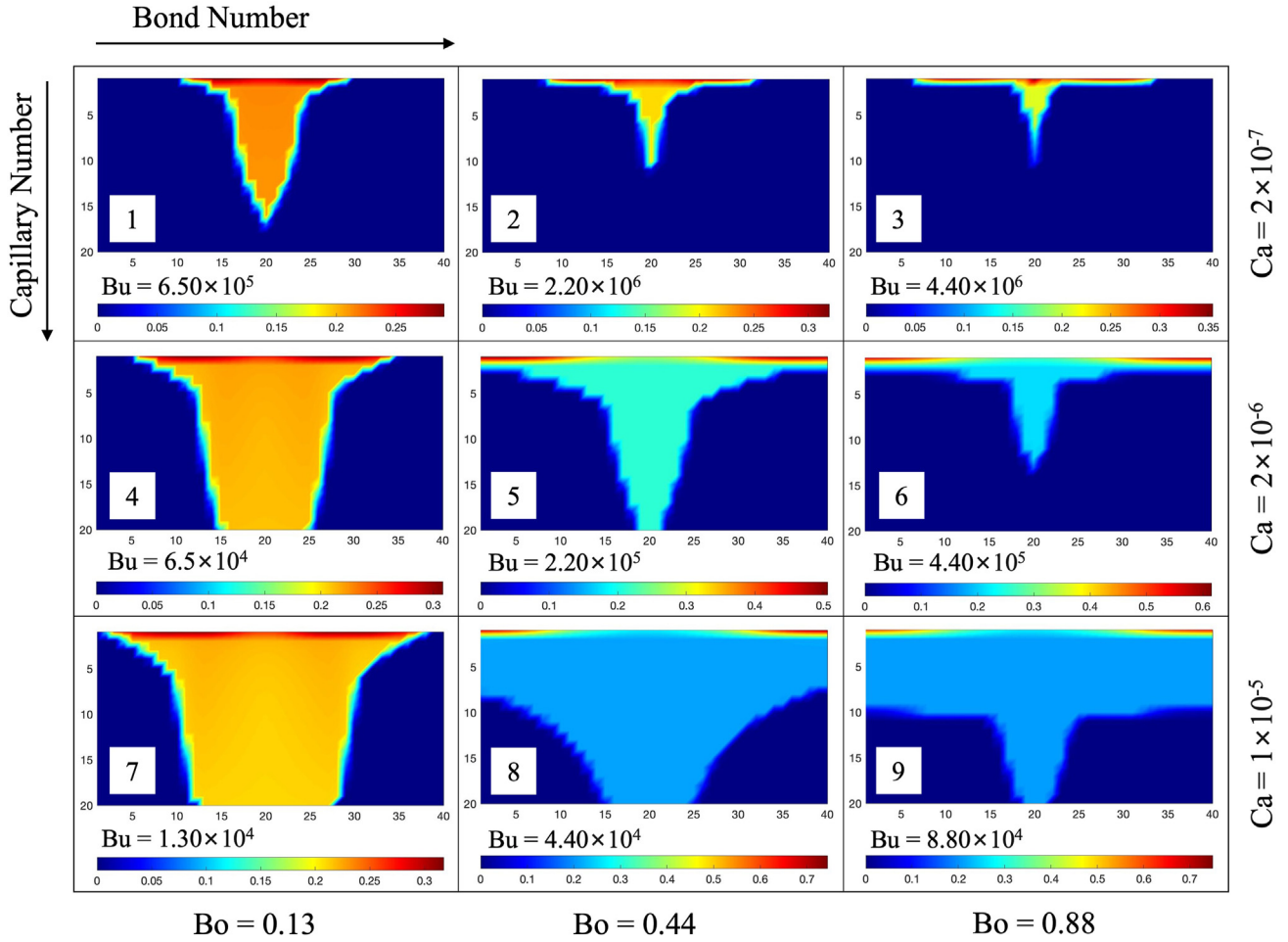


Fig. 8 – Visualisation of hydrogen plume at different capillary, Bond, and Buoyancy numbers at the end of the second withdrawal. Each row presents 3 cases at a constant capillary number. The quantitative difference between these cases is also shown in Table 4.

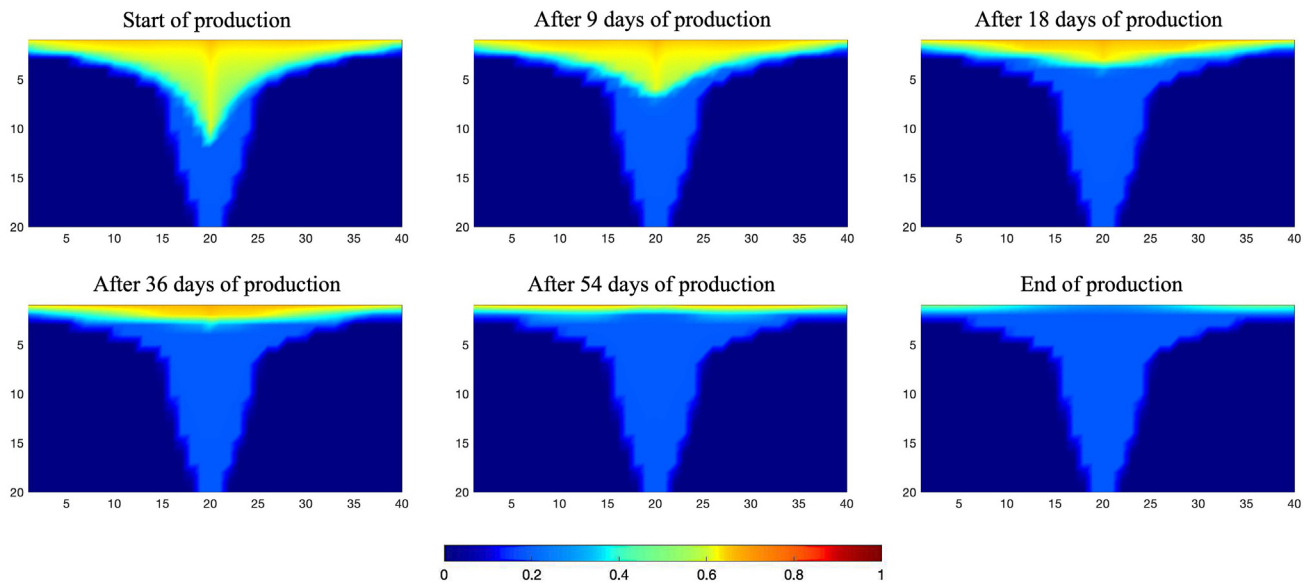


Fig. 9 – An example of the formation of water upconing at six time snapshots during the second cycle of  $H_2$  extraction at rate of 0.13 PV/year ( $Bo = 0.44$  and  $Ca = 2 \times 10^{-6}$ ). Beginning with the second withdrawal time and continuing until the time upconing limits the  $H_2$  withdrawal.



## 5. Conclusions

The inclusion of full hysteretic behaviour of capillary-pressure and relative-permeability functions in two-phase flow simulators impose high computational complexities and are time consuming. Therefore, the first objective of this study was to investigate the sole impact of hysteresis in capillary pressure curves in terms of scanning curve generations, while ignoring the hysteresis in relative permeability. Moreover, this study looked through the effects of operational flow rates and aquifer characteristics in terms of dimensionless parameters (namely capillary number and Bond number) on UHS performance. Analysing the results, the following conclusions are drawn:

- The flow behaviour and effectiveness of the UHS operation were not considerably impacted by the addition of capillary pressure scanning curves in the model. Exclusion of  $P_c$ - $S_w$  scanning curves slightly underestimates the remaining hydrogen saturation.
- The impact of capillary pressure on the bottom-hole pressure and withdrawal rate can become significant at late-time withdrawals, when the water is flowing to the well more considerably, and exclusion of capillary pressure clearly overestimates the bottom-hole capillary pressure.
- The UHS efficiency is highly impacted by the operational flow rate, and aquifer characteristics.
- At high capillary numbers, due to the high pressure drop, water bypasses hydrogen and the resulting water upconing ceases the hydrogen withdrawal.
- Aquifer permeability, particularly in the vertical direction, influences the efficiency of UHS. When Bond number presents a small value, gravity segregation is very slow; the interfaces become very diffuse, and the cut-off constraint is satisfied more quickly compared to high permeability scenarios.

## 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.

## Acknowledgements

The authors thank the University of Manchester for funding the PhD studentship of Farzaneh Nazari through the President's Doctoral Scholar Award. The authors thank Prof. Knut Andreas Lie and Dr. Olav Møyner from the SINTEF group for their support via email communications. Rouhi Farajzadeh

thanks Shell Global Solutions International B.V., The Hague, Netherlands for granting permission to publish this paper.

## Nomenclature

### Letters

|              |                                             |
|--------------|---------------------------------------------|
| $P_c$        | Capillary Pressure, Pa                      |
| $P_e$        | Entry Capillary Pressure, Pa                |
| $k_r$        | Relative Permeability                       |
| $k_r^{max}$  | endpoint relative permeability              |
| $n$          | Brooks-Corey $P_c$ exponent                 |
| $m$          | Brooks-Corey $k_r$ exponent                 |
| $S_w$        | Water Saturation                            |
| $S_{rw}$     | residual Water Saturation                   |
| $S_w^{max}$  | maximum attainable wetting-phase saturation |
| $S_w^{hyst}$ | wetting-phase turning point saturation      |
| $S_g$        | Gas Saturation                              |
| $q_g$        | Gas Velocity, m/s                           |
| $t^i$        | Timestep Number                             |
| $K$          | Absolute Permeability, $m^2$                |
| $r$          | Characteristic Pore Radius, m               |
| $q$          | Flow rate, $m^3/s$                          |
| $Ca$         | Capillary Number                            |
| $Bo$         | Bond Number                                 |
| $Bu$         | Buoyancy Number                             |
| $\epsilon$   | Killough [1] Curvature Parameter            |
| $\phi$       | Porosity                                    |
| $\mu_g$      | Gas Viscosity, Pa.s                         |
| $\sigma$     | Interfacial Tension, N/m                    |
| $\theta$     | Contact Angle, $^\circ$                     |

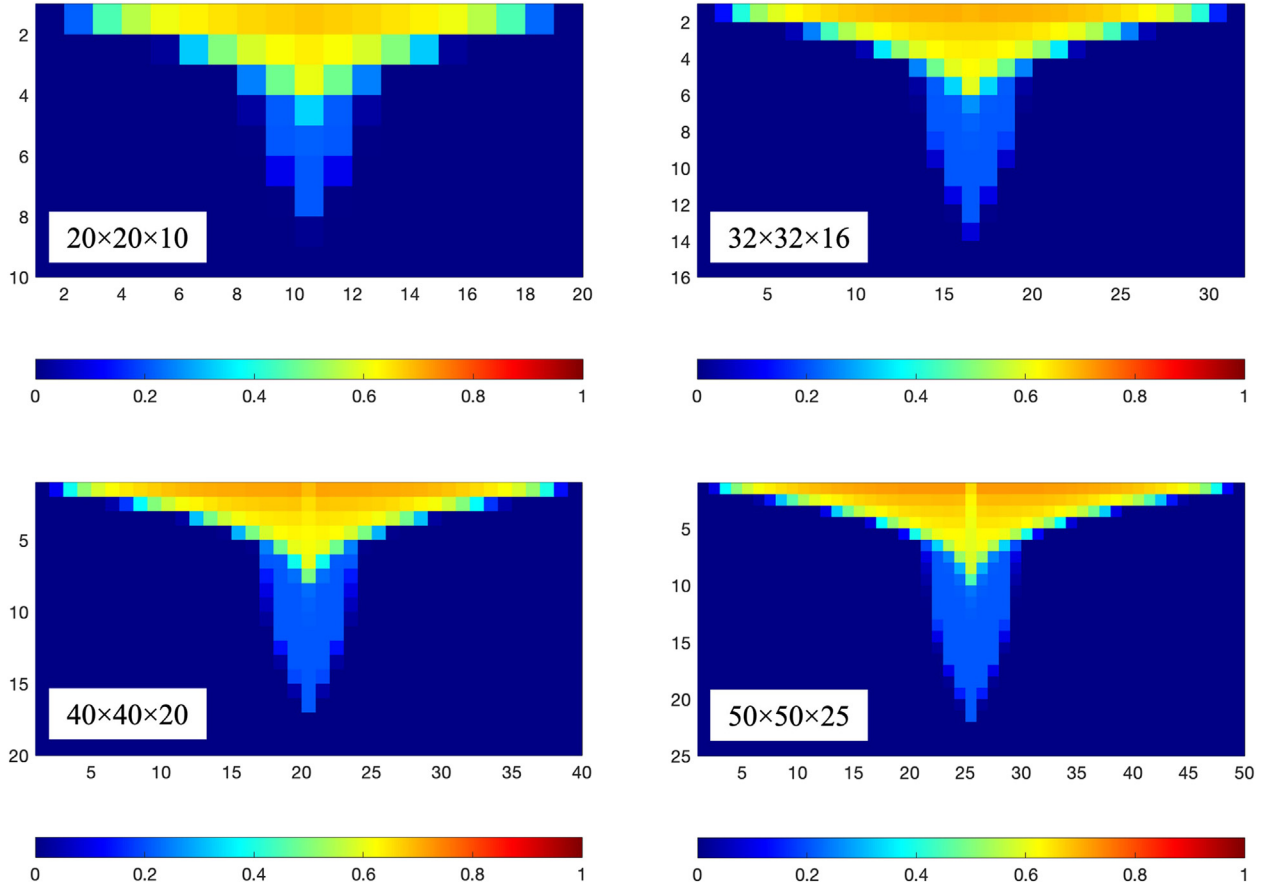
### Abbreviations

|       |                                            |
|-------|--------------------------------------------|
| UHS   | Underground Hydrogen Storage               |
| BC    | Brooks-Corey                               |
| IFT   | Interfacial Tension                        |
| AD-OO | Object-Oriented, Automatic-Differentiation |
| PV    | Pore Volume                                |

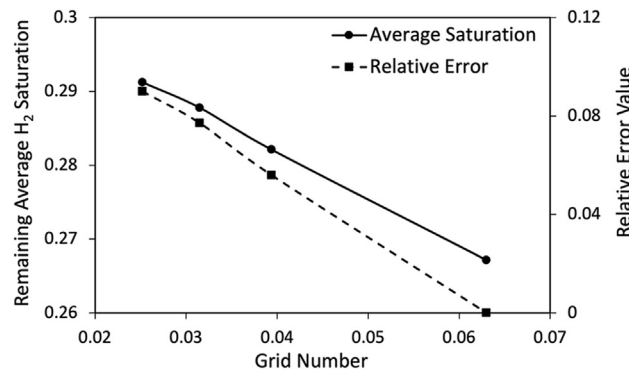
## Appendix A. Sensitivity Analysis

To determine the ideal number of grids for the simulations, a sensitivity analysis regarding the number of grid units is being carried out. Note that the grid aspect ratio is kept constant at 5 in all sensitivity simulations. Figure Appendix A.1 shows the  $H_2$  saturation profiles of four sensitivity analysis cases. To achieve these results, we have considered a single cycle of  $H_2$  injection-withdrawal with rate of 0.015 PV/year. The  $H_2$  was injected for six months and produced for three months after the injection. The results of the sensitivity cases are being compared in terms of an independent parameter, namely the remaining average  $H_2$  saturation. To derive this saturation value, we have considered only the grid blocks which demonstrates  $H_2$  saturation values over 5%. Figure Appendix A.2 presents the error

plot of the conducted sensitivity analysis. The error values are calculated with respect to the point with the lowest number of grids. It can be seen that from the point with  $40 \times 40 \times 20$  grids, the average saturation and error values are approaching a plateau. Therefore, we have considered the  $40 \times 40 \times 20$  case for the rest of the simulations.



**Figure Appendix A.1:** The results of sensitivity analysis conducted regarding the number of grids. The grid aspect ratio in all cases is equal 5. The results illustrates the  $H_2$  saturation profile after 6-months injection with rate of 0.015 PV/year. The X and Y axes demonstrate the grid numbers in the corresponding directions.



**Figure Appendix A.2:** The grid sensitivity results as a function of remaining average  $H_2$  saturation ( $S_g > 0.05$ ) after a 3-months withdrawal with rate of 0.015 PV/year. The x-axis demonstrates the grid number derived based on  $1/\sqrt[3]{n_x n_y n_z}$ . The secondary y-axis also shows the average saturation relative error calculated with respect to the case with lowest number of grids ( $|S_{g,avg}^i - S_{g,avg}^1|/S_{g,avg}^1$ ).

## Appendix B. Operational Flow rate and Pressure Plots

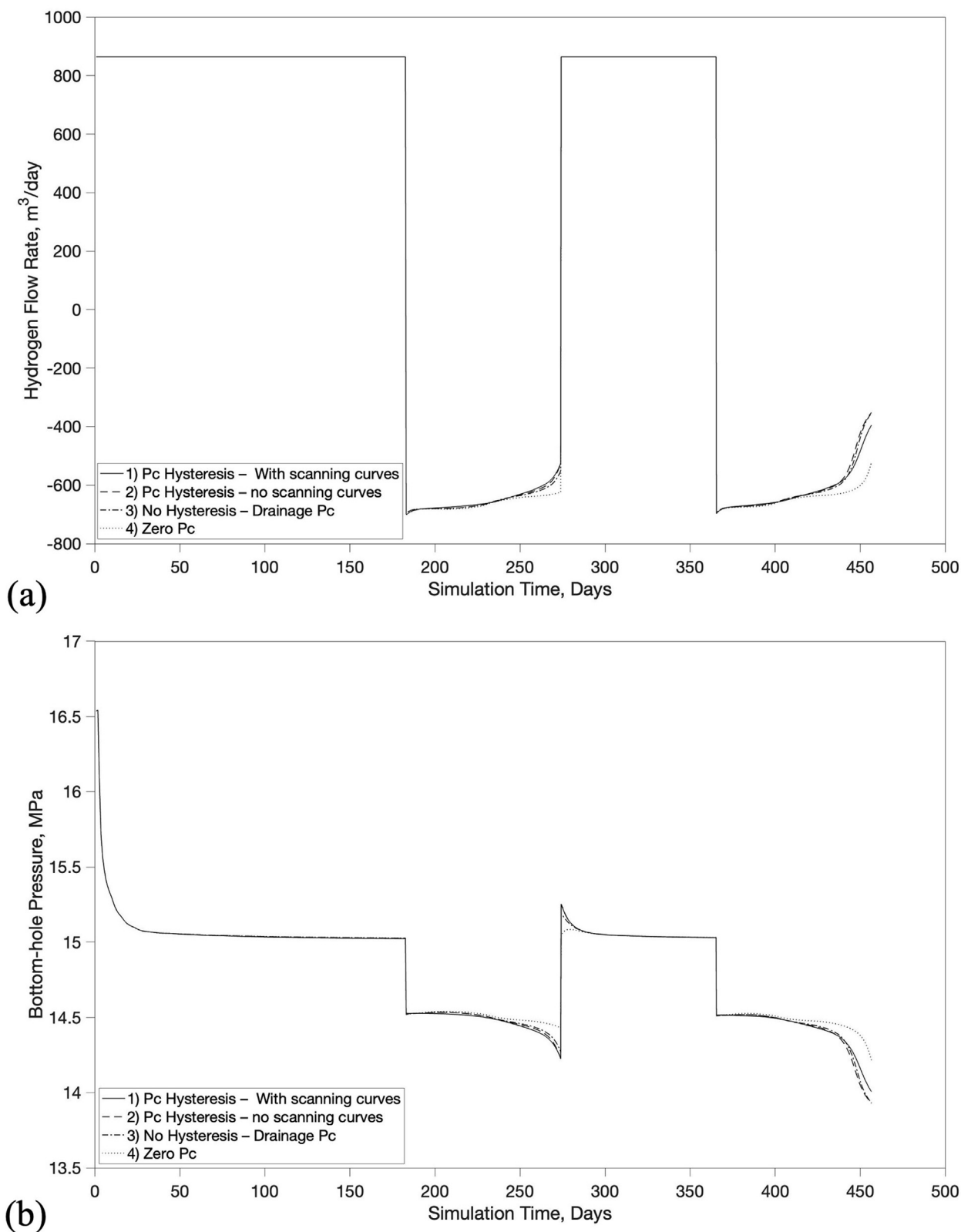
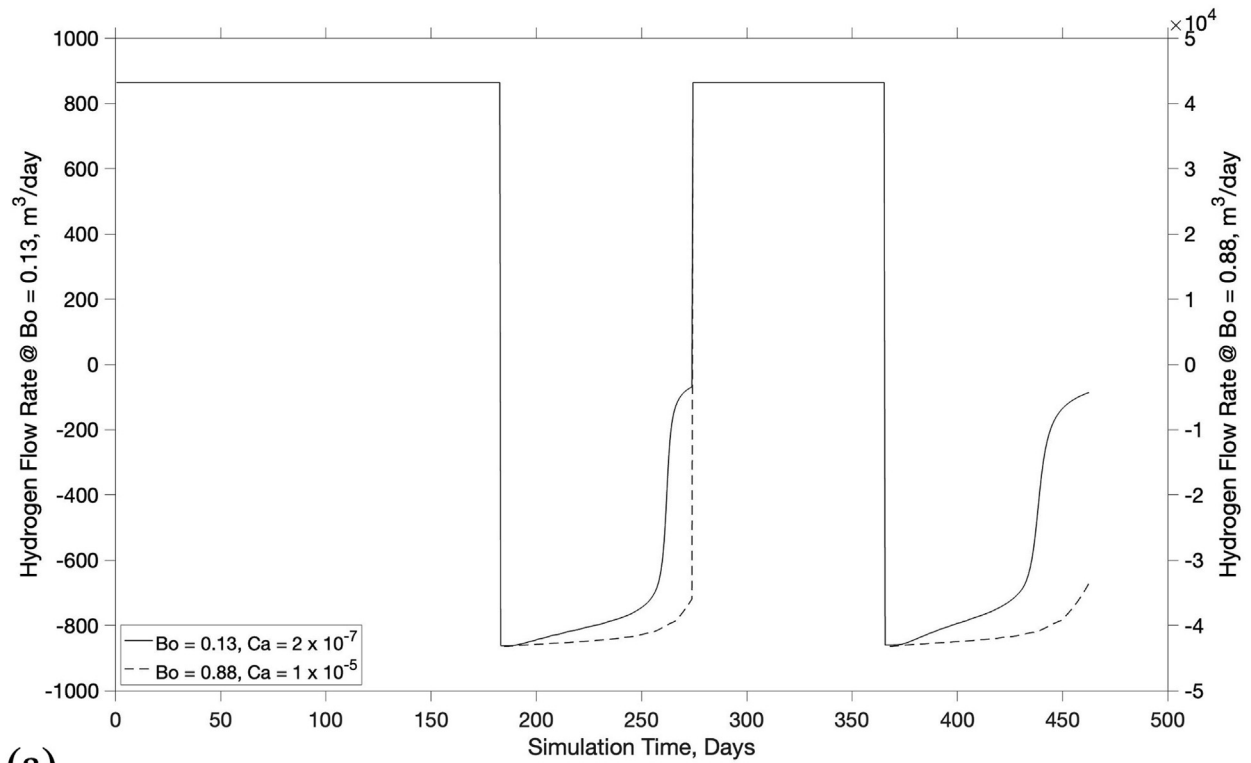
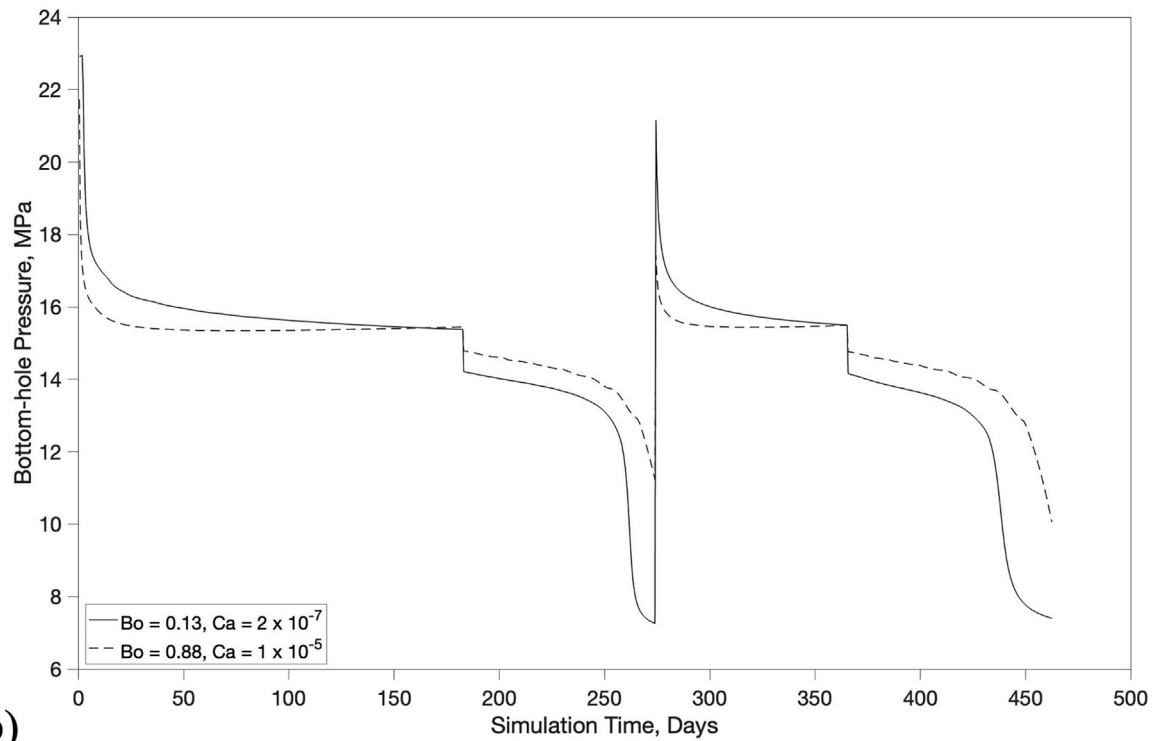


Figure Appendix B.1:  $\text{H}_2$  injection/withdrawal rate (a) and bottom-hole pressure (b) versus simulation time for the four hysteresis cases presented in Table 3.



(a)

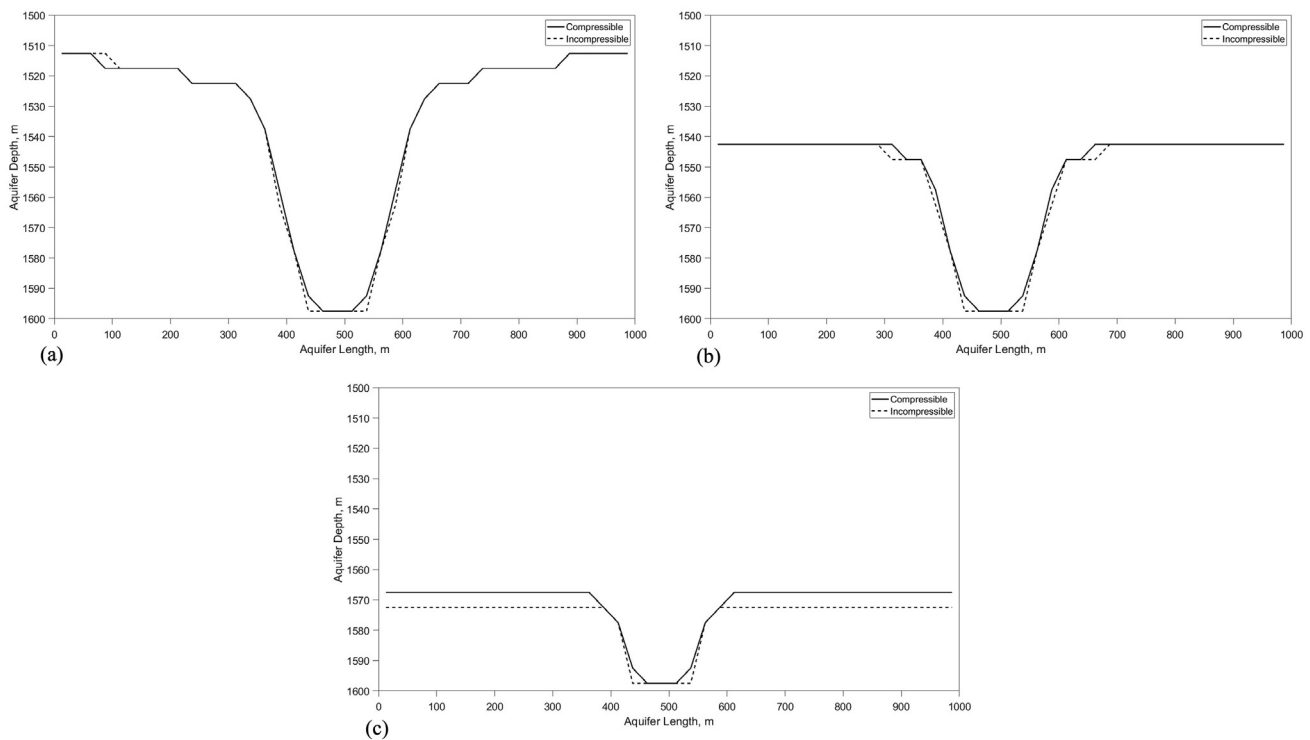


(b)

Figure Appendix B.2: H<sub>2</sub> injection/withdrawal rate (a) and bottom-hole pressure (b) versus simulation time for the the two extreme cases (1 and 9) presented in Table 4.



## Appendix C. Impact of Gas Compressibility



**Figure Appendix C.1:** H<sub>2</sub> injection front after (a) 0.1, (b) 0.3, and (c) 0.5 total injected pore volume for two compressible and incompressible gas phases. These simulations were done based on the Case 5 data presented in Table 4.

## REFERENCES

- [1] Killough J. Reservoir simulation with history-dependent saturation functions. *Soc Petrol Eng J* 1976;16:37–48.
- [2] Lie K-A. An introduction to reservoir simulation using MATLAB/GNU Octave: user guide for the MATLAB Reservoir Simulation Toolbox (MRST). Cambridge University Press; 2019.
- [3] Heinemann N, Alcalde J, Micioc JM, Hangx SJ, Kallmeyer J, Ostertag-Henning C, Hassanpouryouzband A, Thaysen EM, Strobel GJ, Schmidt-Hattenberger C, et al. Enabling large-scale hydrogen storage in porous media—the scientific challenges. *Energy Environ Sci* 2021;14:853–64.
- [4] Hemme C, Van Berk W. Hydrogeochemical modeling to identify potential risks of underground hydrogen storage in depleted gas fields. *Appl Sci* 2018;8:2282.
- [5] Abe JO, Popoola A, Ajenifuja E, Popoola OM. Hydrogen energy, economy and storage: review and recommendation. *Int J Hydrogen Energy* 2019;44:15072–86.
- [6] Singla MK, Nijhawan P, Oberoi AS. Hydrogen fuel and fuel cell technology for cleaner future: a review. *Environ Sci Pollut Control Ser* 2021;28:15607–26.
- [7] Appleby A. Fuel cells and hydrogen fuel. *Int J Hydrogen Energy* 1994;19:175–80.
- [8] Lototskyy MV, Tolj I, Pickering L, Sita C, Barbir F, Yartys V. The use of metal hydrides in fuel cell applications. *Prog Nat Sci: Mater Int* 2017;27:3–20.
- [9] Yartys V, Zavaliy I, Berezovets V, Pirskey Y, Manilevich F, Kytaya A, Verbovitsky Y, Dubov Y, Kutsyi A. Hydrogen generator integrated with fuel cell for portable energy supply. *J Phys: Energy* 2022;5:014014.
- [10] Haseli Y. Maximum conversion efficiency of hydrogen fuel cells. *Int J Hydrogen Energy* 2018;43:9015–21.
- [11] Lin R-H, Xi X-N, Wang P-N, Wu B-D, Tian S-M. Review on hydrogen fuel cell condition monitoring and prediction methods. *Int J Hydrogen Energy* 2019;44:5488–98.
- [12] Zivar D, Kumar S, Foroozesh J. Underground hydrogen storage: a comprehensive review. *Int J Hydrogen Energy* 2021;46:23436–62.
- [13] Tarasov BP, Arbuzov AA, Mozhzhuhin SA, Volodin AA, Fursikov PV, Lototskyy MV, Yartys VA. Hydrogen storage behavior of magnesium catalyzed by nickel-graphene nanocomposites. *Int J Hydrogen Energy* 2019;44:29212–23.
- [14] Pasquini L, Sakaki K, Akiba E, Allendorf MD, Alvares E, Ares JR, Babai D, Baricco M, Von Colbe JB, Berezniatsky M, et al. Magnesium-and intermetallic alloys-based hydrides for energy storage: modelling, synthesis and properties. *Progress in energy* 2022;4:032007.
- [15] Sáinz-García A, Abarca E, Rubí V, Grandia F. Assessment of feasible strategies for seasonal underground hydrogen storage in a saline aquifer. *Int J Hydrogen Energy* 2017;42:16657–66.
- [16] Pan B, Liu K, Ren B, Zhang M, Ju Y, Gu J, Zhang X, Clarkson CR, Edlmann K, Zhu W, et al. Impacts of relative permeability hysteresis, wettability, and injection/withdrawal schemes on underground hydrogen storage in saline aquifers. *Fuel* 2023;333:126516.
- [17] Bo Z, Boon M, Hajibeygi H, Hurter S. Impact of experimentally measured relative permeability hysteresis on

- reservoir-scale performance of underground hydrogen storage (uhs). *Int J Hydrogen Energy* 2023;48(36):13527–42.
- [18] Amid A, Mignard D, Wilkinson M. Seasonal storage of hydrogen in a depleted natural gas reservoir. *Int J Hydrogen Energy* 2016;41:5549–58.
- [19] Ozarslan A. Large-scale hydrogen energy storage in salt caverns. *Int J Hydrogen Energy* 2012;37:14265–77.
- [20] Caglayan DG, Weber N, Heinrichs HU, Linßen J, Robinius M, Kukla PA, Stolten D. Technical potential of salt caverns for hydrogen storage in europe. *Int J Hydrogen Energy* 2020;45:6793–805.
- [21] Dopffel N, Jansen S, Gerritse J. Microbial side effects of underground hydrogen storage—knowledge gaps, risks and opportunities for successful implementation. *Int J Hydrogen Energy* 2021;46:8594–606.
- [22] Chen Y, Niasar V, Ma L, Xie Q. Effect of salinity, mineralogy, and organic materials in hydrogen wetting and its implications for underground hydrogen storage (uhs). *Int J Hydrogen Energy* 2023. <https://doi.org/10.1016/j.ijhydene.2023.05.085>. URL: <https://www.sciencedirect.com/science/article/pii/S0360319923023376>.
- [23] Pfeiffer WT, Bauer S. Subsurface porous media hydrogen storage—scenario development and simulation. *Energy Proc* 2015;76:565–72.
- [24] Pfeiffer W, Al Hagrey S, Köhn D, Rabbel W, Bauer S. Porous media hydrogen storage at a synthetic, heterogeneous field site: numerical simulation of storage operation and geophysical monitoring. *Environ Earth Sci* 2016;75:1–18.
- [25] Feldmann F, Hagemann B, Ganzer L, Panfilov M. Numerical simulation of hydrodynamic and gas mixing processes in underground hydrogen storages. *Environ Earth Sci* 2016;75:1–15.
- [26] Hagemann B, Rasoulzadeh M, Panfilov M, Ganzer L, Reitenbach V. Mathematical modeling of unstable transport in underground hydrogen storage. *Environ Earth Sci* 2015;73:6891–8.
- [27] Hagemann B, Rasoulzadeh M, Panfilov M, Ganzer L, Reitenbach V. Hydrogenization of underground storage of natural gas. *Comput Geosci* 2016;20:595–606.
- [28] Rubin E, De Coninck H. *Ippc special report on carbon dioxide capture and storage*. vol. 2. UK: Cambridge University Press. TNO; 2004. p. 14. Cost Curves for CO<sub>2</sub> Storage, Part.
- [29] Joekar-Niasar V, Doster F, Armstrong R, Wildenschild D, Celia MA. Trapping and hysteresis in two-phase flow in porous media: a pore-network study. *Water Resour Res* 2013;49:4244–56.
- [30] Wang Y, Vuik C, Hajibeygi H. Analysis of hydrodynamic trapping interactions during full-cycle injection and migration of co<sub>2</sub> in deep saline aquifers. *Adv Water Resour* 2022;159:104073.
- [31] Juanes R, Spiteri E, Orr Jr F, Blunt M. Impact of relative permeability hysteresis on geological co<sub>2</sub> storage. *Water Resour Res* 2006;42.
- [32] Carlson FM. Simulation of relative permeability hysteresis to the nonwetting phase. In: *SPE annual technical conference and exhibition*. OnePetro; 1981.
- [33] Kleppe J, Delaplace P, Lenormand R, Hamon G, Chaput E. Representation of capillary pressure hysteresis in reservoir simulation. In: *SPE annual technical conference and exhibition*. OnePetro; 1997.
- [34] Tan T. Representation of hysteresis in capillary pressure for reservoir simulation models. *J Can Petrol Technol* 1990;29.
- [35] Land CS. Calculation of imbibition relative permeability for two-and three-phase flow from rock properties. *Soc Petrol Eng J* 1968;8:149–56.
- [36] Larsen J, Skauge A. Methodology for numerical simulation with cycle-dependent relative permeabilities. *SPE J* 1998;3:163–73.
- [37] Spiteri EJ, Juanes R, Blunt MJ, Orr FM. A new model of trapping and relative permeability hysteresis for all wettability characteristics. *SPE J* 2008;13:277–88.
- [38] Steffy D, Barry D, Johnston C. Influence of antecedent moisture content on residual lnapi saturation. *Soil Sediment Contam* 1997;6:113–47.
- [39] Mualem Y. A conceptual model of hysteresis. *Water Resour Res* 1974;10:514–20.
- [40] Aghabozorgi S, Sohrabi M. A comparative study of predictive models for imbibition relative permeability and trapped non-wetting phase saturation. *J Nat Gas Sci Eng* 2018;52:325–33.
- [41] Aghabozorgi S, Sohrabi M. Generalised model for simulation of two-and three-phase cycle-dependent hysteresis in sandstones. *Fuel* 2022;310:122328.
- [42] Akbarabadi M, Piri M. Relative permeability hysteresis and capillary trapping characteristics of supercritical co<sub>2</sub>/brine systems: an experimental study at reservoir conditions. *Adv Water Resour* 2013;52:190–206.
- [43] Zulfiqar B, Vogel H, Ding Y, Golmohammadi S, Küchler M, Reuter D, Geistlinger H. The impact of wettability and surface roughness on fluid displacement and capillary trapping in 2-d and 3-d porous media: 2. combined effect of wettability, surface roughness, and pore space structure on trapping efficiency in sand packs and micromodels. *Water Resour Res* 2020;56:e2020WR027965.
- [44] Spiteri E, Juanes R, Blunt MJ, Orr FM. Relative permeability hysteresis: trapping models and application to geological co<sub>2</sub> sequestration. In: *SPE annual technical conference and exhibition*. OnePetro; 2005.
- [45] Ide ST, Jessen K, Orr Jr FM. Storage of co<sub>2</sub> in saline aquifers: effects of gravity, viscous, and capillary forces on amount and timing of trapping. *Int J Greenh Gas Control* 2007;1:481–91.
- [46] Gershenzon NI, Ritzi Jr RW, Dominic DF, Mehnert E, Okwen RT. Capillary trapping of co<sub>2</sub> in heterogeneous reservoirs during the injection period. *Int J Greenh Gas Control* 2017;59:13–23.
- [47] Gershenzon NI, Soltanian M, Ritzi Jr RW, Dominic DF. Co<sub>2</sub> capillary-trapping processes in deep saline aquifers. In: *EGU general assembly conference abstracts*; 2014. p. 3935.
- [48] Gershenzon NI, Soltanian M, Ritzi Jr RW, Dominic DF. Influence of small scale heterogeneity on co<sub>2</sub> trapping processes in deep saline aquifers. *Energy Proc* 2014;59:166–73.
- [49] Gershenzon NI, Ritzi Jr RW, Dominic DF, Soltanian M, Mehnert E, Okwen RT. Influence of small-scale fluvial architecture on co<sub>2</sub> trapping processes in deep brine reservoirs. *Water Resour Res* 2015;51:8240–56.
- [50] Yang Z, Chen Y-F, Niemi A. Gas migration and residual trapping in bimodal heterogeneous media during geological storage of co<sub>2</sub>. *Adv Water Resour* 2020;142:103608.
- [51] Boon M, Hajibeygi H. Experimental characterization of h<sub>2</sub>/water multiphase flow in heterogeneous sandstone rock at the core scale relevant for underground hydrogen storage (uhs). *Sci Rep* 2022;12:14604.
- [52] Group CM. User's guide gem, advanced compositional reservoir simulator. Computer Modeling Group; 2020.
- [53] Lysy M, Fernø MA, Ersland G. Effect of relative permeability hysteresis on reservoir simulation of underground hydrogen storage in an offshore aquifer. *J Energy Storage* 2023;64:107229.
- [54] Lysy M, Føyen T, Johannesen EB, Fernø M, Ersland G. Hydrogen relative permeability hysteresis in underground storage. *Geophys Res Lett* 2022;49:e2022GL100364.
- [55] Lie K-A, Møyner O. Advanced modelling with the MATLAB reservoir simulation toolbox. Cambridge University Press; 2021.

- [56] Nazari F, Farajzadeh R, Niasar VJ. Critical parameters controlling wettability in hydrogen underground storage-an analytical study. *JCIS Open* 2022;8:100063.
- [57] Plug W-J, Bruining J. Capillary pressure for the sand-co<sub>2</sub>-water system under various pressure conditions. application to co<sub>2</sub> sequestration. *Adv Water Resour* 2007;30:2339–53.
- [58] Leverett M. Capillary behavior in porous solids. *Transactions of the AIME* 1941;142:152–69.
- [59] Brooks RH, Corey AT. Properties of porous media affecting fluid flow. *J Irrigat Drain Div* 1966;92:61–88.
- [60] Guo Y. Importance of capillary hysteresis on coreflood experiments. In: *IOR 1995-8th European symposium on improved oil recovery*. EAGE Publications BV; 1995. p. 107.
- [61] Debbabi Y, Jackson MD, Hampson GJ, Salinas P. Impact of the buoyancy-viscous force balance on two-phase flow in layered porous media. *Transport Porous Media* 2018;124:263–87.
- [62] Shook M, Li D, Lake LW. Scaling immiscible flow through permeable media by inspectional analysis, *In Situ*;(United States)vol. 16; 1992.
- [63] Francke H, Thorade M. Density and viscosity of brine: an overview from a process engineers perspective. *Geochemistry* 2010;70:23–32.
- [64] Hilsenrath J, Touloukian Y. The viscosity, thermal conductivity, and Prandtl number for air, o<sub>2</sub>, n<sub>2</sub>, no, h<sub>2</sub>, co, co<sub>2</sub>, h<sub>2</sub>o, he, and a. *Trans Am Soc Mech Eng* 1954;76:967–83.
- [65] Sari A, Al Maskari NS, Saeedi A, Xie Q. Impact of surface roughness on wettability of oil-brine-calcite system at sub-pore scale. *J Mol Liq* 2020;299:112107.
- [66] Chow YF, Maitland GC, Trusler JM. Interfacial tensions of the (co<sub>2</sub>+ n<sub>2</sub>+ h<sub>2</sub>o) system at temperatures of (298 to 448) k and pressures up to 40 mpa. *J Chem Therm* 2016;93:392–403.
- [67] Lenhard R, Parker J, Mishra S. On the correspondence between brooks-corey and van genuchten models. *J Irrigat Drain Eng* 1989;115:744–51.
- [68] Kashif M, Cao Y, Yuan G, Asif M, Javed K, Mendez JN, Khan D, Miruo L. Pore size distribution, their geometry and connectivity in deeply buried paleogene es1 sandstone reservoir, nanpu sag, east China. *Petrol Sci* 2019;16:981–1000.