
Research article

Permeability evolution of compacted montmorillonite under cyclic hydrogen loading: Insights from coupled transport and microstructural analysis

Walid Oueslati^{1,2,*}

¹ LR19ES20: Ressources, Materials and Ecosystem (RME), Faculty of Sciences of Bizerte, University of Carthage, Bizerte, 7021, Tunisia

² Physics Department, Faculty of Sciences of Bizerte, University of Carthage, Bizerte, 7021, Tunisia

* **Correspondence:** Email: walidoueslati@ymail.com; walid.oueslati@fsb.ucar.tn; Tel: +216-22-144-337.

Abstract: Underground hydrogen storage (UHS) is increasingly considered a key component of future energy systems, yet its long-term safety critically depends on the integrity of geological sealing formations subjected to cyclic pressure variations. In this study, the response of compacted Na-SWy-2 montmorillonite to cyclic hydrogen loading was investigated using a multi-scale experimental approach combining permeability measurements with X-ray diffraction (XRD), mercury intrusion porosimetry (MIP), and scanning electron microscopy (SEM). The results showed a progressive increase in apparent hydrogen permeability with cycle number, with permeability increasing by a factor of approximately 2–3 after ten cycles. XRD analyses indicated that the interlayer spacing remained stable, suggesting preservation of the clay crystal structure. In contrast, MIP and SEM observations were consistent with an increase in macroporosity and modifications of pore connectivity at the aggregate scale. The observed permeability evolution was interpreted as resulting from coupled hydro-mechanical processes associated with cyclic loading. However, contributions from additional mechanisms, such as partial desaturation and pore-water redistribution, cannot be fully excluded under the experimental conditions. These findings provide mechanistic insight into a potential degradation mechanism affecting the sealing behavior of compacted clay under cyclic hydrogen loading. The observed increase in permeability, together with the development of macroporosity and enhanced pore connectivity, suggests a progressive reduction in sealing efficiency under the investigated conditions.

Because the experimental conditions differ substantially from field-scale environments, the results should be interpreted as evidence of early-stage hydro-mechanical evolution rather than as an assessment of long-term caprock suitability, engineering reliability, or field-scale performance.

Keywords: underground hydrogen storage (UHS); cyclic pressure loading; compacted montmorillonite; gas permeability evolution; fabric-controlled transport; caprock integrity

1. Introduction

1.1. Hydrogen as a key vector in the energy transition

The large-scale integration of renewable energy sources, such as wind and solar power, requires efficient and flexible energy storage solutions capable of compensating for their intrinsic intermittency [1]. In this context, hydrogen (H₂) has emerged as a key energy vector due to its high gravimetric energy density and its potential to enable seasonal-scale energy storage [2]. Unlike electrochemical batteries, which are limited in capacity and storage duration, underground hydrogen storage (UHS) [3] provides a viable pathway for storing substantial quantities of energy over long periods using geological formations. Subsurface storage concepts, including salt caverns, depleted hydrocarbon reservoirs, and deep saline aquifers, are being reconsidered for hydrogen storage. However, the successful deployment of UHS requires a thorough understanding of the interactions between hydrogen and the geological barriers responsible for long-term containment [4].

1.2. Containment challenges: Hydrogen leakage risk

One of the principal challenges associated with underground hydrogen storage is the risk of gas leakage [5]. Hydrogen molecules are significantly smaller and lighter than methane or carbon dioxide, resulting in higher mobility and diffusivity in porous media. Additionally, hydrogen exhibits very low viscosity, which further enhances its ability to migrate through pore networks and microfractures [3–5]. From an operational perspective, UHS systems are subjected to repeated injection and withdrawal cycles, leading to cyclic variations in pore pressure within both the storage reservoir and the overlying caprock [6–8]. These pressure fluctuations may induce mechanical and microstructural changes in low-permeability sealing formations, potentially compromising their long-term sealing efficiency. Understanding how caprock materials respond to such cyclic hydrogen loading is therefore a critical prerequisite for safe and reliable UHS operations [9,10].

1.3. Clay barriers as caprocks: The role of smectites

Clay-rich formations are widely recognized as effective natural barriers against fluid and gas migration due to their extremely low intrinsic permeability, high capillary entry pressure, and capacity for self-sealing. Among clay minerals, smectites, and particularly montmorillonite, play a central role in controlling these properties [11–13].

Montmorillonite-rich materials exhibit a layered crystal structure with expandable interlayers, enabling them to accommodate water molecules and exchangeable cations. These characteristics impart strong swelling behavior and the ability to close fractures or pores generated by mechanical stress. As a result, compacted bentonites are commonly considered the ultimate sealing materials in a variety of subsurface applications, including nuclear waste repositories and geological storage systems. Specifically, recent structural and XRD modeling studies have demonstrated that mechanical constraints and thermo-hydraulic gradients directly control montmorillonite hydration states, swelling capability, and micro-porosity evolution [14–17]. Furthermore, coupled stress-environmental factors have been shown to dictate the long-term ion-exchange performance, contaminant removal efficiency, and structural stability of smectites in containment barriers [18–20]. The reference material SWy-2 montmorillonite, supplied by the Clay Minerals Society, is a well-characterized sodium montmorillonite frequently used as a model material in experimental studies. Despite extensive research on its behavior in aqueous environments [20–22], the response of compacted SWy-2 to cyclic exposure to hydrogen gas remains poorly constrained [23].

1.4. Knowledge gap: Effects of cyclic hydrogen pressure on clay microstructure

To date, most experimental and numerical investigations of gas–clay interactions have entailed carbon dioxide, CO₂, or methane, CH₄, primarily in the context of geological carbon storage or natural gas reservoirs [23–26]. In these systems, chemical reactions, adsorption effects, and multiphase flow mechanisms have been extensively documented.

In contrast, hydrogen is a chemically inert and typically dry gas in subsurface storage scenarios, suggesting fundamentally different interaction mechanisms with clay minerals. In particular, the impact of cyclic hydrogen pressure on the microstructural stability of compacted clay barriers has received very limited attention [27,28]. Repeated pressurization and depressurization may induce mechanical fatigue of the clay skeleton, promote pore network rearrangement, or lead to partial interlayer dehydration, all of which could influence gas transport properties.

In practice, underground hydrogen storage operations are typically characterized by seasonal or moderate cycling regimes, often corresponding to a limited number of cycles per year depending on operational strategies. However, over long operational lifetimes, storage systems may be subjected to repeated pressure variations. Investigating multiple cycles at the laboratory scale therefore provides valuable insight into the early-stage mechanisms governing the response of clay barriers to cyclic loading.

However, the mechanisms controlling permeability evolution under cyclic gas loading remain complex and may involve coupled hydro-mechanical processes, including desaturation effects, suction variations, and pore-water redistribution. These processes are difficult to isolate experimentally and must be considered when interpreting gas transport behavior in partially saturated clays. In addition to clay-rich caprocks, significant attention has been devoted to hydrogen storage in salt formations, particularly in salt caverns [29–32]. These systems benefit from the ductile and self-healing properties of rock salt, which contribute to maintaining cavern integrity under cyclic injection and withdrawal operations.

Researchers [33,34] have investigated the feasibility of underground hydrogen storage in highly impure rock salt formations, as well as the mechanical stability of salt caverns subjected to high-frequency pressure cycling. These works highlight the importance of cyclic loading effects on storage system integrity, including potential fatigue, deformation, and permeability evolution. Although I focus on clay-rich materials, both storage systems share a common challenge related to

repeated pressure fluctuations. Therefore, understanding the response of clay barriers under cyclic hydrogen loading provides complementary insights into the broader assessment of underground hydrogen storage reliability.

While researchers have investigated gas transport in clays for CO₂ and methane storage, experimental investigations addressing cyclic hydrogen loading in compacted clay materials remain limited. In particular, the coupling between cyclic pressure variations, permeability evolution, and microstructural changes has not been extensively documented.

1.5. Objectives of the study

My primary objective of this study is to provide mechanistic insight into the evolution of gas transport properties in compacted montmorillonite SWy-2 subjected to cyclic hydrogen loading. Attention is given to the evolution of transport properties under repeated injection and withdrawal conditions representative of underground hydrogen storage operations. To achieve this objective, I the progressive evolution of hydrogen permeability and gas breakthrough pressure during successive pressure cycles, enabling the cumulative effects of cyclic loading to be quantified. In parallel, post-test microstructural changes are systematically characterized using a multi-scale analytical approach. Variations in the basal spacing d_{001} are assessed by XRD analysis to evaluate potential interlayer modifications, while changes in pore size distribution are investigated by mercury intrusion porosimetry to detect the development of larger transport pathways. Scanning electron microscopy is further employed to document fabric-scale reorganization and the formation of localized microstructural features associated with cyclic mechanical perturbation. On the basis of these combined observations, I seek to identify the dominant mechanisms governing hydrogen migration in compacted montmorillonite, with particular emphasis on distinguishing advective transport through evolving pore networks from diffusion-controlled processes at the interlayer scale.

Finally, the implications of the observed behavior are discussed in terms of possible permeability degradation mechanisms and their consequences for the sealing performance of smectite-rich clay barriers subjected to cyclic hydrogen loading.

Overall, the results provide new insight into the early-stage evolution of transport properties in compacted clay subjected to cyclic hydrogen loading and contribute to the identification of potential degradation mechanisms affecting sealing efficiency under repeated pressure variations.

2. Materials and methods

2.1. Material characterization

The clay material investigated in this study was SWy-2 montmorillonite, a sodium-rich smectite reference material supplied by the Clay Minerals Society. SWy-2 is derived from Wyoming bentonite and is extensively documented in the literature [14–22] as a representative analogue for smectite-dominated caprocks and engineered clay barriers. Its widespread use enables direct comparison with other studies related to geological storage and sealing performance.

Prior to sample preparation, the mineralogical composition of the raw material was verified by powder X-ray diffraction under ambient conditions to confirm the predominance of montmorillonite and the absence of significant secondary mineral phases. The initial basal spacing d_{001} was determined

to establish a reference crystallographic state before compaction and hydrogen exposure. The physico-chemical properties of SWy-2, including cation exchange capacity, specific surface area, and grain density, are consistent with values reported in the Clay Minerals Society data repository and previous experimental studies [35–37]. These intrinsic properties directly control interlayer behavior, adsorption potential, and fluid transport mechanisms, and therefore constitute key parameters for interpreting gas–clay interactions under cyclic loading.

2.2. Sample preparation

The SWy-2 powder was first oven-dried at 60°C for 48 hours to remove free and weakly bound water while preserving the clay’s structural integrity. After cooling in a desiccator, deionized water was gradually added to reach a target gravimetric water content representative of partially hydrated clay under subsurface conditions. Based on the initial water content ($w = 15\%$) and the measured dry density of the compacted specimens, the initial degree of saturation was estimated assuming a grain density of 2.7 g cm^{-3} . This resulted in an initial degree of saturation in the range of approximately 60–70%, indicating partially saturated conditions consistent with gas permeability testing. The moistened powder was sealed and stored for a minimum of seven days to enable moisture homogenization throughout the material [38–42]. However, no direct measurements of suction or water content evolution were performed after sample installation. Therefore, potential changes in degree of saturation during gas injection and cycling were not quantified. Compacted specimens were prepared using uniaxial static compaction in a stainless steel mold. The compaction procedure was carefully controlled to achieve dry densities representative of highly compacted clay barriers encountered in caprock formations or engineered sealing systems. The applied stress was increased gradually to minimize fabric disturbance and ensure reproducibility between samples. Cylindrical specimens with a diameter of 38 mm and a height between 10 and 15 mm were obtained, providing a geometry well suited for gas flow testing in an oedometer configuration. Once compacted, samples were immediately sealed to prevent moisture redistribution or desiccation prior to installation in the testing cell (Figure 1).

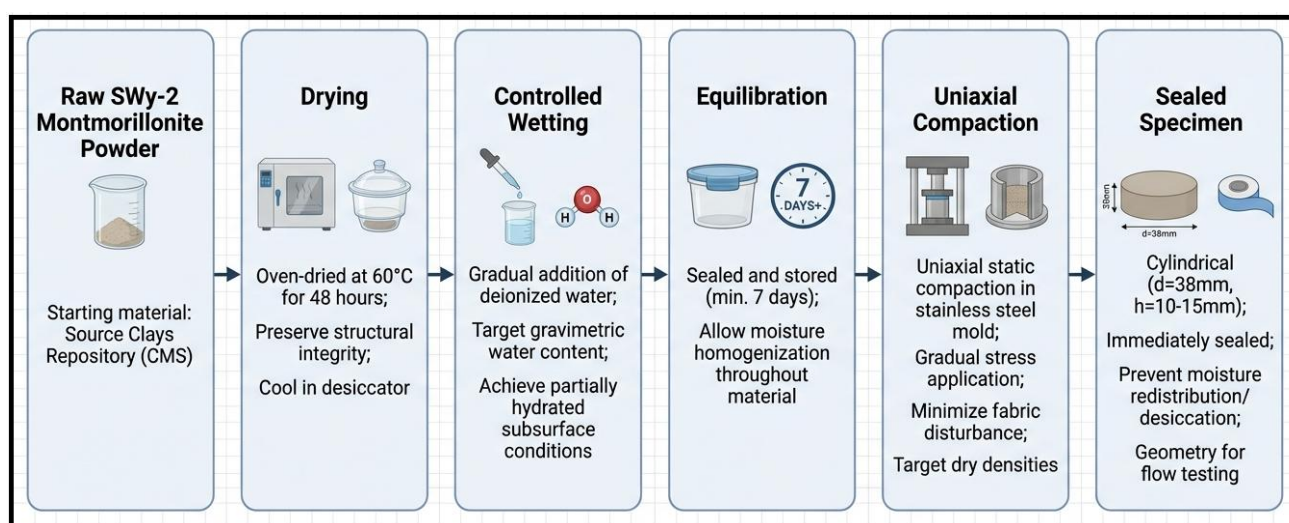


Figure 1. Experimental workflow.

2.3. High-pressure oedometer cell for hydrogen testing

Gas transport experiments were conducted using a high-pressure oedometer-type cell designed for hydrogen permeability testing. The oedometer configuration was selected because it ensures zero lateral strain conditions ($\epsilon_r = 0$), forcing one-dimensional vertical deformation, which closely represents the mechanical lateral restraint of clay barriers in deep geological settings [43]. The compacted clay specimen was placed between two rigid porous plates that ensured uniform gas distribution at the inlet and outlet while preventing particle migration. Axial stress was applied through a rigid piston and maintained constant throughout the test, enabling controlled simulation of in situ vertical stress conditions [44–46]. All components in direct contact with hydrogen were manufactured from hydrogen-compatible stainless-steel alloys to avoid embrittlement or gas contamination. The oedometer cell was equipped with high-precision pressure transducers at the inlet and outlet, enabling accurate monitoring of hydrogen pressure gradients across the specimen. Axial displacement was continuously measured using a linear variable differential transformer (LVDT), enabling detection of any volumetric or structural changes during cyclic pressurization. Hydrogen flow rates were measured at the outlet using a gas flow sensor calibrated for very low flow conditions. It should be noted that the mechanical boundary conditions corresponded to a one-dimensional strain state under controlled axial stress, meaning that confinement was provided passively by the rigid oedometer ring rather than through active fluid confining pressure (σ_3). As a result, the stress path differed from isotropic triaxial conditions encountered in some deep geological formations. A photograph of the experimental setup used for cyclic hydrogen permeability testing is shown in Figure 2.

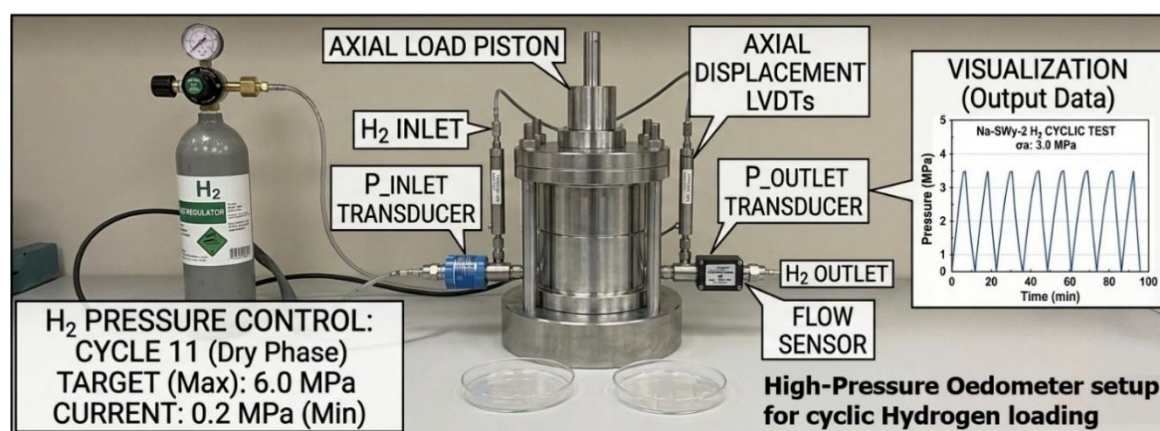


Figure 2. High-pressure oedometer setup for cyclic hydrogen permeability testing. A compacted Na-SWy-2 montmorillonite specimen is subjected to constant axial loading while hydrogen gas is injected under cyclic pressure (0.2–6 MPa). Pressure transducers, a flow sensor, and displacement sensors (LVDTs) enable simultaneous measurement of pressure gradients, gas flow, and axial deformation.

The experiments were conducted under rigid lateral restraint, as the oedometer ring prevents radial expansion during axial loading. Therefore, the stress state represents one-dimensional mechanical confinement (zero lateral strain) rather than isotropic fluid-confining triaxial conditions typically encountered in geological formations.

2.4. Cyclic hydrogen pressure loading protocol

After installation in the odometer cell, a low initial axial stress was applied to ensure mechanical contact and specimen stability. Hydrogen gas with high purity was then introduced following a cyclic pressure protocol designed to simulate realistic underground hydrogen storage operations. Each test consisted of repeated injection–withdrawal cycles, during which hydrogen pressure was progressively increased to a predefined maximum value representative of storage pressures and subsequently decreased to a minimum value corresponding to operational withdrawal conditions [47] (Figure 3). Hydrogen gas was injected under controlled cyclic pressure conditions ranging from 0.2 MPa to 6.0 MPa, corresponding to the pressure range investigated in this study. Pressurization and depressurization were applied at a constant rate of approximately $0.05 \text{ MPa min}^{-1}$ to ensure quasi-static loading conditions. At each maximum pressure level, a stabilization period of 12 hours was applied prior to permeability measurement to ensure steady-state gas flow through the specimen. During testing, the maximum gas pressure (up to 6 MPa) exceeded the applied axial stress (3–5 MPa), leading to a non-conventional stress path. Under such conditions, the effective stress could not be rigorously defined using classical formulations, particularly due to partial saturation and the absence of pore water pressure measurements. Pressurization and depressurization rates were kept sufficiently low to avoid transient or dynamic effects, ensuring quasi-static conditions [47]. At each maximum pressure level, a holding period was imposed to allow the system to reach steady-state flow conditions. This cyclic loading protocol was repeated multiple times on the same specimen to capture cumulative effects associated with pressure fluctuations, such as microstructural fatigue, pore network rearrangement, or changes in effective gas transport pathways.

2.5. Hydrogen Permeability and Breakthrough Pressure Determination

Breakthrough pressure and permeability measurements were performed using the same experimental setup. Breakthrough pressure was determined during the pressurization phase when gas flow was first detected at the outlet, while permeability measurements were subsequently carried out under steady-state flow conditions within the same testing configuration. Hydrogen permeability was determined using a steady-state gas flow approach once continuous hydrogen flow was detected at the outlet of the specimen. The gas breakthrough pressure was defined as the inlet pressure at which measurable and stable hydrogen flow was first recorded, indicating the establishment of a connected gas pathway across the compacted clay. During the first loading cycle, breakthrough corresponded to the initial formation of a connected gas pathway across the specimen. For subsequent cycles, breakthrough pressure was interpreted relative to previously established pathways, which may remain partially open after unloading. As a result, the observed decrease in breakthrough pressure with increasing cycle number reflected a progressive facilitation of gas transport rather than the repeated formation of new pathways (Table 1).

Hydrogen permeability was determined under steady-state conditions using a formulation of Darcy's law for compressible gas flow: $K_{\text{eff}} = (Q \cdot \mu \cdot L) / (A \cdot (P_{\text{in}}^2 - P_{\text{out}}^2))$, where Q is the volumetric flow rate measured at the outlet, μ is the dynamic viscosity of hydrogen, L is the specimen thickness, A is the cross-sectional area, and P_{in} and P_{out} are the inlet and outlet gas pressures, respectively. Steady-state conditions were assumed when flow rates stabilized for a given pressure level.

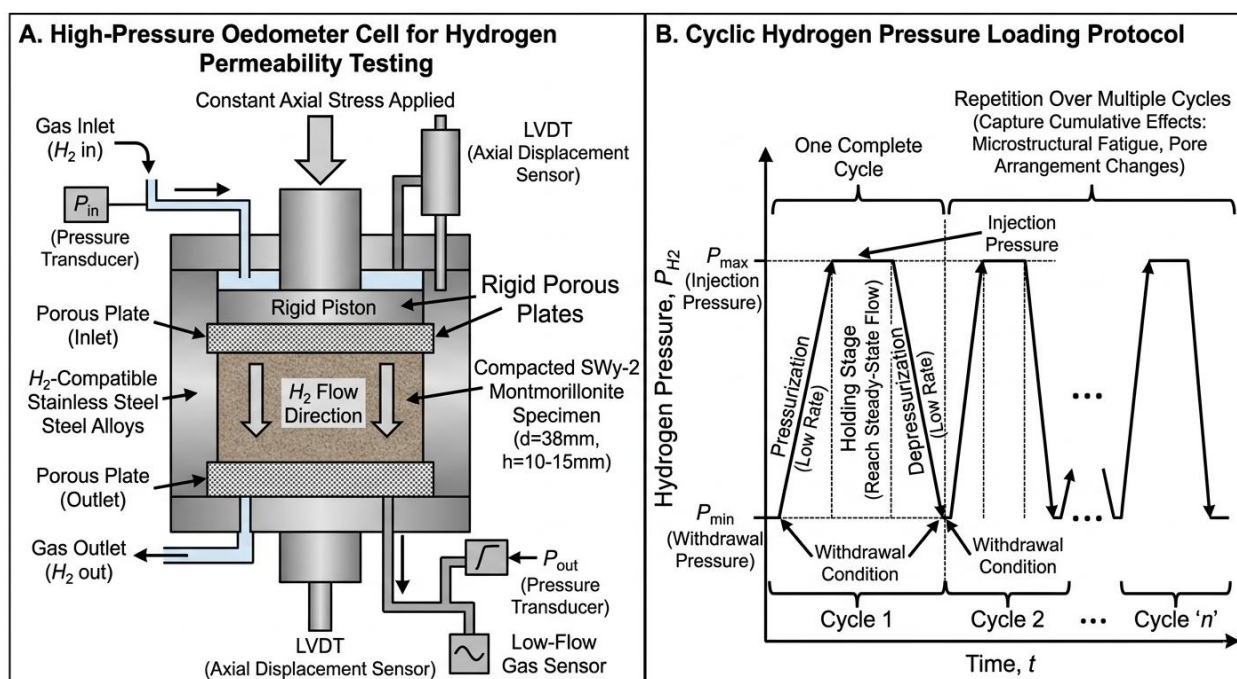


Figure 3. High-Pressure Odometer Apparatus and Cyclic Loading Protocol (Schematic representation of the experimental setup and loading protocol). (A) provides a detailed cross-sectional schematic of the high-pressure oedometer cell, including the sample dimensions ($d = 38$ mm, $h = 10 - 15$ mm), H₂-compatible stainless-steel alloys, and all requested instrumentation. (B) illustrates the cyclic loading protocol, showing how multiple injection-withdrawal cycles are repeated to capture cumulative effects such as microstructural fatigue.

The compacted specimens had a nominal diameter of 38 mm and an average thickness of 12 mm. The corresponding cross-sectional area used in the permeability calculations was:

$$A = \pi(d/2)^2 = 1.134 \times 10^{-3} \text{ m}^2, \text{ where } d \text{ is the specimen diameter.}$$

For transparency and reproducibility purposes, the parameters used for the apparent permeability calculations are reported in Table 2. The calculated permeability values therefore correspond to apparent gas permeability under the imposed experimental conditions [48–51]. Permeability measurements were systematically performed after each pressure cycle, enabling the evolution of transport properties to be assessed as a function of cyclic loading history. Changes in permeability and breakthrough pressure were interpreted as indicators of either mechanical degradation or preservation of the sealing capacity of the compacted montmorillonite (Table 1). No correction for gas slippage (Klinkenberg effect) was applied in this study, and potential leakage or non-ideal gas effects were not explicitly corrected. As a result, the reported values correspond to apparent gas permeability rather than intrinsic permeability, representative of the specific experimental conditions. This may lead to a slight overestimation of absolute permeability values, particularly at low gas pressures and in fine-grained porous materials such as compacted clays. However, these effects do not affect the relative evolution of permeability with cycle number, which remains my primary focus of this study.

Table 1. Summary of test conditions and evolution of hydrogen breakthrough pressure (Pb) and effective gas permeability (Keff) for compacted SWy-2 montmorillonite subjected to cyclic hydrogen pressure loading under constant axial stress.

Specimen ID	ρ_d (g/cm ³)	w (%)	σ_a (MPa)	Cycle Number (N)	Pb (MPa)	K _{eff} (m ²)
SWy2-01	1.60	15.0	3.0	1	4.00	1.50×10^{-20}
				3	3.85	1.95×10^{-20}
				5	3.70	2.80×10^{-20}
				10	3.40	4.50×10^{-20}
SWy2-02	1.70	15.0	5.0	1	5.50	8.00×10^{-21}
				3	5.35	9.80×10^{-21}
				5	5.15	1.25×10^{-20}
				10	4.80	2.10×10^{-20}

Table 2. Parameters used for apparent permeability calculations and corresponding flow rates reconstructed from the permeability equation.

Specimen	Cycle	P _{in} (MPa)	P _{out} (MPa)	L (mm)	Diameter (mm)	A (m ²)	Q ($\times 10^{-12}$ m ³ /s)	K _{eff} (m ²)
SWy2-01	1	4.00	0.10	12	38	1.134×10^{-3}	0.75	1.50×10^{-20}
SWy2-01	3	3.85	0.10	12	38	1.134×10^{-3}	0.92	1.95×10^{-20}
SWy2-01	5	3.70	0.10	12	38	1.134×10^{-3}	1.25	2.80×10^{-20}
SWy2-01	10	3.40	0.10	12	38	1.134×10^{-3}	1.82	4.50×10^{-20}
SWy2-02	1	5.50	0.10	12	38	1.134×10^{-3}	0.62	8.00×10^{-21}
SWy2-02	3	5.35	0.10	12	38	1.134×10^{-3}	0.75	9.80×10^{-21}
SWy2-02	5	5.15	0.10	12	38	1.134×10^{-3}	0.95	1.25×10^{-20}
SWy2-02	10	4.80	0.10	12	38	1.134×10^{-3}	1.50	2.10×10^{-20}

The outlet flow rates reported in Table 2 were reconstructed from the permeability equation using the measured permeability values, specimen geometry, and pressure conditions. Because the original flow-rate recordings were not archived in a format suitable for direct reporting, reconstructed flow rates are provided solely to improve transparency and reproducibility of the permeability calculations. These values should therefore be regarded as derived quantities rather than directly reported experimental measurements.

The breakthrough pressure (Pb) is defined as the inlet pressure at which a continuous and stable gas flow is first detected at the outlet. During the first loading cycle, this corresponds to the initial formation of a connected gas pathway through the specimen.

For subsequent cycles, breakthrough pressure is interpreted relative to the previously established

pathways, which may remain partially open. As a result, the decrease in Pb with cycle number reflects the progressive facilitation of gas transport rather than the repeated formation of new pathways.

2.6. Post-test microstructural characterization

The experimental program involved two cyclically loaded specimens subjected to different compaction and confinement conditions. A summary of the specimens and the characterization procedures performed after testing is provided in Table 3.

Table 3. Experimental specimen matrix and post-test characterization program.

Specimen ID	SWy2-01	SWy2-02
Dry density (g/cm ³)	1.60	1.70
Water content (%)	15	15
Diameter (mm)	38	38
Thickness (mm)	12	12
Hydrogen cycles	10	10
Permeability	Yes	Yes
XRD	Yes	Yes
MIP	Yes	Yes
SEM	Yes	Yes
Purpose	Cyclic hydrogen test	Cyclic hydrogen test

Following permeability testing, each specimen was subdivided for post-test XRD, MIP, and SEM analyses. The initial (pre-test) state used for comparison corresponds to measurements obtained on the material prior to cyclic hydrogen loading.

At the conclusion of the cyclic hydrogen tests, specimens were carefully extracted from the odometer cell and preserved to minimize any alteration prior to analysis. Prior to microstructural characterization, specimens were subjected to drying and handling procedures required for MIP and SEM analyses. It should be noted that such preparation steps, including drying, cutting, and exposure to vacuum (in the case of SEM), may induce microstructural alterations such as shrinkage, microcracking, or modification of pore geometry. Therefore, the observed microstructural features may not fully represent the in situ state of the material under hydrogen loading conditions.

Subsequent microstructural investigations were performed to identify the structural changes induced by hydrogen exposure and cyclic loading using three complementary techniques:

- X-ray Diffraction (XRD): Analyzed to detect variations in the basal spacing (d_{001}) of montmorillonite layers, providing insights into possible interlayer dehydration or rehydration processes.
- Mercury Intrusion Porosimetry (MIP): Utilized to quantify changes in the pore size distribution and to identify the potential development of larger transport pathways [45–48].
- Scanning Electron Microscopy (SEM): Employed to provide qualitative observations of clay fabric reorganization, pore connectivity, and microcrack formation (Figure 4).

Finally, all post-test results were compared with the initial state of the material obtained prior to

cyclic hydrogen loading, allowing qualitative assessment of the microstructural changes associated with cyclic pressurization.

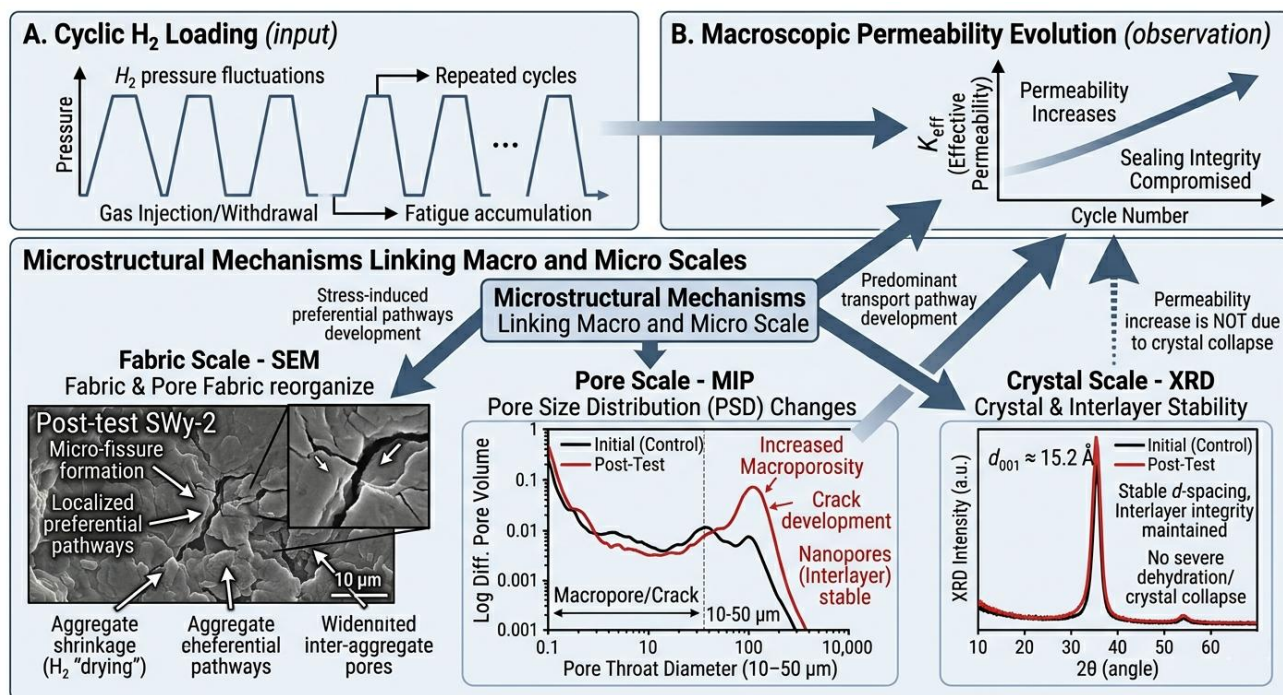


Figure 4. Schematic overview of the analytical framework adopted in this study. The figure summarizes the experimental techniques (permeability measurements, XRD, MIP, and SEM) and the scales of observation investigated. It is intended solely as a graphical summary of the experimental workflow and does not represent experimental data.

3. Results

3.1. Evolution of effective hydrogen permeability under cyclic pressure loading

Figure 5 presents the evolution of the effective hydrogen permeability, K_{eff} , of compacted Na-SWy-2 montmorillonite as a function of the number of cyclic hydrogen pressure loadings under constant axial stress conditions. The results show a systematic and progressive modification of gas transport properties with increasing cycle number for all tested conditions.

For all tested conditions, hydrogen permeability remains within the typical range of very low permeability values expected for highly compacted smectite-rich materials, with K_{eff} initially on the order of 10^{-21} – 10^{-20} m². However, a monotonic increase in permeability is observed as cyclic loading proceeds, with no evidence of stabilization or recovery during unloading phases. This continuous evolution indicates that permeability changes accumulate progressively with repeated pressurization rather than resulting from isolated or transient events [43–51]. For the clay compacted at a dry density of approximately 1.60 g cm⁻³ and subjected to an axial stress of 3 MPa, K_{eff} increases from about 1.5×10^{-20} m² during the first cycle to approximately 4.5×10^{-20} m² after ten cycles, corresponding to a threefold increase relative to the initial state. In contrast, for the clay compacted at a higher dry density (≈ 1.70 g cm⁻³) and subjected to a higher axial stress of 5 MPa, absolute

permeability values remain systematically lower throughout the test. Under these conditions, K_{eff} increases from roughly $8.0 \times 10^{-21} \text{ m}^2$ to $2.1 \times 10^{-20} \text{ m}^2$ between the first and tenth cycles, representing an increase by a factor of approximately 2.6.

Although absolute permeability values depend on the compaction level and applied axial stress, the normalized permeability evolution (Figure 5b) reveals comparable relative trends for both confinement conditions. In both cases, the increase in K_{eff} follows a gradual and cumulative progression with cycle number, without any abrupt change that would indicate macroscopic damage or failure of the compacted clay. This behavior indicates that cyclic loading induces incremental modifications to gas transport pathways rather than triggering sudden permeability breakthroughs.

The influence of dry density and axial stress is clearly reflected in the magnitude of permeability, with higher confinement conditions leading to lower K_{eff} values at all stages of cyclic loading. Nevertheless, the persistence of a systematic permeability increase under both stress states indicates that cyclic hydrogen pressurization affects the transport properties of the clay over a wide range of mechanical confinement conditions.

Overall, the results demonstrate that cyclic hydrogen pressure loading leads to a progressive degradation of sealing efficiency at the macroscopic scale, expressed by a cumulative increase in effective gas permeability. Importantly, this evolution occurs without any abrupt transition or loss of mechanical stability, providing a quantitative basis for linking permeability changes to the microstructural observations presented in Section 3.2.

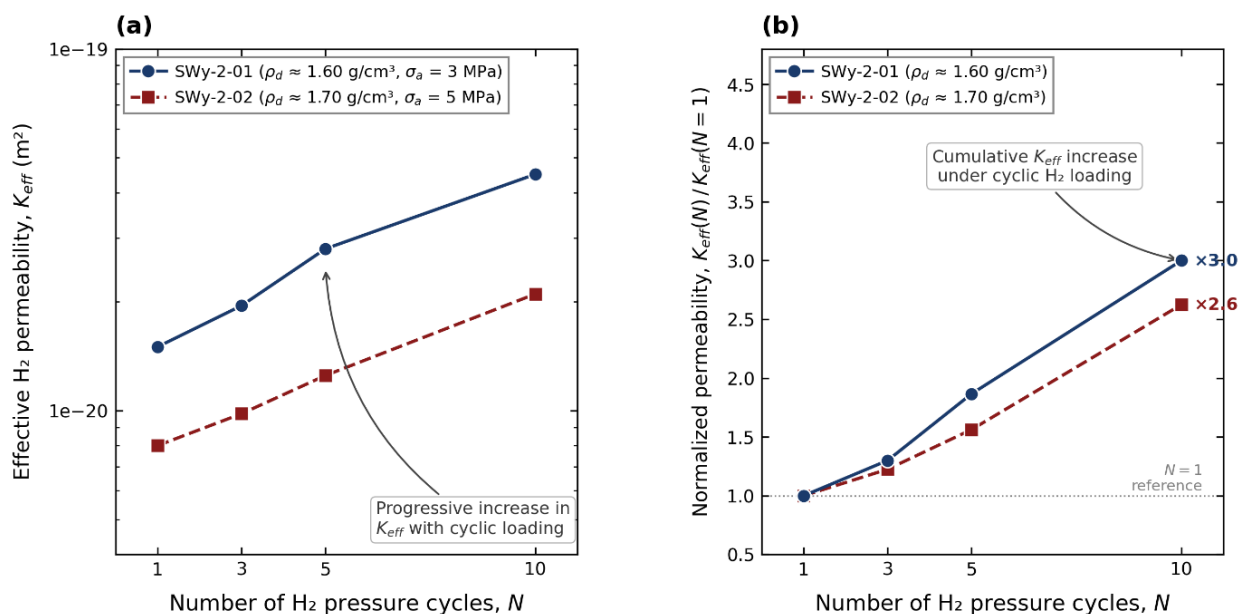


Figure 5. Evolution of effective hydrogen permeability and normalized permeability increase of compacted Na-SWy-2 montmorillonite under cyclic pressure loading.

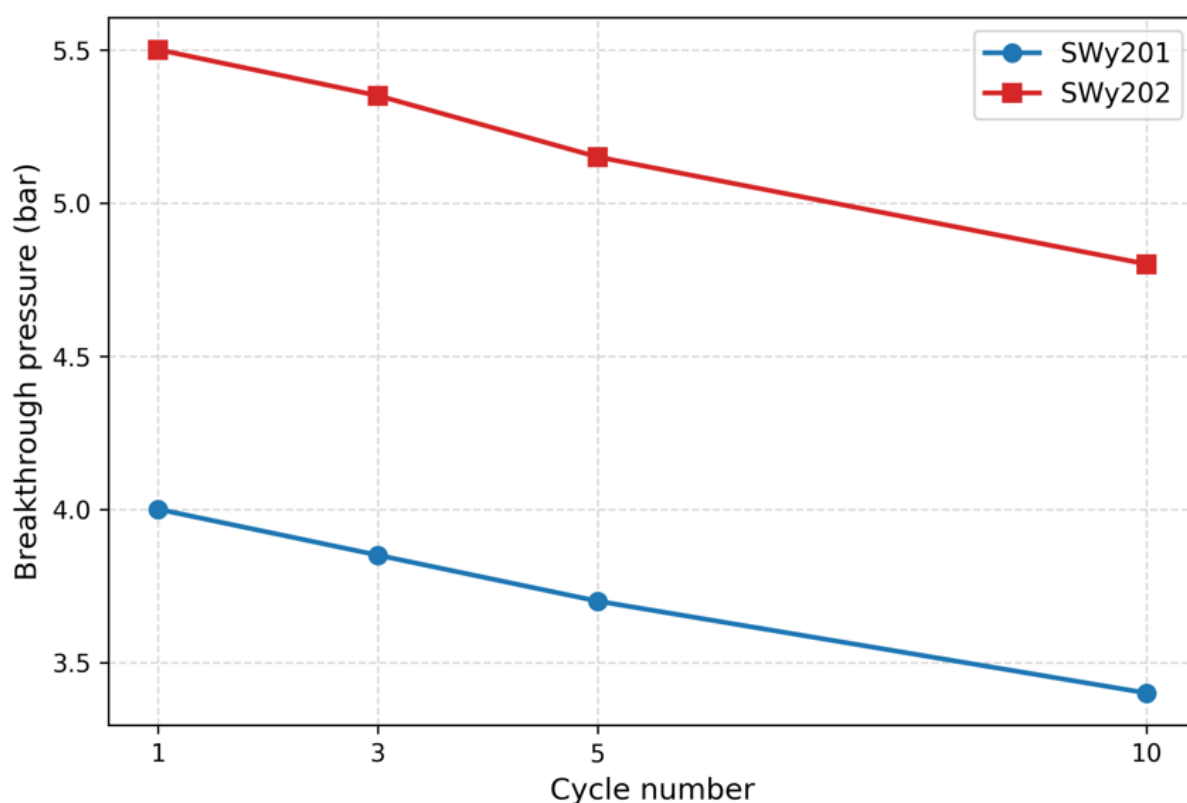


Figure 6. Evolution of hydrogen breakthrough pressure (P_b) as a function of cycle number for the two tested specimens. Breakthrough pressure decreases progressively with cyclic loading, indicating a reduction in resistance to gas migration and a progressive facilitation of transport through the clay barrier.

The evolution of hydrogen breakthrough pressure (P_b) during cyclic loading is shown in Figure 6. For both specimens, P_b decreases progressively with increasing cycle number, indicating that gas transport becomes progressively easier as loading history accumulates. This trend is consistent with the observed increase in permeability and the microstructural changes identified by MIP and SEM analyses.

3.2. Microstructural evolution after cyclic hydrogen loading

Post-test microstructural investigations were conducted to evaluate the effects of cyclic hydrogen pressure loading on the internal structure of compacted SWy-2 montmorillonite. Changes at the crystal, pore, and fabric scales were assessed using XRD analysis (Figure 7), mercury intrusion porosimetry (MIP), and scanning electron microscopy (SEM), respectively.

XRD patterns obtained before and after cyclic hydrogen exposure are characterized by a dominant basal ($00l$) reflection, indicative of the layered smectite structure. The (001) reflection is centered at approximately $7.1\text{--}7.2^\circ$ 2θ (Cu $K\alpha$), corresponding to an interlayer spacing of about $12.3\text{ \AA}$, which is characteristic of the 1W hydration state of Na-montmorillonite [15–20]. This basal spacing remains unchanged after ten cycles of dry hydrogen loading. Higher-order basal reflections are very weak and poorly resolved, consistent with turbostratic stacking, structural disorder, and the effects of compaction under confined conditions. No additional reflections or peak shifts are observed after testing, and only minor variations in peak intensity are detected. These results indicate

that cyclic exposure to hydrogen does not induce interlayer expansion, collapse, or crystallographic degradation of the montmorillonite structure.

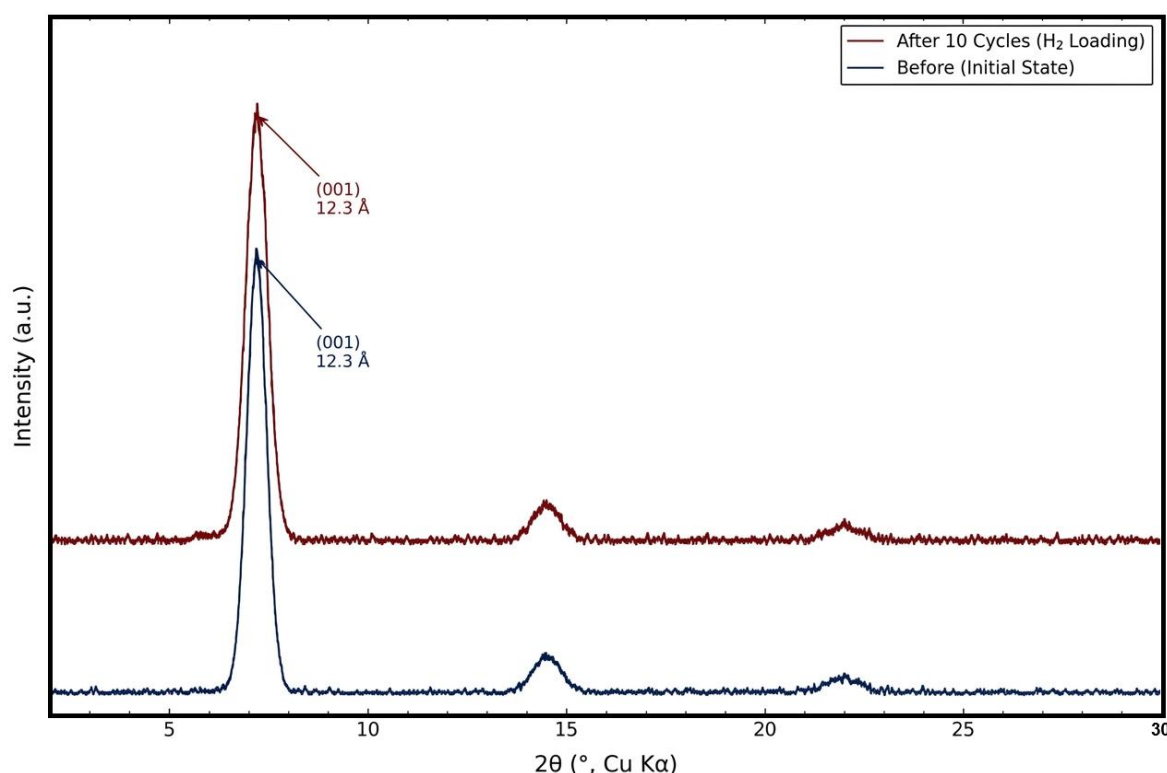


Figure 7. XRD patterns of compacted Na-SWy-2 montmorillonite before and after cyclic dry hydrogen loading, highlighting the stability of the basal (001) reflection corresponding to the 1W hydration state.

Mercury intrusion porosimetry (MIP) data reveal discernible modifications in the pore size distribution following cyclic hydrogen loading [52–54]. In its initial state, the pore system is dominated by micro- and mesopores, reflecting the dense fabric of the compacted clay. Following cyclic testing, a selective increase in pore volume is observed within the macropore size range, whereas the domain associated with finer pores remains largely unchanged. This evolution corresponds to a redistribution of the pore network toward larger pore throats without a concomitant loss of the smaller pore structures (Figure 8).

Nevertheless, it should be noted that MIP intrinsically measures accessible pore throat sizes rather than actual pore body diameters. Furthermore, these results must be interpreted with caution, as they depend on simplified assumptions regarding pore geometry (e.g., cylindrical pore models) and may be influenced by microstructural artifacts induced during the mandatory pre-test drying phase.

Scanning electron microscopy observations provide direct evidence of fabric-scale changes. The control specimen, corresponding to the initial compacted state of SWy-2, exhibits a dense and homogeneous fabric composed of closely packed plate-like aggregates with limited visible inter-aggregate voids. Pore spaces appear small and poorly connected, consistent with a low-permeability microstructure. In contrast, the specimen subjected to ten cycles of dry hydrogen loading exhibits localized microstructural alterations. At the macroscopic scale, these changes are visible as surface

cracking and minor structural degradation, as documented in the post-test photographs (Figure 9). However, it should be noted that part of the observed cracking may be influenced by drying and sample preparation effects, meaning these features must be interpreted as indicative of microstructural trends rather than definitive in situ damage.

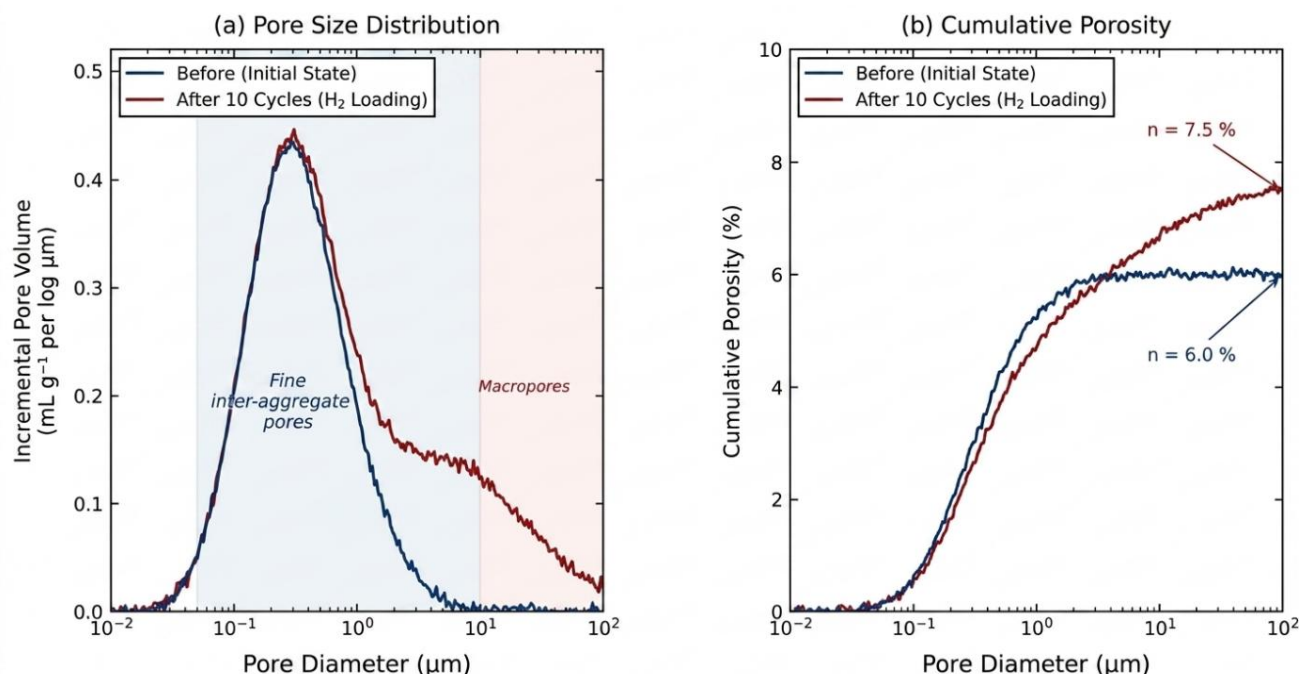


Figure 8. Mercury intrusion porosimetry (MIP) results for compacted Na-SWy-2 montmorillonite before and after ten cycles of dry hydrogen loading. (a) Incremental pore size distribution showing a limited contribution of fine inter-aggregate pores in the initial state and an increased contribution from larger pore throats after cyclic loading. Pore diameters correspond to accessible pore throats intruded by mercury. (b) Cumulative intruded porosity indicating a moderate increase from approximately 6.0 % to 7.5 % after hydrogen cycling.

SEM images show the development of micro-fissures and widened inter-aggregate pores, primarily along aggregate boundaries. Several preferential pathways are visible, characterized by elongated voids and discontinuities within the clay fabric. These features are spatially localized and do not form pervasive fracture networks. The clay aggregates remain intact, and no evidence of platelet collapse, grain fragmentation, or pervasive structural breakdown is observed. The characteristic size of the observed openings and fissures is on the order of a few micrometers, consistent with the increased macropore contribution identified by MIP measurements. Overall, the SEM observations confirm that cyclic hydrogen loading induces fabric reorganization and pore connectivity enhancement at the aggregate scale, without altering the fundamental layered structure of the montmorillonite (Figure 10).

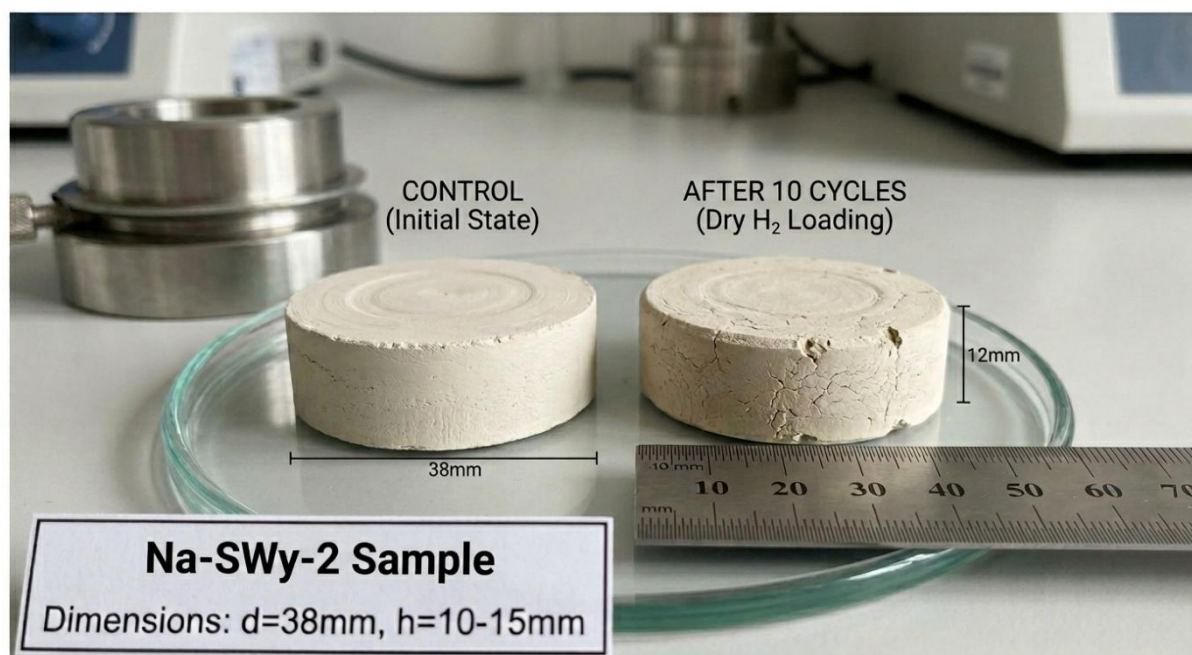


Figure 9. Photographs of compacted Na-SWy-2 montmorillonite specimens before and after cyclic hydrogen loading. The control specimen (left) corresponds to the initial compacted state and exhibits a dense and relatively smooth surface morphology. The specimen subjected to ten cycles of dry hydrogen loading (right) shows visible surface cracking and structural alteration at the macroscopic scale. Both samples have a diameter of approximately 38 mm and a height of about 10–15 mm. These observations are consistent with a modification of pore connectivity and fabric organization, in agreement with the observed increase in gas permeability. However, it should be noted that part of the observed cracking may be influenced by drying and sample preparation effects. Therefore, these features are interpreted as indicative of possible microstructural evolution rather than direct evidence of in situ processes occurring during hydrogen loading.

Taken together, the XRD, MIP, and SEM results demonstrate that cyclic exposure to dry hydrogen primarily affects the pore network and fabric organization of compacted SWy-2 montmorillonite, while preserving its crystallographic integrity and interlayer hydration state. These microstructural observations provide a direct experimental basis for the macroscopic evolution of hydrogen permeability described in Section 3.1.

It must be emphasized that the observed microstructural features are derived from post-test analyses under non in situ conditions. As a result, part of the observed fissure development and pore enlargement may be influenced by sample preparation procedures. Therefore, these observations should be interpreted as indicative of microstructural trends rather than as direct evidence of in situ processes occurring during cyclic hydrogen loading.

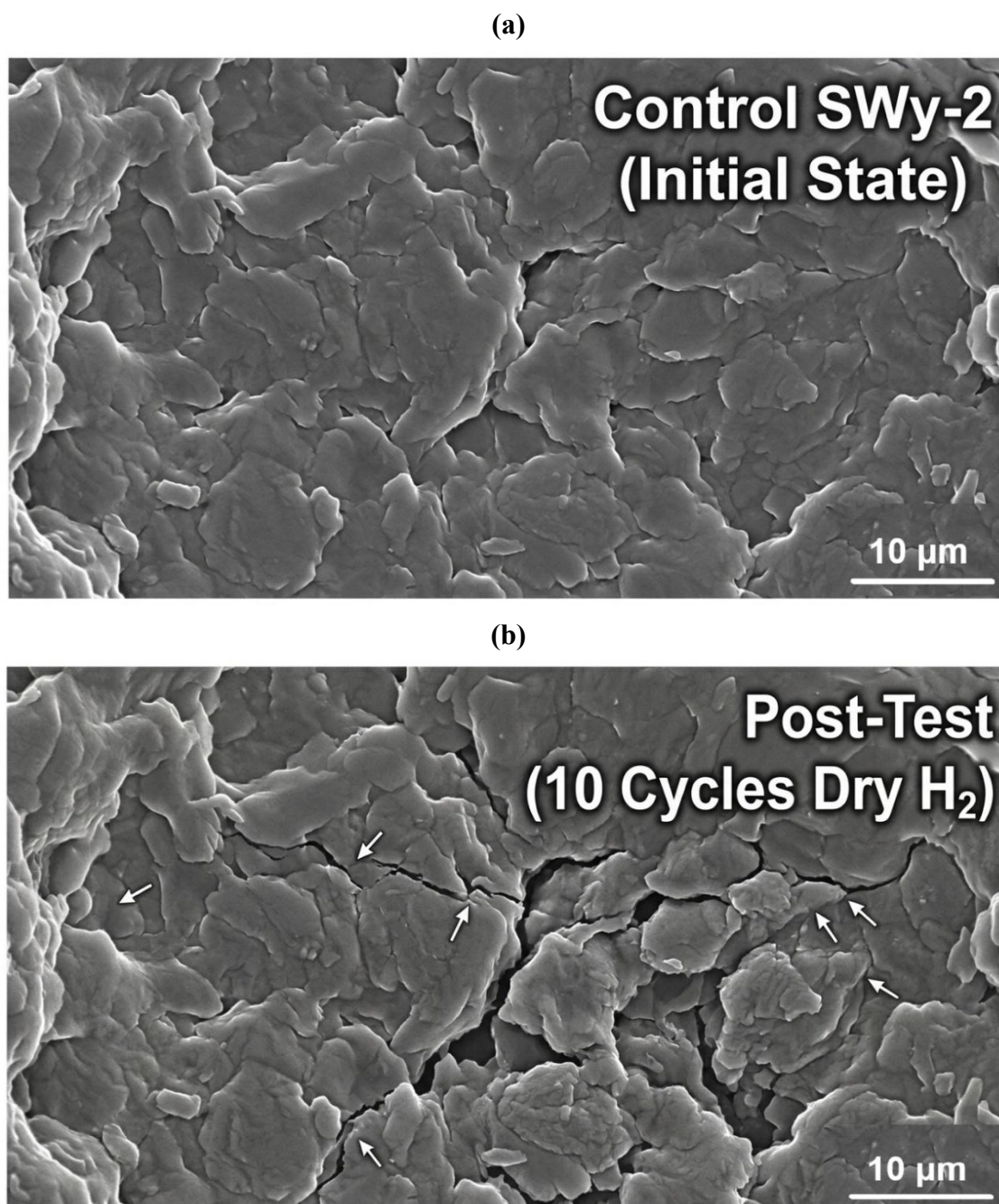


Figure 10. Scanning electron microscopy images of compacted Na-SWy-2 montmorillonite in the initial state (a) and after ten cycles of dry hydrogen loading (b), illustrating fabric-scale pore and fissure development.

4. Discussion

4.1. Control of hydrogen transport by fabric-scale evolution under cyclic loading

The evolution of hydrogen permeability observed during cyclic pressure loading provides evidence that gas transport in compacted Na-SWy-2 montmorillonite is governed primarily by microstructural modifications occurring at the pore and fabric scales rather than by changes in

mineralogical properties. As shown in Figure 5, the effective hydrogen permeability increases progressively with the number of pressure cycles for all confinement conditions while remaining within the range expected for smectite-rich sealing materials [6,8,28].

The monotonic increase in K_{eff} , combined with the absence of abrupt permeability jumps, indicates that transport properties evolve cumulatively rather than through sudden macroscopic damage or failure [7,10]. This gradual response suggests that cyclic pressurization promotes incremental modifications of gas pathways, consistent with a progressive reorganization of the clay fabric. The normalized permeability evolution (Figure 5b) further reinforces this interpretation by showing comparable relative trends under different confinement conditions, despite differences in absolute permeability magnitude.

The interpretation of microstructural evolution must be considered in light of the hydro-mechanical loading conditions. As illustrated in Figure 11, the cyclic loading path includes phases where gas pressure exceeds the applied axial stress. Under partially saturated conditions, such configurations result in a non-classical stress path in which the effective stress cannot be uniquely defined. This supports a qualitative interpretation of fabric opening and microstructural rearrangement mechanisms rather than a strictly defined stress-driven process.

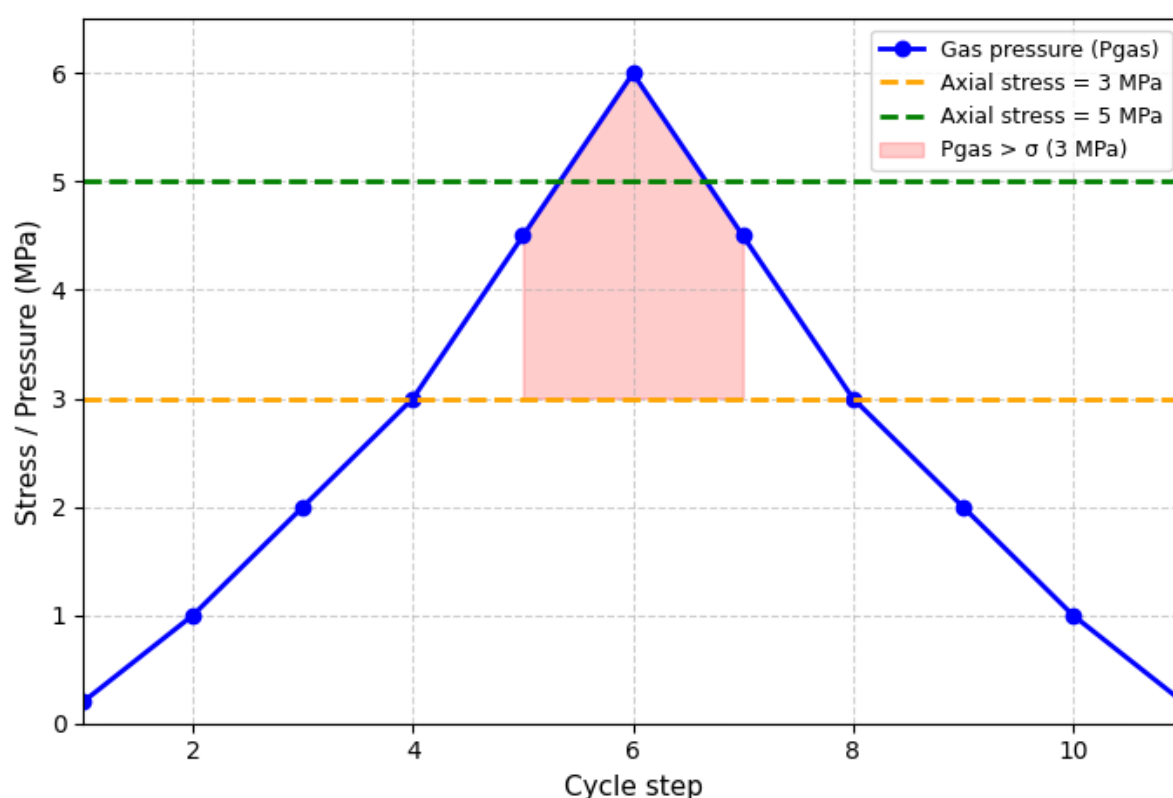


Figure 11. Schematic representation of the hydro-mechanical loading path applied during the cyclic hydrogen tests. Gas pressure (P_{gas}) was varied between 0.2 and 6 MPa while axial stress was maintained constant at 3 or 5 MPa. The figure is provided to illustrate the loading conditions used in the experiments and does not represent direct experimental measurements.

The specimens were tested under partially saturated conditions, with an initial degree of saturation estimated at approximately 60–70%. Under such conditions, gas breakthrough pressure and effective permeability are known to be strongly influenced by capillary effects and pore water distribution [42,46,49]. While the absolute values of P_b and K_{eff} are therefore dependent on the initial saturation state, the progressive permeability increase observed under cyclic loading reflects relative changes in pore connectivity rather than variations in saturation. Although no direct evidence of large-scale drainage was observed during testing, potential changes in saturation state cannot be excluded. Consequently, the observed trends are interpreted primarily in terms of fabric-scale microstructural evolution, while acknowledging possible contributions from coupled hydro-mechanical processes.

While the observed trends are consistent with fabric-scale microstructural evolution, alternative coupled processes may also contribute. In particular, dry gas injection under partially saturated conditions may induce local desaturation, suction variations, and pore-water redistribution, which are known to affect gas transport properties in clays.

Because such processes were not independently controlled in this study, the observed permeability evolution cannot be uniquely attributed to cyclic hydrogen loading alone, but should instead be interpreted as the combined effect of coupled hydro-mechanical mechanisms.

At the crystal scale, XRD results demonstrate the persistence of a basal spacing of approximately 12.3 Å, characteristic of the 1W hydration state of Na-montmorillonite, throughout cyclic hydrogen exposure [11,23,27]. The absence of any detectable shift in the (001) reflection indicates that hydrogen pressurization does not induce interlayer collapse, expansion, or dehydration under the investigated conditions. As summarized in Table 2, this structural stability rules out interlayer processes as a controlling factor for permeability evolution.

In contrast, pore-scale investigations reveal clear changes in the accessible pore network. Mercury intrusion porosimetry highlights an increase in macropore volume following cyclic loading, while the finer pore domains remain largely unchanged [43–45]. SEM observations suggest the presence of localized micro-fissure-like features and enlarged pores at the aggregate scale. However, given the potential influence of sample preparation, these features cannot be considered direct proof of in situ fissure development. Instead, they are interpreted as consistent with a progressive increase in pore connectivity, as reflected by the permeability evolution.

Together, these results indicate that hydrogen migration is dominated by advection through evolving fabric-scale pathways rather than diffusion through interlayer spaces.

The interpretation of microstructural evolution must be considered in light of the specific hydro-mechanical conditions of the experiments. The specimens were tested under partially saturated conditions and subjected to gas pressures that locally exceeded the applied axial stress. Under such conditions, the concept of effective stress is not uniquely defined, as capillary effects and gas pressure contribute to the mechanical response of the porous medium. In addition, the absence of suction measurements prevents quantitative evaluation of stress partitioning between the solid skeleton and pore fluids. Therefore, the proposed mechanisms of fabric opening and micro-fissure development should be interpreted as conceptual processes consistent with the observed permeability evolution and microstructural trends, rather than as strictly defined stress-driven mechanisms. It should be noted that the experimental investigation was conducted on a limited number of specimens representative of two distinct compaction and confinement conditions. While no statistical repetition was performed, the permeability measurements obtained during each loading cycle were internally consistent and repeatable within the resolution of the experimental setup. Steady-state flow conditions were

systematically reached prior to permeability estimation, and the progressive trends observed in effective permeability were reproducible over successive cycles [6].

My objective of this study was not to establish statistical variability, but to identify and interpret the dominant mechanisms controlling hydrogen permeability evolution under cyclic loading. The consistency of the observed permeability increases across confinement levels, combined with the multi-scale microstructural evidence, supports the mechanistic nature of the conclusions drawn.

4.2. Link between macroscopic permeability increase and microstructural changes

The relationship between macroscopic permeability evolution and microstructural modification can be qualitatively assessed by comparing the permeability measurements with the post-test MIP and SEM observations. Specimens exhibiting larger increases in permeability also display greater development of macroporosity and enhanced pore connectivity at the aggregate scale. Although the available dataset does not enable a rigorous statistical correlation, the consistency between transport measurements and microstructural observations supports the interpretation that permeability evolution is associated with progressive changes in the connected pore network [55,56]. These observations provide qualitative support for the hypothesis that fabric-scale pore opening contributes to the observed permeability evolution under cyclic hydrogen loading.

Importantly, SEM images evidence that the increase in macroporosity remains moderate and spatially localized. This suggests that permeability enhancement results from the progressive connectivity of existing pores and micro-fissures, rather than from pervasive damage or large-scale cracking [57–59]. In this context, the progressive decrease in breakthrough pressure (P_b) with cycle number can be interpreted as evidence of facilitated gas transport through previously formed pathways. Rather than representing repeated pore network creation, the evolution of (P_b) suggests a memory effect in the clay microstructure, where earlier cycles reduce the resistance to subsequent gas migration. Such behavior explains why sealing efficiency decreases gradually instead of collapsing abruptly, even after multiple pressure cycles.

Although direct pressure–flow records are not reported here, the progressive decrease in breakthrough pressure and increase in permeability are consistent with a progressive facilitation of gas transport through the connected pore network.

The multi-scale synthesis presented in Table 4 highlights a clear hierarchy of responses: While the crystal structure remains unaffected, pore-scale and fabric-scale features evolve in a manner that directly influences macroscopic transport properties. This decoupling between mineralogical integrity and hydraulic performance is particularly relevant for evaluating the long-term behavior of clay-rich caprocks subjected to repeated gas cycling [60–63] (Table 4).

The number of pressure cycles investigated in this study remains limited compared to the number of cycles that may be experienced during long-term or seasonal underground hydrogen storage operations. However, my objective of this work was not to reproduce operational timescales, but to identify the early-stage mechanisms governing permeability evolution under cyclic loading. The monotonic and progressive nature of the permeability increase observed over the first ten cycles indicates that fabric-scale reorganization occurs incrementally and without abrupt mechanical damage [64,65]. From a mechanistic perspective, these initial cycles are sufficient to reveal the dominant deformation and transport processes, particularly the development of preferential pathways controlled by microstructural fatigue. Although direct extrapolation of permeability values to large numbers of

cycles cannot be achieved from the dataset, the absence of catastrophic permeability jumps suggests a progressive trend that may evolve toward stabilization or asymptotic behavior at higher cycle numbers. Future studies involving extended cyclic loading would be required to quantify long-term permeability trajectories under storage-relevant conditions.

Table 4. Multi-scale synthesis of the effects of cyclic hydrogen loading on compacted Na-SWy-2 montmorillonite.

Scale	Technique	Key observation	Effect of cyclic H ₂ loading	Implication for seal integrity
Crystal scale	XRD	Basal spacing $d_{001} \approx 12.3 \text{ \AA}$ (1W Na-montmorillonite)	No measurable change	Preserved mineralogical integrity
Pore scale	MIP	Dominant inter-aggregate pores; Limited microporosity	Increase in macropore volume ($> 1 \text{ \mu m}$)	Moderate reduction of sealing efficiency
Fabric scale	SEM	Dense aggregate fabric in initial state	Localized micro-fissures and widened pore throats	Development of preferential gas pathways
Macroscopic scale	Gas permeability	Very low initial H ₂ permeability (10^{-21} – 10^{-20} m^2)	Progressive increase of K_{eff} ($\times 2$ – 3)	Cumulative degradation without failure

4.3. A physically based interpretation of permeability evolution under cyclic hydrogen loading

The experimental observations obtained in this study can be interpreted within a physically consistent theoretical framework that links macroscopic permeability evolution to microstructural changes induced by cyclic pressure loading. This framework does not aim at predictive modeling, but rather at establishing a mechanistic coherence between basic transport laws and the observed evolution of the clay microstructure.

At the macroscopic scale, hydrogen transport through the compacted clay specimens is quantified using an effective permeability derived from steady-state gas flow measurements. For compressible fluids, permeability evaluation is based on Darcy's law expressed for gas flow, which can be written as:

$$K_{\text{eff}} = \frac{Q \mu L}{A (P_{\text{in}}^2 - P_{\text{out}}^2)}$$

where Q is the volumetric flow rate, μ is the dynamic viscosity of hydrogen, L is the specimen length, A is its cross-sectional area, and P_{in} and P_{out} are the inlet and outlet pressures, respectively [66–68]. Within this formulation, any systematic variation of K_{eff} directly reflects a modification of the connected pore network controlling gas transport.

From a pore-scale perspective, permeability is classically related to the geometry and connectivity of the pore network. In porous media, simplified expressions such as those derived from Carman–Kozeny-type relationships highlight the dependence of permeability on effective porosity and characteristic pore throat size:

$$K \propto \frac{\phi_{\text{eff}}^3}{(1 - \phi_{\text{eff}})^2} d_{\text{char}}^2$$

where ϕ_{eff} represents the effective connected porosity participating in flow, and d_{char} is a characteristic pore or pore-throat dimension. This relationship does not imply that permeability can be

quantitatively predicted from porosity measurements alone, but it underscores the strong sensitivity of gas transport to modest changes in pore connectivity and pore size [69].

In this study, X-ray diffraction analyses demonstrate that the interlayer spacing of Na-montmorillonite remains stable under cyclic hydrogen loading, maintaining a basal spacing characteristic of the 1W hydration state. This suggests that interlayer processes are unlikely to be the dominant mechanism controlling the observed permeability evolution [70,71]. Consequently, variations in K_{eff} must primarily originate from modifications of extra-layer porosity.

Mercury intrusion porosimetry and SEM observations show that cyclic pressurization leads to a progressive increase in macroporosity and the development of localized micro-fissures at the aggregate scale. These features correspond to an increase in the effective connected porosity ϕ_{eff} and to an enlargement of characteristic pore throats d_{char} , both of which are expected, based on the above considerations, to promote an increase in permeability. The qualitative correlation observed between normalized permeability increase and macroporosity development supports this interpretation and provides experimental consistency between macroscopic transport behavior and microstructural evolution [72,73].

The cumulative nature of permeability increase with cycle number further suggests that cyclic loading acts as a progressive mechanical perturbation of the clay fabric. Under constant axial stress, repeated pressurization and depressurization do not induce sudden structural failure, but rather incremental rearrangements at aggregate contacts and pore throats. Such behavior is consistent with a fatigue-like response, in which microstructural changes accumulate gradually while preserving overall mechanical stability [74].

Within this theoretical framework, the evolution of hydrogen permeability under cyclic loading can be described as a fabric-controlled, advective transport process driven by progressive reorganization of the pore network. Importantly, this process occurs without mineralogical degradation or interlayer alteration, indicating that sealing performance degradation is governed by microstructural connectivity rather than by fundamental changes to the clay crystal structure. This distinction is critical for assessing the long-term integrity of smectite-rich barriers in underground hydrogen storage applications.

4.4. Impact of cyclic pressure loading and mechanical fatigue of the clay skeleton

The observed permeability evolution can be interpreted as a manifestation of mechanical fatigue within the compacted clay skeleton. Under constant axial stress conditions, repeated pressurization and depressurization cycles likely induce stress redistribution at aggregate contacts and pore throats, leading to progressive fabric rearrangement. Over successive cycles, these incremental adjustments accumulate, enhancing pore connectivity and facilitating gas flow. Unlike brittle materials, compacted smectite-rich clays exhibit a ductile and time-dependent response, enabling them to accommodate cyclic loading through gradual microstructural adaptation rather than through sudden fracture. This behavior is consistent with the absence of macroscopic deformation or failure during permeability testing and supports the interpretation of cyclic fatigue as a key controlling mechanism [75,76].

The number of cycles investigated in this study exceeds typical annual operational conditions in underground hydrogen storage. However, the objective is not to reproduce field-scale cycling frequency, but to accelerate and reveal the fundamental mechanisms controlling permeability evolution under repeated loading. From a mechanistic perspective, the first cycles are particularly important, as they correspond to the initial adaptation of the clay microstructure to pressure variations. The observed

permeability increase over ten cycles therefore provides insight into early-stage processes that may occur over longer timescales in real systems.

Dry hydrogen injection may further accentuate this process by locally reducing capillary forces at pore throats, particularly in partially saturated conditions, thereby promoting the opening of preferential pathways without destabilizing the interlayer structure. The combined mechanical and hydraulic effects of cyclic loading may contribute to a gradual increase in pore connectivity and a corresponding reduction in sealing efficiency (Figure 12).

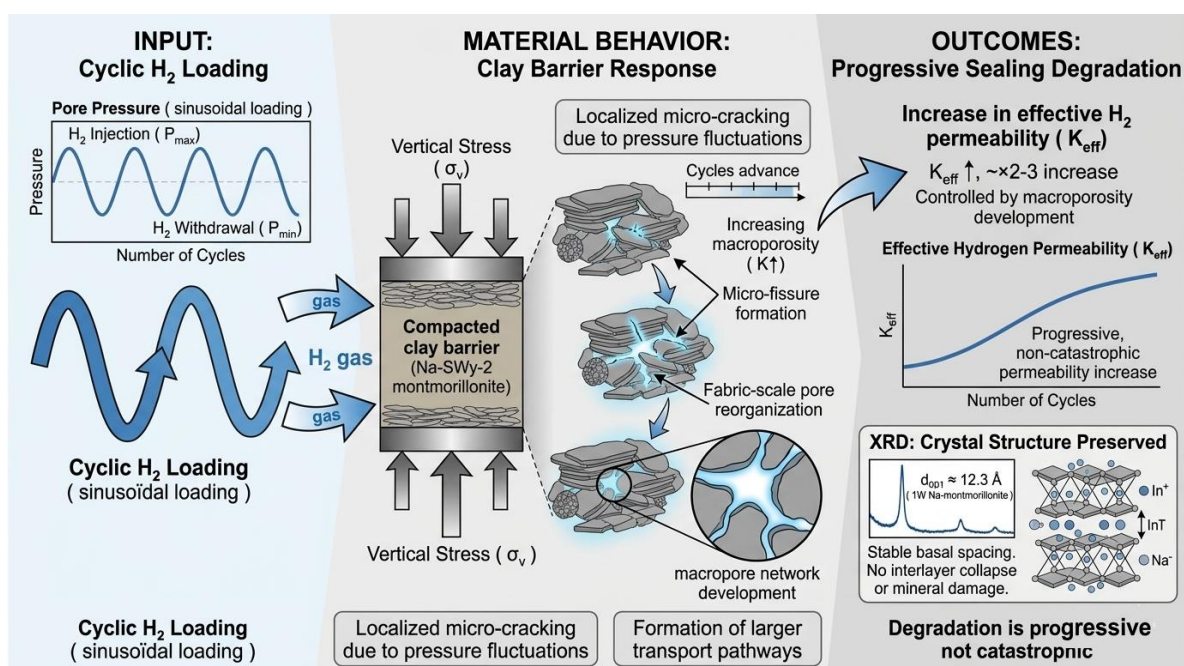


Figure 12. Interpretative conceptual model summarizing the experimental observations obtained in this study. The schematic synthesizes the proposed relationships between cyclic hydrogen loading, permeability evolution, and microstructural changes observed by XRD, MIP, and SEM. It is intended as a qualitative interpretation derived from the experimental results and does not represent direct measurements.

4.5. Implications of cyclic hydrogen loading for clay-based sealing systems

The results obtained in this study indicate that compacted montmorillonite subjected to cyclic hydrogen loading may undergo progressive modifications in its transport properties despite the apparent preservation of mineralogical integrity observed by XRD. The coexistence of stable interlayer spacing and increasing permeability suggests that permeability evolution is primarily associated with changes in pore connectivity and fabric organization rather than crystallographic alteration.

Within the investigated experimental conditions, the simultaneous increase in apparent permeability, decrease in breakthrough pressure, development of macroporosity, and appearance of micro-fissure-like features are collectively consistent with a potential degradation mechanism affecting sealing efficiency during cyclic hydrogen loading. Although the absolute permeability values remain low, the monotonic evolution observed over successive cycles indicates that repeated pressure fluctuations can progressively facilitate gas transport through the compacted clay.

These observations highlight the importance of considering cyclic hydro-mechanical effects when evaluating the behavior of clay-based sealing materials exposed to hydrogen [77–80]. My results suggest that mineralogical stability alone is insufficient to assess sealing performance, as significant changes in transport properties may occur without detectable alteration of the clay crystal structure.

The results obtained in this study can also be compared with recent investigations on hydrogen storage in salt formations. In salt caverns, the evolution of storage performance is largely controlled by the viscoplastic and self-healing behavior of rock salt. In contrast, clay-rich materials rely primarily on low permeability and capillary sealing, which may be progressively altered by modifications in pore connectivity during cyclic loading. Although the governing mechanisms differ, both systems are potentially sensitive to repeated pressure variations and associated hydro-mechanical processes.

It should be emphasized that the experiments were conducted under simplified laboratory conditions using a limited number of loading cycles and partially saturated specimens. Consequently, the observed permeability evolution should be interpreted as evidence of a possible early-stage degradation process rather than as a direct indicator of long-term field behavior. No direct conclusions can therefore be drawn regarding the engineering reliability, long-term suitability, or field-scale performance of clay-rich caprocks for underground hydrogen storage.

Instead, the principal contribution of this work is the identification of mechanisms that may contribute to sealing-efficiency degradation under cyclic hydrogen loading and that warrant further investigation under more representative hydro-mechanical conditions.

4.6. Geochemical considerations on H₂–Clay interactions: Insights from PHREEQC modeling

To assess the potential contribution of geochemical processes, simplified PHREEQC equilibrium calculations were performed. The simulations indicated no significant changes in pH, redox conditions, or mineral stability under the investigated conditions. These results are consistent with the absence of detectable mineralogical changes observed by XRD and suggest that the observed permeability evolution is primarily associated with physical rather than geochemical processes [81,82].

4.7. Experimental limitations and implications for interpretation

The results presented in this study provide insight into the early-stage evolution of gas transport properties in compacted montmorillonite subjected to cyclic hydrogen loading. However, several limitations of the experimental framework must be considered when interpreting the results. First, the experiments were conducted under partially saturated conditions, with an initial degree of saturation of approximately 60–70%, and no direct monitoring of suction evolution was performed during testing. Consequently, the respective contributions of cyclic loading, desaturation, capillary effects, and pore-water redistribution cannot be fully separated.

Second, the mechanical boundary conditions correspond to a one-dimensional odometer configuration with constant axial stress and rigid lateral restraint. Although this setup provides a controlled and reproducible loading framework, it does not reproduce the full three-dimensional stress state encountered in geological formations. In addition, gas pressure locally exceeds the applied axial stress during part of the loading cycle, resulting in a complex hydro-mechanical stress path.

Third, no dedicated control experiments involving inert gases or uncycled specimens were performed. Therefore, the specific contribution of cyclic hydrogen loading cannot be completely

isolated from other coupled hydro-mechanical processes that may occur during testing.

Fourth, the reported permeability values correspond to apparent gas permeability because no correction for gas slippage (Klinkenberg effect) was applied. Although this limitation primarily affects the absolute magnitude of permeability, it introduces uncertainty when comparing the measured values with intrinsic permeability reported in the literature.

Fifth, MIP and SEM analyses were performed on specimens extracted after testing and subjected to drying and preparation procedures. Consequently, some of the observed pore enlargement and micro-fissure-like features may be influenced by preparation-induced artifacts. These observations should therefore be considered as indicative of possible microstructural evolution rather than direct evidence of in situ processes.

Finally, the experimental program was limited to ten pressure cycles and two testing conditions. While this enables identification of early-stage trends in permeability evolution, the results cannot be extrapolated directly to long-term underground hydrogen storage operations involving more complex geological, hydraulic, and mechanical conditions.

Within these limitations, the observed increase in permeability, reduction in breakthrough pressure, and changes in pore structure are consistent with the existence of a potential degradation mechanism affecting sealing efficiency under cyclic hydrogen loading. However, further investigations involving extended cycling, controlled saturation conditions, and additional control experiments are required to better constrain the relative contribution of the mechanisms involved.

5. Conclusions

This study provides experimental insight into the response of compacted Na-SWy-2 montmorillonite to cyclic hydrogen loading using a multi-scale approach combining gas permeability measurements with XRD, MIP, and SEM.

The results show a progressive increase in apparent hydrogen permeability with cycle number, while remaining within the low-permeability range characteristic of compacted clay materials. XRD analyses indicate that the interlayer spacing of montmorillonite remains stable throughout cyclic loading, suggesting preservation of mineralogical integrity. In contrast, MIP and SEM observations are consistent with modifications in pore structure and connectivity at the aggregate scale, although these features must be interpreted with caution due to potential sample preparation effects.

The observed permeability evolution is interpreted as resulting from coupled hydro-mechanical processes induced by cyclic pressure variations. However, the respective contributions of mechanisms such as partial desaturation, suction changes, pore-water redistribution, and microstructural rearrangement cannot be fully isolated within the experimental framework.

The combined increase in apparent permeability, decrease in breakthrough pressure, development of macroporosity, and appearance of micro-fissure-like features indicate that cyclic hydrogen loading may progressively alter transport pathways within compacted montmorillonite. Within the investigated experimental conditions, these observations should be interpreted as evidence of a potential degradation mechanism affecting sealing efficiency under repeated pressure fluctuations rather than as an assessment of long-term material performance.

It is important to emphasize that the experimental conditions investigated in this study differ significantly from those encountered in field-scale underground hydrogen storage systems, particularly with respect to stress state, saturation conditions, geological heterogeneity, and number

of pressure cycles. Consequently, no direct conclusions can be drawn regarding the engineering reliability, long-term suitability, or field-scale performance of clay-rich caprocks for underground hydrogen storage. Overall, the principal contribution of this work is the identification of early-stage hydro-mechanical processes that may contribute to sealing-efficiency degradation under cyclic hydrogen loading. Further investigations incorporating extended cycling, controlled hydro-mechanical boundary conditions, and in situ monitoring techniques are required before any assessment of long-term applicability can be established.

Author contributions

W.O.: Formal analysis, investigation, Methodology, Data curation, Conceptualization, Supervision, Writing—Original draft, Writing—Review and editing, Project administration.

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Data availability statement

The authors confirm that the data supporting the findings of this study are available within the article. Source data are available from the corresponding author upon request.

Use of AI tools declaration

The authors declare they have not used Artificial Intelligence (AI) tools in the creation of this article.

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Conflict of interest

The authors declare no conflict of interest.

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