Geochemical interactions in geological hydrogen Storage: The role of sandstone clay content

Geochemical Interactions in Geological Hydrogen Storage: The Role of Sandstone Clay Content

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Abstract

Hydrogen holds promise as a clean energy alternative, crucial for achieving global decarbonization goals and net-zero carbon emissions. Its low volumetric energy density necessitates underground storage in sandstone formations to maintain year-round supply. The efficacy of such storage hinges on the geochemical interplay between hydrogen and the host sandstone. Despite the slow reaction rates in sandstone, the influence of its clay composition on hydrogen interaction remains underexplored. In this study, we specifically investigate the geochemical interactions of hydrogen with clay-bearing sandstone formations under controlled conditions, simulating storage scenarios. This study evaluates the impact of clay on hydrogen-sandstone geochemistry after 75 days of injection at 1500 psi and 75°C into Berea and Bandera gray sandstone cores, utilizing microcomputed tomography to assess changes in pore structure. Our results reveal that, even in sandstones with high clay content, there is negligible alteration in porosity and mineral content over storage time, indicating stability in these formations. These findings provide crucial insights for selecting suitable geological formations for hydrogen storage, supporting the global shift

towards sustainable energy systems Our study contributes to the global efforts in decarbonization by providing essential guidance on the feasibility of using clay-bearing sandstone formations for efficient and sustainable hydrogen storage.

Keywords: Hydrogen; clay; sandstone; geochemical reactions; underground hydrogen storage.

1. Introduction

Hydrogen is considered a prominent energy carrier, poised to plays a significant part in the energy transition from carbon-based fuel to clean energy and attainment of global decarbonization pursuit [1-3]. While renewable energy sources have been favored as alternatives to fossil fuels, their inherent intermittency and seasonal variability limit their reliability [4, 5]. To counter this, green hydrogen, produced via surplus renewable energy, presents a solution by offering storable energy that can bridge gaps in supply [6, 7].

Nevertheless, there are several hitches that are associated with surface storage, H_2 is highly flammable with high tendency to mix with air or O_2 when stored at the surface facilities [8, 9]. It will need just 5% of O_2 for combustion, unlike fossil fuel that will require around 12% to burn. Its flammability in oxygen has been estimated to vary between 4 to 94%, suggesting that the occurrence of explosions is likely if surface H_2 storage is not meticulously managed. H_2 also high diffusivity, thus the general cost of surface storage installation and maintenance could be quite outrageous [10, 11].

As a result, subsurface geological formations, such as saline aquifers and depleted hydrocarbon reservoirs, have emerged as preferable options for hydrogen storage. [12, 13]. These settings offer a safer and more economical alternative, essential for the hydrogen economy, by facilitating a stable grid and ensuring hydrogen's availability during peak demands [14, 15].

Notably, underground storage mitigates explosion risks due to the reduced oxygen levels. While salt caverns, historically utilized for town and natural gas storage, offer a straightforward structure and high withdrawal rates, their capacity and durability under cyclic injection remain limited [15, 16]. Additionally, their geographic distribution and proximity to hydrogen production sites present logistical challenges [17].

On the other hand, depleted oil, and gas reservoirs, with their vast storage capacities and impermeable caprocks, offer a promising solution for hydrogen storage, utilizing existing infrastructure and drawing upon experiences from methane and CO_2 sequestration [17-19]. However, the translation of these practices to hydrogen underground storage (UHS) is not straightforward due to hydrogen's unique properties such as lower density, reactivity, and volatility, which differ markedly from CO_2 and CH_4. These differences raise concerns about caprock integrity and containment security, particularly in carbonate and sandstone formations where chemical, geochemical, and microbial interactions might compromise the storage system [20, 21].

Recent research has focused on wettability of rock-brine-H_2 systems and rock-fluid interfacial interactions [22-24]. Results generally showed that contact angle increased, whereas H_2/rock IFT generally decreased with pressure, salinity, and presence of organic acids [25-27]. On the other hand, high temperature and nanofluids treatment of rock surfaces were identified as favorable conditions for enhancing the residual trapping of H_2 in the host-rock (quartz, sandstone, calcite, carbonate, basalt, and organic-rich shale), and capillary trapping capacities of caprocks (mica and low total organic carbon shale) [28-31]

The geochemical interaction of hydrogen with carbonate and sandstone rocks has also been fairly investigated, suggesting inert behavior in the latter, thereby supporting the feasibility

of sandstone as a storage medium [32, 33]. Most studies have observed only minor geochemical reactions between H_2 and sandstone under various pressures and temperatures, with implications for reservoir rock permeability. For instance, Yekta and his co-researchers [32, 33] conducted numerical and experimental assessment of abiotic reactions due to sandstone-H_2 interactions and found slight reactions between H_2 and sandstone. The reaction was monitored from 6 weeks to 6 months at pressure of 10 MPa and temperature of 100–200 ◦C using water saturated and dry sandstone samples.

They further performed core flooding experiments to evaluate the impact of changes in rock mineralogy on the reservoir rock total permeability at shallow downhole conditions (20 ◦C, 5.5 MPa), and deep subsurface conditions (45 ◦C, 10 MPa). Their results showed that relative permeability curves were same at both experimented conditions due to minor change in viscosity of H_2 with changing temperature and pressure, compared to CO_2 and N_2. Flesch et al. [34] assessed the impacts of injected H_2 on sandstone substrates from Germany, they observed noticeable alteration in porosity and permeability of Triassic and Permian sandstones samples, but no occurrence of chemical reactions was observed in Tertiary sandstones after 42 days. These findings generally indicated that H_2 would likely behaves as inert gas in sandstone formations, suggesting that successful hydrogen storage in sandstone sites is feasible.

Conversely, carbonate minerals have demonstrated a propensity to react with H_2, potentially affecting storage integrity. In particular, the presence of iron could accelerate reactions, leading to the transformation of minerals such as pyrite to pyrrhotite and influencing hydrogen loss. Specifically, Truche et al. [35] showed that pyrite could play a major role in precipitation of pyrrhotite in an alkaline surrounding at high temperatures ($> 90^oC$) and elevated pressures (50–100 bar). They emphasized that interaction between H_2 and the host reservoir rock could be

quickened in the presence of Fe. Hassannayebi et al. [36] model predictions suggest that geochemical interactions are quite slow in the short time, although such reactions could result in precipitation and dissolution at the long run. They attributed H_2 loss and increased pH to the reduction of pyrite to pyrrhotite.

Bo et al. [37] established from their geochemical model results that H_2 loss from H_2/calcite/bacteria interactions is initiated by calcite and not by clay minerals and silicate. They also reported that the influence of pressure and temperature on H_2 loss in brine solution is negligible, but that calcite plays a major role in considerable H_2 loss during UHS. Gholami [16] conducted series of kinetic and thermodynamic geochemical modeling to assess the geochemical reactions from H_2 injection into porous media. The model results were calibrated against experimental data to demonstrate that the dissolution of halite, anhydrate and carbonates can take place in the long term, resulting in hydrogen sulfide formation, as well as calcite precipitation, and pore structure blockage over time because of scale formation.

They proposed that pyrite could reduce to pyrrhotite probably at temperature beyond 90 ◦C which could promote hydrogen sulfide formation, even when microbial action is non-existence. However, halite (scale) formation only commences at the end of 10 years, suggesting that halite dissolution in porous formation is a very slow phenomenon. In another study, Al-Yaseri et al [17] reported that gas chromatography could only detect traces of CH_4 (0.018 %), whereas no H_2S was spotted from H_2/shale reaction after 11.5 weeks likely because the reaction temperature and period was not sufficient for noticeable CH_4 production.

Despite these insights, the specific interactions between hydrogen and the clay components within sandstone formations remain underexplored. This gap is critical, as clay minerals are prevalent in many potential geological storage sites, and their reactivity with hydrogen could

significantly influence the efficacy of UHS. Therefore, this study conducts a detailed assessment of hydrogen's interaction with clay-rich sandstone to delineate the potential geochemical reactions that may occur. By comparing computed tomography (CT) scans of clay-rich Bandera gray sandstone micro-structures with Berea sandstone samples, both before and after exposure to pressurized hydrogen, we aim to illuminate the changes in pore structure that could inform the feasibility of hydrogen storage in such geological settings.

2.0 Materials and methods

Berea and clay-rich Bandera gray sandstones were selected for this study as they are representative of common sandstone formations. The mineral composition of the two samples as identified by X-ray diffraction (XRD), is presented in Table 1. The research grade H_2 gas (99.99% purity) was sourced from Air Liquide Saudi Arabia. The sandstones compositions were determined using a Bruker-AXS D8 instrument (Bruker, Germany) (scanning parameters 0−90°,2θ,accuracy in peak positions≤0.01 2θ,Bragg−Brentanoconfiguratio). To commence the experiments, the core samples (4 by 91.2 mm) were cleansed with methanol-toluene solution (50% each) by aging for 1 day. The samples were then encased in heat-shrink polytetrafluoroethylene (PTFE) jacket to prevent H_2 flow into porous media. Subsequently, the core plugs were positioned in the reaction cell (1 diameter and 12 cm long, 316 stainless steel) and vacuumed for 60 minutes to eliminate air from the system [38].

Table 1. XRD identification of Berea and Bandera Gray sandstone samples

Composition	Quartz	Kaolinite	Albite	Dolomite	Microcline	Illite	Muscovite	Clinochlore	Chlorite
Bandera Gray sandstone	58.6	1.9	18.9	4.4	-	4.2	-	10.7	1.1
Berea sandstone	90.2	2.8	-	-	5.8	-	1.1	-	-

Following this stage, the samples were partially saturated (30%) with sea water (with composition shown in Table 2) and the 99.99% pure pressurized H_2 was injected at 1500 psi ±1 psi and 75ºC ± 0.1 ºC for a duration of 75 days. Changes in the rock's pore structures were assessed using CT scan images taken before and after hydrogen injection. The HeliScan micro-CT instrument, provided by Thermo Fisher Scientific (Czech Republic), was utilized for the scans. Pore labelling and grain identification were performed based on the threshold values obtained from the micro-CT scan tomograms. The image segmentation and filtering were carried out using PERGEOS SOFTWARE (FEI, Thermo Fisher Scientific).

Table 2. Composition of seawater brine used for saturating the rock samples

Ions	Na^+	Ca^{2+}	Mg^{2+}	SO_4^{2-}	Cl^-	HCO_3^-	Total
Concentration (ppm)	18,300	650	2110	4,290	32,200	120	57,670

Smoothening and noise removal from the raw data were accomplished using a nonlocal means filter. Image segmentation was performed using gradient marker-based watershed tool (PERGEOS SOFTWARE). Porosities in both 2D and 3D were derived from the segmented data of each core sample. Alterations in the core pore structures were deduced from changes in porosity observed before and after the injection of pressurized hydrogen into the rock samples. The set of

scans—Dry (before H_2 treatment) and after 75 days of H_2 treatment for the Berea and Bandera Gray sandstones—were processed using PerGeos software (FEI-Thermo Fisher).

This process began by aligning their orientations and registering two images at a time, using the dry sample scan as the reference. Multiple iterations of the image registration process were carried out to ensure that unchanged regions of the sample would cancel out in the differencing step. Normalized mutual information was employed as the metric for image registration, aiming to minimize entropy between the histograms of the model and reference, thereby maximizing mutual information. The transformed image was resampled using Lanczos interpolation.

After linearly scaling the attenuation histograms of the 3D registered datasets to match, their difference was calculated. Gradient Marker-based Watershed and Top-Hat Segmentation were applied to the image derived from the difference between the pre- and post-treatment images, enabling the labelling of pores and grains by selecting their thresholds. Subsequently, the volume fractions of each label were calculated to determine the differences in porosity and clay content.

3.0 Result and discussion

Previous research on UHS [33, 37, 39-41] has explored the geochemical and biochemical interactions between injected hydrogen and the host sandstone during underground hydrogen storage (UHS), yet the specific reactions involving clay-rich sandstone remain under-documented in the literature. This study addresses this gap by comparing the interaction of hydrogen with clay-rich Bandera Gray sandstone to that of Berea sandstone. Presented in Figures 1-8, the results delineate the extent of these interactions. As illustrated in Figures 1-4, the pre- and post-interaction images exhibit remarkable consistency, indicating negligible geochemical reaction between the hydrogen and the Berea sandstone matrix.

The comparative analysis commenced with the examination of 2D and 3D micro-CT images of Berea sandstone cores both before and after 75 days of hydrogen exposure. As illustrated in Fig 1-4, the pre- and post-interaction images exhibit remarkable consistency, indicating negligible geochemical reaction between the hydrogen and the Berea sandstone matrix. This observation is further corroborated by the microstructural stability in the micro-CT images, with negligible changes in pore structure post-injection (see Table 3). For instance, the porosity of Berea sandstone showed a minimal increase from 11.27% to 11.32%, the clay content altered slightly from 1.28% to 1.35%, and the grain content exhibited negligible variation, shifting from 87.43% to 87.57%. These data clearly showed that the injection of pressurized H_2 into the Berea sandstone demonstrated no significant reaction with the porous medium.

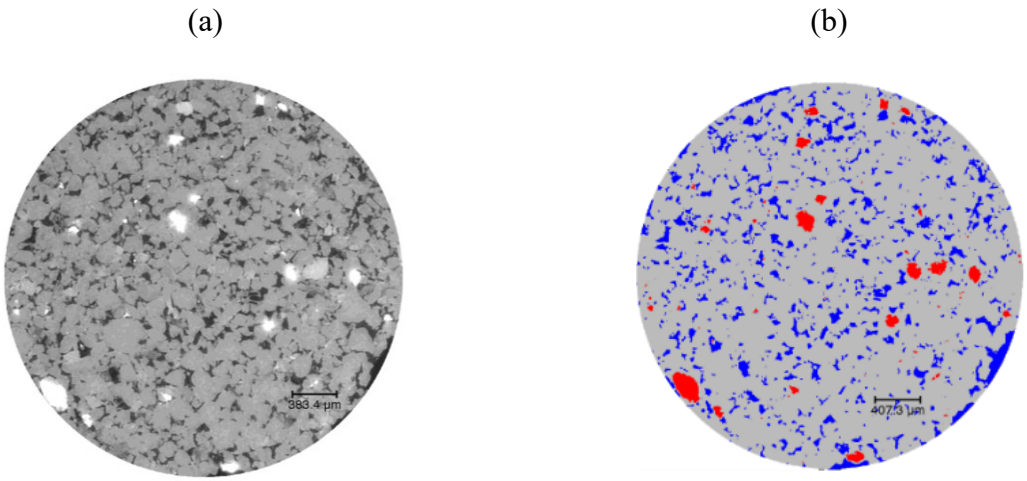


Fig. 1. Images of Berea sandstone images prior to hydrogen injection, including (a) a 2D raw image and (b) a 2D segmented image, which establish a baseline for the sandstone's initial condition.

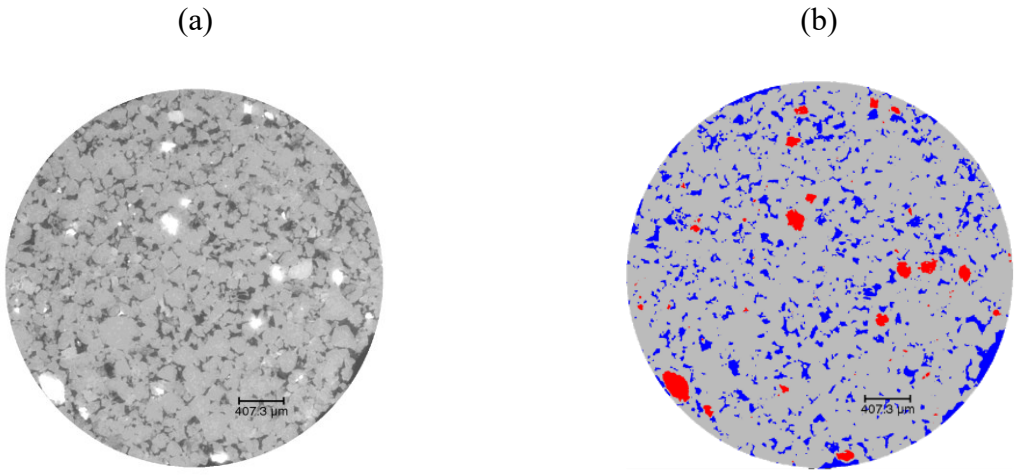


Fig. 2. Images of Berea sandstone after pressurized H_2 injection showing (a) 2D raw image and (b) 2D segmented image.

Generally, there is a good match between the results reported in literature for sandstone-H_2 interactions and results obtained in this study. Labus and Tarkowski [42] conducted geochemical studies to evaluate the possibility of H_2 sub-surface storage in mudstone, claystone, and sandstone samples from Polish lowlands. Their research highlighted goethite as a primary mineral in H_2 consumption, with sandstone displaying minimal porosity alteration, especially when contrasted with mudstone and claystone.

Supporting these findings, Yekta et al. [33] posited that limited reaction is expected between H_2 and sandstone reservoirs. Similarly, Hassanpouryouzband et al. [39] demonstrated through more than 250 batch reaction experiments that the loss of H_2 from geochemical interactions between host-sandstone reservoir rock and injected H_2 does not occur during UHS. This study corroborates the view that sandstone, with its inherent high storage capacity and resilience to geochemical changes, emerges as a preferable candidate for UHS over carbonate formations.

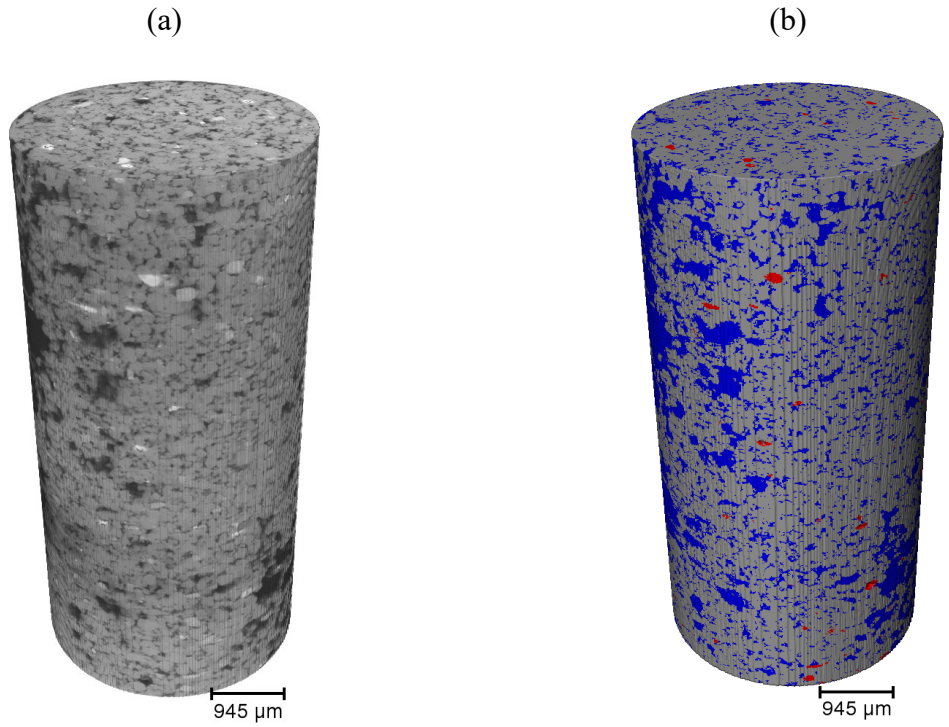


Fig. 3. Images of Berea sandstone before pressurized H_2 injection showing (a) 3D raw image and (b) 3D segmented image.

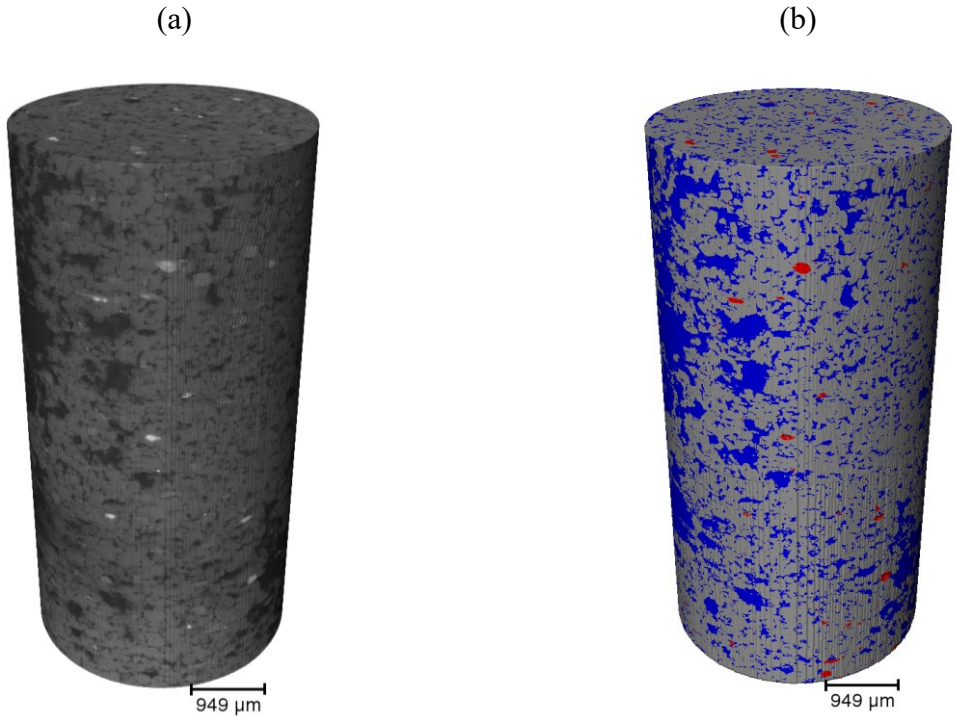


Fig. 4. Images of Berea sandstone after pressurized H_2 injection showing (a) 3D raw image and (b) 3D segmented image.

Table 3. Composition of Berea sandstone before and after H_2 injection

	porosity [%]	Clay [%]	Grains [%]
Berea Sandstone (Before)	11.27	1.28	87.43
Berea Sandstone (After)	11.32	1.35	87.57

Results presented in Figure 5-8 showed that there was no obvious discrepancy between the 2D and 3D images of the clay-rich Bandera Gray sandstone before and after injection of H_2. The micro-CT images were further compared with the changes in pore structure shown in Table 4. Generally, there were no distinct changes in porosity, proportion of clays and grains composition after the cores were pressurized with H_2. Porosity of Bandera Gray was 10.16% and 10.23% before and after interaction with H_2. Clay composition remained almost unchanged at 4.81% and 4.87% pre and post H_2 injection respectively whereas there was no significant change in grain percentage at 85.02% pre hydrogen injection and 85.16% post H_2 injection.

Our results generally showed that there is no dissolution or expansion of clays and quartz composition of the sandstone rocks in the early stages of H_2 injection into Berea sandstone and clay-rich Bandera Gray sandstone formation. These results are consistent with findings from previous studies [43, 44]. Ho et al. [43] conducted Nuclear Magnetic Resonance (NMR) experiments to investigate the NMR response of injected H_2 into Berea sandstone samples. They observed no adsorption hysteresis in Berea sandstone, suggesting that H_2 loss is unlikely during UHS in sandstone formation. Similarly, Ho et al. [44] conducted meta-dynamics molecular simulations to compute the free energy landscape of H_2 intercalation into montmorillonite interlayers. The simulation prediction showed that H_2 intercalation into hydrated interlayers is

thermodynamically unfavorable, suggesting that the loss and leakages of H_2 arising from H_2 intercalation into clay interlayer during UHS is very limited, if occur at all.

With reference to the results obtained in this research, it appears that sandstone formations with a modest quantity of clay in their structure could be a good alternative for UHS. However, it is suggested to investigate the interactions between H_2/brine system and clay rich sandstone for more than 75 days experimented in this research to ensure that such storage sites are suitable for geological hydrogen storage for longer period.

(a) (b)

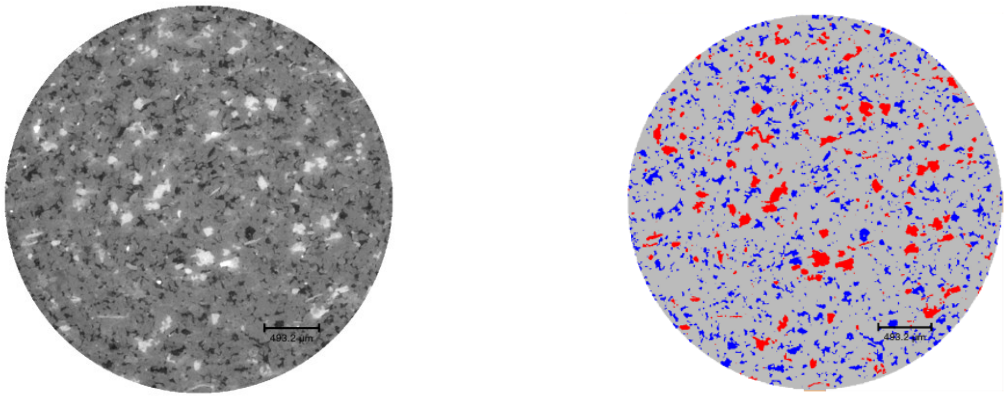
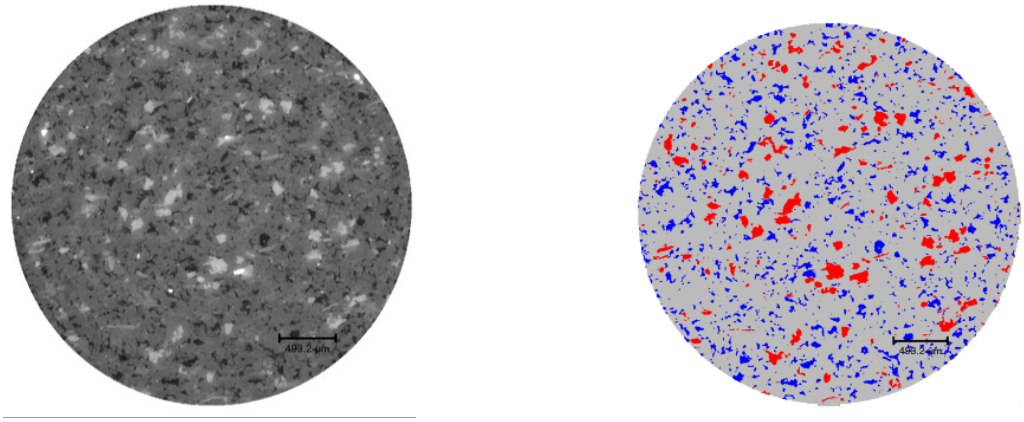


Fig. 5. Images of Bandera Gray sandstone before pressurized H_2 injection showing (a) 2D raw image and (b) 2D segmented image.

(a) (b)

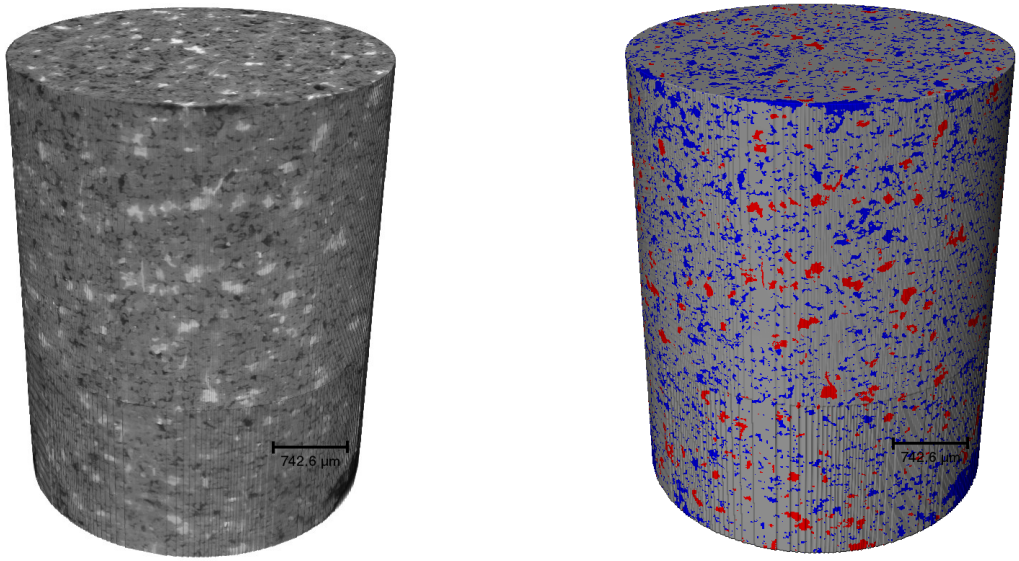


251

252 **Fig. 6.** Images of Bandera Gray sandstone after pressurized H_2 injection showing (a) 2D raw

253 image and (b) 2D segmented image.

(a) (b)



254 **Fig. 7.** Images of Bandera Gray sandstone before pressurized H_2 injection showing (a) 3D raw

255 image and (b) 3D segmented image.

	porosity [%]	Clay [%]	Grains [%]
Bandera gray (Before)	10.16	4.81	85.02
Bandera gray (After)	10.23	4.87	85.16

(a) (b)

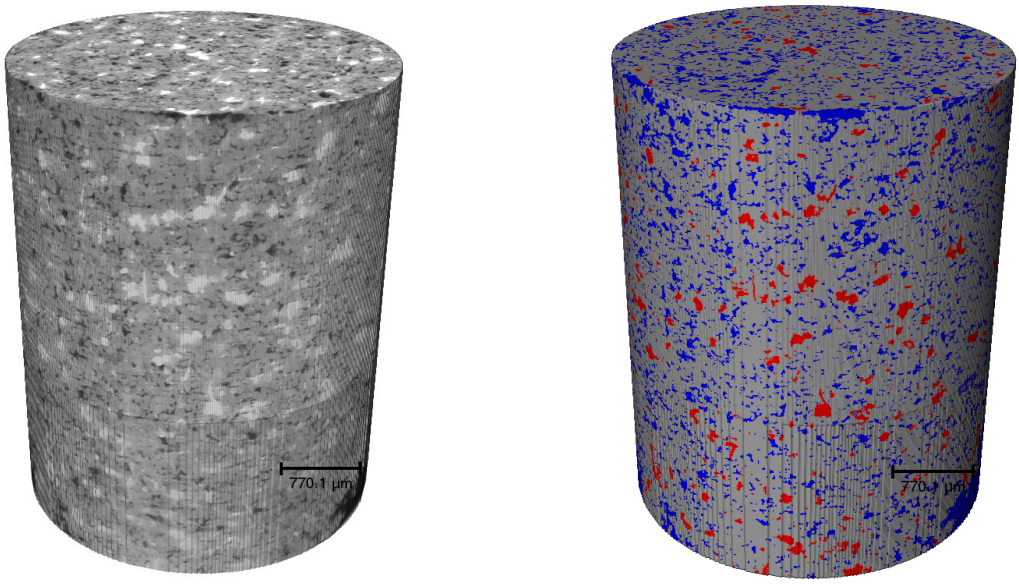


Fig. 8. Images of Bandera Gray sandstone after pressurized H_2 injection showing (a) 3D raw image and (b) 3D segmented image.

4. Conclusions

Sandstone formations are increasingly recognized as favorable geo-storage media for geological hydrogen storage. However, a comprehensive understanding of the rock/brine/H_2 interactions is imperative to determine their suitability for UHS. Prior investigations into these reactions have indicated negligible interactions between hydrogen and sandstone minerals. Building on prior research, our study uniquely examines the impact of clay content in Berea and Bandera Gray sandstones on H2/brine/rock interactions during geological hydrogen storage, an

aspect previously underexplored. We simulated downhole conditions by injecting pressurized hydrogen into brine-saturated cores at 75°C and 1500 psi over a period of 75 days, utilizing micro-computed tomography to analyze the core samples' pore structures and morphology before and after treatment.

The experiments revealed no discernible morphological changes or porosity alterations in Berea and Bandera Gray sandstones, underscoring the inert nature of sandstone—even with a substantial clay presence—when interacting with hydrogen. Our findings suggest that clay-rich sandstone formations offer a more stable option for hydrogen storage than previously thought, paving the way for more efficient and sustainable energy storage solutions. Accordingly, we conclude that clay-rich sandstone formations are promising candidates for hydrogen storage. The potential for injectivity impairment or clay precipitation appears minimal within the short-term storage duration assessed. These findings hold significant potential for enhancing our understanding and selection of optimal porous media for industrial-scale hydrogen storage. Given the minimal changes observed in the sandstone formations, our research indicates that these materials could be effectively utilized in industrial-scale hydrogen storage, supporting the transition to cleaner energy sources.

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