

X-ray Core-Flooding Experiments to Study H₂ and Cushion Gas Residues in Highly Permeable Sandstone Formations

Introduction

Sandstone formations have been proven suitable for carbon dioxide (CO₂) and natural gas (CH₄) storage applications, which can be useful for evaluating their suitability for hydrogen storage (Reitenbach et al., 2015; Tarkowski 2017). Nonetheless, given the distinct physical and chemical properties of hydrogen such as molecular weight, density, viscosity, and diffusivity compared to CO₂ and CH₄, more studies are needed to understand the behavior of hydrogen during storage applications. Recent studies strongly suggested that sandstone rocks have low reactivity to hydrogen, for instance, only minor changes were reported in Buntsandstein sandstone after 6 months of H₂ injection, suggesting very low interactions between hydrogen and rock minerals (Ebrahimiyecka 2017). Similarly, Flesch et al. (2018) observed minor alterations in sandstone petrophysical properties after 6 weeks of hydrogen treatment, suggesting no reactivity with hydrogen. Even at elevated pressure (1450-2900 psi) and temperature (200° C) conditions, the reactivity of sandstone is found limited suggesting a low risk of hydrogen loss (Yekta et al., 2018; Hassanpouryouzband et al., 2022). Geochemical models also predicted that no significant reactivity between hydrogen and sandstone rocks could occur due to the natural stability of silicate minerals (Labus and Tarkowski, 2022).

The combination of X-ray scanning with core-flooding offers significant advantages in scanning samples to accurately detect the saturation levels of brine, as well as gas saturation and residuals. This is crucial for assessing the suitability of subsurface formations, particularly during hydrogen injection and withdrawal cycles. Other methods employed in core-flooding experiments often entail high uncertainty when calculating saturation profiles, particularly during multi-phase flow scenarios. For instance, Jackson et al. (2020) conducted steady-state core flooding experiments utilizing micro-CT imaging on sandstone rocks. While this method proves useful for immiscible fluid applications, uncertainties arise during gaseous flow. Additionally, multiple micro-CT images are necessary to precisely capture fluid saturation profiles. Furthermore, our method applies vertical orientation of the core holder, which allows for better control over low flow rates during gas flooding and gravitational drainage effects, aspects not typically addressed in conventional core-flooding systems. However, one of the disadvantages of this method is that it is only suitable for two-phase fluid flow, where one phase is liquid (e.g., brine). This is because the X-ray sensor can only detect liquid phase (i.e. brine), which makes this technique unsuitable for gas-gas flow scenarios.

This study stands out from previously performed studies due to three distinctive features. Firstly, it offers an experimental assessment of gas flow characteristics in sandstone reservoirs across various gas types. Secondly, the incorporation of the X-ray technique assists in evaluating the gas saturations and gas residuals after the core-flooding experiment. Lastly, it will assist in understanding the role of capillary pressure and gas displacement efficiency during hydrogen storage.

Method

The x-ray gas core-flooding experiments are designed to study the gas residuals and displacement efficiency in sandstone reservoirs. The core sample was placed inside the core holder and kept in a vertical position. Vertical orientation is ideal for controlling the low flow rate during gas flooding and controlling gravitational drainage effects. Also, this setup orientation mimics the fluid flow from an injection well to a production well. The integrated software was used to pressurize the system; the pore (injection) pressure was kept constant at 500 psi, to ensure gas phase injection during the experiment so that all gases can be compared fairly. A confining pressure of about 1500 psi was set, which was always higher than the pore pressure, to avoid fluid bypassing the core sample, while the back pressure was set at 500 psi. All experiments were conducted at room temperature of 25 °C. The core holder was vacuumed before conducting the experiments to remove trapped air inside the sample and the core holder.

Figure 1 shows the flowchart of the experiment in steps. Upon pressurizing the system, the first step was saturating the sample with the seawater brine. The brine was injected from the bottom of the core

holder using the pump at a flow rate of 0.1 cc/min to ensure full saturation of the sample. In the second step, the pump was used to inject the specified gas into the fully saturated sample at a constant flow rate of 2cc/min. A similar injection rate was used during all experiments, therefore any observed differences in displacement behaviors and gas residual would be attributed to the gas type and not the rock or flow properties. Five pore volumes of each gas were injected, which was sufficient to achieve stabilized gas saturation. In the next step, the X-ray was applied to detect gas saturation within the sample. In step number 4, the same seawater brine was re-injected into the core sample, then X-ray was applied in the last step to get the gas residual. The X-ray transparent records the software the brine saturation throughout the sample as a function of distance. The experiments were repeated several times to ensure reliable and concise results as much as possible.

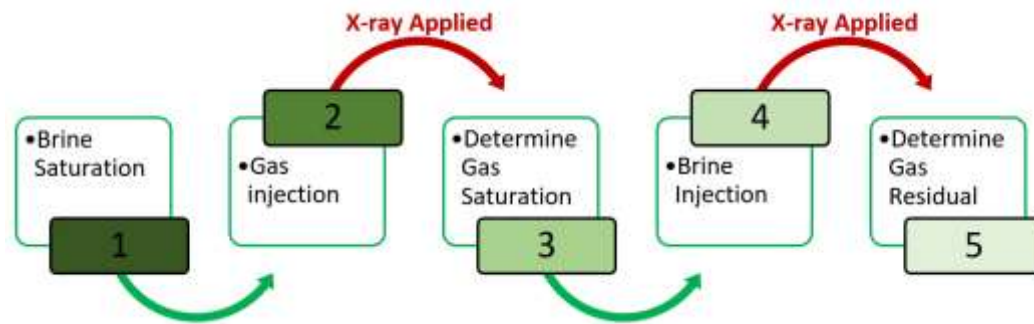


Figure 1 Flowchart showing the work procedure. Red arrows indicate the integration of X-rays at this step.

Conclusions

- 1- The used sandstone sample from Bentheimer formations displayed high consistency in the pore size distribution with macropores dominance and initial water-wet characteristics. However, the capillary pressure profile showed low brine saturation at only 200 psi, which describes the very low capillary pressure in the sample due to the high permeability and large pore radius, despite the strong water-wetting behavior of the sample.
- 2- Gas core-flooding experiments showed that CO₂ exhibited slightly the highest average gas saturation by $\approx 40\%$, while the average gas saturation for H₂ and CH₄ were almost similar ranging between 25-30%. However, the gas residuals in the sample were zero for all gases. These outcomes suggest that the gas residuals in this sandstone sample are unaffected by the gas type, indicating the high potential for gas recovery (especially for hydrogen during withdrawal cycles), and the high displacement efficiency in sandstone rocks.
- 3- The reported zero gas residuals indicate that the wettability of the rock did not change by gas injection, and it appears that the wettability and capillary pressure are not the controlling factors for gas storage in this sandstone sample. This finding suggests that the structural trapping mechanism appears to be the dominant trapping mechanism for shallow high-permeable sandstone rock.
- 4- The calculated capillary numbers at constant flowrate (2 cc/min) were in very low values ($\times 10^{-8}$), suggesting that all gases entered the pores immediately during the injection, and acted as a non-wetting phase. This outcome suggests that the usage of other gases such as CO₂ and CH₄ as cushion gases may not be necessary due to the low capillary pressure and high permeability in this sandstone sample.

References

- Reitenbach V, Ganzer L, Albrecht D, Hagemann B (2015) Influence of added hydrogen on underground gas storage: a review of key issues. *Environ Earth Sci* 73:6927–6937.
- Tarkowski R (2017) Perspectives of using the geological subsurface for hydrogen storage in Poland. *Int J Hydrogen Energy* 42:347–355.

- Ebrahimiyehta A. (2017) Characterization of geochemical interactions and migration of hydrogen in sandstone sedimentary formations : application to geological storage. Université d'Orléans, 2017.
- Flesch S, Pudlo D, Albrecht D, Jacob A, Enzmann F (2018) Hydrogen underground storage— Petrographic and petrophysical variations in reservoir sandstones from laboratory experiments under simulated reservoir conditions. *Int J Hydrogen Energy* 43:20822–20835.
- Yekta AE, Pichavant M, Audigane P (2018) Evaluation of geochemical reactivity of hydrogen in sandstone: Application to geological storage. *Applied Geochemistry* 95:182–194
- Hassanpouryouzband A, Adie K, Cowen T, Thaysen EM, Heinemann N, Butler IB, Wilkinson M, Edlmann K (2022) Geological Hydrogen Storage: Geochemical Reactivity of Hydrogen with Sandstone Reservoirs. *ACS Energy Lett* 2203–2210.
- Labus K, Tarkowski R (2022) Modeling hydrogen – rock – brine interactions for the Jurassic reservoir and cap rocks from Polish Lowlands. *Int J Hydrogen Energy* 47:10947–10962.
- Jackson SJ, Lin Q, Krevor S (2020) Representative Elementary Volumes, Hysteresis, and Heterogeneity in Multiphase Flow From the Pore to Continuum Scale. *Water Resour Res.*
-