https://doi.org/10.36375/prepare_u.iiche.a407

[International Conference on Energy Transition: Challenges and Opportunities at Chemical Engineering Congress (CHEMCON-2023) – On the occasion of Platinum Jubilee Year of Indian Institute of Chemical Engineers]

# Hydrogen Geo-storage – A Review on Storage and Recovery from Carbonate Reservoirs

Anooja Sara Mathew[a], Vishnu Chandrasekharan Nair[b]

[a]*Research Scholar, Deysarkar Centre of Excellence in Petroleum Engineering, IIT Kharagpur – 721302, India*

[b]*Assistant Professor, , Deysarkar Centre of Excellence in Petroleum Engineering, IIT Kharagpur – 721302, India*

## Abstract

Hydrogen has emerged as a promising renewable alternative for addressing our energy needs and advancing towards the goal of achieving net-zero carbon emissions. Large scale generation of hydrogen is a way forward to the storage of renewable energy and securing the energy economy for a future perspective. Within this context, underground hydrogen storage in depleted reservoirs, saline aquifers and salt caverns have garnered increasing attention due to its potential to securely and cost-effectively store hydrogen on a large scale. However, a primary challenge in the domain of hydrogen geo-storage lies in achieving efficient hydrogen extraction from porous media after extended storage periods. In an ideal scenario, the volume of hydrogen recovered should equal the volume initially injected. Due to their abundance and suitability of geological features for storage, formations, once served as a source of fossil fuels are being explored as potential sites for the geo-storage of hydrogen. Although there are many wettability studies on sandstone reservoirs, limited number of studies are available for carbonate rocks. Hence this study provides qualitative insights into the hydrogen trapping efficiency as well as effective recovery from depleted carbonate reservoirs. Also, the challenges associated with the storage phenomena in depleted reservoirs has been addressed here.

*Keywords: Hydrogen Geo-storage; Depleted Reservoirs; Carbonate formations; Residual Hydrocarbon Saturation.*

## 1. Introduction

The rising demand for energy due to increase in population along with the policies for decarbonization have pushed the industries and researchers to develop alternate sources of carbon free energies. Hydrogen has evolved from this as a potential carrier of energy as well as means for storage of renewable energy for future. The production and consumption of renewable energies fluctuate and in order to maintain a continuous flow of energy upon varying demands, storage of the same is required. The idea behind green hydrogen is the production of hydrogen from surplus renewable energies through the process of electrolysis. In 2021, hydrogen demand soared to 94 million tonnes (Mt), exceeding the levels before pandemic (91 Mt in 2019) and contributing to about 2.5% of the total energy consumption of the world. As revealed during the recent COP26 summit in Glasgow, hydrogen plays a pivotal role in the global vision for 2050, with nearly 30 countries unveiling their hydrogen roadmaps [1]. For the Indian Government also, the hydrogen economy is an important consideration. In the budget speech of 2021, the finance minister launched the National Hydrogen Mission for producing hydrogen from green power sources [2]. As India aspires to attain energy self-sufficiency by 2047 and reach a state of Net Zero emissions by 2070, we acknowledge the vital significance of Green Hydrogen. India, endowed with abundant renewable energy resources, is well-positioned to manufacture Green Hydrogen on a global scale. The National Green Hydrogen Mission is designed to outline a thorough strategy for building a Green Hydrogen ecosystem and

https://doi.org/10.36375/prepare_u.iiche.a407

stimulating a coordinated approach to address the prospects and hurdles within this emerging sector [3].

The density of hydrogen is approximately 8 times less than that of methane and 22 times less than that of carbon dioxide. This implies that achieving the same mass of hydrogen gas will create the need for more storage space and higher pressure[4]. Once it is massively produced, comes the requirement of large volumes for storage of hydrogen. Hence comes the idea of geo-storage of hydrogen as surface storage methods have practical difficulties. Currently, there are four operational methods for underground natural gas storage: salt caverns, exhausted gas fields, aquifers, and hard rock caverns equipped with linings [1]. These sites are being explored as the potential storage options for hydrogen underground. Fig.1. shows the hydrogen economy including generation, transportation and underground storage of energy from the excess electricity from the renewables and the recovery of hydrogen during peak energy demands.

The chosen criteria for hydrocarbon reservoirs and saline aquifers encompass the following factors: reservoir capacity, the composition of the overlying rock layers, geological activity, depth of occurrence, exploration progress, mineral composition, and the thickness of the overlying rock formations [5]. The lower viscosity of hydrogen compared to methane and carbon dioxide poses a significant challenge for its containment during geological storage. Additionally, the high mobility and extensive diffusion of $H_2$ result in the reduction of residual amount of $H_2$ remaining within the porous media during extraction [4]. Even though it is important to highlight that Underground Hydrogen Storage (UHS) can leverage the well-developed technologies and extensive knowledge base associated with geological storage of natural gas and carbon dioxide which have been established over several decades, more experimental investigations have to be done with different combinations of existing and expected reservoir conditions in this area which is still at its infancy to evolve.

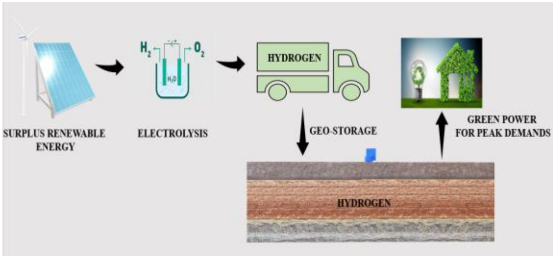


**Figure.1:** Hydrogen Economy

## 2. Depleted Reservoirs for Hydrogen Geo-storage

In this era of extensive exploration into large-scale green hydrogen production methods, the quest for viable storage solutions for green hydrogen has also garnered significant interest. Due to the knowledge of existing infrastructure for the extraction of hydrocarbons, depleted hydrocarbon reservoirs serve as the best option among all the practiced geo-storage options for hydrogen. According to the capital cost assessment, depleted oil and gas reservoirs are found to be the most cost-effective storage option, with an estimated cost of approximately $1.29 per kilogram [6].

Oil and gas reservoirs are well known for their vast area, high porosity, reservoir tightness and the presence of impermeable caprock that seals the contained gas. These properties are making them the suitable candidates for underground hydrogen storage [5]–[7]. Geological traps are where natural gas and petroleum deposits are found. Hydrocarbons get stored within the pore spaces of rocks like carbonates or sandstones, which are referred to as reservoirs. The rocks that act as seals for these traps prevent hydrocarbons from migrating excessively. These are the most prevalent types of porous geological underground materials and have good history of natural gas extraction operations over a century [5]. Most of the hydrocarbon reservoirs are sandstone and carbonate reservoirs. Study of different combinations of reservoir conditions and parameters have been conducted on the hydrogen storage in sandstone reservoirs but, there exists a lack of data in the storage of hydrogen in carbonate formations. Hence this paper mainly focuses on the studies conducted on carbonate reservoirs in the context of hydrogen geo-storage.

One of the parameters that plays a pivotal role in the selection of sites for hydrogen storage in

https://doi.org/10.36375/prepare_u.iiche.a407

underground sites is wettability. It's characterized by the ability of the fluid to stay in contact with a solid surface when other immiscible fluids are present. The wettability of hydrogen within reservoir rock relies on the attributes of the reservoir rock, the fluid in the pores, and hydrogen. This wettability can be predicted by assessing the contact angle through measurements [7]. Assessing the potential of hydrogen geo-storage from a wettability perspective is an emerging research area that requires thorough investigation. Due to the lack of literatures to quantify the $H_2$ wettability of carbonate reservoirs, Hosseini et al.(2022) conducted study on calcite minerals and found out high temperatures, low pressures, low organic acid concentrations and low salinity as the suitable conditions for reducing the risk of hydrogen storage projects in the carbonate reservoirs[8]. Their results also show that at standard conditions, the calcite/hydrogen/water system remained strongly water wet which became intermediately wet with higher physical and thermal conditions. This aligns with the results of the study conducted by Aghaei et al. (2023) where the wettability of carbonate rocks was not altered due to the presence of hydrogen instead the surfaces remained water wet[9]. Their work suggests the future need of studies on geochemical interactions of $H_2$-rock brine system, snap-off and residual trapping of $H_2$ that may have influence in the injectivity and recovery efficiencies. Atomistic molecular dynamics simulations were conducted by Yaseri et al. (2023) to investigate the wettability of the hydrogen-water system on quartz and calcite minerals. These simulations were executed using the high-performance open-source software, GROMACS. Sandstone and limestone reservoirs exhibited complete water-wettability (contact angle of 0 degrees) when exposed to hydrogen, irrespective of variations in pressure, temperature, or salinity. They suggested to further validate the wettability outcomes and recommended to explore $H_2$ core flooding under diverse pressure, temperature, and salinity conditions in future research [10].

The recovery of hydrogen from the underground storage sites upon the varying demand is as much important as its containment. In an ideal condition, the recovered volume of hydrogen should be equal to the injected volume. Losses of different kind can occur during the short - term and long - term storage of hydrogen due to rock-fluid interactions. The interfacial tension (IFT) between the reservoir rock and the fluid governs how the fluid spreads, thereby influencing the interaction between hydrogen and the rock [7]. The measurement of IFT through experimental studies are challenging due to the limitations in measuring the interfacial tension between the rock surface and the liquid or gas fluid phases. Only the IFT between liquid and the gas can be measured in laboratory. Another work of Hosseini et al. (2023) have combined the Neumann's equation and Young's equation to determine the rock – fluid interfacial tension in a carbonate formation. Their studies found out the IFT between calcite/H2/water as a function of temperature, pressure, organic acid concentrations and salinity. The study results emphasize the importance of interfacial tension between the rock and the fluid phases in the context of UHS. The value of IFT was found to decrease with increase in pressure, salinity and organic acid concentration, but increased with temperature [11].

Hou et al. (2023) measured the zeta potential on the surface of carbonate rocks in different salinity solutions in order to find the effect of divalent and monovalent ions on alteration of hydrogen wettability. They also studied the effect of pressure and temperature parameters on the advancing contact angles and receding contact angles in a system of carbonate rock/ hydrogen/ brine [12]. The results indicated the increase in contact angle caused by the divalent ions which showed an increased zeta potential. Also, it was seen that the contact angle increased with pressure and decreased with temperature.

## 3. Challenges Associated with Hydrogen Geo-storage in Depleted Reservoirs

Many reservoirs which once served as the potential source of hydrocarbons have now been considered as the suitable storage options for hydrogen. Even though the presence of existing gaseous hydrocarbon mixtures in the depleted reservoirs are considered as the beneficial component of cushion gas which maintains the pressure during the injection and withdrawal stages [4] of hydrogen underground, the presence of the same can affect the purity of hydrogen stored. Not every reservoir identified for underground hydrogen storage needs to be entirely depleted, as some residual gases may persist within it. These native gas remnants within the reservoir have the potential to mix with hydrogen. The degree to which multiphase, multicomponent interactions occur within the reservoir can introduce contaminants into the stored hydrogen. To ensure the secure operation of UHS, it is imperative that the reservoir is devoid of contaminants [5]. However, the residual oil saturations if present in these reservoirs are detrimental as it interacts with the stored hydrogen. The dynamic interaction between the remaining oil saturation and hydrogen storage involves a spectrum of intricate procedures, encompassing factors such as competition for pore space, capillary entrapment, and limitations related to diffusion. Oil fields are prone to exhibit intricate multiphase fluid flow dynamics when interacting with hydrogen, leading to reduced utilization of pore space and increased storage expenses. Additionally, they lack the advantage of pre-existing cushion gas [13]. Hence, the studies considering the effect of oil in UHS are very limited in numbers. Bechara et al. (2023) have conducted studies on the effect of residual oil saturations on unconventional shale reservoirs during the injection, storage and recovery of hydrogen from them. These experimental studies have found out that there is possibility of the production of unwanted amount of oil along with the extraction of hydrogen which indeed creates the need of additional separation procedures for hydrogen. They have also found an additional increase in weight of oil saturated sample on exposure to hydrogen indicating the formation of some chemical compounds [14], [15]. Geochemical aspects of these storage mechanisms have to be properly evaluated under conditions of reservoir temperature and pressure. Also, effects on long term storage of hydrogen has to be studied in future in order to convert depleted oil reservoirs into suitable sites for geo-storage of hydrogen.

Studies conducted by Zeng et al. (2022) on the Majiagou carbonate reservoir in China showed the reduction of hydrogen due to calcite dissolution and geochemical reactions occurring in carbonate rocks. They found out by the geochemical modelling studies that the loss of hydrogen in 0.5 years was about 6.5% and 7.65% at 5 years but, it can reach approximately up to 81.1% over 500 years [17]. This quantifies the suitability of such reservoirs for the short-term storage of hydrogen. Geochemical reactions between rock minerals and the introduced hydrogen may lead to an overestimation of Underground Hydrogen Storage capacity in carbonate formations if not appropriately considered. It is essential to safeguard the stability and integrity of the storage rocks and uphold the formation pressure to guarantee effective hydrogen injection and recovery during peak periods of demand. However, research on the interactions between hydrogen and the reservoir rocks mainly sandstone and carbonate and their impact on the geological storage of hydrogen in these formations is notably scarce. The experimental findings by Yaseri et al. (2023) using micro CT indicated that the expansion of calcite was more pronounced than the dissolution during the initial 75 days of hydrogen injection into carbonate reservoirs, which primarily composed of calcite[16].

The significant impact of hydrogen's subsurface hydrodynamic behavior is a greater concern, particularly when considering depleted reservoirs and aquifers, and it applies universally to all underground hydrogen storage methods. The occurrence of hydrogen in underground areas has also been observed to accelerate geochemical reactions, presenting another concern related to UHS in depleted reservoirs and aquifers [5]. It's important to emphasize that while these capacity assessments are accurate, there's still uncertainty regarding whether hydrogen can be injected into and retrieved from storage reservoirs in a manner similar to natural gas. Consequently, the true hydrogen storage capacity of gas fields remains unconfirmed. Continual effective evaluations of designated storage sites will aid in determining the feasibility of seasonal hydrogen injection and withdrawal. Rigorous risk assessments for abandoned wells and faults, along with thorough dynamic simulations of the hydrogen storage process, can significantly reduce the risk of hydrogen leakage through these potentially vulnerable pathways [13]. However, achieving the effective storage of pure hydrogen in depleted hydrocarbon reservoirs and aquifers has not been accomplished due to the unclear understanding of microbial and geochemical processes during underground hydrogen storage. The storage capacity, as well as the ability to inject and withdraw $H_2$, are influenced by the reactions between hydrogen and the rock within the storage formations [16].

## 4. Conclusion

Hydrogen will continue to increase its demand in the upcoming period of time as a carbon free carrier of energy. With increased demand, increased storage options are also required. Research has identified underground hydrogen storage in depleted reservoirs as a viable and cost-effective choice for hydrogen storage. But due to lack of data in the domains of reactions and interactions of hydrogen under reservoir conditions, it is difficult to predict the efficiency of storage and recovery of hydrogen from UHS sites and it leaves an unclear picture especially in the case of carbonate reservoir. Increased pressure temperature conditions as well as increased periods of storage need to be studied in order to quantify the appropriateness of storage sites. Additionally, residual oil saturations remaining in depleted reservoirs may result in inadvertently retrieving undesirable amounts of oil along with the hydrogen. Thus, understanding the interaction between the remaining oil and stored hydrogen in these reservoirs, and its impact on storage efficiency and subsequent hydrogen recovery, is crucial. Further research in this direction can be expected in future.

## Declaration of Competing Interest

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

## References

[1] INTERNATIONAL ENERGY AGENCY, “Global Hydrogen Review 2022,” OECD, Oct. 2022. doi: 10.1787/a15b8442-en.

[2] Energy Transition Advisory Committee Ministry of Petroleum & Natural Gas Government of India (ETAC), “THE GREEN SHIFT - The low carbon transition of India’s Oil & Gas sector.”

[3] M. of N. and R. E. Goverment of India, “National Green Hydrogen Mission Ministry of New and Renewable Energy,” no. January, 2023, [Online]. Available: https://www.iea.org/reports/india-energy-outlook-2021

[4] N. S. Muhammed *et al.*, “Hydrogen storage in depleted gas reservoirs: A comprehensive review,” *Fuel*, vol. 337, no. September 2022, p. 127032, Apr. 2023, doi: 10.1016/j.fuel.2022.127032.

[5] H. Bin Navaid, H. Emadi, and M. Watson, “A comprehensive literature review on the challenges associated with underground hydrogen storage,” *Int. J. Hydrogen Energy*, vol. 48, no. 28, pp. 10603–10635, Apr. 2023, doi: 10.1016/j.ijhydene.2022.11.225.

[6] E. I. Epelle *et al.*, “Perspectives and prospects of underground hydrogen storage and natural hydrogen,” *Sustain. Energy Fuels*, vol. 6, no. 14, pp. 3324–3343, 2022, doi: 10.1039/D2SE00618A.

[7] M. S. A. Perera, “A review of underground hydrogen storage in depleted gas reservoirs: Insights into various rock-fluid interaction mechanisms and their impact on the process integrity,” *Fuel*, vol. 334, no. August 2022, p. 126677, Feb. 2023, doi: 10.1016/j.fuel.2022.126677.

[8] M. Hosseini, J. Fahimpour, M. Ali, A. Keshavarz, and S. Iglauer, “Hydrogen wettability of carbonate formations: Implications for hydrogen geo-storage,” *J. Colloid Interface Sci.*, vol. 614, pp. 256–266, May 2022, doi: 10.1016/j.jcis.2022.01.068.

[9] H. Aghaei, A. Al-Yaseri, A. Toorajipour, B. Shahsavani, N. Yekeen, and K. Edlmann, “Host-rock and caprock wettability during hydrogen drainage: Implications of hydrogen subsurface storage,” *Fuel*, vol. 351, no. April, p. 129048, Nov. 2023, doi: 10.1016/j.fuel.2023.129048.

[10] A. Al-Yaseri, S. Abdel-Azeim, and J. Al-Hamad, “Wettability of water-H2-quartz and water-H2-calcite experiment

and molecular dynamics simulations: Critical assessment," *Int. J. Hydrogen Energy*, vol. 48, no. 89, pp. 34897–34905, Nov. 2023, doi: 10.1016/j.ijhydene.2023.05.294.

[11] M. Hosseini, M. Ali, J. Fahimpour, A. Keshavarz, and S. Iglauer, "Calcite–Fluid Interfacial Tension: H 2 and CO 2 Geological Storage in Carbonates," *Energy & Fuels*, vol. 37, no. 8, pp. 5986–5994, Apr. 2023, doi: 10.1021/acs.energyfuels.3c00399.

[12] J. Hou, S. Lin, M. Zhang, and W. Li, "Salinity, temperature and pressure effect on hydrogen wettability of carbonate rocks," *Int. J. Hydrogen Energy*, vol. 48, no. 30, pp. 11303–11311, Apr. 2023, doi: 10.1016/j.ijhydene.2022.05.274.

[13] J. Mouli-Castillo, N. Heinemann, and K. Edlmann, "Mapping geological hydrogen storage capacity and regional heating demands: An applied UK case study," *Appl. Energy*, vol. 283, no. August 2020, p. 116348, Feb. 2021, doi: 10.1016/j.apenergy.2020.116348.

[14] E. Bechara *et al.*, "Effect of Hydrogen Interaction with Crude Oil on the Extraction of Hydrogen if Stored in Depleted Shale Reservoirs: An Experimental Study Analyzing the Outcome of Cyclic Hydrogen Injection into Oil-Saturated Core Samples," in *Day 2 Thu, June 29, 2023*, SPE, Jun. 2023. doi: 10.2118/213851-MS.

[15] E. Bechara, T. Gamadi, A. Hussain, and H. Emadibaladehi, "Effect of Hydrogen Exposure on Shale Reservoir Properties and Evaluation of Hydrogen Storage Possibility in Depleted Unconventional Formations," in *Proceedings of the 10th Unconventional Resources Technology Conference*, Tulsa, OK, USA: American Association of Petroleum Geologists, 2022, pp. 1–15. doi: 10.15530/urtec-2022-3723858.

[16] A. Al-Yaseri, H. Al-Mukainah, N. Yekeen, and A. S. Al-Qasim, "Experimental investigation of hydrogen-carbonate reactions via computerized tomography: Implications for underground hydrogen storage," *Int. J. Hydrogen Energy*, vol. 48, no. 9, pp. 3583–3592, Jan. 2023, doi: 10.1016/j.ijhydene.2022.10.148.

[17] L. Zeng, A. Keshavarz, Q. Xie, and S. Iglauer, "Hydrogen storage in Majiagou carbonate reservoir in China: Geochemical modelling on carbonate dissolution and hydrogen loss," *Int. J. Hydrogen Energy*, vol. 47, no. 59, pp. 24861–24870, Jul. 2022,doi:10.1016/j.ijhydene.2022.05.247.