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Towards a fundamental understanding of geological hydrogen storage

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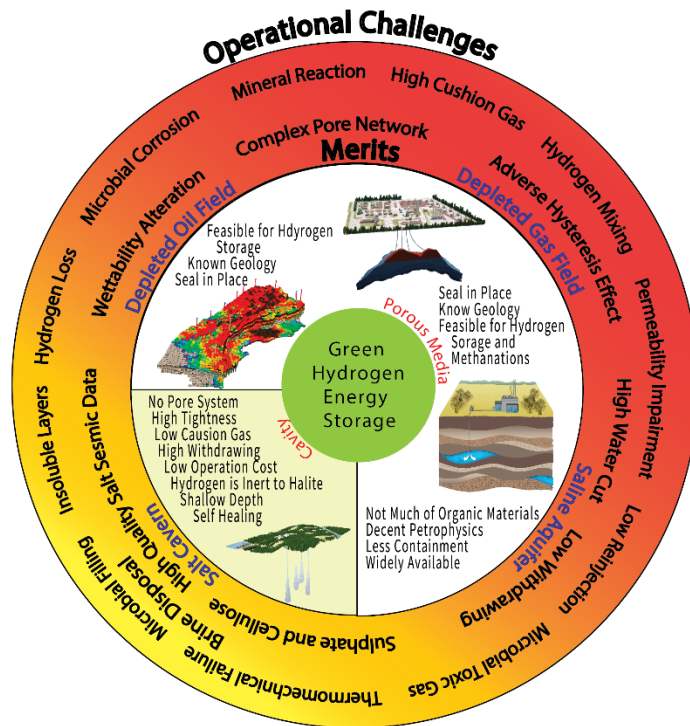
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28 **Table of content entry**



29

30 This review is unique in its presentation and reported distinctive information which could
 31 attract scientific community to devise an optimized plan for geological hydrogen storage
 32 industry.

33

34 Abstract

35 Geological H_2 storage plays a central role to enable the successful transition to the renewable
36 H_2 economy and achieve net-zero emission in the atmosphere. Depleted oil and gas
37 reservoirs are already explored with extensive reservoir and operational data. However,
38 residual hydrocarbons can mix with injected H_2 in the reservoirs. Furthermore, low density
39 and high diffusivity of H_2 may establish H_2 leakage from the reservoirs via fault pathways.
40 Interestingly, H_2 can be consumed by microorganisms, which results in pore-network
41 precipitation, plugging and partial permeability impairment. Therefore, stored H_2 may be lost
42 in the formations if storage scenario is not planned cautiously. While salt caverns are safe and
43 commercially proven geo-rock for H_2 storage, they have low-storage capacity compared to
44 depleted gas reservoirs. Moreover, salt structures (e.g., domel, bedded) and microorganisms
45 activities in the salt cavern are limiting factors, which can influence the storage process.
46 Accordingly, we discuss challenges and future perspectives of hydrogen storage in different
47 geological settings. We also highlight geographical limitations with diverse microbial
48 communities and theoretical understanding of abiotic transformation (in terms of rock's
49 minerals i.e., mica, and calcite) for geological H_2 storage. Regarding the fundamental behavior
50 of H_2 in the geological settings, it is less soluble in formation water, therefore it may achieve
51 less solubility trapping compared to CO_2 and CH_4 . Furthermore, H_2 gas could attain higher
52 capillary entrance pressures in porous media over CH_4 and CO_2 due to higher interfacial
53 tension. Additionally, the low viscosity of H_2 may facilitate its injection and production but H_2
54 may establish the secondary trapping and viscous fingering. Thus, this review documented a
55 blend of key information for the amendment of subsurface H_2 storage at an industrial scale.

56 **Key words:** Energy transformation, Hydrogen, geological, storing, reproducing, net-zero

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59

60

61 1 Introduction

62

63 In 2020, the world's primary energy consumption and carbon emissions from energy use are
64 declined by 4.5% and 6.3%, respectively. This was mainly pushed by a record reduction in oil
65 consumption (9.1 million barrels per day) and considerable falls in natural gas use by 2.3%¹.
66 In contrary to this, the energy demand growth rate was positive in 2019². The rise of COVID-
67 19 and the uncertain schedule of partial/full lockdowns constrained the economical growth ³
68 which results in a decline in the global demand for coal (down to 1.7 %) in 2019 ². Moreover,
69 the demand for natural gas decreased to 2%, and oil demand reduced to 20% in 2020 Q1 ⁴.
70 Overall, CO_2 emission went down (which was 2.1% in 2018, and 0.5% in 2019) due to the
71 massive impact of COVID-19 onto the consumption of fossil fuels around the globe. The
72 negative trend of CO_2 emission was observed in different regions such as Europe (-8%), China
73 (-8%), and US (-9%) ². Nevertheless, the global primary energy demand is 84% and still would
74 be delivered by fossil fuels. CO_2 emission growth by 0.5 % cannot be persistent anymore
75 because developed nations will once again gain momentum to retain their position in the
76 global energy supply market ².

77 European Union Energy Security and Strategy (EUESS) affirmed that the suppression of
78 greenhouse gases down to 20 to 30% by 2020 is key to protect climate change and
79 environment. Therefore, there is a pressing need to increase the participation of renewable
80 energy to 20% in the primary global energy demand ⁵. Additionally, Energy Road Map Goals
81 (ERMG) 2050 set objectives to mitigate greenhouse gases emission (GHGE) by 80 to 95% ⁶.
82 Consequently, renewable energy production would be given a priority and 55% of energy
83 would be projected to produce from renewable energy systems alone in Europe by 2050 ⁵.
84 Additionally, South-East Asia has projected to achieve the 35 to 40 GW (35%) of renewable
85 energy target by 2025 using wind and solar energy's lower costs ⁷. In 2020, the US has grown
86 renewable energy consumption, reaching a record high 12% of the total US energy demand
87 ⁸. Renewable energy sources are found better with diverse applications ⁹, in particular,
88 minimum electricity bills ¹⁰, and low maintenance costs ¹¹. For example., solar system is the
89 green transformation of energy from sunlight into power through direct photovoltaic effect
90 or concentrated solar power system ¹². Additionally, the energy from the sun would not run
91 out unlike fossil fuels and it provides more heat energy than our need ¹³. Moreover, solar

92 energy can be produced without emitting CO_2 for commercial production of electricity ^{14, 15}.
93 In the same line, researchers have a concentrated focus on the utilization of wind energy using
94 wind turbines because of its overwhelming merits such as sustainability ¹⁶, cost-effectiveness
95 ¹⁷, and clean energy ¹⁸. Moreover, tidal energy projects were reviewed to optimize electricity
96 production because of their low maintenance cost ^{19, 20}, renewable behaviour ^{21, 22}, zero CO_2
97 emission ²³. In need of renewable energy, energy experts and researchers have been
98 exploring the cost-effective and environmentally friendly solar and wind energy systems
99 based on geographical locations for the generation of green H_2 energy ^{24, 25}. Renewable H_2
100 energy is zero-carbon fuel and can be utilized in fuel cells, internal combustion engines and
101 power generation. H_2 is booming and can contain major energy supply businesses around the
102 globe ²⁶. The research and development community (R&DC) has realised that transformation
103 of energy from fossil fuels economy to hydrogen economy is inevitable because H_2 does not
104 generate GHGE after burning ²⁷. The need of H_2 is considerably increasing and multiplied
105 three times since 1975, and it will keep increasing. Around 98% of H_2 is produced from the
106 fossil fuels sources, including natural gas (6%) and coal (2%) ²⁸. Nevertheless, generation of
107 H_2 from the fossil fuels is causing 830 million tonnes of CO_2 emission per year (sum of total
108 CO_2 emissions of Indonesia and the UK) ²⁹. Figure 1 summarises the sustainable carbon free
109 hydrogen energy system which enable and combine all the phases including renewable
110 energy source, renewable hydrogen generation, hydrogen storage and hydrogen supply.

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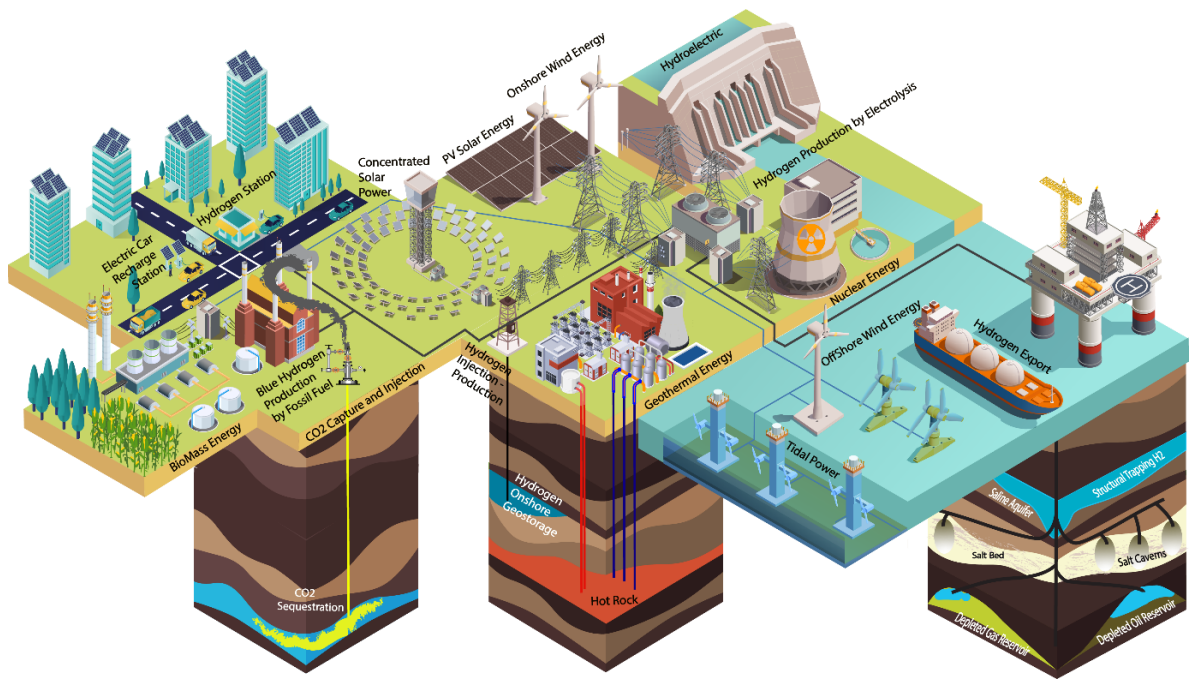


Figure 1 Graphical representation of entire carbon-free and sustainable hydrogen energy production and supply chain mainly comprising H_2 geostorage in depleted gas reservoirs and salt caverns

112

113 Scientific community has made significant progress to generate H_2 using electrolysis reaction
 114 using nanocomposite rods^{30, 31}. The electrocatalysis reaction requires energy thus supply of
 115 energy from solar parks and windmills can be used to produce 'green H_2 '. For example., 9.1
 116 ton of water produced 1.1 ton of H_2 , and 8 ton of O_2 without CO_2 emission using 39.4 MWh
 117 solar energy³². H_2 can be used for passenger cars, prime movers, and buses, nevertheless a
 118 flexible, cost-effective and environmental friendliness storage system is needed so that H_2
 119 can be stored and reproduced adequately to compensate for fluctuating energy demand³³
 120 without impacting the atmosphere. Energy yield of H_2 is higher (e.g., $120 \text{ MJ}\cdot\text{kg}^{-1}$ energy
 121 density)³⁴ than CH_4 ($45 \text{ MJ}\cdot\text{kg}^{-1}$)³⁴. However, surface storage of H_2 is a challenge^{35, 36} because
 122 of the low volumetric density of H_2 which is $0.0838 \text{ kg}\cdot\text{m}^{-3}$ at ambient pressure and 20°C .
 123 Additionally, liquid H_2 with $70.8 \text{ kg}\cdot\text{m}^{-3}$ is not an adequate choice for long-term storage. Liquid
 124 H_2 can continuously be boiled-off resulting inefficient energy storage system and may rise H_2
 125 leakage and H_2 loss problems³⁷.

126 Underground H_2 storage could preclude multiple technical issues that constraint its
 127 widespread use³⁸. For example., residual oil in the depleted reservoirs may influence H_2

128 purity. Moreover, microorganism can consume H_2 which may result in loss of stored H_2 and
129 precipitation in the pore-system due to release of by-products, including H_2S and the acids ³⁹,
130 ⁴⁰ Additionally, deep saline aquifer H_2 storage system can cause the problems of mineral
131 dissolution and high water cut during the withdrawing period of H_2 ⁴¹. Thus, **different**
132 **mechanisms of formation damages can occur due to fines mobilisation and migration**
133 **during hydrogen injection/withdrawal** ⁴²⁻⁴⁴. Nevertheless, H_2 storage in salt caverns is a
134 proven technology ⁴⁵, owing to its inexpensive investment ⁴⁶, enhanced sealing properties ⁴⁷
135 and minimum gas cushion requirements ⁴⁸. However, microorganisms in particular sulphate
136 reducing bacteria (SRB) can develop the risk of H_2S release as a by-product in the salt caverns
137 ⁴⁹. Moreover, H_2S release and dispersive behaviour of H_2 may catalyse the H_2 embrittlement
138 in casing and well completion assembly, which result in H_2 leakage ⁵⁰⁻⁵².

139 Biogeochemical, physiochemical, hydrodynamics and microbial activities can influence the
140 underground H_2 storage ⁵³. Effect of H_2 storage in the sandstone rock was found inert with
141 minimum mineral dissolution equilibria at the variable reservoir temperature conditions.
142 Nevertheless, literature rarely reported the hydrogeochemical effect of H_2 on shale (mica)
143 and carbonate (calcite). Meanwhile, biotic transformation due to microbial activity can
144 influence the gas mixing and propagation of microbial communities in the subsurface
145 formation ⁵³. Importantly, SRB have a significant effect on H_2 storage in the porous media as
146 well as in salt caverns. SRB may consume H_2 gas in their metabolic system and yield gases
147 (e.g., CH_4 , CO_2 , H_2S).

148 Methanogenesis, homoacetogenesis and sulphate reduction are three major processes which
149 lead to H_2 consumption. Among 42 water test samples (depleted gas fields), 32 samples
150 depict microbial activity at >122 °C temperature, and >4.4 M $NaCl$ salinity ⁵⁴. Thus, it was
151 recommended that monitoring of geochemical and microbial activities are essential for the
152 sustainability of H_2 underground storage ³⁹. In the context of storage security and
153 withdrawing capacity, H_2 has a low density, low viscosity and high diffusivity when compared
154 to CO_2 and CH_4 . Thus, these fundamental and thermophysical behavior of H_2 may cause loss
155 of H_2 , and critically impact H_2 withdrawal capacity. But, this could be prevented by cautiously
156 planning the injection and withdrawal scenarios in the porous media ³⁷. Additionally, high
157 bouncy column pressure of H_2 may have a considerable impact on geo-structural trapping ⁵⁵.
158 Therefore, investigators believed that H_2 can be safely stored at initial reservoir conditions in

159 both offshore and onshore depleted gas fields. In depleted gas fields, caprocks could provide
160 sufficient sealing corresponding to high interfacial tension between water+ H_2 and capillary
161 entrance pressures ³⁷.

162

163

164 Figure 2 summarizes the effect of geological formations, and microorganisms on the UGHS in
165 the different mediums, including salt caverns, saline aquifers, and depleted gas reservoirs.
166 Herein, we have discussed both porous reservoir rock (depleted oil/gas, and aquifers) and
167 cavity reservoir rock (salt cavern) and quantified adequate liabilities for H_2 biogeochemical
168 storage security ⁵⁶. To achieve that: (i) H_2 phase behavior, abiotic and biotic geological
169 transformations have been provided which is very limited in the literature. (ii) Application,
170 principles and energy conversion and management of green H_2 have been discussed. (iii)
171 Comparative analysis of depleted oil, depleted gas, saline aquifer reservoirs and salt cavern
172 have been provided relative to storing characteristics, withdrawing capacity, underground
173 microbial life and realistic experience. (iv) Occurrence of possible geological trapping and
174 mechanism have been critically presented. (v) Salt cavern and its H_2 storage capacity,
175 operational challenges, possible solutions and recommendations are comprehensively
176 evaluated and concluded.

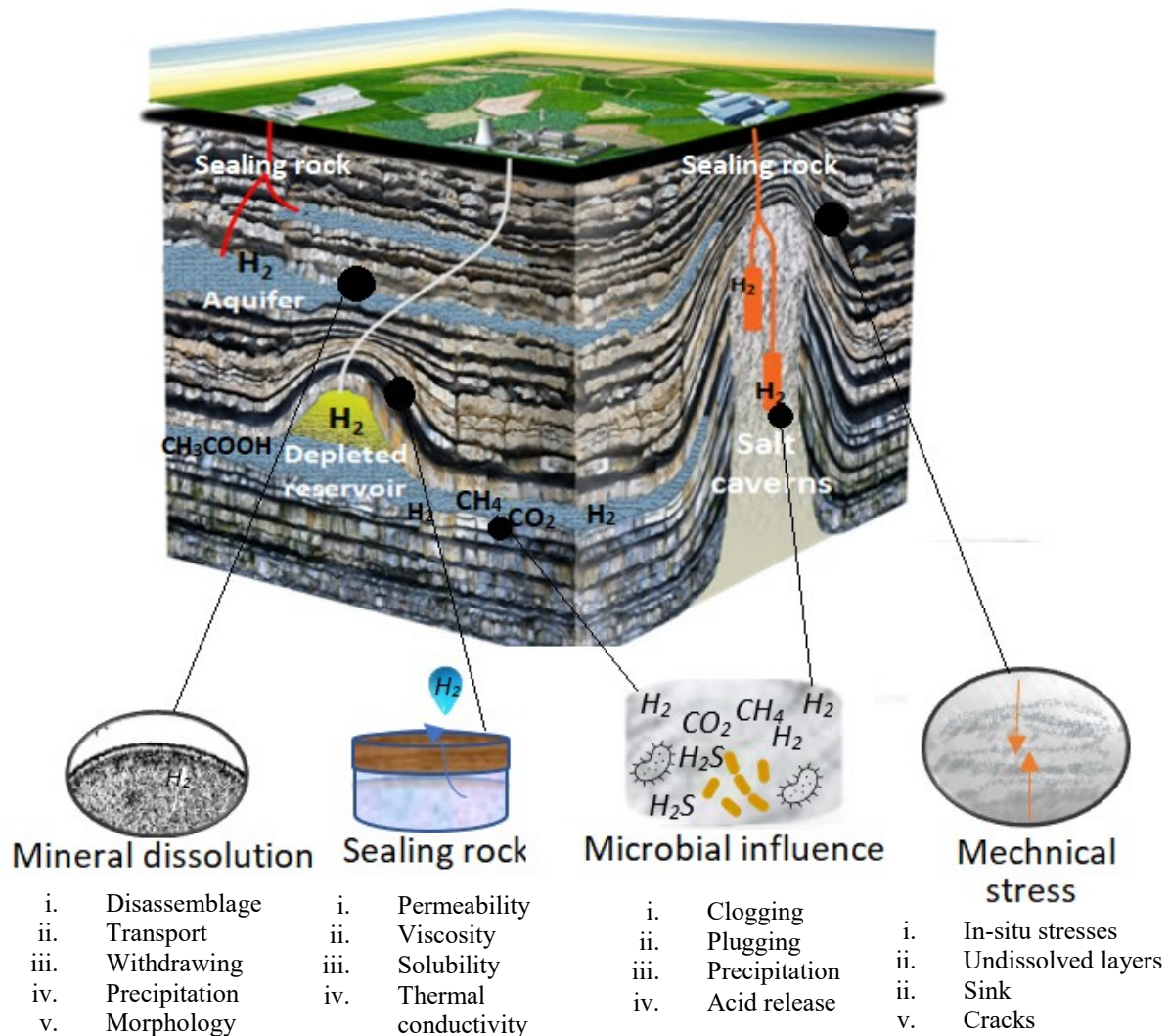


Figure 2 UGHS can be influenced by several factors, e.g., mineralogical alteration, sealing rock ability, microbial influence (natural, indigenous, and anthropogenic) and mechanical stresses

177 **2 Abiotic and biotic transformation of the geological rocks**
 178

179 The geological rocks may be influenced by abiotic (e.g., mineralogy, temperature, pressure,
 180 salinity, and gas composition etc) ⁵⁷⁻⁷⁶ and biotic (both indigenous and anthropogenic
 181 microbial life) transformations in the presence of H_2 ^{39, 54, 77}. The porous formation has a
 182 significant capacity to store H_2 gas ⁷⁸. However, the interactions of H_2 with rock-forming
 183 mineral (units) mainly quartz, calcite, and mica are rarely discussed in the literature, which
 184 can unveil important information regarding H_2 underground storage security at large green-

185 scale (LGS) in depleted reservoir ^{79,80}. H_2 associated geochemical alterations were not
186 observed in sandstone at variable injection pressure (1 to 20 MPa), reservoir temperature
187 (20 to 100 °C) and salinity (0 to 10 weight%) ⁸¹. H_2 gas has a very small molecular size ⁸², is
188 less dense (less dense than air) ⁸³ and has a strong diffusion rate⁸⁴ to such an extent that
189 hydrogen can travel between the structure of clathrate hydrates ⁸⁵. It can easily spread in
190 the formation cracks or natural fractures and migrate up to impact trapping structures ⁴⁹. In
191 fact, H_2 can react and form compounds. H_2 exists in two distinctive oxidation states (+1, -1)
192 thus it can act as oxidising and reducing agents ⁸⁶. H_2 can induce redox reaction and alter
193 mineral assemblage of rock resulting in mineral dissolution and precipitation ⁵³. Certainly,
194 the petrophysical properties (e.g., porosity and permeability) can be influenced by
195 mineralogical transformations and affect H_2 production/injection cycle security.

196 For instance, sulphur species can be easily reduced by H_2 at a very low oxidation state ⁸⁷.
197 Therefore, pyrite (commonly found in the veins of quartz and sedimentary rocks) becomes
198 thermodynamically unstable in the presence of H_2 . Thus, pyrite reduces to pyrrhotite and
199 H_2S as depicted in equation 1 ⁸⁸;






201 To further explore the phenomenon, researchers evaluated that pyrrhotite covered the pyrite
202 particles when the temperature increased from 90 °C to 190 °C in the presence of H_2 at 116
203 psi partial pressure of H_2 . Thus, reduction in pyrite caused sulphide and precipitation in the
204 system and impairment of rock's flowing properties. Importantly, the amount of sulphide has
205 further reduced the pH of the solution which contains HS^- , $H_2S_{(aq)}$, $H_2S_{(g)}$ ⁸⁷ and can cause
206 mineral dissolution. Researchers carried out numerical analysis and experimental studies to
207 obtain geochemical reactivity of H_2 with sandstone which typically contains quartz, K-
208 feldspars, and other rock-forming units. H_2 could not bring any mineralogical variation in the
209 quartz nevertheless very minor alteration was obtained in hematite, and muscovite
210 proportions ⁸⁹. However, iron was released from the muscovite that did not impact the
211 permeability and porosity of the rock. Thus, sandstone was referred to as 'abiotic free
212 mineral' and illustrated no transformation during H_2 storage ⁸⁹. However, the effect of H_2 is
213 yet to be explored on hydrogeochemical alteration of calcite and mica. Moreover, biotic

214 mineral transformations have been noticed in several studies for H_2 storage in the subsurface
215 ^{39, 54, 77}.

216 Subsurface formation holds diverse types of archaea and bacteria, here collectively
217 mentioned as microbes ³⁹. The potential microbial presence was observed $1-17 \times 10^7$ cells ml⁻¹
218 ¹ in formation water samples, which can consume <0.01 to 3.2% of H_2 ⁹⁰. The increasing
219 concentration of injected H_2 in the subsurface may trigger the microbe's growth ⁷⁷.
220 Additionally, pH of formation water and brine may influence microbial growth through
221 metabolism system and redox reaction. Methanogens, homoacetogens, and sulphate
222 reducers are habited to 6.5 to 7.5 pH. However, the growth of most methanogens and
223 sulphate reducers were not found at 4 to 9.5 pH ^{54, 91}. Biotic mineral transformation can
224 adversely affect gas withdrawing, gas injection, formation permeability reduction and H_2 loss
225 ⁷⁷. SRB, methanogens and IRB can consume inorganic kosmotropic components include *NaCl*,
226 *KCl* and sulphate sources (e.g., *Na₂SO₄*, *FeSO₄*, *K₂SO₄*, *S*) in the presence of stored H_2 and
227 metabolically release by-products (e.g., H_2S , acids, CH_4 , and CO_2) and ensured acidic
228 behaviour ⁹². SRB has induced carbonate precipitation and influenced the contamination
229 security ⁹³. Most SRB including halophilic are permissive to growth and could induce stress in
230 the presence of O_2 and high salinity. Additionally, SRB consumes sulphate (as an electron
231 acceptor) in the presence of H_2 (as an electron donor). Additionally, H_2 energy-based
232 microorganisms lead by methanogens archaea ⁹⁴. Methanogens consumed CO_2 and H_2 in the
233 presence of inorganic minerals components e.g., nitrite, sulphate and iron oxide and
234 metabolically released CH_4 and water ⁹⁵. Additionally, homoacetogens coupled H_2 oxidation
235 to CO_2 reduction resulting acetate acid ⁹⁶, which can cause corrosion problems ⁹⁷.
236 Additionally, variation in gas composition, clogging near injection wellbore, and biofilm
237 growth have been observed nevertheless growth of microbes and their influence on the
238 petrophysical property are not reported ^{97, 98}. However, chaotropy agents (e.g., *MgCl₂*,
239 *FeCl₃*, *CaCl₃*, *LiCl*, and *LiBr*) have ability to limit microbial life in the depleted oil/gas
240 reservoirs, saline aquifers, and salt caverns for H_2 storage projects. Table 1 presents the
241 influence of microorganism reactions over H_2 storage security in equations 2 to 5.

242

243 Table 1 Biotic transformation and impacts for H_2 geostorage

Hydrogenotrophic microbes	Reaction and possible mineralization	Impact	Study
SRB (Bacteria) 	$SO_4^{2-} + 4H_2 + 2H^+ \rightleftharpoons H_2S + 4H_2O \quad 3$ <p>H_2 reacts with anhydrite and other inorganic sulphate sources and reduces sulphate to sulphide.</p>	<ul style="list-style-type: none"> • H_2S release • High sulphide release • Gas mixing • Iron corrosion • pH reduction • Hydrogen embrittlement • Mineral precipitation 	99-107
Iron reducing bacteria (Bacteria) 	$3Fe_2O_3 + 5H_2 \rightleftharpoons 2Fe_3O_4 + H_2O \quad 4$ <p>Microbes can reduce passive film, e.g., ferric components on metal surfaces.</p> <p>By-product water can release and occupy interstitial pore space of sandstone causing excess water saturation and mineral dissolution</p>	<ul style="list-style-type: none"> • Low sulphide reduction • Metal corrosion • Carbon steel corrosion • Mineral dissolution 	108, 109
Methanogens (Archaea) 	$CO_2 + 4H_2 \rightleftharpoons CH_4 + 2H_2O \quad 5$ <p>In an exceptional case, microbes attach to the edges of kaolinite clay and prevent CH_4 flow. Aluminium ions from kaolinite can be toxic to methanogens growth.</p>	<ul style="list-style-type: none"> • CH_4 flow suppresses in Kaolinite • Injection reduces • Gas mixing • Withdrawing and injection capacity reduces • Permeability and porosity can alter 	110-112

244

245 Overall, H_2S increased the acidic behaviour in the porous media and causing mineral
 246 precipitation ¹¹³, and formation clogging ¹¹⁴. H_2 reinjection would be challenging, if the rate
 247 of mineral precipitation is higher than the rate of mineral dissolution in the subsurface ⁵³.

248 Literature hardly provides information regarding biogeochemical behavior of microbes with
249 rock-forming minerals (e.g., mica, and calcite) in the presence of H_2 under reservoir
250 conditions ^{39, 53, 55}.

251 **3 Review of H_2 geological storage in depleted oil/gas, and saline aquifer**

252
253 H_2 storage in the depleted oil systems seems to be adequate because there is already
254 availability of natural space to be filled ¹¹⁵. Moreover, the location of cap rock is already
255 identified during seismic survey with reliable structural and sealing depth information ¹¹⁶. The
256 geological and mineralogical data of depleted oil and gas reservoirs are already known from
257 well-logging and drilled cuttings ¹¹⁷. Furthermore, a great deal of information relative to
258 behaviour depleted oil reservoirs could be available to plan and design the feasibility of H_2
259 storage ¹¹⁸. Additionally, it has been evaluated that one giant depleted gas field can restrain
260 sufficient seasonal H_2 storage capacity for most of the countries around the globe. For
261 example., North Sea Lemen field in the UK has 833 TWh storage capacity which is sufficient
262 to fulfil the entire seasonal energy demand ³⁷.

263 Nevertheless, in depleted oil and natural gas reservoirs, a slow seasonal process can be
264 achieved and therefore continuous turnover is difficult because of multiphase flow in the
265 porous media ³⁹ and overall performance of flexibility of H_2 storage and withdrawal capacity
266 are slightly fair in the depleted oil which present different types of reservoirs including saline
267 aquifer, and depleted oil/gas reservoirs.

268 Additionally, evaporation of formation water in residual oil resulting contaminants which can
269 reduce injectivity of gas besides the depleted oil field is seemed to be previously habituated
270 with gas storage mechanism ¹¹⁹. Additionally, evaporation of liquid media can increase the
271 gas moisture content which can influence gas dehydration process cost at the surface.
272 Moreover, humid shaly sand or reservoir rocks with high montmorillonite content may
273 activate the process of the in-situ stresses and at a certain stage, it might have degraded
274 formation potential to withdraw the gas ¹²⁰.

275

276

277 Therefore, there is a pressing need to properly analyse the feasibility of H_2 in the depleted
278 hydrocarbon reservoir because hydrogen can:

279 ■ Regarding biotic transformation: Microbes can consume carbon energy from residual
280 hydrocarbon ¹²¹ and speed up their growth process in the presence of H_2 . Moreover,
281 sulphate is found in a considerable quantity in depleted oil/gas fluid wells. Thus, H_2
282 injection may reuse sulphate because of increased microbial growth ¹²².

283 ■ Regarding abiotic transformation: H_2 may dissolve reservoir rock-forming units at
284 high water saturation. H_2 may react with sulphur-based minerals and produce abiotic
285 H_2S , the process can deplete H_2 purity and may catalyse the kinetics of gas mixing ¹²³.
286 However, it is difficult to comment either biotic or abiotic is a major source of by-
287 product gases and H_2 /by-product gas mixing ³⁹.

288 However, H_2 storage in the depleted natural gas reservoirs is advantageous when compared
289 to its storage in saline deep aquifers because depleted gas fields have residual gas saturation.
290 Therefore, a lower volume of injected H_2 may achieve residual status in the depleted natural
291 gas reservoirs ¹²⁴. Moreover, cushion gas requirement would be less in the depleted gas
292 reservoir due to residual gas saturation. In the case of saline aquifer, since pore-spaces of
293 aquifers are not filled with gas therefore initially it is necessary to fill the pore-space of aquifer
294 ¹²⁵, which increases the cushion gas requirements. Aquifers have been used in Europe for
295 natural gas storage. Aquifers are permeable porous geological rock formations that contain
296 fresh water and sometimes a high concentration of brine water ¹²⁶. The permeable and
297 porous formations of aquifers are mostly sandstone and carbonate ¹²⁷. The impermeable cap
298 rocks (such as anhydrite layer, tight shale and salt) of aquifers are pivotal in ensuring the gas
299 trap and storage process ¹²⁸.

300 In principle, rigorous exploration work is carried out to determine storing and sealing capacity
301 of gas in aquifer reservoirs ¹²⁹. Several storage wells are required to be drilled through the
302 caprock into aquifer reservoirs. Aquifer well is drilled in multiple phases and account for both
303 cementing and casing cost^{130, 131}. The aquifer rock contains brine or water which displace in
304 the pore system by pumping H_2 ¹³². Thus, pressure can rise significantly due to compression
305 in closed system aquifers thereby injection volume of gas in comparison to the formation

306 breakdown pressure limit is small⁸⁰. Ultimately both factors, such as an increase in pressure
307 and permeability of aquifer¹³³ can be used to measure the maximum H_2 injection rates¹³².

308 Aquifers have the ability to store a large volume of gas¹³⁴ but there are some issues such as
309 chemical and biological activities (such as *biofilms* caused by anthropogenic activities) in the
310 vicinity of wellbore^{135, 136}. It is possible that during the reproducing/withdrawing period, high
311 bottom hole flowing pressure (p_{wf}) (due to over volume of cushion gas) can push the H_2 gas
312 back towards the wellbore and extend injection/pumping time¹³⁷.

313 The exploration cost of aquifer formation is high which is aimed to verify the size of the
314 storage media and relative areal extent and structural trapping^{138, 139}. Moreover, aquifer
315 reservoirs are continuously examined using monitoring wells and storage wells^{140, 141}.
316 Additionally, aquifer reservoirs require a large volume of cushion gas during the phase of
317 commissioning which is unreproducible during the phase of operation and decommissioning.
318 Therefore, CO_2 can be used as a cushion gas in aquifer reservoirs. Through this approach, a
319 loss of injected gas can be mitigated in the deep saline aquifers. In aquifers, there might be a
320 chance of the mineralogical activity where evaporated water encourages the generation of
321 H_2S due to the presence of sulphide minerals and H_2 which may reduce the hydrogen purity
322 (via abiotic process)¹⁴². The feasibility of the H_2 geo-storage in the depleted oil and gas
323 reservoir and deep saline aquifers is given in the supplementary information in **Table S1**.

324 Overall, volumetric capacity, recovery efficiencies and rates of depleted reservoirs are not
325 evaluated adequately and required in-depth investigation to formulate H_2 injection program
326³⁷. Accurate evaluation of these factors is possible with history matching of numerical
327 modelling and operational data during field-scale H_2 geostorage projects¹⁴³.

328 **3.1 Underground H_2 geo-structural trapping**

329

330 Developing large-scale H_2 storage in geological formations is required essential information
331 of cushion gas requirement, storage capacity and sealing security. The overlying cap rock (e.g.,
332 shale) characterizes adequate permeability to mitigate H_2 leakage in the complex media
333 (gas/liquid/rock).

334 As aqueous phase (water, and organic traces) and non-aqueous (H_2 , CO_2 , H_2S and CH_4) may
335 basically act like hydrophilic and hydrophobic to the rock's surface and interstitial pore-
336 network ¹⁴⁴. The wettability alteration is influenced by the flow of the different phases,
337 surface morphology of rock, relative permeability, different phases saturation and capillary
338 pressure ¹⁴⁵. Therefore, H_2 trapping mechanism may possibly be affected via withdrawing,
339 re/injection, formation contamination (fines migration), bacterial based formation
340 precipitation and clogging, and mineral dissolution in pore-network system ^{146, 147}. H_2 is high
341 diffusive gas and therefore the main risk associated with its storage is leakage via
342 underground natural pathways (such as., seepages, faults, and fractures) and drilled
343 wellbores. H_2 has a small molecular diameter ¹⁴⁸ (when compared to CO_2) ¹⁴⁹ and therefore,
344 it may quickly buoyant upward under certain reservoir conditions. Investigators believed that
345 uplift migration of H_2 can be mitigated through high capillary entrances and structural
346 trapping processes ³⁷. To date, the literature lacks to provide convenient information on the
347 H_2 trapping in subsurface media through the processes and leaves several open research
348 questions for the scientific community ¹⁵⁰.

349

- 350 • Importantly, structural trapping is a principle mechanism for underground natural and
351 anthropogenic fluid storage (Figure 3). Nevertheless, the physical properties of H_2
352 (e.g., high diffusivity, light weight, and low solubility etc.,) and chemical kinetics
353 (oxidation and reduction) may result in H_2 leakage from fault paths. Moreover,
354 information of H_2 reaction with impermeable strata (e.g., shale and mica) is
355 anonymous to date except for H_2 reaction with Kaolinite and Smectite. These clays
356 may mitigate H_2 flow in the narrow pore-network of cap rock (e.g., shale) and ensure
357 molecular H_2 structural trapping ¹⁰³.

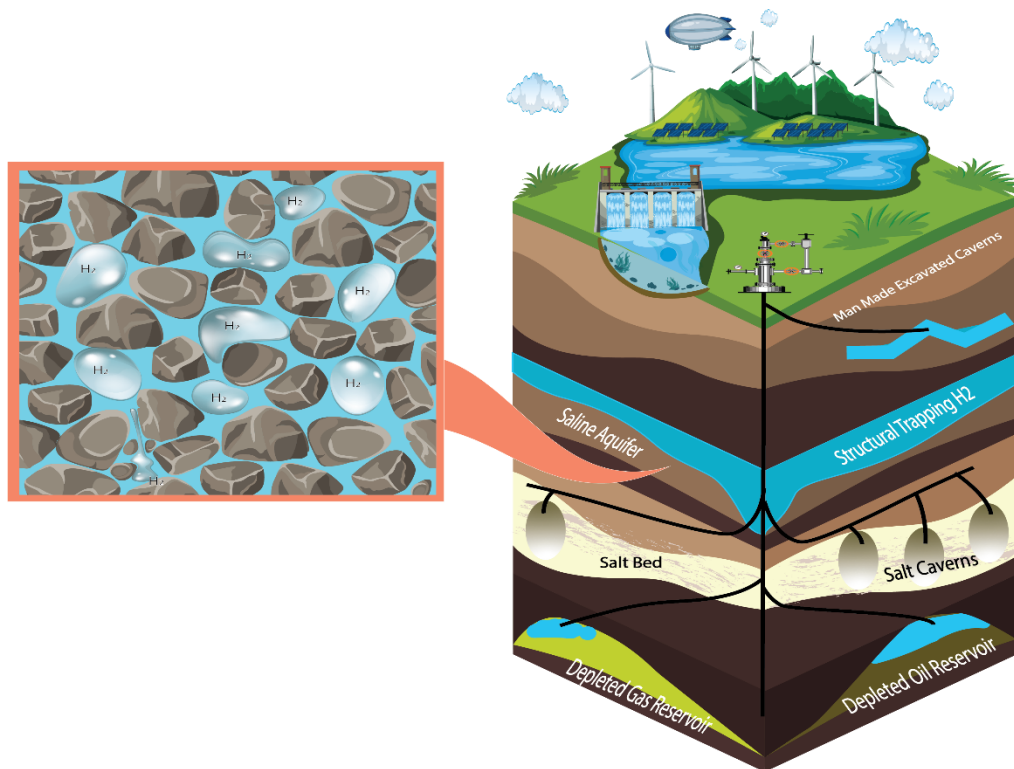


Figure 3 Structural trapping, capillary trapping, and mineral trapping have been illustrated in aquifer storage rock system (right). H_2 molecule tends to migrate upward to achieve residual trapping and structural trapping (left)

358

359 H_2 gas bubble may trap in the pore system due to the influence of capillary forces and grain's
 360 morphology resulting in an impact on residual trapping ¹⁵¹. In our earlier study, we modelled
 361 the interfacial tension of H_2 and CH_4 at storage conditions where higher capillary entry
 362 pressure is predicted for H_2 ³⁷. Moreover, morphology and size of pores in the rock can
 363 influence the phenomenon. In typical water-saturated porous formation, interfacial tension
 364 and density behaviour of H_2 are different when compared to H_2+CH_4 (H_2 +cushion gas) which
 365 is discussed in our present study as illustrated in Figure 4a. Interfacial tension of H_2 is higher
 366 when compared to H_2+CH_4 which will increase the column height (e.g., capillary entry
 367 pressure) and thus water-saturated caprock would be able to sufficiently seal the stored H_2
 368 (Figure 4a). Accordingly, hydrogen could be stored at a higher pressure than initial pre-
 369 production pressures in depleted gas fields ³⁷. However, it is important to maintain stored
 370 pressure lower than pre-production pressure to reduce the chance of geomechanical failure.
 371 Nevertheless, H_2 has less viscosity and density when compared to CH_4 and CO_2 which can
 372 cause H_2 viscous fingering (Figure 4 b and c). Moreover, thermal conductivity of H_2 is high in

373 geological locations. Synergetic effects of these fundamental properties may kinetically
 374 influence the presence of H_2 in the porous media and influence the H_2 storage security.

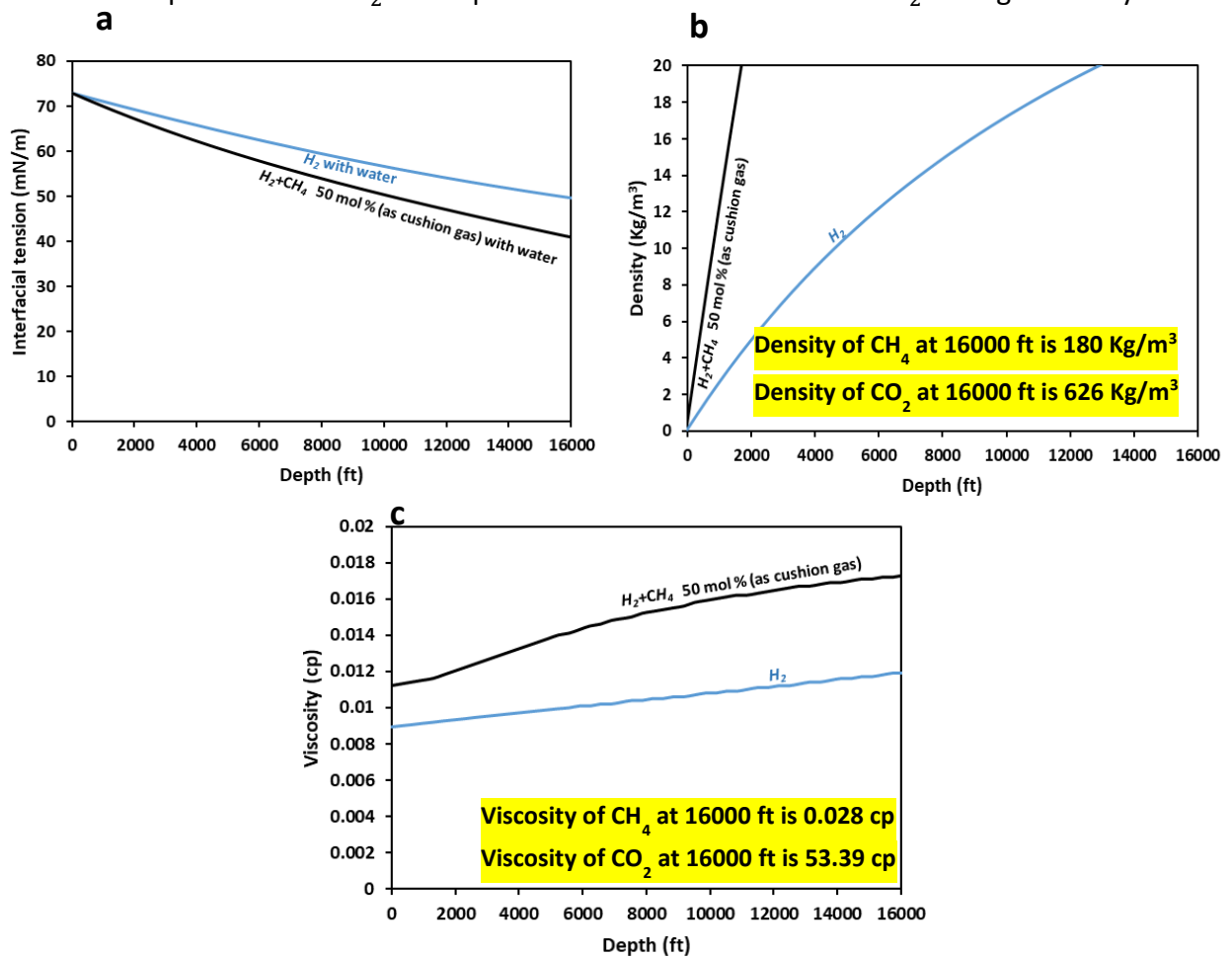


Figure 4 a) H_2 , and H_2+CH_4 (50mole%) interfacial tension along with water versus similar conditions b) H_2 , and H_2+CH_4 (50mole%) density versus depth, c) Viscosity of H_2 , and H_2+CH_4 (50 mole%) versus the depths. Equation of states and auxiliary models in the studies¹⁵²⁻¹⁵⁵ were used to measure fundamental properties of the fluids and the techniques^{37, 156} were used to measure the interfacial tension of the pristine gas and the mixed gas system.

375 H_2 is less soluble and lighter when compared to CH_4 and CO_2 thus it may achieve less
 376 solubility trapping. Solubility of H_2 , CO_2 and CH_4 in the water as illustrated in Figure 5a and
 377 b. Additionally, the adsorption process of gases (e.g., CH_4 , N_2 and CO_2) in the pore-network
 378 system can increase with increase in the kinetic diameter of the gas molecule¹⁵⁷. Assuming
 379 the same principle for H_2 , the kinetic diameter of H_2 is less than CO_2 and CH_4 thus H_2 may
 380 achieve less adsorption in the pore-network system. Table S2 in the supplementary
 381 information provides the kinetic diameter of the gases and pore size of storage rock (e.g.,

382 sandstone and carbonate) and sealing rock (shale). The molecular size of gases is nano
 383 compared to reservoir rocks' pore sizes which are micro in size (excluding cap rock). Thus,
 384 gases can easily flow in pore-network with non-disruptive pore wall-gas molecule collision.
 385 Therefore, diffusion at molecular level is very likely and cause leakage in the micron size pore-
 386 network system ¹⁵⁸. In contrast, shale contains nano size pores (less than 2 nm) which may
 387 dominate collision between H_2 molecules and pore wall to neglect molecular diffusivity and
 388 achieve structural trapping of H_2 . Diffusion of H_2 was determined in different gases system
 389 (e.g., H_2 in CH_4 and H_2 in CO_2) using Chen and Othmer method and it was observed that H_2
 390 diffusivity was high when compared diffusivity of CO_2 in CH_4 (Figure 5c). Consequently, H_2
 391 may achieve uplift migration and tends to achieve cap rock trapping or structural trapping.
 392 Nevertheless, high diffusivity of H_2 could raise leakage in low pressure zones and fault path
 393 leading H_2 trapping insecurity.

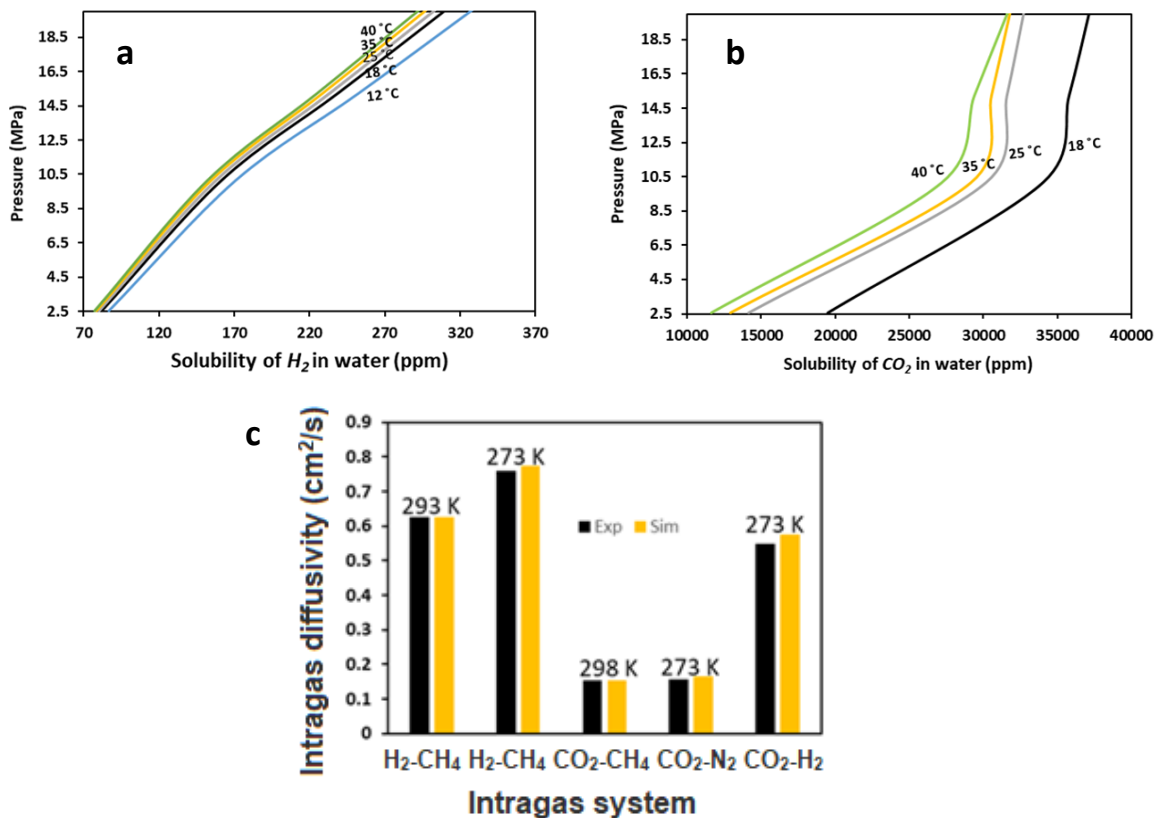


Figure 5 (a) Solubility of H_2 in water at variable pressure and temperature was determined using Valderrama-Patel-Teja (VPT) equation of state used in the study ¹⁵⁹ (b) Solubility of CO_2 in water data at variable pressure and temperature was taken from the study ¹⁶⁰. Solubility of H_2 is significantly less when compared to solubility of CO_2 and CH_4 in water. (c) Diffusivity of H_2 in CO_2 and CH_4 and diffusivity of CO_2 in CH_4 are measured using Chen-Othmer technique ¹⁶¹ at variable pressures and temperatures

394

395 The trapping behaviour of sandstone (aquifer quartz surface) for H_2 at realistic temperature
396 and pressure conditions show that the rock surface was weakly/intermediate water-wet but
397 after the addition of organic component onto the rock surface, the wettability of H_2 was
398 increased ¹⁶². Nevertheless, organic components in the porous media may raise problems e.g.,
399 (i) organic components mixing with stored H_2 and (ii) influence on the microbiological life (as
400 microbes may consume organic components as an energy source) at pore-scale level and
401 increase their activity ^{39, 55}. Moreover, H_2 storage and structural trapping can be influenced
402 by the mineralogy of sealing rock and hysteresis effect ^{55, 163}. The careful analysis of hysteresis
403 can help to adequately predict H_2 injection (primary drainage), H_2 gas production (secondary
404 imbibition) and H_2 re-injection (secondary drainage) ¹⁶³. The recent simulation study
405 quantified the trapping behaviour of H_2 /brine in sandstone formation via interpreting the
406 hysteresis effect ¹⁶³. However, they observed that cyclic hydrogen storage in the underground
407 formation resisted the hysteresis effect which has been previously used to examine the gas
408 storage (in particular CO_2 , and CH_4) mechanism at continuum scale via capillary pressure and
409 relative permeability information. These both properties are sensitive to contact angle in the
410 H_2 storage system. Additionally, high frequent production/reinjection cycles may cause
411 pressure and temperature variations. These underground variations may weaken sealing
412 capacity of cap rock, minimize fracture pressure result in in loss of cushion gas and working
413 gas. Therefore, detailed study of the storage rock via pore network imaging (e.g., mineral
414 precipitation), kinetic rates and sensitive analysis of the modified hysteresis effect can provide
415 necessary information of H_2 residual saturation after secondary imbibition ^{163, 164} which is
416 essential for the process of H_2 production from geological formation. Operational parameters
417 and feasibility of H_2 storage in different storage systems are given in **Error! Reference source**
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419 Table 2 Comparison of underground porous reservoir rocks for hydrogen storage

Parameters	Depleted gas reservoir	Depleted oil reservoir	Saline aquifer	Reference
H_2 storage experience	Medium	No	Medium	119, 132
Exploration cost	Medium	Medium	High	165
Hydrogen mixing with other components/ and loss	High	High	Low	166
Hydrogen withdrawing capacity	Low	Low	Low	33
Presence of microorganism	Low to high	Low to high	Medium	39
Storing media	Porous	Porous	Porous	138, 167
Bottom hole flowing temperature (°F)	150	150		
Bottom hole flowing pressure (psi)	150 to 500	250 to 500		168
Areal extent (sq.km)	Varies significantly	Varies significantly	Varies significantly	
Interval thickness (m)	25 to 50	25 to 50		169
Typical storage capacity (Gt)	675 to 9000		1000 to 10,000	
Salinity (ppm)	Varies	Varies	1500000 to 1900000	
pH	7	7		39
Water cut	30 to 70%	30 to 70%	80 to 90%	169

420

421 **4 Prospect of underground H_2 storage in salt formation**

422

423 In comparison to H_2 storage in the porous rock, its storage in salt cavern has been less
 424 challenging due to the reason that salt layers are impermeable and inert to H_2 . Moreover,
 425 withdrawal capacity of H_2 is more efficient in the salt cavern when compared to porous rock
 426 as could be used as huge storage tanks. Nevertheless, field-scale H_2 storage in the salt cavern
 427 has several challenges. Gas storing capacity and salt stability in salt caverns are key factors for
 428 the consideration of H_2 storage ^{48, 170}. Large pillars of thick salts layers of caverns (both top
 429 and bottom layers) presented efficient H_2 storing capacity ¹⁷¹. Realistic experience of salt
 430 cavern H_2 storage found satisfactory relative to high cavern pressure potential and

431 homogeneity in the salt formation ¹⁷². The large salt caverns are constructed in salt domes
432 (with uniform layers of salt) near the Gulf Coast in the US ¹⁷³. However, the geology of salt
433 caverns located in Western States, North-eastern, and Midwestern is not homogenous and
434 as such inadequate for storing H_2 in the US¹⁷⁴. In US, Germany, and the UK, H_2 salt cavern
435 realistic storage projects provided four decades of experience. This experience can be utilized
436 for the establishment of LGS H_2 storage capacity ¹⁷⁵. The large volume of the cavern can
437 provide a high flow rate of H_2 . Capsule type salt cavern is considered more appropriate
438 choice for H_2 storage because of its better height and adequate stability (e.g., under low
439 tensile and overburden stresses) ¹⁷⁶. The storing media has adequate stability to high pressure
440 and low-temperature conditions ^{33, 177}. Moreover, smooth operational proceedings of the
441 project are very pivotal for its successful completion ¹⁷⁸⁻¹⁸⁰. Salt cavern H_2 sealing capacity is
442 high however mechanical and mineralogical properties of the salt cavern are important
443 factors ¹⁸¹ to endure compressive and tensile failure. Moreover, low working pressure may
444 reduce the risk of mechanical failure and sink risk, however, it would increase the expenses
445 and later required high pressure H_2 injection operation to maintain the in-situ pressure of the
446 cavern for less injection time and optimum delivery of H_2 ¹²⁴. However, salt cavern provided
447 low storage capacity and did not provide long-term storage solution when compared to
448 depleted oil and gas reservoirs. Operational parameters for underground hydrogen storage
449 in salt cavern are provided in Table S3.

450

451 **4.1 Salt cavern UGHS challenges and solutions**

452

453 Selection of salt cavern for the H_2 storage depends on the depth, purity, composition, and
454 wall thickness of the salt cavern ¹⁸². Continuation, distribution, and thickness of the rocks
455 (evaporate type) varied within the basin area which is further based on the formation of the
456 salt structures such as pillows, salt stocks and diapir. Investigators found that average salt
457 structures can store around 214 kWh and 458 kWh per m^3 ^{49 183}. Nevertheless, salt dome
458 structures have more energy storage density such as 210 GWh when compared to a salt
459 bedded structure which could provide around 65 to 160 GWh energy storage capacity ¹⁸³.

460 Salt beds and domes are naturally developed due to movement between the substrate and
461 overlying strata in the presence of low-density salt; the mechanism is known as 'halokinesis'
462 ¹⁸⁴. This mechanism could be influenced by buoyancy, gravity difference, salt spreading,
463 thermal convection and differential loading forces ¹⁸⁵⁻¹⁸⁷. Buoyancy forces and low-density
464 salt layers are primary reasons for the formation of a salt dome in comparison to the effect
465 of tectonic activity ¹⁸⁸ as depicted in Figure 6 **Error! Reference source not found.**

466 Typically, salt caverns are made up of halite salt along with anhydrite salt (layers). Both halite
467 and anhydrite remain as the main sources of salt cavern's brine composition. Adequate salt
468 diaper which is composed of 99% halite ($NaCl$) and less than 1% of anhydrite ($CaSO_4$) salt and
469 other impurities ¹⁸⁹. However, the composition of salt bedded layers and salt domes are based
470 on geographical locations and subsurface environment. The uniform layer of halite ($NaCl$)
471 could be an appropriate choice for the construction of salt caverns. Thanks to halite salt due
472 to its high thermal conductivity, self-healing properties, impermeable behaviour and plastic
473 properties, it could establish potential sealing to mitigate H_2 leakage in the system ¹⁹⁰.
474 Importantly, halite could not react with H_2 . However, there were some common impurities
475 such as quartz (SiO_2), anhydrite ($CaSO_4$), dolomite ($CaMg(CO_3)_2$), pyrite (FeS_2), gypsum
476 ($CaSO_4 \cdot 2H_2O$), and calcite ($CaCO_3$) could be found in the salt layer which may react ¹⁹¹ but
477 requires further investigations. Moreover, some common ions impurities are also observed
478 e.g., as Fe^{+3} , Mg^{+2} , CO^{-2} , Ca^{+2} , SO_4^{-2} , Fe^{+2} , K^+ , and Cl^- , Ba^{+2} , B^{+3} and Sr^{+2} ⁴⁹ which can enhance
479 microbial activity. Additionally, the mineralogical properties of anhydrite salt are required to
480 be analysed because of its hygroscopic nature. Moreover, the interaction of anhydrite salt
481 with water produces gypsum and the solubility of anhydrite is 140 times less when compared
482 to halite salt e.g., 2.5 g of anhydrite salt requires a litre of water for its absolute solubility ¹⁹²,
483 ¹⁹³ which can cause an inappropriate delay in salt cavern construction. High concentrations of
484 Ca^{+2} , and SO_4^{-2} ions from anhydrite remain in the cavern and it could form H_2S in the presence
485 of H_2 or methane through both biotic and abiotic sulphate reduction. H_2S has corrosive (can
486 develop H_2 embrittlement problems in casing and steel tubulars) and toxic nature which may
487 increase surface processing facility which results in an increase in the overall cost of the
488 project. Hence, H_2S catalyses the H^+ ions embrittlement problems in steel tubular; it reduces
489 the tensile and ductile strength of the tubular and de-velops the corrosion films of FeS and
490 free H^+ on the surface of metal. Thus, free ions of H^+ along with S^{2-} accelerate the corrosive

491 rate via H_2 or H^+ ion trapping in the interstitial spaces of the metal promoting hydrogen
492 embrittlement in the system ¹⁹⁴. Hence, H_2 can cause the embrittlement problems and
493 proceed the H^+ diffusion and its adsorption in the interstitial spaces (metal lattices) ^{195, 196}.
494 Cavern leakages problems are mainly associated with 'Fillings' (e.g., undissolved rock layers)
495 in salt caverns, overburden compressive forces, tensile forces, microbial degradation activity,
496 well casing leakages and H_2 embrittlement (corrosion damage) ¹⁹⁷⁻¹⁹⁹. Figure 6 provides a
497 drawing of a salt cavern and its problems which can influence the UGHS process. Thus, self-
498 healing (natural cavern's wall recovery) and tight permeability of salt cavern's wall (for H_2
499 entrapment in pores) could be two pivotal factors for enhanced H_2 gas storage and security.
500 The permeability of 1 m thick salt layer is evaluated 2 nD at 1 MPa pore pressure value ²⁰⁰
501 nevertheless typical permeability of tight gas reservoir is 1000 nD ²⁰¹. However, in amateur
502 salt structure, mechanism of 'halokinesis' and the contaminations in the salt formation can
503 increase the permeability of salt cavern resulting H_2 leakage and inadequate reinjection of
504 H_2 into salt cavern. Table S4 presents the review of salt cavern performance, challenges, and
505 adequate solutions.

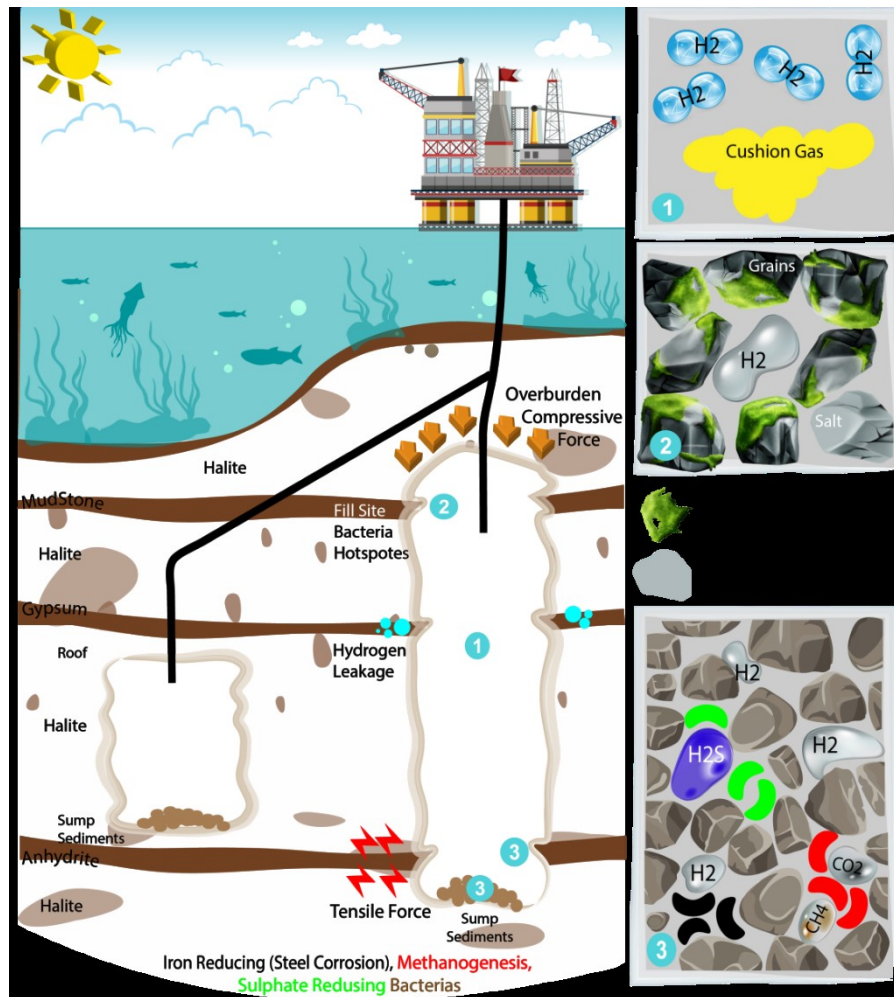


Figure 6 Salt caverns challenges have been illustrated at a microscopic level: 1) Cushion gas (e.g., N_2 and CO_2) is recognised as inert gas. Nevertheless, H_2 withdrawing cycles may ensure in-situ thermal variation and can impact H_2 mixing with the inert gas¹³². 2). Salt has natural self-healing properties²⁰². However, deposition of microbes on salt crystals and acids release from microbes can influence the self-healing behaviour of salt cavern. Consequently, the process may degrade rock salt stability, rise permeability, microcracks and creep damage^{190, 203}. 3). Anthropogenic drilling fluid additives waste (e.g., polymers and carbonate sources) etc., may promote bacterium growth in the cavern's 'Fillings' and sump. Microorganism living in the fillings may consume anhydrite salt causing, H_2 loss, permeability increment, H_2 leaking and H_2S generation in the system^{166, 204}. More importantly, fillings problem is common in bedded salt cavern system and may emerge undissolved rock layers e.g., anhydrite and mudstone as H_2 leakage hot-spots.

506

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511 4.2 Biogeochemical behaviour of H_2 in salt formation

512 Salt formation is inert and inactive to hydrogen ²⁰⁵. However, brine water can stay in the salt
513 cavern's sump. Brine water may evaporate into stored gas ²⁰⁵ and increase moisture content
514 in the gas which can be a challenge and requires surface facilities for dehydration of H_2 .
515 Overall, the amount of contamination in the withdrawn H_2 gas from salt cavern is less when
516 compared to depleted oil and natural gas fields ¹²¹. More importantly, the pore-network in
517 the depleted reservoirs may be plugged due to reactions between minerals and H_2 via
518 microorganism (biotic) and host rock (abiotic) ²⁰⁶. Thus, withdrawing and reinjection capacity
519 of H_2 may be influenced in depleted reservoirs. The practical experience shows that both
520 natural gas and H_2 were stored together in salt caverns successfully for several decades in the
521 UK, the US and Germany ^{79, 207}. The inert nature of salt with hydrogen yields overall high
522 stability, adequate flexibility, improved integrity, and enhanced withdrawing capacity.
523 Therefore, it can be used to contain daily demand or seasonal gas peaks in winter for heating
524 purposes. Moreover, salt cavern H_2 storage is cost-effective, stable and operationally
525 durable, contaminated free, and viable to store gases and liquids at high-pressure conditions
526 ²⁰⁸. However, the main problem is the storage capacity of salt caverns which is comparatively
527 low.

528 Salt caverns are artificially developed by injecting water into salt formations to carry out salt
529 dissolution which is referred to as 'salt mining'. The typical feature of a salt cavern is listed in
530 Table 3. Artificially developed salt caverns have been used for 50 years, and primarily used
531 for the storing purpose of hydrocarbon, in particular methane, and oil, later it has been
532 noticed as a potential storing media for H_e , and H_2 . First time in US, oil and LPG were both
533 stored in salt caverns in 1950. In 1961, the very first salt cavern was used for the storing of
534 natural gas in US ²⁰⁹. Nevertheless, the first salt cavern for the storage of H_2 was developed
535 in the UK in 1971 and it is operational to date. Interestingly, around 300 and 2000 salt caverns
536 are in Germany and North America currently used for the storage of compressed air,
537 compressed H_e , butane, radioactive waste and compressed H_2 ^{208, 210}.

538

539 **Table 3** Typical artificial salt cavern features for H₂ storage

Factor	Configuration	References
Volume	Storing volume 100000 m ³ , and vary with geographical based on the technical as well as geological history, geographical location and behaviour of the salt cavern	210, 211
Salt thickness	At least 200 m	210-212
Depth	At least 500 m to 2000 m	210, 212
Height	At least 400 m	210, 213
Operating pressure	20 MPa is essential for high volumetric storing capacity	210, 212
Mechanical properties	Optimum mechanical stability of salt cavity is key to sustainable H ₂ storing	210-212

540

541 High diffusive behaviour of H₂ and the bacterial process can damage the sealing behaviour of
 542 the cavern and contaminate the stored H₂ purity⁴⁹. Microorganisms are living in the sump's
 543 residual water with indigenous microorganisms communities since the leaching process. The
 544 sump may contain anthropogenic drilling material which may provide energy sources (e.g.,
 545 cellulose, starch, SO₄ and insoluble carbonates) to bacteria²⁰³. Thus, bacteria can start
 546 consuming H₂ and generate H₂S in the presence of carbonate and sulphate via sulphate
 547 reduction²¹⁴. *Desulfovibrio halophilus* and *Desulfovibrio Vulgaris* utilized sulphate for the
 548 acceptance of electrons in the environment of anaerobic metabolism^{215, 216}. Bacterial survival
 549 is based on sulphate reduction such as from sulphate (S[+VI]) to sulphide (S[-II]). Sulphate is
 550 activated before it can accept the electron via enzyme adenosine triphosphate sulfurylase
 551 phenomenon, and thus synergy behaviour of triphosphate sulfurylase+sulphate develops
 552 adenosine 5' phosphosulfate and reduced sulphate to sulphide ultimately produce H₂S in the
 553 presence of H₂²¹⁷. Table 4 illustrates that different types of SRB have the ability to survive
 554 typical reservoir temperature condition. In fact, the growth of these bacteria expedited at
 555 these temperatures and increased sulphate reduction process. Figure 7 depicts that SRB have
 556 been found in salt cavern, depleted hydrocarbon reservoir rocks and saline aquifer which can
 557 generate H₂S in the system^{49, 218-225}. Some storage rock has shown high H₂S generation when
 558 compared to the allowable limit.

559

560

561 Table 4 Reaction and temperature in geological rock for the occurrence of the bacteria

Type of bacteria	Factors	References
Thermochemical sulphate reduction	100 to 180 °C	218
Bacterial sulphate reduction	0 to 60-80 °C Some greater than 80 °C	226
Hyperthermophilic SRB	110 °C	227
Optimal growth temperature for the SRB	38°C, acidic, saline environment. Mostly SRB activity reduces, if concentration of NaCl increases above 50 to 100 g/l, however, SRB activity is found in salt lakes and brine water which reflects sustainability of SRB in high salt concentration system.	218

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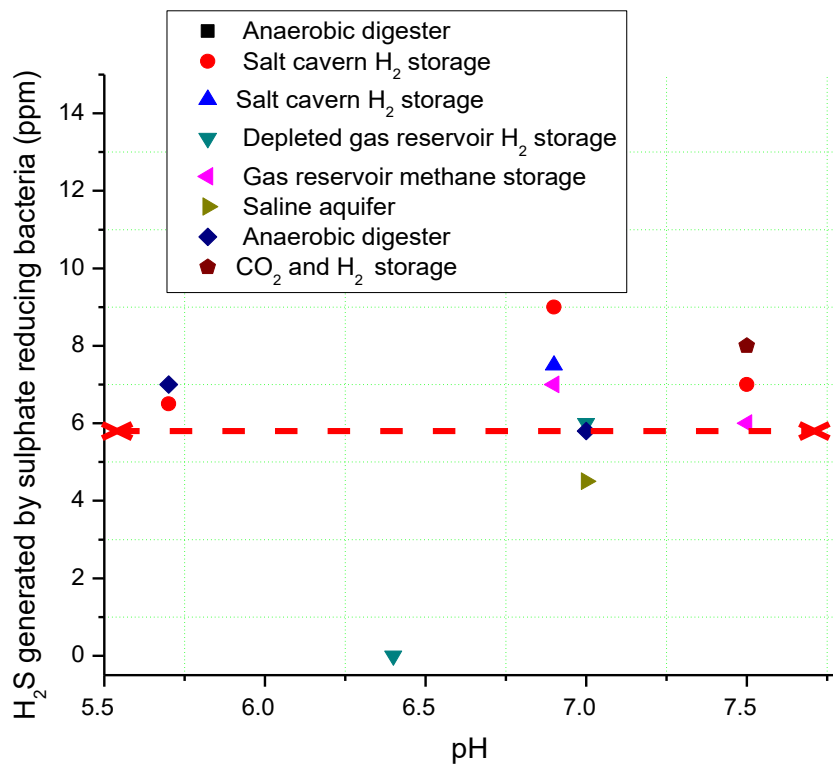


Figure 7 Sulphate reducing bacteria has generated H_2S in the surface and subsurface storage systems. Storage rock has shown high generation of H_2S which is greater than its threshold value (red dashed line) ^{49, 218-225}.

563

564

565 4.3 Progress, and factors influencing the development of salt cavern

566 Selections of fresh water, drilling fluids and completion fluids are important factors during the
567 construction of a salt cavern. Underground salt formations are not sterile and contain multiple
568 types of indigenous, natural, and anthropogenic microorganisms. The supplement source of
569 energy from fresh water (such as carbonates (CO_3^{2-}), and Mg^{+2} , and Ca^{+2} etc.,) and drilling
570 fluids (e.g., barium sulphate ($BaSO_4$), cellulose and starch polymers) may improve the growth
571 of microbes inside the caverns and rise problems. Leaching, debrining, and filling are pivotal
572 phases of underground salt cavern construction^{228, 229} and carried out into geological salt
573 deposits as illustrated Figure 8. The geological salt deposit, in particular, salt domes is selected
574 after careful assessment of the previous exploratory and drilled wellbore data²³⁰. Time period
575 for the development of salt cavern based on the areal extent of salt formation and formation
576 hydraulic diffusivity, however, it might take more than one year to construct a single cavern
577^{177, 231}. The drilled hole which could be used for the injection of H_2 based on the factors,
578 including geological formations, structure, fault locations and depth of the salt caverns (which
579 typically range from 300 m to 2000 m). Like in oil and gas well drilling, the salt cavern wells
580 are also drilled in a telescopic design, however, the diameter of salt cavern wells is larger
581 when compared to conventional oil and gas wells²³². These caverns are cased and cemented
582 adequately so that H_2 gas may not leak and sustain smooth H_2 injection/production cycle²³³.
583 Tightness of the salt cavern well is examined to predict cementing and casing leakages
584 problems²³⁴. Then, brine is displaced towards the surface via injection of H_2 inside the salt
585 cavern during filling phase. Nevertheless, 100% brine recovery is not easy because production
586 tubing did not reach until the bottom of salt cavern²³⁵.

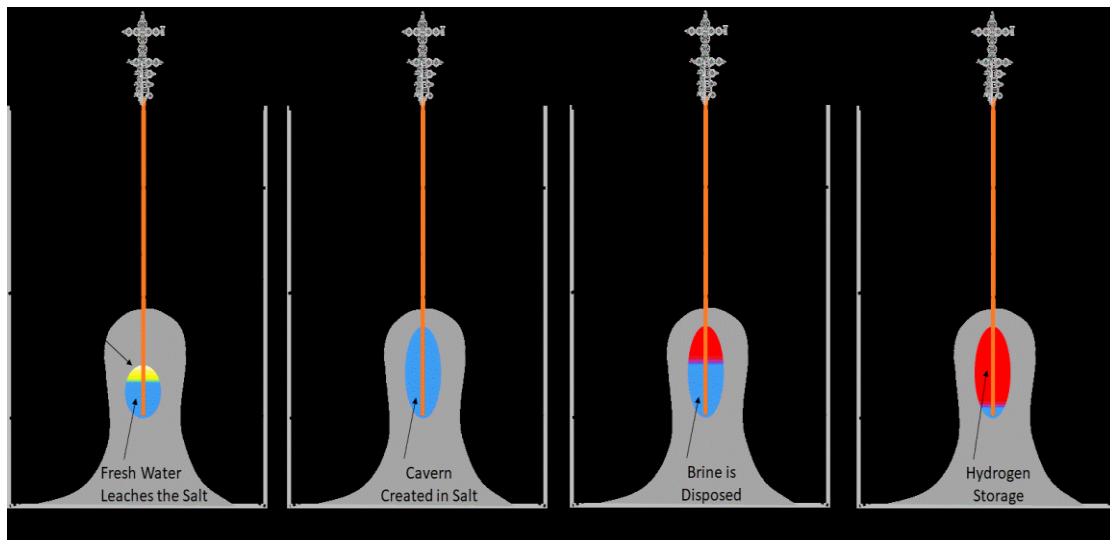


Figure 8 The process of salt cavern development involves multiple steps such as i Salt formation selection ii Leaching, iii Debrining phase and iv Filling

587

588 Design of salt cavern strongly based on the properties of the salt rock deposits and its
 589 structure ²³⁶. Additionally, other factors are also required to be given proper attention during
 590 the formation of salt caverns e.g., depth of salt cavern ¹⁸², height and diameter of salt cavern
 591 ²³⁷. For the geo mechanical stability of the cavern, the salt thickness of both hanging wall as
 592 well as foot wall ^{238, 239} and relative information of the cavern must be explicitly defined. The
 593 minimum safe thickness of the salt cavern is the function of the diameter of the salt cavern.
 594 For instance, it has been suggested that the minimum thickness of the hanging wall must be
 595 equal to 75% of the cavern's diameter; and 20% of the cavern's diameter for the case of foot
 596 wall. Additionally, the least ratio (height to diameter) of the salt cavern must be 0.5 in the
 597 condition of bedded salt structure ²⁴⁰. The capsule-shaped salt caverns have been widely used
 598 for the H_2 storage purpose. The capsule-shaped cavern adopted minimum stress when
 599 compared to a cylindrical or elliptical shaped cavern at 27 MPa overburden pressure and 1200
 600 meter depth ²⁴⁰.

601 In Utah US, researchers are working to store H_2 underground in a vertical salt cylinder which
 602 is 804 m wide and 1609 m deep. This could be the largest salt cavern reservoir to store
 603 underground H_2 in the history of the US. The project could produce around 1000 MW of clean
 604 energy via stored H_2 . The stored H_2 would be enough power to provide electricity to 150,000
 605 households for the period of one year. ²⁴¹.

606 In the first phase of the project, H_2 energy storage would supply around 150,000 MWh of
 607 storing capacity which is 150 times higher energy when compared to the existing lithium-ion
 608 batteries storage system in the US ²⁴¹. Additionally, the cost of underground salt drilling and
 609 maintenance is 10 times less than surface H_2 storage tanks system and it is 20 times less when
 610 compared to hard rock mines expenditures. According to the US Strategic Petroleum Reserves
 611 data, the US has around 60 caverns which are typically 200 ft diameter and 2500 ft tall. Table
 612 5 presents the underground salt cavern H_2 storage projects information.

613 Table 5 Operational H_2 salt cavern projects

Countries	Salt cavern H_2 storage sites	Potential of salt caverns	References
The UK	Teesside (operational)	<ul style="list-style-type: none"> • Elliptically shaped, • Depth of 350-450 m • Volumetric capacity of 210,000 m³ 	33, 205
The US	Operational Moss Bluff, Clemens salt dome, Spindle top	<ul style="list-style-type: none"> • Depth of 800 m (cavern top) • Storing volumes 580,000 m³ capacity 	33, 242
	Salt cavern H_2 stored facility in Texas since 1980s.	<ul style="list-style-type: none"> • 850 m cavern's roof height, • 49 m diameter and 300m height • Storing capacity of 1066 million cubic feet (or 30.2×10⁶ m³) 	

614

615 Information relative to the volumetric capacity of the salt cavern is important to carry out a
 616 suitable H_2 storing program ²⁴³. The method for the measurement of volumetric capacity is
 617 defined in the previous study ³³. Typical cavern construction involved multiple steps such as
 618 in-depth geological investigation which illustrates information of salt domes, inter-bedded
 619 salt structures and their areal extent ³³. Basic parameters are pivotal to be considered e.g.,
 620 cavern roof depth, storage pressure, gas cushion, water volume in the cavern, and surface
 621 pipeline facility. The cavern's thickness of 500 m to 2000 m and height of 500 m to 1200 m
 622 would be an appropriate choice for 7.09 MPa to 19.2 MPa storage pressures ³³ This could be
 623 explained that high thickness salt wall and roof are required which can sustain high injection
 624 pressure and microbial degradation activity in the system ²⁴³.

625 Importantly, the cost of the cavern during gas injection remained consistent until unless the
 626 underground H_2 storage pressure of the system is maintained. Hence, maximum
 627 underground pressure should be maintained $P_{max} > 7$ MPa. This may influence the cost of the
 628 compressor capacity and its power consumption. Moreover, the cost may raise with the depth

629 of the injection well bore and the use of high-grade tubular accessories (both casing and
630 completion). Additionally, it is pivotal that cushion gas is controlled and made consistent
631 around 30% of storing volume³³. Additionally, H_2 withdrawing pressure can be maintained
632 adequately relative to cushion gas volume pressure²⁴⁴.

633 **5 H_2 energy storage capacity in pipelines, gasometer tanks, depleted gas** 634 **reservoir and salt caverns**

635

636 We found that a single Marlin offshore field in Australia could store more than 100 TWh
637 energy storage capacity of H_2 as illustrated in Figure 9a. This is enough energy to fulfil the
638 demand of South Australia which annual demand was recorded 89.61 TWh in 2019 to 2020
639²⁴⁵. Underground H_2 trapping is safe, environmentally friendly and provide massive H_2 energy
640 storage solution when compared to surface H_2 storage system. However, surface liquid H_2
641 storage system may loss and boil off H_2 due to heat transfer from the environment²⁴⁶. Thus,
642 depleted gas reservoirs and salt caverns are apparently adequate choices for long-term and
643 short-term H_2 storage.

644 Salt cavern storage could help to compensate fluctuating energy demand (and sustain
645 multiple cycles)⁵⁵. Figure 9b summarises the energy storage capacity of H_2 increases (and
646 volume of the stored gas decreases) with increase in pressure and density which agrees with
647 typical behaviour of the gases. Generally, the energy storage capacity of H_2 can vary with the
648 volumetric capacity of salt caverns (e.g., 750,000 m³ 500,000 m³) as illustrated in Figure 9b.
649 The graphical illustration shows that the energy storage capacity of H_2 could increase with
650 depth. However, a uniform structure with an adequate salt thickness could impact the
651 withdrawing capacity of H_2 which are geographically constrained.

652

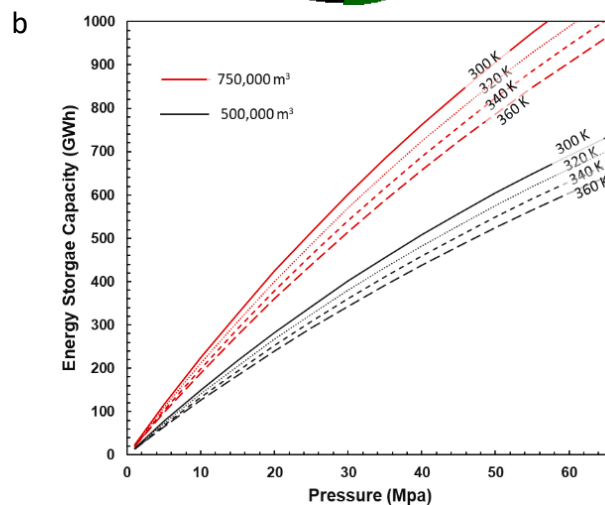
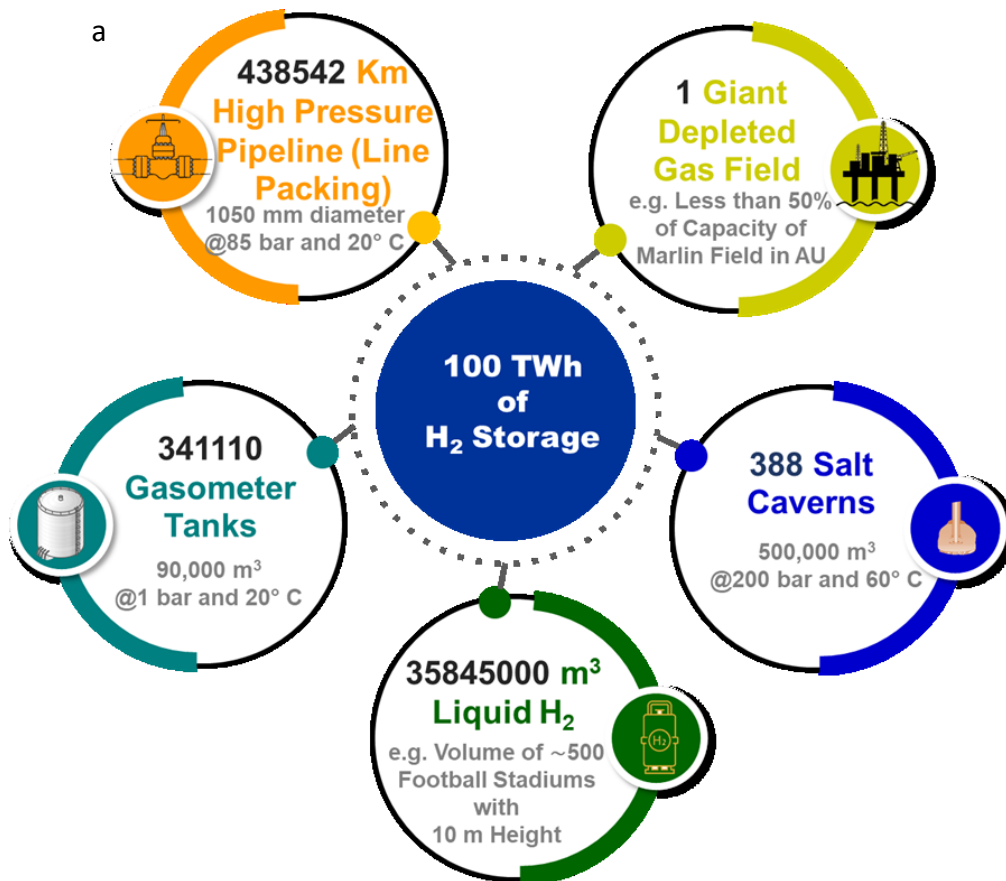


Figure 9 (a) Single Marline offshore depleted gas field could store 100 TWh of H_2 energy which is equivalent to 388 salt caverns capacity (each 90,000 m³). Each surface facility including 341110 gasometer tanks (each 90,000 m³) and 438542 Km line packing (diameter 1050 mm) could store 100 TWh. (b) Energy storage capacity of salt caverns of different volumetric capacities decreases with temperature and increases with pressure. The storage capacities have been calculated using H2CapEs³⁷

654 **6 Conclusions**

655

656 We have drawn the following conclusions from this review study:

- 657 i. H_2 energy and its storage technology are pacing with unprecedented momentum.
658 Thus, the prospect of H_2 energy seems to be very bright due to its overwhelming
659 advantages: clean source of energy, amazing energy density, and transformative
660 nature. H_2 can be produced through green techniques. It has the potential to be used
661 in various practical applications with high energy performance facilitating social
662 growth, economic competitiveness, and environmental benefits. More importantly,
663 developed economies in particular Australia, Germany, the UK, the US have realised
664 that there is a need to scale up H_2 production and its flexible storage system at the
665 LGS to deliver a future featuring “energy accessibility, security, affordability and
666 sustainability”.
- 667 ii. Energy stakeholders have realised that H_2 (as clean energy) has the potential to transit
668 from fossil-based energy economy to H_2 economy. Since the last decade, significant
669 contributions have been made to produce H_2 gas via environmental friendliness and
670 cost-effective way. However, flexible H_2 storage remains a challenge and requires
671 extensive research at an industrial scale.
- 672 iii. Depleted natural gas and oil reservoirs are considered for H_2 storage because of their
673 benefits, such as these are seismically proven large extended areas. Moreover, H_2
674 storage operator will have enough former mineralogical and geological (drilling order
675 data) to understand the appropriate physiochemical behaviour of H_2 and their
676 underground interactions with the grains and in-situ residual saturations.
677 Nevertheless, if H_2 is stored in the depleted natural gas fields, it can contaminate with
678 residual natural gas, and thus when H_2 will be withdrawn from the reservoir it can be
679 used for heating purposes. H_2 storage capacity of one depleted gas reservoir is
680 significantly higher when compared to energy storage capacities of salt caverns,
681 gasometer tanks, and extended pipelines.
- 682 iv. So far geological aquifer formations are proven realistic storing media for CH_4 rather
683 than H_2 . Moreover, the aquifers do not contain residual hydrocarbon saturation which
684 may affect the composition of H_2 . However, the H_2 storage operator is required to

685 fulfil additional exploration costs. Additionally, aquifers may contain microorganisms
686 that can react with H_2 and ensure precipitation mechanism in the porous media; thus,
687 the permeability of the reservoir can be influenced. Additionally, dissolution of H_2 in
688 aquifers may not be negligible because of the huge amount of available water.

689 v. Salt caverns are proven for H_2 storage capacity and security. Nevertheless, salt caverns
690 have very low-storage capacities in several orders of magnitudes (areal extent and
691 height) when compared with depleted gas fields. Additionally, salt cavern wall's
692 thermal stability, microorganism contaminations and H_2 withdrawing ability are
693 challenges to be overcome for its implementation in struggling economies. Thus, by
694 resolving the said challenges in salt caverns, we can explicitly exploit the potential of
695 realistic, clean, high efficiency, futuristic energy storage system.

696 **7 Future road map and recommendations for underground H_2 energy storage**

697

698 Following recommendations are proposed after a thorough analysis of the literature relative
699 to underground H_2 storage case.

700 i. Depleted oil and gas reservoirs with rich exploration, and geological drilling order data,
701 and high storage volume can be used for H_2 storage projects. However, anthropogenic
702 activity, biological transformation, in-situ H_2/CH_4 /cushion gas mixing can influence
703 withdrawing/injection cycles and stored gas quality, and security. At some point,
704 anthropogenic activity can be minimized via adequate drilling fluid additives and
705 completion fluid additives which can inhibit mineralogical alteration and microbial life.
706 Mixed gas can be utilized for heating and other purpose based on BTU of the gas.

707 ii. Multiple H_2 withdrawal/injection cycle may raise in-situ thermal and pressure
708 variation in the reservoir which results in mineral dissolution altering reservoir
709 permeability and sealing capacity. The effect of physical and thermal stresses in the
710 reservoir requires further investigation, coupling abiotic reactions e.g., pyrite can
711 transform to pyrrhotite causing H_2S release. However, H_2 did not occur any
712 mineralogical alteration in sandstone. Nevertheless, the reaction of H_2 on carbonate
713 and shale is yet to be answered.

- 714 iii. Biotic transformation in the storage rock varies with different factors including
715 temperature, brine water activity, salinities and pH. Microbial activity can develop
716 biofilms that can plug H_2 flow in the porous media and impair permeability.
717 Additionally, the indigenous halophilic hyperthermal nature of bacteria can be
718 critically relevant to H_2 storage. Nevertheless, the effect of microbial activity can
719 inhibit at high temperature and high salinity environment and addition of microbial
720 resistant inhibitors
- 721 iv. Tiny H_2 molecules have high dispersion which may develop a gas slippage effect in the
722 salt caverns and H_2 can diffuse through the cavern and can raise the permeability
723 damage resulting problem during withdrawing the H_2 . Thus, it is recommended to
724 investigate the permeability of the salt cavern formation using H_2 gas. The
725 aforementioned case can help us to understand the optimized case of the gas cushion,
726 well performance and sustainability of the overall project.
- 727 v. Literature review unveiled that salt caverns consist of minerals such as calcite, pyrite,
728 anhydrite and halite. Thus, halokinesis is an unpredictable natural condition and it may
729 increase the permeability of the salt layer and develop the problems of the gas
730 breaching in the cavity. Thus, 90% of salt cavern problems are associated with H_2
731 leakages and their migration towards wellbore either resulting in corrosion (from H_2
732 embrittlement) or casing damage (salt creeping) and H_2 losses.

733 Moreover, injection/production cycles of H_2 can influence the geomechanical characteristics
734 of salt caverns. For this issue, it is recommended that the wellbore can be drilled with tailor-
735 made (morphology can be tuned) nanomaterial which may invade into the wall of the
736 wellbore and permanently seal the walls, moreover nanomaterial can be added in the cement
737 slurry which can adequately seal the annulus between the formation and casing to constrain
738 the exposure of H_2 in the wellbore. The storing capacity of the salt cavern wells (alone) has
739 been rarely investigated in the laboratory thus there is a need to examine the behaviour of
740 H_2 with steel tubular (such as casing and tubing) to counter the problems of hydrogen
741 embrittlement so that technology can be geared up for the LGS purposes. Salt caverns contain
742 SRB in the bottom of the cavern's sump and fillings. These bacteria can produce H_2S and can
743 alter the purity of the stored H_2 . Further, H_2S may catalyse the process of H_2 embrittlement
744 and corrosion failure in the completion string. Thus, we recommend examining the behaviour

745 of these microorganisms and their underground activity so that these microorganisms can be
746 neutralized from further imparting the H_2 storage process.

747 Additionally, microorganism activity and colonization can weak the sealing capacity of
748 the salt's wall and H_2 pressure in the salt cavern. Thermal energy can influence the
749 stability of the salt cavern's wall. Thus, there is a further need to examine and test the
750 strength of the wall under these realistic conditions (such as pressure and
751 temperature).

752 **8 'Supporting information**

753 Supporting information is available free of charge via the Internet at <http://pubs.acs.org/>.

754 **9 Acknowledgement**

755

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757 **10 Abbreviations and units**

758

Abbreviations

BP	British Petroleum
EIA	Environmental impact assessment
EU ESS	European Union Energy Security and Strategy
GHGE	Greenhouse gases emissions
IRB	Iron-reducing bacteria
IRENA	International renewable energy agency
LGS	Large green-scale
LPG	Liquefied petroleum gas
nD	Nanodarcy
Q1	First quarter
SRB	Sulphate reducing bacteria
UGHS	Underground hydrogen storage

Units

MJ·kg ⁻¹	Megajoules per kilogram
cm/s	centimeters per second
cp	centipoise
GWh	Gigawatt-hours
kg·m ⁻³	kilogram per cubic metre
mg/l	milligrams per litre
MPa	Megapascal
MJ·kg ⁻¹	Megajoules per kilogram
MWh	Megawatt-hour
ppm	parts per million

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762 11 Reference

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