

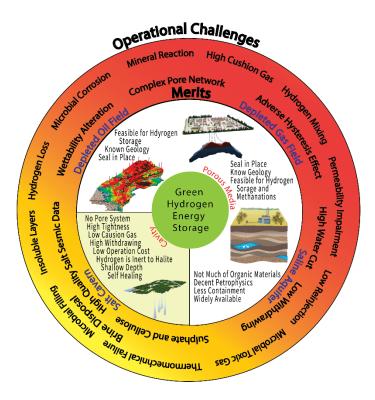
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Toward a Fundamental Understanding of Geological Hydrogen Storage

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Authors	Aftab, Adnan; Hassanpouryouzband, Aliakbar; Xie, Quan; Machuca, Laura L.; Sarmadivaleh, Mohammad
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1	Towards a fundamental understanding of geological hydrogen storage
2 3	Adnan Aftab ^{1,2,3} *, Aliakbar Hassanpouryouzband ⁴ , Quan Xie ¹ , Laura L. Machuca ⁵ , [,] Mohammad Sarmadivaleh ¹ *
4 5	¹ Curtin University, Discipline of Petroleum Engineering, 26 Dick Perry Avenue, 6151 Kensington, Australia
6 7	² Petroleum Engineering Department, Mehran UET, SZAB, Khairpur Mir's Campus, 66020 Sindh, Pakistan Pakistan
8 9	³ Energy Resources and Petroleum Engineering, King Abdullah University of Science and Technology KAUST, Thuwal, 23955-6900, Saudi Arabia
10 11	⁴ School of Geosciences, University of Edinburgh, Grant Institute, West Main Road, Edinburgh EH9 3FE, U.K.
12	⁵ Curtin Corrosion Centre, Curtin University, Bentley, Western Australia 6102, Australia
13 14 15	Corresponding Emails: <u>adnan.aftab@postgrad.curint.edu.au</u> <u>mohammad.sarmadivaleh@curtin.edu.au</u>
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- 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.

34 Abstract

Geological H_2 storage plays a central role to enable the successful transition to the renewable 35 H_2 economy and achieve net-zero emission in the atmosphere. Depleted oil and gas 36 37 reservoirs are already explored with extensive reservoir and operational data. However, residual hydrocarbons can mix with injected H_2 in the reservoirs. Furthermore, low density 38 and high diffusivity of H_2 may establish H_2 leakage from the reservoirs via fault pathways. 39 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 in the formations if storage scenario is not planned cautiously. While salt caverns are safe and 42 commercially proven geo-rock for H_2 storage, they have low-storage capacity compared to 43 44 depleted gas reservoirs. Moreover, salt structures (e.g., domel, bedded) and microorganisms activities in the salt cavern are limiting factors, which can influence the storage process. 45 Accordingly, we discuss challenges and future perspectives of hydrogen storage in different 46 47 geological settings. We also highlight geographical limitations with diverse microbial 48 communities and theoretical understanding of abiotic transformation (in terms of rock's minerals i.e., mica, and calcite) for geological H₂ storage. Regarding the fundamental behavior 49 of H_2 in the geological settings, it is less soluble in formation water, therefore it may achieve 50 less solubility trapping compared to CO_2 and CH_4 . Furthermore, H_2 gas could attain higher 51 capillary entrance pressures in porous media over CH_4 and CO_2 due to higher interfacial 52 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.

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

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61 **1** Introduction

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In 2020, the world's primary energy consumption and carbon emissions from energy use are 63 declined by 4.5% and 6.3%, respectively. This was mainly pushed by a record reduction in oil 64 65 consumption (9.1 million barrels per day) and considerable falls in natural gas use by 2.3%¹. In contrary to this, the energy demand growth rate was positive in 2019². The rise of COVID-66 19 and the uncertain schedule of partial/full lockdowns constrained the economical growth ³ 67 68 which results in a decline in the global demand for coal (down to 1.7 %) in 2019². Moreover, the demand for natural gas decreased to 2%, and oil demand reduced to 20% in 2020 Q1⁴. 69 Overall, CO_2 emission went down (which was 2.1% in 2018, and 0.5% in 2019) due to the 70 massive impact of COVID-19 onto the consumption of fossil fuels around the globe. The 71 negative trend of CO_2 emission was observed in different regions such as Europe (-8%), China 72 (-8%), and US (-9%)². Nevertheless, the global primary energy demand is 84% and still would 73 be delivered by fossil fuels. CO₂ emission growth by 0.5 % cannot be persistent anymore 74 75 because developed nations will once again gain momentum to retain their position in the 76 global energy supply market ².

European Union Energy Security and Strategy (EUESS) affirmed that the suppression of 77 greenhouse gases down to 20 to 30% by 2020 is key to protect climate change and 78 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 would be projected to produce from renewable energy systems alone in Europe by 2050⁵. 83 84 Additionally, South-East Asia has projected to achieve the 35 to 40 GW (35%) of renewable energy target by 2025 using wind and solar energy's lower costs ⁷. In 2020, the US has grown 85 renewable energy consumption, reaching a record high 12% of the total US energy demand 86 87 ⁸. Renewable energy sources are found better with diverse applications ⁹, in particular, minimum electricity bills ¹⁰, and low maintenance costs ¹¹. For example., solar system is the 88 green transformation of energy from sunlight into power through direct photovoltaic effect 89 or concentrated solar power system ¹². Additionally, the energy from the sun would not run 90 out unlike fossil fuels and it provides more heat energy than our need ¹³. Moreover, solar 91

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

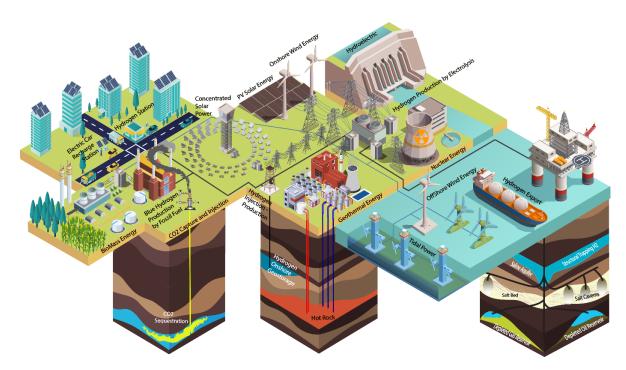


Figure 1 Graphical representation of entire carbon-free and sustainable hydrogen energy production and supply chain mainly comprising H_2 geostorage in depleted gas reservoirs and salt caverns

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Scientific community has made significant progress to generate H_2 using electrolysis reaction 113 using nanocomposite rods ^{30, 31}. The electrocatalysis reaction requires energy thus supply of 114 115 energy from solar parks and windmills can be used to produce 'green $H_{2'}$. For example., 9.1 ton of water produced 1.1 ton of H_2 , and 8 ton of O_2 without CO_2 emission using 39.4 MWh 116 solar energy 32 . H_2 can be used for passenger cars, prime movers, and buses, nevertheless a 117 flexible, cost-effective and environmental friendliness storage system is needed so that H_2 118 can be stored and reproduced adequately to compensate for fluctuating energy demand ³³ 119 without impacting the atmosphere. Energy yield of H_2 is higher (e.g., 120 MJ·kg⁻¹ energy 120 density) ³⁴ than CH_4 (45 MJ·kg⁻¹) ³⁴. However, surface storage of H_2 is a challenge ^{35, 36} because 121 of the low volumetric density of H_2 which is 0.0838 kg·m⁻³ at ambient pressure and 20 °C. 122 Additionally, liquid H_2 with 70.8 kg·m⁻³ is not an adequate choice for long-term storage. Liquid 123 H_2 can continuously be boiled-off resulting inefficient energy storage system and may rise H_2 124 leakage and H_2 loss problems ³⁷. 125

126 Underground H_2 storage could preclude multiple technical issues that constraint its 127 widespread use ³⁸. 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 precipitation in the pore-system due to release of by-products, including H_2S and the acids ^{39,} 129 130 40 Additionally, deep saline aquifer H_2 storage system can cause the problems of mineral dissolution and high water cut during the withdrawing period of H_2 ⁴¹. Thus, different 131 mechanisms of formation damages can occur due to fines mobilisation and migration 132 during hydrogen injection/withdrawal 42-44. Nevertheless, H_2 storage in salt caverns is a 133 proven technology ⁴⁵, owing to its inexpensive investment ⁴⁶, enhanced sealing properties ⁴⁷ 134 135 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 in casing and well completion assembly, which result in H_2 leakage ⁵⁰⁻⁵². 138

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 minimum mineral dissolution equilibria at the variable reservoir temperature conditions. 141 142 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 influence the gas mixing and propagation of microbial communities in the subsurface 144 formation ⁵³. Importantly, SRB have a significant effect on H_2 storage in the porous media as 145 well as in salt caverns. SRB may consume ${\it H}_2$ gas in their metabolic system and yield gases 146 147 (e.g., *CH*₄, *CO*₂, *H*₂*S*).

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 sustainability of H_2 underground storage ³⁹. In the context of storage security and 152 with drawing capacity, ${\it H}_2$ has a low density, low viscosity and high diffusivity when compared 153 to CO_2 and CH_4 . Thus, these fundamental and thermophysical behavior of H_2 may cause loss 154 of H_2 , and critically impact H_2 with drawal capacity. But, this could be prevented by cautiously 155 156 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 both offshore and onshore depleted gas fields. In depleted gas fields, caprocks could provide sufficient sealing corresponding to high interfacial tension between water+ H_2 and capillary entrance pressures ³⁷.

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

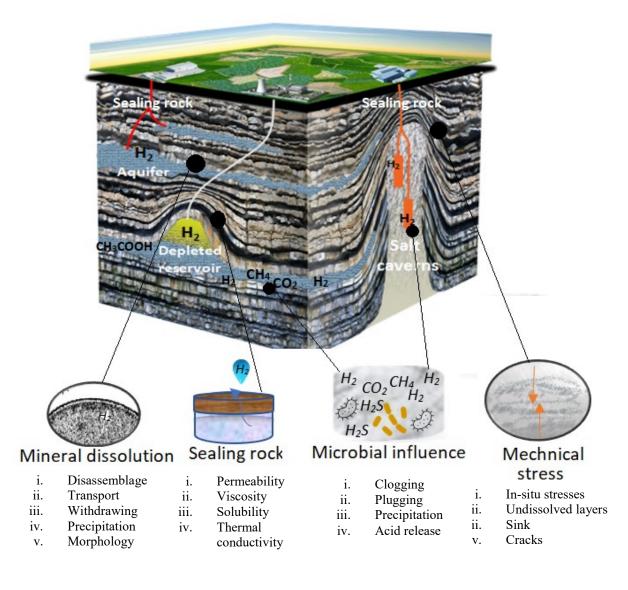


Figure 2 UGHS can be influenced by several factors, e.g., mineralogical alteration, sealing rock ability, microbial influence (natural, indigenous, and anthropogenic) and mechanical stresses

2 Abiotic and biotic transformation of the geological rocks

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- 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-

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

195 mineralogical transformations and affect H_2 production/injection cycle security.

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

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$$FeS+(1-x) H_2 = FeS+H_2S$$
 1

201 To further explore the phenomenon, researchers evaluated that pyrrhotite covered the pyrite particles when the temperature increased from 90 °C to 190 °C in the presence of H_2 at 116 202 psi partial pressure of H_2 . Thus, reduction in pyrite caused sulphide and precipitation in the 203 system and impairment of rock's flowing properties. Importantly, the amount of sulphide has 204 further reduced the pH of the solution which contains HS^- , $H_2S_{(aq)}$, $H_2S_{(g)}^{87}$ and can cause 205 mineral dissolution. Researchers carried out numerical analysis and experimental studies to 206 207 obtain geochemical reactivity of H_2 with sandstone which typically contains quartz, Kfeldspars, and other rock-forming units. H_2 could not bring any mineralogical variation in the 208 209 quartz nevertheless very minor alteration was obtained in hematite, and muscovite proportions⁸⁹. However, iron was released from the muscovite that did not impact the 210 permeability and porosity of the rock. Thus, sandstone was referred to as 'abiotic free 211 mineral' and illustrated no transformation during H_2 storage ⁸⁹. However, the effect of H_2 is 212 213 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}.

Subsurface formation holds diverse types of archaea and bacteria, here collectively 216 mentioned as microbes ³⁹. The potential microbial presence was observed 1-17*10⁷ cells ml⁻ 217 ¹ in formation water samples, which can consume <0.01 to 3.2% of H_2 ⁹⁰. The increasing 218 concentration of injected H_2 in the subsurface may trigger the microbe's growth ⁷⁷. 219 Additionally, pH of formation water and brine may influence microbial growth through 220 221 metabolism system and redox reaction. Methanogens, homoacetogens, and sulphate reducers are habited to 6.5 to 7.5 pH. However, the growth of most methanogens and 222 sulphate reducers were not found at 4 to 9.5 pH ^{54, 91}. Biotic mineral transformation can 223 adversely affect gas withdrawing, gas injection, formation permeability reduction and H_2 loss 224 225 ⁷⁷. SRB, methanogens and IRB can consume inorganic kosmotropic components include *NaCl*, KCl and sulphate sources (e.g., Na_2SO_4 , $FeSO_4$, K_2SO_4 , S) in the presence of stored H_2 and 226 metabolically release by-products (e.g., H_2S , acids, CH_4 , and CO_2) and ensured acidic 227 228 behaviour ⁹². SRB has induced carbonate precipitation and influenced the contamination security ⁹³. Most SRB including halophilic are permissive to growth and could induce stress in 229 the presence of O_2 and high salinity. Additionally, SRB consumes sulphate (as an electron 230 acceptor) in the presence of H_2 (as an electron donor). Additionally, H_2 energy-based 231 microorganisms lead by methanogens archaea 94 . Methanogens consumed CO_2 and H_2 in the 232 233 presence of inorganic minerals components e.g., nitrite, sulphate and iron oxide and metabolically released CH_4 and water ⁹⁵. Additionally, homoacetogens coupled H_2 oxidation 234 235 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 petrophysical property are not reported ^{97, 98}. However, chaotropy agents (e.g., MgCl₂, 238 FeCl₃, CaCl₃, LiCl, and LiBr) have ability to limit microbial life in the depleted oil/gas 239 240 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.

243 Table 1 Biotic transformation and impacts for H_2 geostorage

Hydrogeno trophic microbes	Reaction and possible mineralization	Impact	Study
SRB (Bacteria)	$SO_4^{2-} + 4H_2 + 2H^+ \rightleftharpoons H_2S + 4H_2O$ 3 H_2 reacts with anhydrite and other inorganic sulphate sources and reduces sulphate to sulphide.	 <i>H</i>₂<i>S</i> release High sulphide release Gas mixing Iron corrosion pH reduction Hydrogen embrittleme nt Mineral precipitation 	99-107
Iron reducing bacteria (Bacteria)	$3Fe_2O_3 + 5H_2 \rightleftharpoons 2Fe_3O_4 + H_2O_4$ Microbes can reduce passive film, e.g., ferric components on metal surfaces. By-product water can release and occupy interstitial pore space of sandstone causing excess water saturation and mineral dissolution	 Low sulphide reduction Metal corrosion Carbon steel corrosion Mineral dissolution 	108, 109
Methanoge ns (Archaea)	$CO_2+4H_2 \rightleftharpoons CH_4+2H_2O$ 5 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.	 CH₄ flow suppresses in Kaolinite Injection reduces Gas mixing Withdrawing and injection capacity reduces Permeability and porosity can alter 	110-112

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Overall, H_2S increased the acidic behaviour in the porous media and causing mineral precipitation ¹¹³, and formation clogging ¹¹⁴. H_2 reinjection would be challenging, if the rate of mineral precipitation is higher than the rate of mineral dissolution in the subsurface ⁵³. Literature hardly provides information regarding biogeochemical behavior of microbes with rock-forming minerals (e.g., mica, and calcite) in the presence of H_2 under reservoir conditions ^{39, 53, 55}.

251 **3** Review of H_2 geological storage in depleted oil/gas, and saline aquifer 252

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

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

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

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

- Regarding biotic transformation: Microbes can consume carbon energy from residual hydrocarbon ¹²¹ and speed up their growth process in the presence of H_2 . Moreover, sulphate is found in a considerable quantity in depleted oil/gas fluid wells. Thus, H_2 injection may reuse sulphate because of increased microbial growth ¹²².
- Regarding abiotic transformation: H_2 may dissolute reservoir rock-forming units at high water saturation. H_2 may react with sulphur-based minerals and produce abiotic H_2S , the process can deplete H_2 purity and may catalyse the kinetics of gas mixing ¹²³. However, it is difficult to comment either biotic or abiotic is a major source of byproduct gases and H_2 /by-product gas mixing ³⁹.

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

In principle, rigorous exploration work is carried out to determine storing and sealing capacity of gas in aquifer reservoirs ¹²⁹. Several storage wells are required to be drilled through the caprock into aquifer reservoirs. Aquifer well is drilled in multiple phases and account for both cementing and casing $cost^{130, 131}$. The aquifer rock contains brine or water which displace in the pore system by pumping H₂ ¹³². Thus, pressure can rise significantly due to compression in closed system aquifers thereby injection volume of gas in comparison to the formation breakdown pressure limit is small ⁸⁰. Ultimately both factors, such as an increase in pressure and permeability of aquifer ¹³³ can be used to measure the maximum H_2 injection rates ¹³².

Aquifers have the ability to store a large volume of gas ¹³⁴ but there are some issues such as chemical and biological activities (such as *biofilms* caused by anthropogenic activities) in the vicinity of wellbore ^{135, 136}. It is possible that during the reproducing/withdrawing period, high bottom hole flowing pressure (p_{wf}) (due to over volume of cushion gas) can push the H_2 gas back towards the wellbore and extend injection/pumping time ¹³⁷.

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

Overall, volumetric capacity, recovery efficiencies and rates of depleted reservoirs are not evaluated adequately and required in-depth investigation to formulate H_2 injection program ³⁷. Accurate evaluation of these factors is possible with history matching of numerical modelling and operational data during field-scale H_2 geostorage projects ¹⁴³.

328 **3.1** Underground H₂ geo-structural trapping

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

As aqueous phase (water, and organic traces) and non-aqueous (H_2 , CO_2 , H_2S and CH_4) may 334 basically act like hydrophilic and hydrophobic to the rock's surface and interstitial pore-335 336 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 pressure ¹⁴⁵. Therefore, H₂ trapping mechanism may possibly be affected via withdrawing, 338 re/injection, formation contamination (fines migration), bacterial based formation 339 precipitation and clogging, and mineral dissolution in pore-network system $^{146, 147}$. H_2 is high 340 diffusive gas and therefore the main risk associated with its storage is leakage via 341 underground natural pathways (such as., seepages, faults, and fractures) and drilled 342 wellbores. H_2 has a small molecular diameter ¹⁴⁸ (when compared to CO_2) ¹⁴⁹ and therefore, 343 344 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 trapping processes ³⁷. To date, the literature lacks to provide convenient information on the 346 H_2 trapping in subsurface media through the processes and leaves several open research 347 questions for the scientific community ¹⁵⁰. 348

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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 (e.g., high diffusivity, light weight, and low solubility etc.,) and chemical kinetics 352 (oxidation and reduction) may result in H_2 leakage from fault paths. Moreover, 353 information of H_2 reaction with impermeable strata (e.g., shale and mica) is 354 anonymous to date except for H_2 reaction with Kaolinite and Smectite. These clays 355 may mitigate H_2 flow in the narrow pore-network of cap rock (e.g., shale) and ensure 356 molecular H_2 structural trapping ¹⁰³. 357

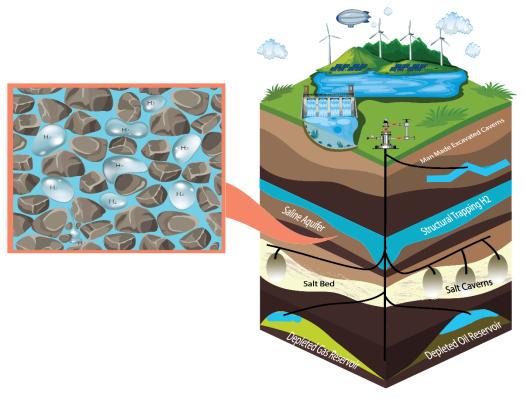
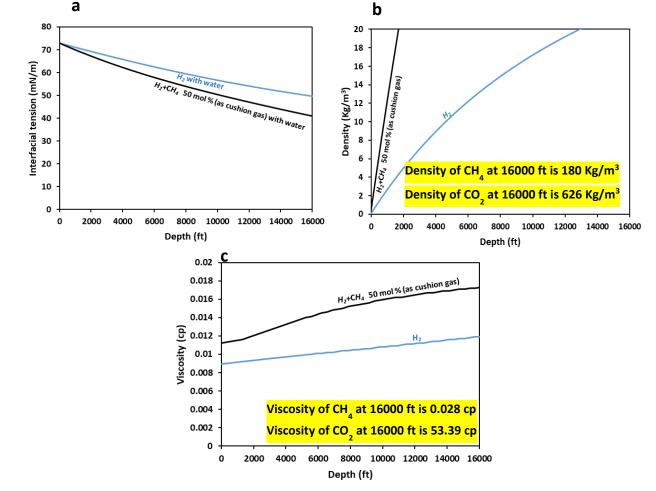


Figure 3 Structural trapping, capillary trapping, and mineral trapping have been illustrated in aquifer storage rock system (right). H_2 molecule tends to migrate upward to achieve residual trapping and structural trapping (left)

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359 H_2 gas bubble may trap in the pore system due to the influence of capillary forces and grain's morphology resulting in an impact on residual trapping ¹⁵¹. In our earlier study, we modelled 360 the interfacial tension of H_2 and CH_4 at storage conditions where higher capillary entry 361 pressure is predicted for H_2 37 . Moreover, morphology and size of pores in the rock can 362 363 influence the phenomenon. In typical water-saturated porous formation, interfacial tension and density behaviour of H_2 are different when compared to H_2 + CH_4 (H_2 +cushion gas) which 364 365 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 pressure) and thus water-saturated caprock would be able to sufficiently seal the stored H_2 367 (Figure 4a). Accordingly, hydrogen could be stored at a higher pressure than initial pre-368 production pressures in depleted gas fields ³⁷. However, it is important to maintain stored 369 370 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



influence the presence of e 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 wat er 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 studies ¹⁵²⁻¹⁵⁵ 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.

 H_2 is less soluble and lighter when compared to CH_4 and CO_2 thus it may achieve less solubility trapping. Solubility of H_2 , CO_2 and CH_4 in the water as illustrated in Figure 5a and b. Additionally, the adsorption process of gases (e.g., CH_4 , N_2 and CO_2) in the pore-network system can increase with increase in the kinetic diameter of the gas molecule ¹⁵⁷. Assuming the same principle for H_2 , the kinetic diameter of H_2 is less than CO_2 and CH_4 thus H_2 may achieve less adsorption in the pore-network system. Table S2 in the supplementary information provides the kinetic diameter of the gases and pore size of storage rock (e.g.,

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

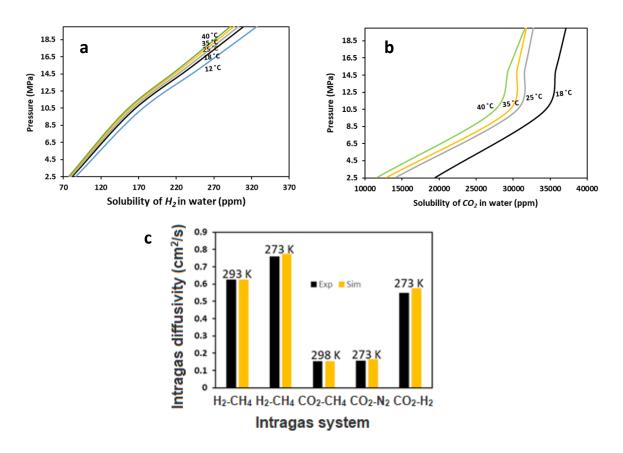


Figure 5 (a) Solubility of H_2 in water at variable pressure and temperature was determined using Valderrama-Patel-Teja (VPT) equation of state used in the study ¹⁵⁹ (b) Solubility of CO2 in water data at variable pressure and temperature was taken from the study ¹⁶⁰. Solubility of H_2 is significantly less when compared to solubility of CO_2 and CH_4 in water. (c) Diffusivity of H_2 in CO_2 and CH₄ and diffusivity of CO_2 in CH_4 are measured using Chen-Othmer technique ¹⁶¹ at variable pressures and temperatures

394

The trapping behaviour of sandstone (aquifer quartz surface) for H_2 at realistic temperature 395 396 and pressure conditions show that the rock surface was weakly/intermediate water-wet but after the addition of organic component onto the rock surface, the wettability of H_2 was 397 increased ¹⁶². Nevertheless, organic components in the porous media may raise problems e.g., 398 399 (i) organic components mixing with stored H_2 and (ii) influence on the microbiological life (as microbes may consume organic components as an energy source) at pore-scale level and 400 increase their activity $^{39, 55}$. Moreover, H_2 storage and structural trapping can be influenced 401 402 by the mineralogy of sealing rock and hysteresis effect ^{55, 163}. The careful analysis of hysteresis 403 can help to adequately predict H₂injection (primary drainage), H₂ gas production (secondary imbibition) and H_2 re-injection (secondary drainage) ¹⁶³. The recent simulation study 404 quantified the trapping behaviour of H_2 /brine in sandstone formation via interpreting the 405 hysteresis effect ¹⁶³. However, they observed that cyclic hydrogen storage in the underground 406 407 formation resisted the hysteresis effect which has been previously used to examine the gas storage (in particular CO_2 , and CH_4) mechanism at continuum scale via capillary pressure and 408 409 relative permeability information. These both properties are sensitive to contact angle in the H_2 storage system. Additionally, high frequent production/reinjection cycles may cause 410 411 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 gas. Therefore, detailed study of the storage rock via pore network imaging (e.g., mineral 413 precipitation), kinetic rates and sensitive analysis of the modified hysteresis effect can provide 414 necessary information of H_2 residual saturation after secondary imbibition ^{163, 164} which is 415 essential for the process of H₂ production from geological formation. Operational parameters 416 and feasibility of H₂ storage in different storage systems are given in Error! Reference source 417 418 not found.

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	
рН	7	7		39
Water cut	30 to 70%	30 to 70%	80 to 90%	169

Table 2 Comparison of underground porous reservoir rocks for hydrogen storage

420

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

422

In comparison to H₂ storage in the porous rock, its storage in salt cavern has been less 423 challenging due to the reason that salt layers are impermeable and inert to H_2 . Moreover, 424 withdrawal capacity of H_2 is more efficient in the salt cavern when compared to porous rock 425 426 as could be used as huge storage tanks. Nevertheless, field-scale H_2 storage in the salt cavern has several challenges. Gas storing capacity and salt stability in salt caverns are key factors for 427 the consideration of H_2 storage ^{48, 170}. Large pillars of thick salts layers of caverns (both top 428 and bottom layers) presented efficient H_2 storing capacity 171 . Realistic experience of salt 429 cavern H₂ storage found satisfactory relative to high cavern pressure potential and 430

homogeneity in the salt formation ¹⁷². The large salt caverns are constructed in salt domes 431 (with uniform layers of salt) near the Gulf Cost in the US¹⁷³. However, the geology of salt 432 433 caverns located in Western States, North-eastern, and Midwestern is not homogenous and as such inadequate for storing H_2 in the US¹⁷⁴. In US, Germany, and the UK, H_2 salt cavern 434 realistic storage projects provided four decades of experience. This experience can be utilized 435 for the establishment of LGS H_2 storage capacity ¹⁷⁵. The large volume of the cavern can 436 provide a high flow rate of H_2 . Capsule type salt cavern is considered more appropriate 437 choice for H_2 storage because of its better height and adequate stability (e.g., under low 438 tensile and overburden stresses) ¹⁷⁶. The storing media has adequate stability to high pressure 439 440 and low-temperature conditions ^{33, 177}. Moreover, smooth operational proceedings of the project are very pivotal for its successful completion $^{178-180}$. Salt cavern H_2 sealing capacity is 441 442 high however mechanical and mineralogical properties of the salt cavern are important factors ¹⁸¹ to endure compressive and tensile failure. Moreover, low working pressure may 443 444 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 cavern for less injection time and optimum delivery of H_2 ¹²⁴. However, salt cavern provided 446 447 low storage capacity and did not provide long-term storage solution when compared to depleted oil and gas reservoirs. Operational parameters for underground hydrogen storage 448 in salt cavern are provided in Table S3. 449

450

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

452

Selection of salt cavern for the H_2 storage depends on the depth, purity, composition, and wall thickness of the salt cavern ¹⁸². Continuation, distribution, and thickness of the rocks (evaporate type) varied within the basin area which is further based on the formation of the salt structures such as pillows, salt stocks and diapir. Investigators found that average salt structures can store around 214 kWh and 458 kWh per m³ ⁴⁹ ¹⁸³. Nevertheless, salt dome structures have more energy storage density such as 210 GWh when compared to a salt bedded structure which could provide around 65 to 160 GWh energy storage capacity ¹⁸³. Salt beds and domes are naturally developed due to movement between the substrate and overlying strata in the presence of low-density salt; the mechanism is known as 'halokinesis' ¹⁸⁴. This mechanism could be influenced by buoyancy, gravity difference, salt spreading, thermal convection and differential loading forces ¹⁸⁵⁻¹⁸⁷. Buoyancy forces and low-density salt layers are primary reasons for the formation of a salt dome in comparison to the effect of tectonic activity ¹⁸⁸ as depicted in Figure 6**Error! Reference source not found.**.

Typically, salt caverns are made up of halite salt along with anhydrite salt (layers). Both halite 466 467 and anhydrite remain as the main sources of salt cavern's brine composition. Adequate salt diaper which is composed of 99% halite (NaCl) and less than 1% of anhydrite ($CaSO_4$) salt and 468 other impurities ¹⁸⁹. However, the composition of salt bedded layers and salt domes are based 469 470 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 to its high thermal conductivity, self-healing properties, impermeable behaviour and plastic 472 properties, it could establish potential sealing to mitigate H_2 leakage in the system ¹⁹⁰. 473 474 Importantly, halite could not react with H_2 . However, there were some common impurities 475 such as quartz (SiO₂), anhydrite ($CaSO_4$), dolomite ($CaMg(CO_3)_2$), pyrite (FeS_2), gypsum $(CaSO_4 \cdot 2H_2O)$, and calcite $(CaCO_3)$ could be found in the salt layer which may react ¹⁹¹ but 476 477 requires further investigations. Moreover, some common ions impurities are also observed e,g., as Fe⁺³, Mg⁺², CO⁻², Ca⁺², SO₄⁻², Fe⁺², K⁺, and Cl⁻, Ba⁺², B⁺³ and Sr^{+2 49} which can enhance 478 479 microbial activity. Additionally, the mineralogical properties of anhydrite salt are required to 480 be analysed because of its hydroscopic 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,} ¹⁹³ which can cause an inappropriate delay in salt cavern construction. High concentrations of 483 Ca^{+2} , and SO_4^{-2} ions from anhydrite remain in the cavern and it could form H_2S in the presence 484 of H_2 or methane through both biotic and abiotic sulphate reduction. H_2S has corrosive (can 485 develop H_2 embrittlement problems in casing and steel tubulars) and toxic nature which may 486 487 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 the tensile and ductile strength of the tubular and de-velops the corrosion films of FeS and 489 free H^+ on the surface of metal. Thus, free ions of H^+ along with S^{2-} accelerate the corrosive 490

rate via H_2 or H^+ ion trapping in the interstitial spaces of the metal promoting hydrogen 491 492 embrittlement in the system ¹⁹⁴. Hence, H₂ can cause the embrittlement problems and proceed the H^+ diffusion and its adsorption in the interstitial spaces (metal lattices) ^{195, 196}. 493 Cavern leakages problems are mainly associated with 'Fillings' (e.g., undissolved rock layers) 494 in salt caverns, overburden compressive forces, tensile forces, microbial degradation activity, 495 well casing leakages and H₂ embrittlement (corrosion damage) ¹⁹⁷⁻¹⁹⁹. Figure 6 provides a 496 drawing of a salt cavern and its problems which can influence the UGHS process. Thus, self-497 healing (natural cavern's wall recovery) and tight permeability of salt cavern's wall (for H_2 498 entrapment in pores) could be two pivotal factors for enhanced H_2 gas storage and security. 499 500 The permeability of 1 m thick salt layer is evaluated 2 nD at 1 MPa pore pressure value ²⁰⁰ nevertheless typical permeability of tight gas reservoir is 1000 nD ²⁰¹. However, in amateur 501 502 salt structure, mechanism of 'halokinesis' and the contaminations in the salt formation can increase the permeability of salt cavern resulting H_2 leakage and inadequate reinjection of 503 H_2 into salt cavern. Table S4 presents the review of salt cavern performance, challenges, and 504 505 adequate solutions.

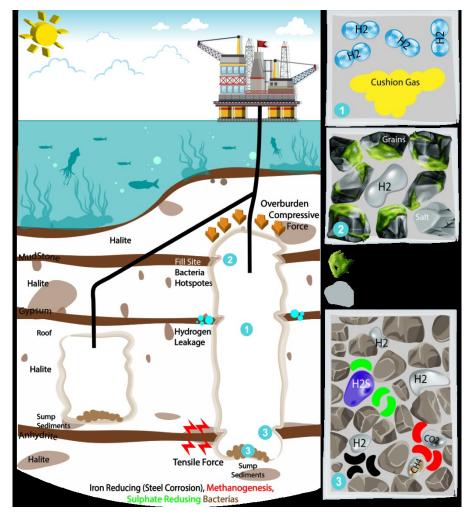


Figure 6 Salt caverns challenges have been illustrated at a microscopic level: 1) Cushion gas (e.g., N_2 and CO_2) is recognised as inert gas. Nevertheless, H_2 withdrawing cycles may ensure in-situ thermal variation and can impact H_2 mixing with the inert gas ¹³². 2). Salt has natural self-healing properties ²⁰². 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.

511 **4.2** Biogeochemical behaviour of H_2 in salt formation

Salt formation is inert and inactive to hydrogen ²⁰⁵. However, brine water can stay in the salt 512 cavern's sump. Brine water may evaporate into stored gas ²⁰⁵ and increase moisture content 513 in the gas which can be a challenge and requires surface facilities for dehydration of H_2 . 514 Overall, the amount of contamination in the withdrawn H_2 gas from salt cavern is less when 515 516 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 microorganism (biotic) and host rock (abiotic) ²⁰⁶. Thus, withdrawing and reinjection capacity 518 of H_2 may be influenced in depleted reservoirs. The practical experience shows that both 519 natural gas and H₂ were stored together in salt caverns successfully for several decades in the 520 UK, the US and Germany ^{79, 207}. The inert nature of salt with hydrogen yields overall high 521 522 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 durable, contaminated free, and viable to store gases and liquids at high-pressure conditions 525 ²⁰⁸. However, the main problem is the storage capacity of salt caverns which is comparatively 526 527 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 Table 3. Artificially developed salt caverns have been used for 50 years, and primarily used 530 531 for the storing purpose of hydrocarbon, in particular methane, and oil, later it has been noticed as a potential storing media for H_e , and H_2 . First time in US, oil and LPG were both 532 stored in salt caverns in 1950. In 1961, the very first salt cavern was used for the storing of 533 natural gas in US $^{\rm 209}$. Nevertheless, the first salt cavern for the storage of H_2 was developed 534 in the UK in 1971 and it is operational to date. Interestingly, around 300 and 2000 salt caverns 535 536 are in Germany and North America currently used for the storage of compressed air, compressed H_e , butane, radioactive waste and compressed H_2 ^{208, 210}. 537

Factor	Configuration	References
Volume	Storing volume 100000 m ³ , and vary with geographical	210, 211
	based on the technical as well as geological history,	
	geographical location and behaviour of the salt cavern	
Salt	At least 200 m	210-212
thickness		
Depth	At least 500 m to 2000 m	210, 212
Height	At least 400 m	210, 213
Operating	20 MPa is essential for high volumetric storing capacity	210, 212
pressure		
Mechanical	Optimum mechanical stability of salt cavity is key to	210-212
properties	sustainable H_2 storing	

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

540

High diffusive behaviour of H_2 and the bacterial process can damage the sealing behaviour of 541 the cavern and contaminate the stored H_2 purity ⁴⁹. Microorganisms are living in the sump's 542 residual water with indigenous microorganisms communities since the leaching process. The 543 544 sump may contain anthropogenic drilling material which may provide energy sources (e.g., cellulose, starch, SO₄ and insoluble carbonates) to bacteria ²⁰³. Thus, bacteria can start 545 consuming H_2 and generate H_2S in the presence of carbonate and sulphate via sulphate 546 reduction ²¹⁴. Desulfovibrio halophilius and Desulfovibrio Vulgaris utilized sulphate for the 547 acceptance of electrons in the environment of anaerobic metabolism ^{215, 216}. Bacterial survival 548 is based on sulphate reduction such as from sulphate (S[+VI]) to sulphide (S[-II]). Sulphate is 549 activated before it can accept the electron via enzyme adenosine triphosphate sulfurylase 550 551 phenomenon, and thus synergy behaviour of triphosphate sulfurylase+sulphate develops adenosine 5' phosphosulfate and reduced sulphate to sulphide ultimately produce H_2S in the 552 presence of H_2 ²¹⁷. Table 4 illustrates that different types of SRB have the ability to survive 553 typical reservoir temperature condition. In fact, the growth of these bacteria expedited at 554 555 these temperatures and increased sulphate reduction process. Figure 7 depicts that SRB have been found in salt cavern, depleted hydrocarbon reservoir rocks and saline aquifer which can 556 generate H_2S in the system ^{49, 218-225}. Some storage rock has shown high H_2S generation when 557 558 compared to the allowable limit.

559

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

Type of bacteria	Factors	References
Thermochemical	100 to 180 °C	218
sulphate		
reduction		
Bacterial sulphate	0 to 60-80 °C	226
reduction	Some greater than 80 °C	
Hyperthermophilic	110 °C	227
SRB		
Optimal growth	38°C, acidic, saline environment.	218
temperature for	Mostly SRB activity reduces, if concentration of NaCl increases	
the SRB	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.	

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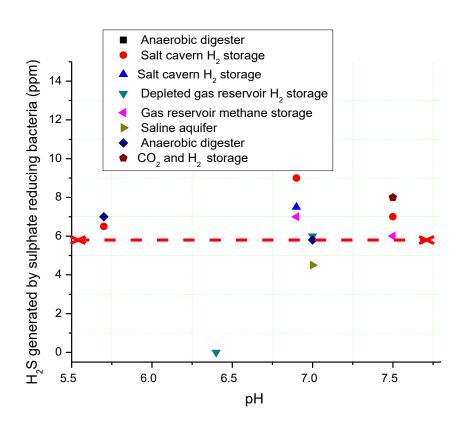


Figure 7 Sulphate reducing bacteria has generated H_2S in the surface and subsurface storage systems. Storage rock has shown high generation of H_2S which is greater than its threshold value (red dashed line) ^{49, 218-225}.

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

Selections of fresh water, drilling fluids and completion fluids are important factors during the 566 construction of a salt cavern. Underground salt formations are not sterile and contain multiple 567 types of indigenous, natural, and anthropogenic microorganisms. The supplement source of 568 energy from fresh water (such as carbonates (CO_3^{2-}) , and Mg^{+2} , and Ca^{+2} etc.,) and drilling 569 fluids (e.g., barium sulphate (BaSO₄), cellulose and starch polymers) may improve the growth 570 of microbes inside the caverns and rise problems. Leaching, debrining, and filling are pivotal 571 phases of underground salt cavern construction ^{228, 229} and carried out into geological salt 572 deposits as illustrated Figure 8. The geological salt deposit, in particular, salt domes is selected 573 after careful assessment of the previous exploratory and drilled wellbore data ²³⁰. Time period 574 for the development of salt cavern based on the areal extent of salt formation and formation 575 hydraulic diffusivity, however, it might take more than one year to construct a single cavern 576 ^{177, 231}. The drilled hole which could be used for the injection of H_2 based on the factors, 577 including geological formations, structure, fault locations and depth of the salt caverns (which 578 typically range from 300 m to 2000 m). Like in oil and gas well drilling, the salt cavern wells 579 580 are also drilled in a telescopic design, however, the diameter of salt cavern wells is larger when compared to conventional oil and gas wells ²³². These caverns are cased and cemented 581 adequately so that H_2 gas may not leak and sustain smooth H_2 injection/production cycle ²³³. 582

Tightness of the salt cavern well is examined to predict cementing and casing leakages problems ²³⁴. Then, brine is displaced towards the surface via injection of H_2 inside the salt cavern during filling phase. Nevertheless, 100% brine recovery is not easy because production tubing did not reach until the bottom of salt cavern ²³⁵.

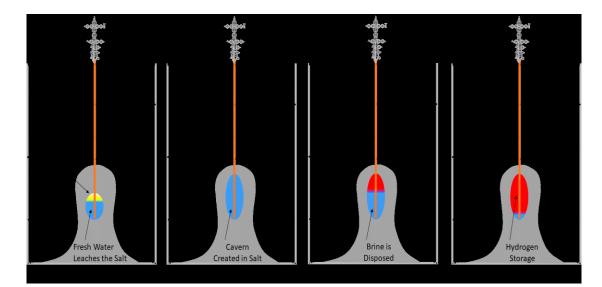


Figure 8 The process of salt cavern development involves multiple steps such as i Salt formation selection ii Leaching, iii Debrining phase and iv Filling

587

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

In Utah US, researchers are working to store H_2 underground in a vertical salt cylinder which is 804 m wide and 1609 m deep. This could be the largest salt cavern reservoir to store underground H_2 in the history of the US. The project could produce around 1000 MW of clean energy via stored H_2 . The stored H_2 would be enough power to provide electricity to 150,000 households for the period of one year. ²⁴¹. In the first phase of the project, H_2 energy storage would supply around 150,000 MWh of storing capacity which is 150 times higher energy when compared to the existing lithium-ion batteries storage system in the US ²⁴¹. Additionally, the cost of underground salt drilling and maintenance is 10 times less than surface H_2 storage tanks system and it is 20 times less when compared to hard rock mines expenditures. According to the US Strategic Petroleum Reserves data, the US has around 60 caverns which are typically 200 ft diameter and 2500 ft tall. Table 5 presents the underground salt cavern H_2 storage projects information.

613 Table 5 Operational H₂ salt cavern projects

Countries	Salt cavern H_2 storage sites	Potential of salt caverns	References
The UK	Teesside (operational)	 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	 Depth of 800 m (cavern top) Storing volumes 580,000 m³ capacity 	33, 242
	Salt cavern H_2 stored facility in Texas since 1980s.	 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 suitable H_2 storing program ²⁴³. The method for the measurement of volumetric capacity is 616 defined in the previous study ³³. Typical cavern construction involved multiple steps such as 617 618 in-depth geological investigation which illustrates information of salt domes, inter-bedded salt structures and their areal extent ³³. Basic parameters are pivotal to be considered e.g., 619 cavern roof depth, storage pressure, gas cushion, water volume in the cavern, and surface 620 pipeline facility. The cavern's thickness of 500 m to 2000 m and height of 500 m to 1200 m 621 would be an appropriate choice for 7.09 MPa to 19.2 MPa storage pressures ³³ This could be 622 623 explained that high thickness salt wall and roof are required which can sustain high injection pressure and microbial degradation activity in the system ²⁴³. 624

Importantly, the cost of the cavern during gas injection remained consistent until unless the underground H_2 storage pressure of the system is maintained. Hence, maximum underground pressure should be maintained $P_{max} > 7$ MPa. This may influence the cost of the compressor capacity and its power consumption. Moreover, the cost may raise with the depth of the injection well bore and the use of high-grade tubular accessories (both casing and completion). Additionally, it is pivotal that cushion gas is controlled and made consistent around 30% of storing volume ³³. Additionally, H_2 withdrawing pressure can be maintained adequately relative to cushion gas volume pressure ²⁴⁴.

5 H₂ energy storage capacity in pipelines, gasometer tanks, depleted gas reservoir and salt caverns

635

636 We found that a single Marlin offshore field in Australia could store more than 100 TWh energy storage capacity of H_2 as illustrated in Figure 9a. This is enough energy to fulfil the 637 demand of South Australia which annual demand was recorded 89.61 TWh in 2019 to 2020 638 ²⁴⁵. Underground H_2 trapping is safe, environmentally friendly and provide massive H_2 energy 639 640 storage solution when compared to surface H_2 storage system. However, surface liquid H_2 storage system may loss and boil off H_2 due to heat transfer from the environment ²⁴⁶. Thus, 641 depleted gas reservoirs and salt caverns are apparently adequate choices for long-term and 642 short-term H_2 storage. 643

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

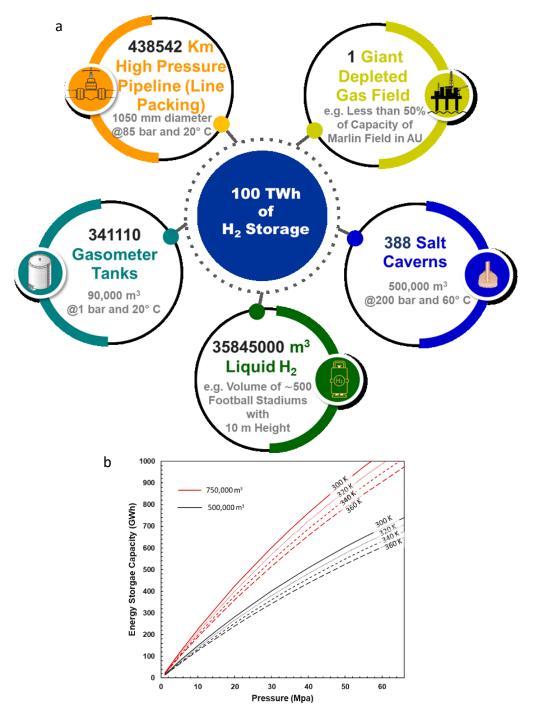


Figure 9 (a) Single Marline offshore depleted gas field could store 100 TWh of H_2 energy which is equivalent to 388 salt caverns capacity (each 90,000 m³). Each surface facility including 34111 gasometer tanks (each 90,000 m³) and 438542 Km line packing (diameter 1050 mm) could store 100 TWh. (b) Energy 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:

i. H_2 energy and its storage technology are pacing with unprecedented momentum. 657 658 Thus, the prospect of H_2 energy seems to be very bright due to its overwhelming advantages: clean source of energy, amazing energy density, and transformative 659 nature. H_2 can be produced through green techniques. It has the potential to be used 660 in various practical applications with high energy performance facilitating social 661 growth, economic competitiveness, and environmental benefits. More importantly, 662 developed economies in particular Australia, Germany, the UK, the US have realised 663 that there is a need to scale up H_2 production and its flexible storage system at the 664 LGS to deliver a future featuring "energy accessibility, security, affordability and 665 666 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.

iii. Depleted natural gas and oil reservoirs are considered for H_2 storage because of their 672 benefits, such as these are seismically proven large extended areas. Moreover, H_2 673 storage operator will have enough former mineralogical and geological (drilling order 674 data) to understand the appropriate physiochemical behaviour of H_2 and their 675 underground interactions with the grains and in-situ residual saturations. 676 Nevertheless, if H_2 is stored in the depleted natural gas fields, it can contaminate with 677 residual natural gas, and thus when H_2 will be withdrawn from the reservoir it can be 678 used for heating purposes. H_2 storage capacity of one depleted gas reservoir is 679 680 significantly higher when compared to energy storage capacities of salt caverns, gasometer tanks, and extended pipelines. 681

iv. So far geological aquifer formations are proven realistic storing media for CH_4 rather than H_2 . Moreover, the aquifers do not contain residual hydrocarbon saturation which may affect the composition of H_2 . However, the H_2 storage operator is required to fulfil additional exploration costs. Additionally, aquifers may contain microorganisms that can react with H_2 and ensure precipitation mechanism in the porous media; thus, the permeability of the reservoir can be influenced. Additionally, dissolution of H_2 in aquifers may not be negligible because of the huge amount of available water.

689v.Salt caverns are proven for H_2 storage capacity and security. Nevertheless, salt caverns690have very low-storage capacities in several orders of magnitudes (areal extent and691height) when compared with depleted gas fields. Additionally, salt cavern wall's692thermal stability, microorganism contaminations and H_2 withdrawing ability are693challenges to be overcome for its implementation in struggling economies. Thus, by694resolving the said challenges in salt caverns, we can explicitly exploit the potential of695realistic, clean, high efficiency, futuristic energy storage system.

696 7 Future road map and recommendations for underground *H*₂ energy storage697

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

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

ii. Multiple H_2 withdrawal/injection cycle may raise in-situ thermal and pressure variation in the reservoir which results in mineral dissolution altering reservoir permeability and sealing capacity. The effect of physical and thermal stresses in the reservoir requires further investigation, coupling abiotic reactions e.g., pyrite can transform to pyrrhotite causing H_2S release. However, H_2 did not occur any mineralogical alteration in sandstone. Nevertheless, the reaction of H_2 on carbonate and shale is yet to be answered. 714iii.Biotic transformation in the storage rock varies with different factors including715temperature, brine water activity, salinities and pH. Microbial activity can develop716biofilms that can plug H_2 flow in the porous media and impair permeability.717Additionally, the indigenous halophilic hyperthermal nature of bacteria can be718critically relevant to H_2 storage. Nevertheless, the effect of microbial activity can719inhibit at high temperature and high salinity environment and addition of microbial720resistant inhibitors

iv. Tiny H_2 molecules have high dispersion which may develop a gas slippage effect in the salt caverns and H_2 can diffuse through the cavern and can raise the permeability damage resulting problem during withdrawing the H_2 . Thus, it is recommended to investigate the permeability of the salt cavern formation using H_2 gas. The aforementioned case can help us to understand the optimized case of the gas cushion, well performance and sustainability of the overall project.

727v.Literature review unveiled that salt caverns consist of minerals such as calcite, pyrite,728anhydrite and halite. Thus, halokinesis is an unpredictable natural condition and it may729increase the permeability of the salt layer and develop the problems of the gas730breaching in the cavity. Thus, 90% of salt cavern problems are associated with H_2 731leakages and their migration towards wellbore either resulting in corrosion (from H_2 732embrittlement) or casing damage (salt creeping) and H_2 losses.

733 Moreover, injection/production cycles of H₂ 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 the exposure of H_2 in the wellbore. The storing capacity of the salt cavern wells (alone) has 738 739 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 SRB in the bottom of the cavern's sump and fillings. These bacteria can produce H_2S and can 742 alter the purity of the stored H_2 . Further, H_2S may catalyse the process of H_2 embrittlement 743 and corrosion failure in the completion string. Thus, we recommend examining the behaviour 744

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

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

752 **8** 'Supporting information

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

754 9 Acknowledgement

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757 **10** Abbreviations and units

758

Abbreviations

BP	British Petroleum
EIA	Environmental impact assessment
EUESS	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
ср	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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