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18 **Abstract**

19 Indubitably, hydrogen demonstrates sterling properties as an energy carrier and is widely anticipated as
20 the future resource for fuels and chemicals. Herein, an updated assessment of progress recorded on the
21 production, transportation, utilization, and storage of hydrogen is examined. Firstly, the numerous
22 routes for the production of hydrogen from renewable and non-renewable sources are systematically
23 demystified. Subsequently, the transportation framework for hydrogen is discussed. Moreover, the
24 industrial application of hydrogen is elucidated and the existing hydrogen storage systems are
25 chronologically analyzed. From the review of the literature, photocatalytic water splitting technology
26 is the most environmentally benign method available for H_2 production. Additionally, underground
27 hydrogen storage in a geologic porous medium offers the largest and most affordable storage capacity
28 for H_2 gas. Nonetheless, the compatibility of H_2 with reservoir fluid in porous media and other safety
29 concerns has to be adequately understood to avoid preventable losses. Finally, the key research gaps
30 were highlighted and potential areas for future research are proffered.

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32 **Keywords**: Hydrogen, production, transportation, utilization, storage, and hydrogen economy.

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Contents

1. Introduction

Presently, fossil fuels play a pivotal role as the primary energy source in modern-day society. Remarkably, the rise of the industrial revolution was facilitated by the advances in the storage capabilities of these fuels. However, as we move into the 21st century, continued reliance on non-renewable fuels as the primary source of energy is unsustainable due to the rapid depletion of its resources and the substantial emission of greenhouse gases caused by the combustion of these fuels [1]. To this end, efforts are being put in place to ensure a sustainable energy transition. This resulted in the development of several environmentally sensitive initiatives such as the 2015 COP21 Agreement in Paris [2], COP26 in Glasgow [3], COP27 in Egypt [4], and the 2030 climate and energy framework. The objectives were (i) to reduce greenhouse gas emissions by 40% compared to the pre-industrialized period, (ii) to attain at least a 32% share of renewable energy, (iii) to increase energy efficiency by at least 32.5% [5]. However, to achieve these targets, decarbonization mainly in the transport, power, heating, steel, and other energy-intensive industries will be required as they are the largest users of fossil fuels. Furthermore, increased energy production from renewable energy sources must be promoted to limit the dependency on fossil fuels (**Fig. 1**) [6–11].
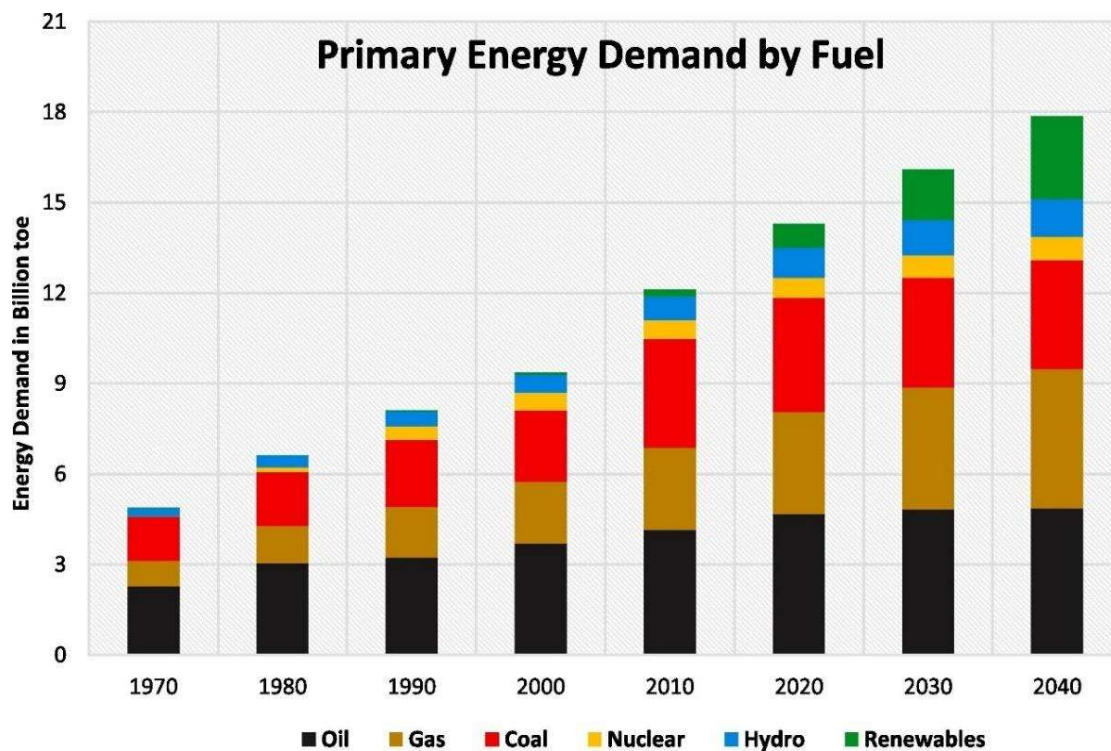
Fig. 1. Past, present, and predicted energy demand for various fuel consumptions [12].

Recently, hydrogen (H_2) has been identified as a renewable energy carrier/vector in a bid to tremendously reduce acute dependence on fossil fuels. **Table 1** shows a comparative

characteristic of H_2 with conventional fuels and indicates the efficiency of a hydrogen economy. The term "Hydrogen economy" refers to a socio-economic system in which hydrogen is utilized as a primary energy carrier, replacing or complementing traditional fossil fuels. A hydrogen economy envisions a transition from carbon-based energy sources to hydrogen-based energy infrastructure, to achieve a more sustainable, clean, and efficient energy system [13]. The successful implementation of a hydrogen economy requires advancements in hydrogen production, transportation (and/or distribution), utilization, and storage technologies, as well as the establishment of supportive policies and infrastructure to enable widespread adoption [14].

Table 1. Hydrogen characteristics with other conventional fuels [15,16].

Properties	Hydrogen ($H_{2(g)}$)	Methane ($CH_{4(g)}$)	Petrol/Gasoline	Diesel
Energy density (MJ/kg)	120.00	55.60	46.40	45.60
Density (kg/m³)	0.089	0.657	715 – 780	875 - 959
Boiling point (°C)	-253	-162	27 - 225	187 - 343
Volume occupied by a unit mass (L/kg)	1123.6	1522.1	1.3 – 1.4	1.0 – 1.1
Volume occupied by unit energy (L/MJ)	100	25	0.03	0.026
Heat of vaporization (KJ/kg)	444	577	348.9	232.6
Low to High heating value (MJ/kg)	120 – 141.9	50 – 55.5	43.4 – 46.4	42.6 – 45.6
Diffusivity in the air (cm²/s)	0.63	0.20	0.002	0.0008
Flammability range (vol% in the air)	4 - 75	5 - 15	1 - 8	0.6 – 7.5
Ignition temperature in air (°C)	585	534	280	210
Ignition Energy (MJ)	0.02	0.30	0.8	20
Flame Velocity (cm/s)	270	34	47	40
Research Octane Number (RON)	> 130	125	87	30
Flash point (°C)	-253	-188	-45	62

Generally, hydrogen is produced from renewable and non-renewable energy sources. However, production from non-renewable sources presently dominates the market due to intermittency and fluctuations inherent in renewable sources. Currently, over 95% of H_2 production is from fossil fuels (i.e., grey H_2) via steam methane reforming (SMR) of natural gas (**Fig. 2**). The retrofitting of SMR with carbon capture and storage (CCS) technology produces blue H_2 [17–19]. On the other hand, green H_2 is produced from the splitting of water molecules via electrolysis with an electricity supply from renewable sources (such as wind, solar, and others)

134 [20–22]. Moreover, brown H_2 is produced from coal gasification processes [23]. In a bid to

135 speed energy transition, there are growing calls to increase H_2 production from renewable

136 sources. Besides, research and development are focusing on cost-effective production

137 strategies.

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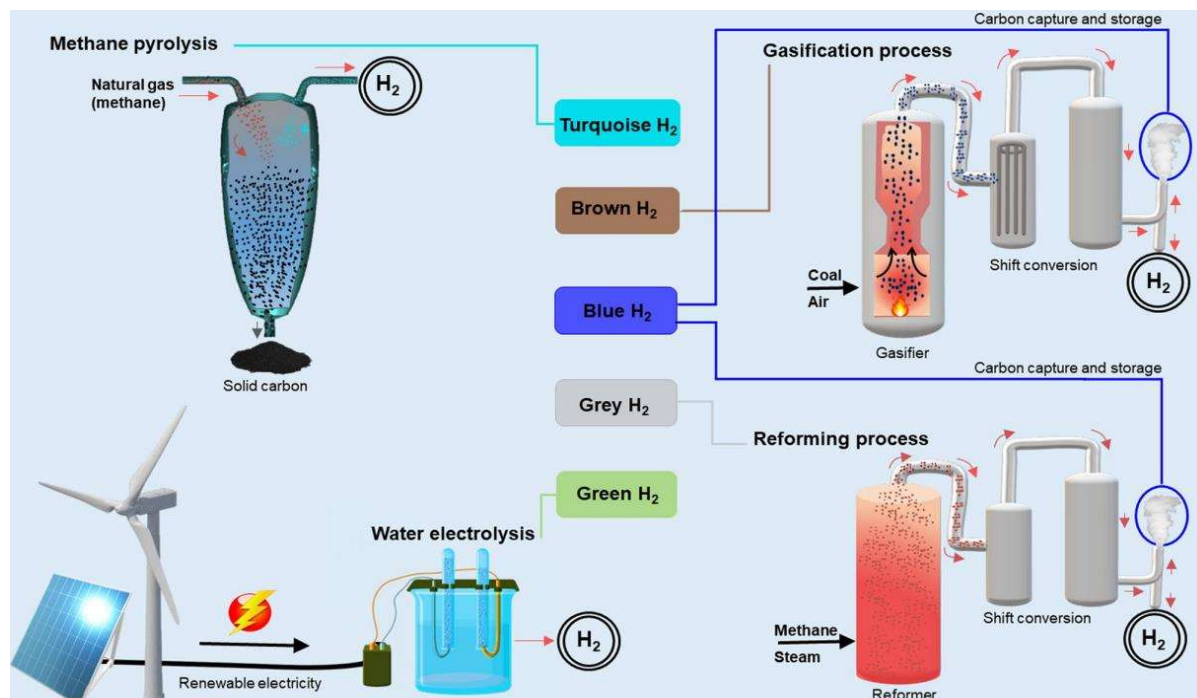
139 **Fig. 2**. Sources of H_2 production and their colors. Utilizing water electrolysis and renewable energy
140 sources like solar or wind energy, green hydrogen is created. Methane steam reforming and coal
141 gasification, respectively, yield grey and brown hydrogen, and when these processes are combined with
142 carbon capture and storage, blue H_2 is created. The pyrolysis of methane results in the production of
143 turquoise H_2, along with solid carbon as a byproduct [22].

144 Apart from its production methods, hydrogen transportation, utilization, and storage play a

145 crucial role in the development and success of the hydrogen economy. Hydrogen transportation

146 involves the development of safe and cost-effective transportation infrastructure, including

147 pipelines, compressed gas cylinders, liquid hydrogen carriers, and fuel cell vehicles to aid

148 availability and accessibility to end users. Meanwhile, the utilization of hydrogen in energy-

149 intensive industries such as transportation, chemical feedstock, iron and steel, cement, heat and

150 power, and others will significantly reduce associated emissions which account for huge

151 environmental pollution. Similarly, to achieve full incorporation of hydrogen in the global

152 energy supply, concerted efforts are required to ensure suitable and sustainable storage of

153 hydrogen to overcome the intermittency and fluctuation witnessed during its production [24–

154 26].

Summarily, hydrogen is courted as a sustainable fuel to reduce global carbon footprint due to its efficiency and environmentally benign nature. Recent advances have witnessed significant contributions to the hydrogen economy. The objective of this review is to holistically examine and provide updated information on hydrogen production, transportation, utilization, and storage. Through an extensive analysis of the current state-of-the-art technological advancements, this review aims to provide insights into the different technologies involved in each stage of the hydrogen value chain. Ultimately, the goal is to contribute to the advancement in understanding sustainable hydrogen systems and facilitate the transition to a carbon-neutral energy future.

2. Hydrogen production for a sustainable economy

Hydrogen production has in recent times been considered as the most important and promising route towards meeting and maintaining a global and sustainable energy requirement. This is because hydrogen is the greenest form of energy devoid of any carbon footprint [27]. According to market projections, hydrogen production has significantly increased in the last few years, and an expected growth rate of $5 - 10\%$ is forecasted by 2050 to meet the global demand, especially in the steel and ammonia industries [28]. To meet this requirement for global hydrogen production, several technologies have over time been discovered utilizing both renewable and non-renewable sources with the most prominent ones captured in **Fig. 3**. Thus, hydrogen productions via these methods are critically discussed in this section.
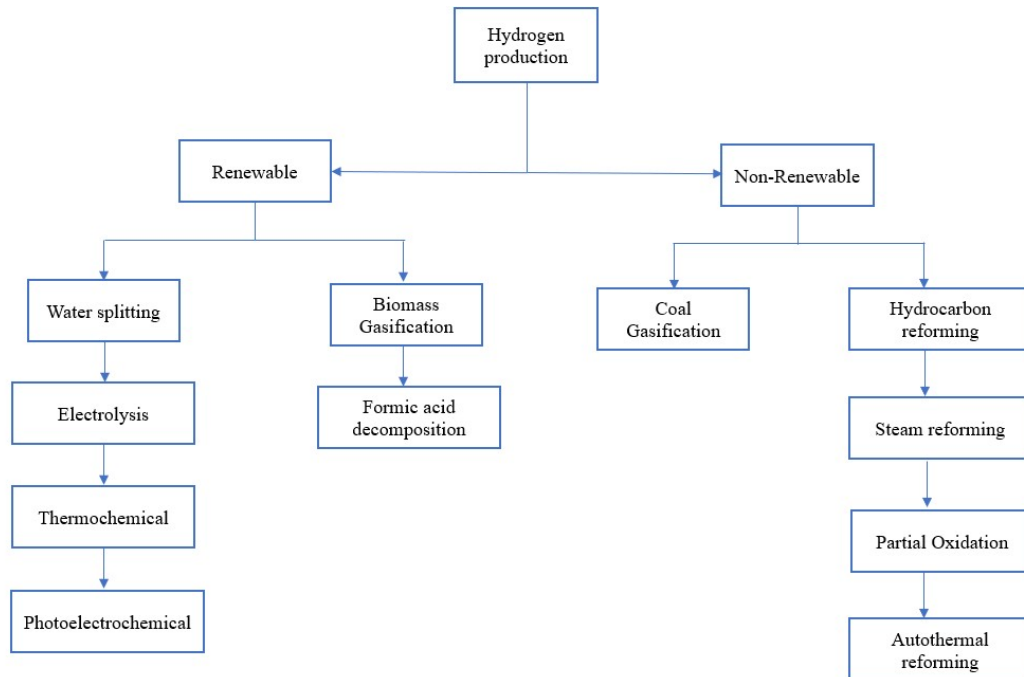

Fig. 3. Schematics for various routes for renewable and non-renewable hydrogen production.

2.1 Water Splitting

Hydrogen production via water splitting is the most sustainable technology, utilizing the most abundant material on earth (water) under the influence of a driving force such as direct electric current, temperature, light, or a hybrid that combines any of the listed [29,30]. The main setback to this process is that it is a highly energy-intensive process, resulting in a minimum energy requirement of 45 kWh/kg of hydrogen sufficiently produced [28]. Water splitting can be achieved mainly by electrolytic, thermochemical, photochemical, or hybrid processes [31,32].

2.1.1 Electrolytic Water Splitting

In this process, water decomposes into hydrogen and oxygen by the passage of direct electric current in an electrolyzer. This process is the most reliable, effective, and established for water splitting [33]. This technology simply converts electricity to chemical energy which is hydrogen and oxygen (an important side product of the process). There are numerous electrolytic water-splitting design technologies classified based on the type of electrolyte. The prominent ones are namely the solid-oxide electrolysis cell (SOEC), proton exchange membrane (PEM) electrolysis cell, and alkaline electrolysis cell (AEC) [30]. The most efficient technology electrically is the SOEC. However, it is the least developed due to the challenges of corrosion, chrome migration, and thermal cycling. The efficiency of the PEM electrolyzer is higher than the AEC but at a higher capital cost [30]. In the PEM electrolyzer, the anode is placed in the water where water is split into hydrogen ions (H^+). The H^+ is then transported to the cathode via the membrane where combination takes place to form H_2.

Electrolytic water splitting occurs in electrolytes that provide the required ionic conductivity between installed electrodes and also within the confines of their porous frameworks. The electrolytes are either acidic or alkaline, and the choice of electrolyte determines the route for convective water transport for H_2 and O_2 evolution [34]. In an alkaline electrolyte, hydroxide ions permeate from the cathode to the anode during the process of electrolysis, while in an acidic electrolyte, the protons permeate but in the opposite direction [34,35].

Hydrogen evolution reaction (HER) is the major half-cell reaction in the electrolysis and it takes place at the cathode to generate H_2 gas via dual electron transfer for proton reduction to yield H_2. The HER is best carried out in acidic or alkaline environments based on mechanisms shown in **Fig. 4**. **Fig. 4(a)** depicts how the total HER reaction ($2H + 2e- \rightarrow H_2$) is obtained from different mechanistic steps which are the Volmer reaction ($H^+ + e- \rightarrow H^*$); Heyrovsky reaction ($H^* + H^+ + e- \rightarrow H_2$) and Tafel reaction ($H^* + H^* \rightarrow H_2$) in an acidic solution. For the alkaline

208   solution; **Fig. 4(b)**, the total HER reaction is obtained from water molecules dissociation since

209   an alkaline electrolyte does not contain H^+. This route also involves the Volmer reaction (H_2O

210   + e- → H* + OH-); Heyrovsky reaction (H* + H_2O + e- → H_2 + OH^-) and Tafel reaction (H* +

211   H* → H_2) in alkaline solution [35,36].

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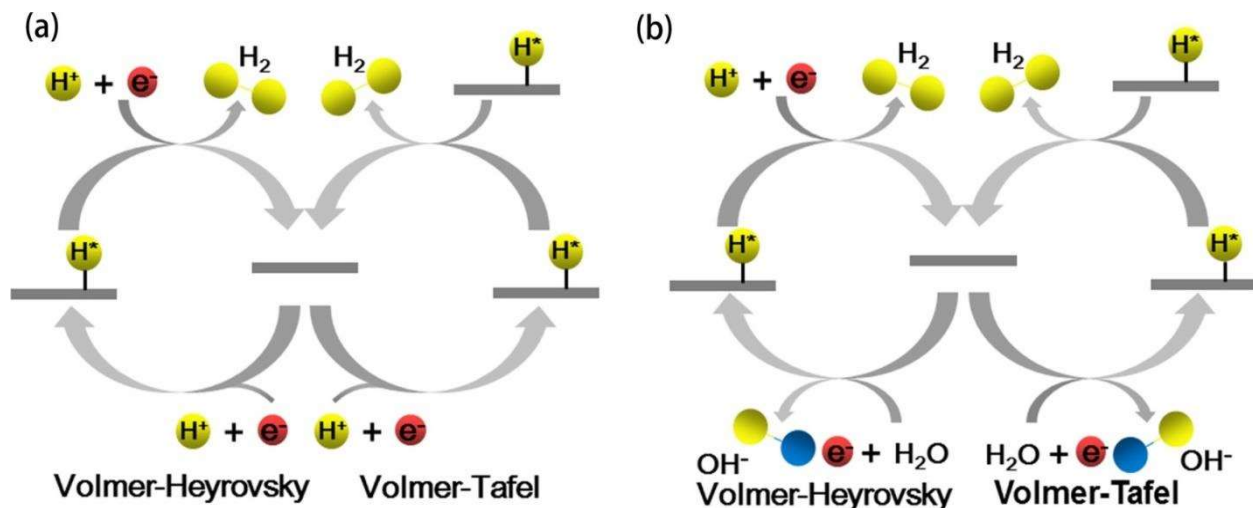
213   **Fig. 4.** Schematic representation of the HER reaction mechanism (a) in acid solution and (b) in

214   alkaline solution (Reproduced with permission from [35]).

215   The introduction of electrocatalysts is crucial for water splitting to curtail high energy demand

216   due to the high overpotentials of the system. Heterogeneous transition metal catalysts have

217   been tested for water splitting with high performance in alkaline solutions. However, many of

218   such catalysts are highly unstable in acidic or oxidative solutions. Thus, the need for

219   modification or further search for novel ones with the required robustness [37]. The application

220   of heterojunctions such as nitrogen-doped Fe_2O_3/$NiTe_2$ [38]; $FeNi_2S_4$/CoFe nanocomposite

221   [39]; core-shell CoS2@N-doped carbon nanoneedle [40]; Pt/Porphyrin MOF nanoparticles

222   [41] have been found to improve the robustness for hydrogen production.

223   **2.1.2   Thermochemical Water Splitting**

224   Thermochemical water splitting (TWS) is a promising and attractive technology for hydrogen

225   production via the splitting of water to form H_2 and O_2. TWS involves repeated two-step

226   thermochemical reactions involving a series of exothermic and endothermic reactions whereby

227   a metallic oxide is either oxidized or reduced, resulting in the thermal dissociation of water into

228   H_2 and O_2 [42]. On reduction, the metal oxide reacts with steam and eventually reoxidizes back

229   taking oxygen away from steam and releasing H_2 gas in the process [43]. Temperature

230   requirements for one-step direct water splitting occur at elevated temperatures higher than 2000

231   °C, requiring a high-powered heat source and high temperature-resistant material. Thus, TWS

232 via two-step or more has been the focus of research over the years [44]. The more the number
233 of steps, the lesser will be the maximum temperature for TWS. However, more steps increase
234 the complexity of the process. The TWS technology is suitable for large-scale industrial H_2
235 production due to its substantial economies of scale [45,46].

236 In theory, thermal energy is the only driving force for this technology. When driven by thermal
237 and other forms of energy such as light or electricity, it is termed a hybrid thermochemical
238 process [47]. TWS process is poorly dependent on catalyst application and as such water is the
239 only substance consumed to produce H_2 while other substances are recycled.

240 The thermal and physio-chemical properties of the redox material strongly influence the
241 performance and overall efficiency of thermochemical cycles. Redox materials are classified
242 as volatile and non-volatile depending on their physical state in the entire process [48]. Ceria-
243 and perovskite-based oxides are among the most promising for thermochemical cycles based
244 on thermal stability, oxygen storage capacity, sinter resistance, and excellent redox potentials
245 [49].

2.1.3   Hydrolysis of Metallic Hydrides

247 Hydrogen production via hydrolysis of metallic hydrides is attracting tremendous attention in
248 recent times as an alternative to existing conventional methods such as fossil-fuel reforming,
249 biomass gasification, water splitting, and biological H_2 production [50–53]. This is due to the
250 low cost of catalyst and energy consumption, high theoretical H_2 yield, and highly simplified
251 operating conditions at elevated gravimetric H_2 capacity of around 10 % w/w [54]. The process
252 involves the reaction of water in the form of vapor or liquid with solid metallic hydrides to
253 generate pure hydrogen. The setbacks to this method of solid-liquid reaction are that the
254 products of the insoluble hydrolysis are highly alkaline with high pH values, which thus inhibit
255 the reaction process [55]. To achieve the required reaction completion, an excess amount of
256 acid is added to the reactant mixture. This is however detrimental to the reactor and other
257 associated equipment. Furthermore, the solid-liquid reaction is inherently associated with low
258 efficiency on a weight basis due to the usage of excessive water-acid mixture requirement for
259 acceptable H_2 yield.

260 Hydrogen production from heavy metal-based hydrides such as $TiFeH_2$, $TiCr_2H_3$, $LaNi_5H_{6.7}$,
261 Mg_2NiH_4, and $NaAlH_4$ which are reversible H_2 storage materials, have been well investigated
262 over the past decade. However, their associated high H-storage capacities are affected by severe
263 H_2 supply operating conditions [56]. Ouyang et al. [57] reported that a theoretical H-capacity

10

264   of 7.6 wt.% could be attained for Mg_2NiH_4 only when heated to temperatures above 300 ℃.

265   Light metallic hydrides such as CaH_2, $NaBH_4$, MgH_2, LiH, $LiBH_4$ are irreversible H-storage

266   materials with higher hydrogen supply efficiency via hydrolysis or alcoholysis [58]. H_2 supply

267   efficiency of light metallic hydrides is however usually blocked due to the development of

268   surface passivation layers, thereby causing slow reaction kinetics and low yield. Nevertheless,

269   it is the most applicable industrially as a result of its low cost. Among numerous sources of

270   hydrolysis H_2 production, sodium borohydride ($NaBH_4$) is widely used as a result of its

271   controllability and safety [59]. $NaBH_4$ and other potential boron hydrides (such as KBH_4 and

272   NH_3BH_3) hydrolysis are accompanied by the presence of biopolymer-based catalysts according

273   to the following equations [60]:

274   $$NaBH_4 + 2H_2O \rightarrow NaBO_2 + 4H_2 \tag{1}$$

275   $$NH_3BH_3 + 2H_2O \rightarrow NH_4 + BO_2^- + 3H_2 \tag{2}$$

276   $$KBH_4 + 2H_2O \rightarrow KBO_2 + 4H_2 \tag{3}$$
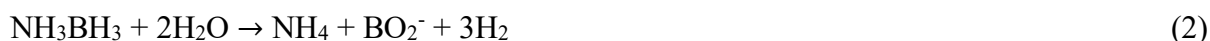
277   The mechanism of those reactions occurs as follows: BH_4^- from $NaBH_4$ and KBH_4 are

278   separated into BH_3^- and H+ active sites. There is then a transfer of electrons from BH_3^- to the

279   active sites. BH_3^- and OH- combine to form $BH_3(OH)^-$. $BH_3(OH)^-$ radicals then released 3H to

280   produce $B(OH)_4^-$. Another H atom is produced from water molecules by electron transfer to

281   active sites. This pathway is similarly followed in the hydrolysis of NH_3BH_3 [60,61]. A

282   summary of hydrides that have been recently tested in recent times for hydrogen production is

283   presented in **Table 2**.

284   **Table 2.** Summary of contemporary hydride precursors for H_2 production by hydrolysis.

Materials	Contribution	Hydride amount (mg)	Hydrogenation conditions	HGR ($mLg^{-1}min^{-1}$)	H_2 Conversion Yield (%)	H_2 Yield ($mL\ g^{-1}$)	Refs.
H-Mg$_3$La	Role of LaH$_3$ on hydrolysis yield and kinetics of MgH$_2$.	250	25 ℃, 21 mins	43.8	88.31	918.4	[62]
H-La$_2$Mg$_{17}$				40.1	60.11	851.7	
H–Mg$_3$Ce	Synergistic catalysis between CeH$_3$ and MgH$_2$ for faster hydrolysis.	200	25 ℃, 15 mins	-	87.0	853	[63]
H-Mg$_3$CeNi$_{0.1}$	Modification of hydrolysis mechanisms by Ni addition.	200 – 300	25 ℃, 1.5 mins	276	84.0	1088	[64]

Materials	Contribution	Hydride amount (mg)	Hydrogenation conditions	HGR (mLg^{-1}min^{-1})	H$_2$ Conversion Yield (%)	H$_2$ Yield (mL g^{-1})	Refs.
H-Mg$_3$La	Role of particle size on hydrolysis rate and H$_2$ yield.	200	25 °C, 1.5 mins	276	-	863	[65]
MgH$_2$	Effects of ball milling, acid addition, and ultrasound on hydrolysis kinetics and yield.	20	40 °C, 30 mins	-	85.0	-	[66]
MgH$_2$-LiH	Ball milling for refined particle size and improved kinetics for hydrolysis.	100	380 °C, 30 mins	-	89.0	1263	[67]
MgH$_2$	Role of ball milling and additives for optimal kinetics and yield of H$_2$.	10 – 15	400°C, 2 h	-	70.0	-	[68]
MgH$_2$-LiH	Role of LiH on hydrolysis of MgH$_2$-LiH composite.	50	25 °C, 50 mins	58	72.0	1870	[69]
MgH$_2$/NaBO$_2$	Optimization of reactor structure for improved MgH$_2$ coverage and hydride hydrolysis.	100	25 °C, 50 mins	-	-	1597.20	[70]
MgH$_2$	Use of aqueous organic acids for controlled H$_2$ generation.	200	20 °C, 5 mins	-	93.59	-	[71]
H–CaMg$_2$Ti$_{0.1}$	Role of ternary alloy on hydrolysis kinetics.	200	80 °C, 20 mins	951.6	-	1055	[72]

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2.1.4 Hydrolysis of Metals/Alloys

The hydrolysis of metals involves a reaction of Group I, II, and XIII metals with pure water to produce metallic hydroxides and H$_2$ generation in the process as follows [73]:

$$M(s) + nH_2O \rightarrow M(OH)n\,(s) + n/2\,H_2\,(g) \qquad\qquad (4)$$

Where M is a potential metal and n is the oxidation state of the metal, M.

In the 2000s, heavy metal combinations such as LaNi$_5$, ZrFe$_2$, etc. have emerged as potential hydrogen-producing materials due to their excellent cycling capability and swift kinetics under

moderate and atmospheric conditions [74]. However, heavy metals have a characteristic low hydrogen sorption approximately ≤ 2 wt.% H_2. To attain the required H_2 target, the focus has shifted to novel lightweight metals. The most popular metals for hydrolysis are Mg, Al, and their respective alloys, owing to their abundance in nature, lightweight, and lower cost [75]. The alloying is carried out by mechanical ball milling at high-energy ball-powder collision kneading and re-fracturing the mixture together [76]. This provides synergy between constituting metals towards ensuring a high yield of H_2, fast hydrogen generation rate, low reaction temperature, and low activation energy [77]. Various approaches have been developed and tested for the overall performance of metal hydrolysis. Notables are hierarchical Al-Mg parent alloy [78]; Mg-Al-Fe composite for seawater hydrolysis [79]; β-phase Mg chips for hydrolysis in $MgCl_2$ solution [73].

2.2   Biomass Gasification

Biomass gasification is an established technological pathway that converts biomass or carbonaceous materials into producer gas, which is then transformed to hydrogen or syn gas on further cleaning and conditioning. This process occurs in a controlled environment under the action of heat, oxygen, and steam [80,81].

The utilization of biomass for gasification is advantageous as the most renewable feedstock because of its less dependency on climate and location, and can also be easily stored or transported. Furthermore, biomass is most widely available of which it accounts for over 10% of the global sources of energy. It is classified among the top energy sources based on the 2011 world's energy consumption [82,83]. The gasification of biomass for H_2 production is classified into two routes namely: steam and supercritical water gasification. Steam gasification converts natural biomass into H_2, CO, CO_2, and CH_4 at high temperatures necessary for the introduction of steam as a gasification agent. Supercritical water gasification involves hydrothermal gasification of biomass in supercritical water, which acts as both a reactant and a reaction medium [84].

Catalytic gasification of biomass is aimed at reducing gasification temperature; and improving yield and selectivity of H_2 and biomass conversation. The major setback of steam gasification over supercritical water gasification is the propensity for tar formation which causes blockage of pipe band fittings. Catalyst application promotes tar cracking and alkali catalyst is the most suitable for both gasification routes [85].

2.2.1 Formic Acid Decomposition

Formic acid (HCOOH) is the simplest member of the carboxylic group produced chemically from the hydrolysis of cellulosic biomass precursors and hydrolysis of methyl formate [86]. It is a very important hydrogen carrier with a potential for large-scale hydrogen production due to its high H_2 density, low toxicity, high flash point, biodegradability, and low technical difficulty [87]. A high yield of hydrogen from formic acid (FA) decomposition is attained via two distinct pathways namely: dehydration (decarbonylation) and dehydrogenation (decarboxylation) [88] as represented by the **Eqs. (5)** and **(6)**, respectively.

$$HCOOH \leftrightarrow CO + H_2O \qquad \Delta G = \text{-12.4 kJ mol}^{-1}; \Delta H = \text{29.2 kJ mol}^{-1} \qquad (5)$$

$$HCOOH \leftrightarrow NCO_2 + H_2 \qquad \Delta G = \text{-12.4 kJ mol}^{-1}; \Delta H = \text{29.2 kJ mol}^{-1} \qquad (6)$$
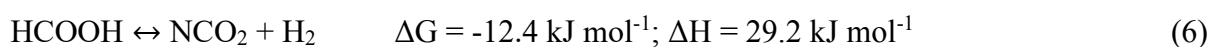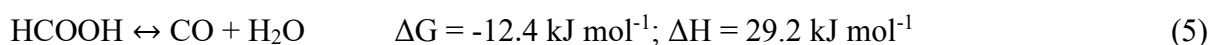
Based on their Gibbs free energy, FA dehydrogenation is more spontaneous with more likelihood of occurrence than the dehydration pathway. It is imperative to boost the selectivity of dehydrogenation reaction over the dehydration pathway to inhibit CO formation which is poisonous to catalysts in fuel cells and other applications [89]. It has been established from the literature that the pathway for FA decomposition (dehydrogenation or dehydration) depends strongly on the surface morphology and chemistry of the catalysts and reaction conditions such as temperature and concentration [90,91]. Homogeneous and heterogeneous catalysts have over the last decade been developed for FA decomposition towards H_2 production. Active metals from the transition series such as Rh, Ru, and Ir, and metallic complexes with phosphine-related compounds and nitrogen-related compounds act as ligands [87].

Homogeneous catalysts showed high performance displaying a turnover frequency (TOF) in the range of $286 - 32200$ h^{-1} at temperatures ranging from $40 - 120$ ℃. However, the catalysts exhibit poor performance in terms of stability, product separation, catalyst regeneration, and adverse environmental problems associated with heavy metal pollution [92,93]. Heterogeneous catalysis has received tremendous research consideration to address the drawbacks of homogeneous catalyst application, dating back to the 1930s. Numerous design configurations have been reported over time for heterogeneous catalysts ranging from single metal atoms [94]; to supported metals [90] and metal complexes [89].

Supported noble metal catalysts with active sites such as Pd, Pt, Rh, Ru, Au, Ag, and Ir have been tested and showed great potential for FA decomposition [91,95]. Among these noble metals, Pd has shown superiority over others in terms of catalytic activity, stability, and H_2

selectivity, especially at low or room temperatures [96,97]. Several support systems have been developed to improve the properties and performance of active sites of noble metals. Carbonaceous materials are the ideal support structure for FA dehydrogenation catalysts. This is a result of their intrinsic properties providing anchoring sites for active metals and their surfaces are easily modified to accommodate basic functional groups needed to boost FA dehydrogenation [88]. Typical examples are namely: Pd supported on activated carbon [91,95]; Pd on amine-functionalized carbon nanotubes [98]; Pd on N-doped graphene [99].

Research outputs on heterogeneous catalytic dehydrogenation of FA have over time shown great promise especially at room temperature for H_2 production. The quest for robust and efficient catalysts suitable for this process has mainly been carried out using trial and error for a combination of active sites, supports, promoters, and other additives. Contemporary cutting-edge research has made concerted efforts towards understanding the complex dehydrogenation mechanism. This will aid the rational design of an efficient catalyst system suitable. Qin et al. [100] synthesized facile Pd nanoparticles supported on carbon black (Pd@Bi/C) with a TOF of $4350h^{-1}$ at 30 ℃. According to results obtained from kinetic studies, in situ FTIR, and DFT calculations, Bi adatoms are responsible for the adsorption of formate intermediate which facilitates the cleavage of C – H bond. This weakens CO and H adsorption on active Pd sites thus, enhancing the selectivity for H_2 production. According to DFT calculations, pyridinic nitrogen-stabilized single-atom catalyst configurations produce more catalytic activity than dispersed nanoparticles [94]. Furthermore, the effect of nitrogen doping on Pd-supported graphene for FA dehydrogenation was studied with the aid of DFT computations [99]. DFT results showed that H_2 generation from FA dehydrogenation occurs via Path I i.e. HCOO (formate) pathway rather than Path II, the COOH (carboxylate) pathway as depicted in **Fig. 5**. The undesired pathway involving HCOOH dehydration to produce CO through COOH (Path IIIA) or formyl (HCO) (Path IIIB) intermediate species is not likely to occur on the Pd-N_3Gr catalyst due to its large activation energy. It was also discovered that active sites on the Pd-N_3Gr surface increased with N-doping [99]. Cai et al. [96] conducted a similar DFT simulation using a Pt-based catalyst.

Fig. 5. Formic acid decomposition pathway over Pd-N$_3$Gr catalyst surface using DFT computation (Reproduced with permission from [99]).

2.3   Hydrocarbon Reforming

Hydrocarbon reforming is a thermal process for H$_2$ production from hydrocarbons. The feedstock for hydrocarbon reforming is predominantly methane in its pure form or natural gas and in some cases other light hydrocarbons such as ethane, ethanol, propane, butane, and their mixtures [19]. Reforming hydrocarbons is an energy-intensive process and is triggered by the presence of components such as carbon dioxide, steam, and oxygen which act as oxidizing agents [101]. Reforming processes that occur in the presence of steam, CO$_2$, and oxygen are termed steam reforming, autothermal reforming, and partial oxidation respectively. Methane is the most widely used hydrocarbon for H$_2$ production simply for the fact that it contains around 25% hydrogen on a weight basis which is the highest for any hydrocarbon.

2.3.1   Steam Reforming

Steam reforming of methane (SRM) is an endothermic catalytic reaction between methane and steam to produce hydrogen-rich synthetic gas (syngas) at elevated temperatures and pressures

401 [102,103]. The SRM reaction - **Eq. (7)** - takes place efficiently at temperatures (750 – 950 ℃)

402 and pressures (14 – 20 bar) [104]. The reaction occurs alongside the water-gas-shift (WGS)

403 reaction, **Eq. (8)**, an exothermic reaction that improves the overall hydrogen yield.

404 $CH_4 (g) + H_2O (g) \leftrightarrow CO (g) + 3H_2 (g)\ \Delta H^o_{298\ k} = 206\ kJ\ mol^{-1}$ (7)

405 $CO (g) + H_2O (g) \leftrightarrow CO_2 (g) + H_2 (g)\ \Delta H^o_{298\ k} = -41\ kJ\ mol^{-1}$ (8)
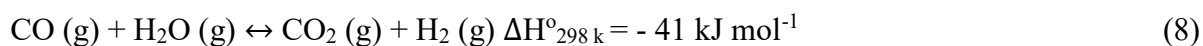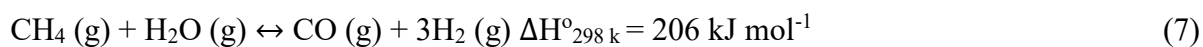
406 SRM is the most mature and efficient technology for H_2 and syngas production. However, the

407 major challenges associated with it are still unresolved [19]. The first challenge is the exorbitant

408 energy cost to meet the highly endothermic SRM reaction conditions and the capital cost of

409 heat transfer and heat-resistant equipment. The second challenge is the short catalyst lifespan

410 attributed to coke deposition, sulfur poisoning, and active site sintering of catalysts during the

411 reaction process [105]. According to a comparative analysis conducted on catalyst performance

412 as reported from the works of [106] and [105], active metals are rated based on their catalytic

413 activities as Fe < Co < Pt = Pd = Ir < Ni < Ru < Rh. Rating based on turnover frequency

414 (TOF) of active sites on silica support is Rh, Ir < Ru < Ni, while Co < Ru = Pt < Ir < Ni <

415 Rh for alumina-supported catalysts.

416 In recent times, concerted efforts have been deployed towards improving the yield of H_2 and

417 overall SRM efficiency. These include advanced technologies such as chemical looping SRM

418 [107]; solid oxide fuel cell (SOFC) SRM [108,109]; photocatalytic SRM [110]; low-

419 temperature SRM [111]; electrolytic SRM [112]; and plasma-assisted SRM [113,114].

420 According to [115], H_2 production from SRM in comparison to steam reforming of other

421 feedstocks such as biogas, methanol, ethanol, glycerin, and biomass has the least installed cost.

422 This is because of the lowest presence of unreacted hydrocarbons in the product gas stream

423 over other methods.

424 SRM is still the most mature and widely used technology for H_2 production. However, stability

425 issues with regard to the catalyst and harsh reaction conditions are still the main bottlenecks

426 affecting its profitability and sustainability [19].

427 **2.3.2 Partial Oxidation**

428 Partial oxidation of methane (POM) is an alternative hydrogen production method in an

429 exothermic process whereby methane or natural gas is converted to CO and H_2 by partial

430 oxidation as shown in **Eq. (9)**. H_2 or syngas is produced by this method at a reduced energy

cost due to its inherent exothermicity and fast kinetics [116]. The reaction produced product syngas at an H_2/CO ratio of 2 which is suitable for H_2 production and ideal for Fischer-Tropsch and methanol synthesis [117,118].

$$CH_4 (g) + 1/2\ O_2 (g) \leftrightarrow CO (g) + 2H_2 (g)\ \Delta H^o_{298} = -35.6\ kJ\ mol^{-1} \qquad (9)$$
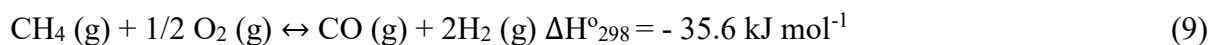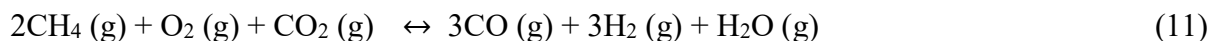
Partial oxidation of methane to CO and H_2 occurs at temperatures in the range of 700 – 900 ºC and atmospheric pressure to attain complete conversion of reactants and good yield of products [119]. Competitive side reactions such as CO disproportionation (Boudouard reaction) and water–gas–shift reaction [**(Eq. (10)** and **Eq. (8)**] occur alongside the POM reaction. These side reactions are both exothermic and proceed at temperatures lower than that of POM. However, they are reversible reactions and can thus be reversed by elevating the temperature in the reactor [120,121].

$$CO (g) \leftrightarrow C (s) + CO_2 (g)\ \Delta H^o_{298} = -173.0\ kJ\ mol^{-1} \qquad (10)$$
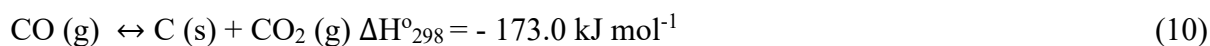
2.3.3 Autothermal Reforming

Autothermal reforming of methane (ARM) is a syngas or H_2 production technology that utilizes the combination of oxygen and steam or CO_2 as oxidizing agents with methane as the feed. The ARM reactions occur in a single reactor chamber such that a syngas H_2/CO stoichiometric ratio of 1:1 is produced when carbon dioxide is used [**(Eq. (11)**]. ARM using steam generates syngas H_2/CO ratio of 2.5:1 as shown in **Eq. (12)**. This reaction technology is most suitable for the production of syngas with variable H_2/CO ratio for generation of cheaper H_2 and certain biofuels such as dimethyl ether (DME) which comprises of H_2/CO ratio of 1:1.

$$2CH_4 (g) + O_2 (g) + CO_2 (g) \leftrightarrow 3CO (g) + 3H_2 (g) + H_2O (g) \qquad (11)$$

$$4CH_4 (g) + O_2 (g) + 2H_2O (g) \leftrightarrow 4CO (g) + 10H_2 (g) \qquad (12)$$

ARM is a good alternative to SRM in the sense that it minimizes the enormous energy demand of highly endothermic SRM via combination with exothermic oxidation reaction [122]. In the quest for a higher production rate of H_2, the ARM can be driven to a state of autothermal limitation by increasing the steam-to-oxygen ratio [123]. Under this circumstance, the exothermicity of methane oxidation alone is insufficient to supply the energy requirement for the optimum yield of syngas. Methane combustion in the reactor augments the system energy by supplying its heat of reaction. However, the occurrence of CH_4 combustion affects the

syngas selectivity. Liu et al. [123] recommended an efficient heat integration of the process via reverse flow reactor configuration as a solution to avoid autothermal limitation at high steam: O_2 feed ratio. Based on the research conducted by Argonne National Laboratory (ANL), it was suggested that ARM should be of simple design and fast response. However, the ARM plant has a higher possibility for explosion, thus affecting its global acceptability as a route for H_2 production [124]. Nevertheless, ARM reactors have been shown to possess very short start-up times, basically less than 5s, and a broad range of flow. Among its notable advantages are improved efficiency, purity of H_2, fuel flexibility in terms of sulfur tolerance, and coke gasification during catalyst regeneration [125].

Various catalyst configurations have been investigated to be promising for heat mitigation and suitable for application on a commercial scale. Based on numerous catalysts surveyed, it has been discovered that Ruthenium (Ru) and Rhodium (Rh) are noble metals with the most promising active sites for ARM. Transition metals such as Ni have shown great potential to the noble metals. Alumina-based supports are excellent for SRM but require some modifications to be applied as ARM catalysts [122]. Doping of alumina with oxides such as La_2O_3, ZrO_2, CeO_2, and combinations has proven to enhance the stability of catalyst active sites and support. In an effort to reduce the cost of noble metal catalysts, Ni-based catalysts exhibit high performance with reasonable coke and sulfur resistance [126].

2.4 Coal Gasification

Coal gasification is a thermal process of producing syn gas comprising mainly of H_2, CO, and trace amounts of CO_2, CH_4, and water vapor from the natural reaction of coal with oxygen, air, or steam [127]. Gasification of coal is a popular technology for applications in power generation over most combustion technologies owing to the global availability of coal and the cleaner nature of the technology. Coal gasification performance strongly depends on the coal type or properties and the gasifier design configuration [128]. It is therefore imperative to have a thorough understanding of the inherent properties of different coal types and how they influence the gasification parameters such as gasification agents (O_2, and H_2O vapor, air), catalyst configuration, temperature, pressure, and reactor design that significantly affect the gasification process efficiency [129,130]. Converted studies have shown that the use of steam and gasification agents produces syngas with higher calorific value as a result of hydrogen syngas being produced [131].

A good number of research contributions have over time been presented on the production of hydrogen-rich syngas from coal gasification in an effort towards improving the gasification performance for higher H_2 yield. Among these the supercritical water coal gasification [132]; co-solvent gasification of coal biomass blend [133]; two-step supercritical de-polymerization and supercritical water gasification of low-quality coal [134]; chemical looping coal gasification [135] and underground coal gasification [129,136]. Most contemporary literature reported success in the production of H_2-rich syngas from coal gasification for energy and power generation. However, the production of pure H_2 gas has been rarely talked about. It is, therefore, necessary to conduct research on coal gasification with an emphasis on gas separation equipment designated to provide information on H_2 purity.

2.5 Concluding remarks

In summary, hydrogen production technologies encompass a wide array of approaches designed to generate hydrogen gas, a versatile element crucial for diverse industrial applications and an emerging clean energy source. These technologies span between renewables (such as water splitting and biomass) and non-renewable (including hydrocarbon reforming and coal gasification). A detailed comparative analysis of these four primary hydrogen production technologies, including their respective merits, drawbacks, development stages, and future prospects, is presented in **Table 3** for reference.

Table 3. Summary on the various hydrogen production technologies.

Technology	Merits	Demerits	Development stage	Prospects
Water Splitting	• Established technology. • Zero emission of CO_2. • O_2 as by-product • Multitude of available raw materials. • Potential use of wastewater.	• Storage and transportation problems. • Low transformation efficiency. • Requires relatively more sunlight. • High capital costs for the experimental setup.	Early to developmental.	• Improved efficiency. • Cost reduction. • Large-scale adoption.
Biomass	• Consumes CO_2 • Produced O_2 as a by-product. • Operates under mild conditions.	• Low yields of H_2. • Sunlight needed. • Large reactor required. • O_2 sensitivity. • High cost of materials.	Developmental	• Biotechnology breakthroughs. • Process design and optimization.

Hydrocarbon reforming	• Highest conversion efficiency. • Existing infrastructure. • Well established technology.	• Use of fossil fuels. • Continuous emission of CO_2.	Mature	• Integration of CCS technology. • Transition to sustainable feedstocks.
Coal gasification	• Abundant natural resources. • Cheap feedstock. • Neutral CO_2.	• Fluctuating H_2 yields because of feedstock impurities. • Seasonal availability and formation of tar.	Mature	• Advances in materials. • Improved reaction efficiency.

510

511

3 Hydrogen transportation for a sustainable economy

Hydrogen transportation refers to the movement of hydrogen from production sites to end-use locations, where it can be employed as a clean energy source. Often, natural gas concepts, in terms of transportation and storage are usually cited as a basis for hydrogen gas in the open literature [18]. For example, the current status of CH_4 (which is the major component of natural gas) with respect to transportation (and/or storage) can be achieved through various technologies such as (i) pipelines; (ii) liquified natural gas (LNG); (iii) compressed natural gas (CNG); (iv) gas to products and (v) underground facilities.

Specifically, **pipelines** are widely recognized as the most convenient and efficient method for transporting natural gas. This technology entails an intricate pipeline network designed to facilitate the transportation of gas from its source to different destinations. These pipelines vary in diameter, typically ranging from 0.152 to 1.422 meters, and are capable of operating at pressures exceeding 49.3 bar [137]. Pipeline transportation is more suitable for relatively shorter distances, typically less than 2000 km [138]. However, for long-distance transportation across oceans, pipeline transportation is not technically and economically feasible. Therefore, **LNG** is the preferred option for transporting natural gas over long distances, particularly those exceeding 2000 km, involving cross-ocean routes, and offshore natural gas production [138]. The technology of LNG involves the cooling of natural gas to a temperature of -162 °C at atmospheric pressure where a single cubic meter of LNG holds approximately 625 cubic meters of natural gas, resulting in a much higher energy density compared to natural gas [139]. This condensed state of its volume under standard conditions facilitates its transportation over long distances (even up to 4023.3 km) [140]. The case of **CNG** technology is particularly beneficial

for offshore gas reserves where pipeline infrastructure is unavailable or LNG costs are exorbitantly high. The CNG process involves compressing natural gas to high pressures (exceeding 137.9 bar) and lowering temperatures [137].

For ease and portability in terms of safety, refilling, and delivery together with the energy density requirements, CH_4 storage and transport are conducted via **gas-to-product** technology which encompasses: (1) gas-to-wire, (2) gas-to-liquids, and (3) gas-to-solids. In the gas-to-wire mode, as described by Rajnauth et al. [137], natural gas is converted into electricity at the source and transmitted to markets through cables. However, this technique is cost-intensive due to direct current transmission. In the gas-to-liquid mode, the conversion of natural gas into liquid hydrocarbons such as methanol or synthetic crude oil takes place, after which they can be transported to their respective destinations. Though this method is suitable for long-distance transportation, its efficiency falls short compared to LNG. In the gas-to-solid mode, natural gas transforms into natural gas hydrates by combining with water under specific conditions of high pressure and low temperature [141,142]. However, looking at a larger capacity and long-term efficiency, **underground storage facilities** are considered to be the most preferred option [143].

In the case of hydrogen, transportation can be achieved through (i) gaseous transport, (ii) liquid transport, (iii) hydrogen carrier materials, and (iv) blending with natural gas [144]. While each method differs, the choice of their usage is contingent upon factors such as distance, volume, infrastructure availability, and specific application requirements.

3.1 Gaseous transport

Gaseous hydrogen transportation entails distributing hydrogen gas in its gaseous form. This method requires the compression of hydrogen at elevated pressures and its conveyance in specialized containers, guaranteeing safety and adherence to regulations [145]. It is a commonly adopted approach due to its cost-effectiveness and the existence of established infrastructure. The primary methods for gas compression encompass mechanical techniques (like piston and diaphragm compressors), and centrifugal compression [146,147]. Other advanced methods include ionic liquid piston compression [148], and electrochemical compression (based on PEM electrolysis principles) [149]. In recent times, there has been a growing adoption of metal hydride hydrogen compressors owing to their notable benefits such as: enhanced safety, environmental compatibility, minimal vibration and noise, effective sealing, absence of friction, efficient hydrogen purification, and reduced maintenance expenses

[150–152]. Overall, after a successful compression, the gaseous hydrogen is typically transported via the use of pipelines and high-pressure tube trailers attached to a compressed cylinder as elaborated below.

3.1.2 Pipeline

Hydrogen transportation via pipelines is one of the primary methods used to transport hydrogen over long distances. It involves the use of dedicated pipelines designed to safely and efficiently carry hydrogen gas from production facilities to distribution points or end-users. Pipelines serve as a means to transport hydrogen by connecting compressors, city stations, and storage facilities. Compressor stations are crucial in the continuous maintenance of hydrogen flow by supplying the necessary energy within the transmission system, ensuring that the pressure and temperature requirements are met [18,144].

Pipeline transportation of hydrogen offers several advantages. For example, it allows for large-scale delivery of hydrogen, making it suitable for meeting the demands of industrial, commercial, and residential users. It provides a continuous and reliable supply of hydrogen, eliminating the need for frequent transportation and storage logistics. Furthermore, its cost-effectiveness for large-scale power plants, environmental friendliness, and the potential for long-term pipeline operation spanning several decades make these methods stand out. Additionally, pipeline infrastructure can be integrated with existing natural gas pipelines, enabling the repurposing of existing infrastructure for hydrogen transportation [153,154].

Some drawbacks are yet associated with pipeline transportation. As an example, hydrogen has different properties than natural gas and requires specific pipeline materials, coatings, and design considerations to prevent hydrogen embrittlement and leakage [155,156]. Safety measures, such as monitoring systems, leak detection technologies, and maintenance protocols, are crucial to ensure the integrity of the pipeline to prevent accidents. Furthermore, the expansion of the hydrogen pipeline infrastructure requires significant investment and planning (such as route selection, regulatory approvals, and coordination with stakeholders). As hydrogen infrastructure continues to develop, the establishment of standardized codes and regulations specific to hydrogen pipeline transportation will be important for ensuring safety and interoperability [155].

Efforts to mitigate hydrogen embrittlement and enhance the performance and durability of hydrogen pipelines are currently focused on research into new materials, coatings, and pipeline designs. Using materials highly resistant to hydrogen embrittlement, like austenitic stainless

steels, nickel-based alloys, and specific aluminum alloys, proves effective in risk reduction [157]. More so, innovations such as the development of high-entropy alloys with unique microstructures prove promising to the resistance of hydrogen embrittlement [158]. Non-metallic materials, such as composite materials or polymers, also have the advantage of eliminating the risk [159]. Surface treatments like nitriding, carburizing, and shot peening can modify material surface properties to improve resistance to hydrogen embrittlement [160]. Protective coatings, such as ceramic or metallic coatings, act as barriers to hydrogen penetration, reducing embrittlement risks in the underlying material [161]. Additionally, addressing impurities like oxygen, sulfur, or water in hydrogen gas through advanced purification technologies like pressure swing adsorption, membrane separation, or cryogenic distillation can further minimize embrittlement risks [162].

3.1.2 Compressed cylinders

Compressed gas cylinders offer a flexible and portable solution for hydrogen delivery to various locations, especially for smaller-scale applications. In this method, hydrogen gas is compressed and stored in high-pressure cylinders constructed from durable materials like steel or composites, capable of withstanding pressures typically ranging from 200 to 700 bars [18,144]. Specially designed high-pressure tube trailers are then employed to safely transport these compressed hydrogen cylinders or vessels at high pressures [163]. These trailers, often equipped with multiple cylinders or tubes on a chassis, primarily transport hydrogen by road to end-users or storage facilities. Transportation can also extend to rail or ship, contingent upon specific needs and the availability of suitable infrastructure [146]. For larger hydrogen volumes, these containers are connected to compressed hydrogen gas tube trailers enclosed within protective frames, with the weight of these containers determining the maximum transportable hydrogen quantity. Therefore, the development of lighter tank materials becomes crucial to enable the transportation of larger hydrogen quantities [18,144]. Upon reaching their destination, the tube trailers are linked to the hydrogen system, facilitating the transfer of compressed hydrogen to storage facilities or directly to end-user applications such as refueling stations or industrial processes.

In terms of advantages, compressed gas cylinders allow for easy transportation of hydrogen to locations where pipeline infrastructure may not be available or feasible. It also provides a convenient option for temporary or mobile applications, such as fueling stations for hydrogen-powered vehicles or remote power generation. On the contrary, compressed gas cylinders have a limited storage capacity, and transporting large quantities of hydrogen would require multiple

cylinders and additional logistics. The weight and size of the cylinders can also present challenges, particularly for long-distance transportation. Proper handling, must be followed to minimize the risks associated with handling high-pressure cylinders and the potential for hydrogen leakage. Furthermore, the availability and accessibility of refueling or exchange facilities for hydrogen cylinders need to be considered to ensure a reliable supply chain. Infrastructure for cylinder refilling or exchange should be established, and regulatory compliance for the transportation of compressed gases must be adhered to.

3.2 Liquid transport

Liquid hydrogen transport involves the movement of hydrogen in a liquid state, typically at extremely low temperatures (-253 °C). Liquid hydrogen provides a means to transport large volumes of hydrogen over long distances more efficiently than the gaseous method. In this method, hydrogen gas is cooled through liquefaction technologies such as the Linde-Hampson cycle, Claude cycle, Brayton cycle, magnetic refrigeration, thermoacoustic, and two-stage mixed refrigerant cycles [146,164]. While these technologies have their operational principle, complexity, cost, scalability, and benefits as detailed by Zhang et al. [146], they allow for a greater hydrogen density, making transportation of hydrogen in liquid form more efficient. From the storage tanks, the liquid hydrogen is then loaded into specialized containers or insulated cryogenic tanks designed to handle the low-temperature environment to minimize heat transfer and maintain the hydrogen in its liquid state [18,144]. These tankers are usually equipped with pressure relief systems to maintain low temperatures and pressures during transit. Typically, transportation of liquid hydrogen to end-users or storage facilities primarily occurs via road [165], although alternative modes like rail or ship may be considered based on infrastructure. Upon reaching the destination, the cryogenic tanker is connected to the hydrogen system of the end-user or storage facility, facilitating the transfer of liquid hydrogen to suitable storage tanks or processing units. For most applications, liquid hydrogen is vaporized and warmed to ambient temperature before use [166].

While cryogenic tankers remain the primary mode of liquid hydrogen transportation, ongoing efforts by researchers and industry experts aim to enhance efficiency, cost-effectiveness, and safety [167]. These endeavors include the exploration of novel materials and designs for tanker insulation, to minimize heat transfer and better maintain the low temperature of liquid hydrogen during transport. Promising approaches involve vacuum insulation panels, composed of a core material like silica aerogel or fumed silica enclosed within a gas-impermeable, vacuum-sealed

barrier [168]. Multilayer insulation, featuring multiple reflective layers separated by spacers or low-conductivity materials, is also under investigation [169]. Additionally, carbon fiber-reinforced polymers (CFRPs) are being assessed for their suitability in both insulation and structural roles within liquid hydrogen tankers [170]. The adoption of lightweight materials in cryogenic tanker construction offers potential benefits, including reduced system weight, potential cost savings, and increased payload capacity [171]. New pressure-relief systems are in development to enhance the safe transport of liquid hydrogen by more efficiently managing internal tanker pressures [172]. Beyond cryogenic tankers, the industry is exploring the use of ships equipped with advanced cryogenic tanks and insulation systems for large-scale, long-distance liquid hydrogen transportation [173].

Transporting hydrogen as a liquid offers several advantages. Liquid hydrogen boasts a higher energy density compared to its gaseous counterpart, allowing for more hydrogen to be transported efficiently in terms of volume and weight. This makes it an ideal choice for long-distance transportation, enabling the movement of larger quantities of hydrogen in a compact form [145]. Similar to gaseous hydrogen, liquid hydrogen transportation plays a vital role in advancing the energy sector and the chemical industry as it is frequently utilized as a propellant in rocket engines, requiring its transport to launch sites [174]. Liquid hydrogen offers flexibility in transportation modes, including road, rail, and ship. Moreover, liquid hydrogen minimizes hydrogen loss due to diffusion or leakage thanks to its cryogenic storage. Handling liquid hydrogen is generally simpler and safer than managing high-pressure gaseous hydrogen. Furthermore, transporting hydrogen in liquid form mitigates the risk of hydrogen embrittlement often observed in high-pressure gas transport pipelines [146]. Notwithstanding, transporting liquid hydrogen also comes with some disadvantages and challenges. For example, maintaining cryogenic temperatures demands costly specialized equipment and insulation [175]. Hydrogen liquefaction is energy-intensive, causing energy losses and higher costs [145,146]. Evaporation during transport leads to hydrogen loss, necessitating continuous refrigeration or venting. Infrastructure like cryogenic tankers and facilities may not be widely available. Although, less prone to diffusion and leakage, safety concerns arise from extreme cold and flammability [176]. Long-distance transport, while possible, may require extra considerations and infrastructure. Ensuring liquid hydrogen purity is vital due to potential contaminants affecting performance and safety. Compliance with safety and environmental regulations can be complex. Specialized equipment and energy needs increase overall transportation costs, and proper handling requires

26

695 personnel training and safety protocols to mitigate risks linked to extreme cold and
696 flammability [166].

3.3 Hydrogen carrier materials

698 Hydrogen carrier materials, also known as hydrogen storage materials or compounds, play a
699 vital role in the storage and transportation of hydrogen gas [145]. Common examples of
700 hydrogen carriers include metal hydrides, ammonia, and liquid organics. These materials
701 enable safer and more versatile hydrogen distribution solutions, contributing to the
702 advancement of hydrogen-based transportation technologies.

3.3.1   Metal hydrides

704 Metal hydrides are a class of hydrogen carrier materials used for the storage and transportation
705 of hydrogen gas. Initially, hydrogen is merged with metal hydride-forming alloys at the
706 production site, leading to an exothermic reaction [177]. This reaction results in the absorption
707 and storage of hydrogen within the metal hydride material. Subsequently, the hydrogen-
708 saturated metal hydride material is transported to its intended destination using conventional
709 transportation modes like trucks, trains, or ships. Upon arrival, the stored hydrogen is released
710 by heating the metal hydride material [178]. The now-depleted metal hydride material can be
711 conveniently transported back to the production site for re-hydrogenation, thus enabling a
712 closed-loop transportation system.

713 Light metal borohydrides such as lithium (Li), sodium (Na), calcium (Ca), and magnesium
714 (Mg) occupy extremely large volumetric and gravimetric hydrogen capacities, therefore they
715 have been widely studied as hydrogen carriers [179–182]. For example, sodium borohydride
716 ($NaBH_4$) ranks as the extensively studied metal hydride option for hydrogen storage and
717 transportation. $NaBH_4$ can release hydrogen through hydrolysis, offering excellent control,
718 high hydrogen purity, substantial gravimetric hydrogen storage capacity, and environmentally
719 friendly by-products [183,184]. However, its primary challenge revolves around its
720 regeneration. Conventionally, the hydrolysis of $NaBH_4$ is often represented by **Eq. (13)**,
721 resulting in spent fuel in the form of hydrated sodium metaborate ($NaBO_2 \cdot xH_2O$) [183,185].

722 $$NaBH_4 + (2 + x)H_2O \rightarrow NaBO_2 . xH_2O + 4H_2 \qquad (13)$$
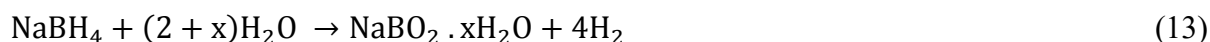
The regeneration of $NaBH_4$ from the hydrolytic product so far has featured high costs and low yields. In an aqueous solution, $NaBO_2$ reacts with CO_2 from the air to produce $Na_2B_4O_7 \cdot 10H_2O$ and $NaCO_3$ through the following chemical reaction displayed in **Eq. (14)**:

$$4NaBO_2.xH_2O + CO_2 + (10 - 4x)H_2O \rightarrow Na_2B_4O_7 . 10H_2O + Na_2CO_3 \tag{14}$$
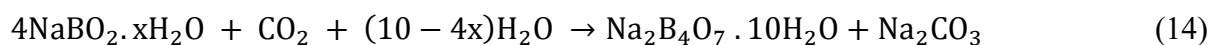
Therefore, utilizing $Na_2B_4O_7 \cdot 10H_2O$, a primary component of the naturally occurring borax mineral, makes it especially attractive to establish a straightforward, and cost-effective method for producing $NaBH_4$.

Presently, two primary raw materials, namely H_2 and metal hydride, have been utilized as hydrogen sources in the (re)generation of $NaBH_4$. For instance, the synthesis of $NaBH_4$ involves annealing $Na_2B_4O_7$ with sodium (Na) and SiO_2 under high-pressure H_2 conditions (above 3 MPa) at elevated temperatures (400–500 °C) [186]. This approach tends to be costly due to the harsh reaction conditions and a significant requirement for sodium metal. Recently, an alternative method for producing $NaBH_4$ has been explored, which involves annealing dehydrated borax ($Na_2B_4O_7$) with lower-cost magnesium (Mg) at high temperatures (550 °C) and under high H_2 pressure (2.5 MPa) [187]. However, this process remains energy-intensive and hazardous. Further optimization has also been achieved through ball-milling $Na_2B_4O_7$ [188] and $NaBO_2$ [189] with magnesium hydrides (MgH_2), resulting in maximum yields of 78% and 76%, respectively. Nonetheless, the use of expensive MgH_2 limits the practicality of mass production via these methods. Moreover, obtaining $Na_2B_4O_7$ by dehydrating $Na_2B_4O_7 \cdot 10H_2O$ requires a high-energy process at temperatures around 600 °C. Similarly, obtaining $NaBO_2$ by dehydrating $NaBO_2 \cdot xH_2O$ is formed at temperatures exceeding 350 °C [190,191].

Some other studies also noted that $NaBH_4$ hydrolytic by-products can take the form of $NaBO_2 \cdot xH_2O$ (x = 2, 4), resulting from drying a $NaBO_2$ aqueous solution at temperatures below 110 °C [190]. Based on this, researchers have also experimented with the ball milling of such by-products with Mg-based alloys (e.g., Mg_2Si, Mg-Al alloy) both with and without Mg, at room temperature under an Argon atmosphere [192,193]. Their study observed that over 74% $NaBH_4$ can be achieved. However, Mg-containing alloys were costlier than Mg itself and also led to the formation of additional by-products like stable metal oxides during the ball-milling process [192,193]. Hence, Mg alone should be used as a reducing agent to optimize this process.

754 In a more recent study, Zhu et al. [184] introduced a novel approach in which H^+ ions within
755 the coordinated water of $Na_2B_4O_7 \cdot xH_2O$ (x = 5, 10) were used as a direct hydrogen source for
756 $NaBH_4$ synthesis. Based on their approach, $Na_2B_4O_7 \cdot xH_2O$ (x = 5, 10) and Na_2CO_3 were
757 combined by exposing an aqueous $NaBO_2$ solution to CO_2 and subsequently drying it at a
758 temperature below 54 °C. Ball-milling this mixture with Mg at 25 °C under atmospheric-argon
759 pressure results in the production of $NaBH_4$ with an impressive yield of 78.9%. In contrast to
760 previous methods that rely on dehydrated $Na_2B_4O_7$/$NaBO_2$, MgH_2, high-pressure H_2, and/or
761 elevated temperatures, their novel approach was more advantageous as it utilizes cost-effective
762 materials and operates under mild reaction conditions. Thus, their one-step approach enables
763 efficient large-scale production and closes the loop for $NaBH_4$ regeneration which is
764 considered a promising cycle pathway for large-scale application of NaBH4 as a hydrogen
765 carrier [184].

766 Consequently, magnesium borohydride ($Mg(BH_4)_2$), which is synthesized through the
767 combination of magnesium and borane, followed by hydrogenation under pressure [194] is
768 another promising hydrogen carrier material. In comparison to $NaBH_4$ which is commonly
769 used in various chemical processes, $Mg(BH_4)_2$ differ based on preparation, chemical
770 composition, reactivity and stability, application, and most importantly, their hydrogen storage
771 capacities [179,182].

772 The above discussion points toward the continuous efforts by researchers to improve the
773 regeneration capacities of these hydrides for efficient hydrogen transport. Their unique
774 properties, such as high hydrogen storage capacity and controllable release, make them
775 valuable candidates in the pursuit of a sustainable hydrogen economy. While challenges remain
776 in terms of material synthesis, regeneration, and scalability, ongoing research and innovation
777 in this field continue to drive progress. As we move forward, collaboration between scientists,
778 engineers, and industry experts will be key to unlocking the full potential of light metal
779 borohydrides and ushering in a new era of clean energy.

780 ### 3.3.2 Ammonia
781 Ammonia serves as a versatile hydrogen carrier material by offering a practical solution for
782 hydrogen storage and transport [164]. Conventionally, this approach uses natural gas or coal
783 as an energy source for ammonia synthesis, releasing millions of tonnes of CO_2 into the
784 atmosphere, accounting for about 1-2% of the global CO_2 emission through the Haber-Bosch
785 process [195]. Other approaches via green technology involve the use of low-carbon renewable
786 energy sources to produce green ammonia through the Haber-Bosch process, combining

787 nitrogen and hydrogen [195]. Ammonia can efficiently store and transport hydrogen due to its

788 high hydrogen content by weight and its ability to form a stable liquid under moderate pressure

789 and temperature conditions. At the destination, ammonia is converted back into hydrogen

790 through a catalytic process, providing a reliable source of hydrogen for various applications,

791 particularly in the chemical processing industry.

792 The future viability of using ammonia as a hydrogen carrier relies heavily on advancements in

793 effective catalysts. While the catalytic synthesis of ammonia has been extensively studied over

794 the past century, ongoing research is dedicated to developing new catalysts that can lower the

795 process's operating temperature and pressure and enhance the conversion of reactants into

796 ammonia. A comprehensive review of various catalyst synthesis and enhancement efforts can

797 be found in the works of [195,196]. Their findings underscore that traditional catalysts like

798 fused-Fe and promoted-Ru, commonly used in the conventional Haber–Bosch process for

799 ammonia synthesis, have seen improvements through the emergence of innovative catalysts,

800 including electride, hydride, amide, perovskite oxide hydride/oxynitride hydride, nitride, and

801 oxide-promoted metals such as Fe, Co, and Ni. These advancements hold promise for the future

802 of ammonia-based hydrogen storage and transportation.

803 ### 3.3.3 Liquid organics

804 Liquid Organic Hydrogen Carriers (LOHCs) are chemical compounds used for the storage and

805 hydrogen transport. LOHCs can reversibly bond with hydrogen molecules through chemical

806 reactions, enabling them to store hydrogen under manageable conditions. LOHCs are

807 composed of two components: a hydrogen-lean organic compound (LOHC−) and a hydrogen-

808 rich organic compound (LOHC+) [197]. The process of hydrogen storage using LOHCs, as

809 described by Modisha et al. [197], involves converting LOHC− into LOHC+ through a

810 catalytic hydrogenation reaction to store hydrogen. Subsequently, the release of hydrogen is

811 achieved by converting LOHC+ back into LOHC− through a catalytic dehydrogenation

812 reaction [198].

813 The primary LOHC compounds under extensive study for storage and transportation

814 encompass benzene and cyclohexane [199,200], toluene and methylcyclohexane [201,202],

815 naphthalene and decalin [203,204], N-ethyl carbazole and perhydro-N-ethyl carbazole (H0-

816 NEC, H12-NEC) [205,206], as well as dibenzyl toluene and perhydro dibenzyl toluene (H0-

817 DBT, H18-DBT) [197]. In many cases, these aromatic compounds undergo dehydrogenation

818 to their corresponding aromatic counterparts in the presence of platinum catalysts at

819 temperatures ranging from 300 °C to 350 °C [207]. Supported catalysts like Pt/TiO$_2$, Pt/Al$_2$O$_3$,

820 Pt/V$_2$O$_5$, Pt/Y$_2$O$_3$, and Pt-Re/Al$_2$O$_3$ are employed for methylcyclohexane dehydrogenation

821 [205].

822 Of the mentioned LOHCs, researchers are actively investigating advanced LOHCs

823 characterized by enhanced hydrogen storage capabilities and improved thermodynamic

824 properties. Among these, NEC stands out as a heterocyclic organic compound exhibiting

825 promise as a hydrogen storage and transportation medium [208,209]. It facilitates hydrogen

826 storage through a reversible hydrogenation-dehydrogenation process, allowing for efficient

827 hydrogen release and uptake. H0-DBT, a liquid organic compound, also has been extensively

828 studied due to its high volumetric and gravimetric hydrogen storage capacity, coupled with

829 ease of dehydrogenation and hydrogenation, rendering it suitable for hydrogen storage and

830 transport [210]. Another compound of interest is H18-DBT, the fully hydrogenated form of

831 dibenzyl toluene, capable of controlled dehydrogenation to release hydrogen [211]. This

832 compound holds promise as an effective LOHC, offering substantial hydrogen storage capacity

833 and favorable thermodynamic characteristics.

834 Hydrogen carriers can facilitate the delivery of hydrogen to remote or off-grid locations where

835 direct pipeline or other transportation methods may not be feasible. The utilization of hydrogen

836 carriers addresses several challenges associated with transporting hydrogen in gaseous or liquid

837 form. For instance, many hydrogen carriers have high energy densities, which enable more

838 efficient transportation and storage compared to gaseous or liquid hydrogen [145]. They also

839 have lower flammability and explosion risks compared to gaseous or liquid hydrogen,

840 enhancing safety during transportation and storage. Certain hydrogen carriers, including

841 LOHCs and ammonia, can leverage pre-existing infrastructure like pipelines, tanker trucks, and

842 ships, thereby minimizing the necessity for extensive infrastructure upgrades [212]. However,

843 the release of hydrogen from these carriers usually entails energy input and may entail

844 conversion losses, influencing the overall energy efficiency of hydrogen as a carrier [213].

845 Hydrogen carrier systems might entail complex chemical processes and necessitate specialized

846 equipment for hydrogen storage and release [214]. As a result, the advancement and adoption

847 of hydrogen carrier technologies can be financially demanding, especially when incorporating

848 novel materials or systems [215].

849

3.4 Blending with natural gas

Hydrogen blending (a more recent method) refers to the practice of mixing hydrogen gas with another fuel, typically natural gas, to create a blended fuel before distribution. The purpose of hydrogen blending is to introduce hydrogen into existing fuel infrastructure and utilize its clean energy properties. In the context of natural gas, blending hydrogen with natural gas can be done in various proportions such as 5 - 15%, and up to higher percentages, such as 50%. The resulting blend is often referred to as "hydrogen-enriched natural gas" or "hydrogen-blended natural gas" [144,155,216]. It is worth noting that blends containing less than 5-15% hydrogen by volume typically pose minor issues, depending on specific pipeline conditions and natural gas compositions [144,217]. However, blending hydrogen in the range of 15% to 50% necessitates more substantial modifications, such as converting large household appliances or enhancing compression capacity along the distribution path for industrial users [144,218].

For example, Li et al. [219] conducted a numerical study to examine the Joule Thompson's (J-T) coefficient of natural gas under various hydrogen blending ratios. Their findings demonstrated a nearly linear decrease in the J-T coefficient of the natural gas-hydrogen mixture as the hydrogen blending ratio increased. Moreover, their research indicated that when the hydrogen blending ratio reached 30% (mole fraction), the J-T coefficient of the natural gas-hydrogen mixture decreased by approximately 40% to 50% compared to pure natural gas [219]. Zhou et al. [220] also investigated an integrated energy system combining hydrogen-blended gas and electricity, analyzing the impact of different hydrogen-blending approaches. Their findings indicated that blending hydrogen into the upper line of the natural gas network is more advantageous than the lower line. Additionally, a concentrated hydrogen blending strategy was found to be superior to a dispersed one [220]. In their research, Zhang et al. [221] developed a mathematical model for the transportation of hydrogen-blended natural gas and investigated its effects on the hydraulic and thermal properties of natural gas pipelines and networks. Their findings indicated that hydrogen blending led to a reduction in pipeline friction resistance and an increase in volume flow rates [221]. Additionally, they observed that as the hydrogen blending ratio increased, the performance of the centrifugal compressor deteriorated, and the operating point for the combined operation of the pipeline and compressor shifted towards higher volume flow rates and lower pressures [221].

Various separation and purification technologies are employed to extract hydrogen from natural gas closer to the point of use. Three methods—pressure swing adsorption, membrane

separation, and electrochemical hydrogen separation—can be applied to extract hydrogen from hydrogen-natural gas blends [144]. While blending hydrogen with a natural gas pipeline network is considered a means of effectively transporting pure hydrogen to the market, increasing the blend levels of hydrogen beyond 50% in natural gas poses numerous challenges, particularly concerning pipeline materials, safety considerations, and the need for adjustments in end-use applications. The substantial costs associated with accommodating higher hydrogen blends within a specific pipeline system must be carefully weighed against the benefits of adding hydrogen as a component in natural gas blends [222]. Notwithstanding, hydrogen blending in natural gas offers several benefits:

- Decarbonization: Hydrogen is a clean-burning fuel that produces minimal or no greenhouse gas emissions when combusted. By blending hydrogen with natural gas, the overall carbon intensity of the fuel mixture is reduced, contributing to the decarbonization of the energy sector.

- Infrastructure Compatibility: Blending hydrogen with natural gas allows for the utilization of existing natural gas infrastructure, including pipelines, storage facilities, and distribution networks. This minimizes the need for significant infrastructure modifications or the creation of separate hydrogen distribution systems.

- Flexibility: Hydrogen blending provides flexibility in adjusting the hydrogen content based on specific requirements and objectives. It allows for incremental increases in hydrogen content over time as technology advances and the availability of hydrogen increases.

- Market Development: Hydrogen blending can serve as an initial step toward the wider adoption of pure hydrogen fuels. It helps build familiarity and experience with hydrogen utilization while supporting the development of a hydrogen market.

- Increase in energy security and cost. Hydrogen blending can have both positive and negative effects on energy security and cost, depending on various factors and the specific context of implementation.

However, there are considerations and challenges associated with hydrogen blending, including the compatibility of existing infrastructure, the potential impact on appliance and combustion equipment performance, safety aspects, and the need for appropriate gas quality standards. Thus, hydrogen blending is considered a transitional approach toward a hydrogen economy, allowing for the gradual integration of hydrogen into existing energy systems while leveraging existing infrastructure and facilitating the transition to cleaner energy sources.

3.5 Concluding remarks

916 Hydrogen transportation technologies encompass the techniques and systems employed in the
917 distribution and conveyance of hydrogen, facilitating its movement from production facilities
918 to end-users or storage locations. These technologies are pivotal components within the
919 hydrogen supply chain. Key hydrogen transportation methods encompass gaseous and liquid
920 transport, hydrogen carriers, and the emerging practice of hydrogen blending. In **Table 4**, a
921 thorough comparative analysis is presented, shedding light on the primary hydrogen
922 transportation approaches. This comparison outlines their respective merits, drawbacks, current
923 developmental status, and prospects for hydrogen transport.

924 **Table 4**. Summary on the various hydrogen transportation technologies.

Technology	Merits	Demerits	Development stage	Prospects
Pipelines	• Large storage capacity. • Low cost of operation. • Efficient and reliable.	• Requires huge initial cost of investment. • Longer distance but limited to geographical reach. • Highly susceptible to leak and sabotage within the host communities.	Mature	• Expansion of pipeline networks. • Improved materials and leak detection technologies. • Improvement in "right of way" in the host community can also improves pipeline safety.
Compressed cylinders	• Simple to implement and expand. • Adaptable routing.	• Limited capacity. • Increased transportation cost. • Safety issues related to elevated pressure.	Mature	• Materials and design improvements to increase storage capacity. • Incorporation of advanced safety measures and infrastructure.
Liquid transport	• Higher energy density. • Established technology.	• Cryogenic storage. • High energy consumption.	Advanced	• Enhanced insulation materials. • Energy-efficient processes.
Carrier materials	• Ease of handling, transportation, and storage.	• Complex chemical processes. • Energy-intensive especially during release.	Developmental	• Advances in carrier materials. • Efficient release methods.
Hydrogen blending	• Reduced carbon emissions.	• Limited hydrogen content. • Infrastructure upgrades.	Developmental	• Sector Integration. • Technological advancements.

	• Utilization of existing infrastructure. • Improved combustion characteristics.	• Compression and separation.		

925

4 Hydrogen utilization for a sustainable economy

Hydrogen, the most abundant element in the universe, holds immense potential as a versatile energy carrier and has diverse applications across various sectors. As shown in **Fig. 6**, it can be used as a clean fuel, an energy storage medium, and a crucial raw material in industrial processes. In the energy sector, hydrogen can be employed in fuel cells to generate electricity, powering a wide range of applications from transportation to residential and commercial power supply. It can be blended with natural gas or used in dedicated hydrogen combustion engines to reduce emissions. Hydrogen also plays a vital role in decarbonizing heavy industries such as steel production, chemical manufacturing, and refineries. Additionally, hydrogen is utilized for fertilizer production, food processing, and as a feedstock in the production of various chemicals, including ammonia and methanol.

937
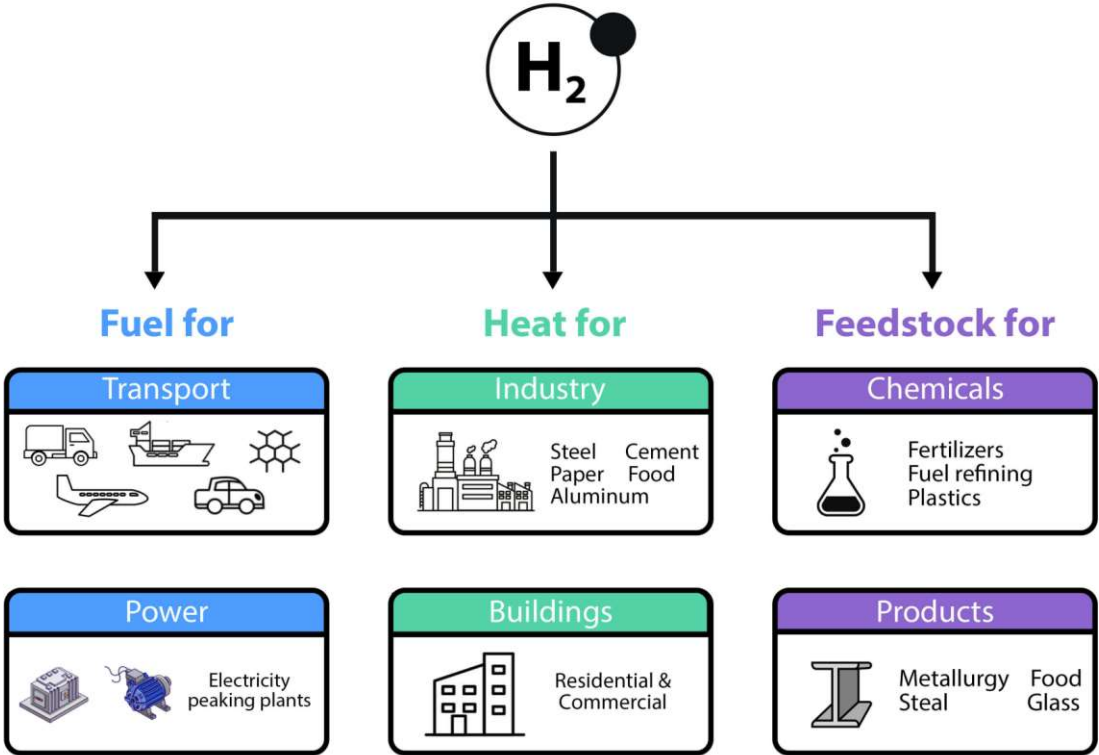
Fig. 6. Main uses of hydrogen as a clean fuel [223].

4.1 Direct fuel for heat and power

Currently, a significant amount of energy generated from conventional technologies is utilized in heating and cooling systems, and for power generation [224]. However, the use of conventional fuels (natural gas, crude oil, and coal) for energy generates a vast quantity of greenhouse gas emissions, thereby causing environmental pollution [225]. Lately, hydrogen has been courted as a sustainable, cost-effective, and efficient alternative to conventional fuels [226]. The utilization of hydrogen in boilers and heating systems was observed to generate heat and infinitesimal emissions compared to natural gas [227]. Moreover, hydrogen was found suitable for use in combustion units for generating power, and in households for cooking [228,229]. An advantage of the recent use of hydrogen in households and industries is its ease of adaptability to existing natural gas infrastructure. This is because pure hydrogen exhibits a Wobbe Index (WI) within the permissible limit of the natural gas safety range for most burners [22]. WI is a generalized metric to determine the compatibility of appliances with different fuel types. Albeit there is a need to install specialized burners in existing combustion units because of the difference in combustion velocity of hydrogen and natural gas [13]. This ensures easy control of the flame, thereby preventing incomplete combustion or overheating.

4.2 Power-to-gas technology

The use of renewable sources (e.g., wind and solar) for generating energy is faced with the challenge of interruption and periodic stopping [230]. Hence, batteries, supercapacitors, flywheels, and compressed air are used for storing produced energy. However, the aforementioned are only suited for short-term storage [231]. To overcome this limitation, hydrogen has lately been identified as an energy vector that can be used to achieve long-term energy storage via power-to-gas technology [232,233]. The newly devised technique is used for converting excess power (electricity) generated during peak periods to hydrogen gas via water electrolysis [234]. The water splitting process by electrocatalytic reaction is conducted in two half-cell reactions namely: (i) hydrogen evolution reaction (HER) taking place at the cathode, and (ii) oxygen evolution reaction (OER) at the anode [235]. The produced hydrogen is stored for subsequent use in natural gas distribution networks, methanation processes, hydrogen combustion engines, or fuel cells [236–238].
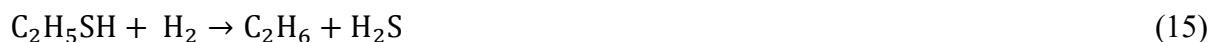
4.3 Fuel cells

Generally, fuel cells are electrochemical devices used to generate electrical energy from chemical energy through redox reactions [239,240]. The major components of a fuel cell are the cathode, anode, and electrolyte [241]. Based on the nature of the electrolyte, the common types of fuel cells are proton exchange membrane fuel cells (PEMFC), solid oxide fuel cells (SOFC), phosphoric acid fuel cells (PAFC), molten carbonate fuel cells (MCFC), direct alcohol fuel cell (DAFC), and alkaline fuel cells (AFC) [242]. Fuel cells operate in similitude to conventional batteries. Nevertheless, as compared to conventional batteries that require charging and storing energy, fuel cells have no charging requirement and generate continuous electricity when the external source of fuel and air is constant [243]. Moreover, they have high energy conversion efficiency (> 60%) and high reliability when compared to the unreliable status of batteries in extreme weather conditions [238,243,244]. Hitherto, the common chemical energy source of fuel cells is derived from hydrocarbons and carbonaceous materials. However, the shortcoming of these energy carriers in fuel cells is that they generate CO_2 as a byproduct. Recently, hydrogen has been utilized as a direct energy carrier in fuel cells to generate eco-friendly energy. The use of hydrogen in fuel cells generates water and heat as byproducts of electricity which makes the energy carrier environmentally benign [245]. Moreover, the utilization of hydrogen fuel cells witnesses lower cost of maintenance and longer life since they have fewer moving parts [246].

The application of fuel cell technology includes electrical power generation and transportation sectors [23,247,248]. Several properties of hydrogen make it desirable for the transportation industry. As compared to hydrocarbons with an energy release of 42.8 MJ/kg, hydrogen has a high energy release of 119.96 MJ/kg during its use [249]. Moreover, hydrogen is characterized by low dynamic viscosity, low molecular weight, and higher specific impulse which makes it desirable in the aerospace industry [249,250]. Also, due to its durability, bunker capability, and adaptability, hydrogen is posited as a good alternative to natural gas and other fuels in the maritime industry [251]. Nonetheless, to properly harness the potential of hydrogen in the transportation industry, storage volume and low-temperature storage requirements need to be addressed [251]. Hydrogen has an extremely low density of around 0.09 kg/m^3 compared to conventional JP − 8 and gasoline with densities of 800 kg/m^3 and 750 kg/m^3, respectively [13,252]. Moreover, in the aerospace industry, the combustion of liquid hydrogen is fazed with the downside property of low ignition energy and high flame velocity [253].

4.4 Hydrocarbon fuel refining and upgrading

Crude oil (especially heavy oil) contains heteroatoms namely sulfur, nitrogen, and oxygen (SNO) [254]. Besides, they also contain significant amounts of metals such as Nickel (Ni), vanadium (V), chromium (Cr), Titanium (Ti), Manganese (Mn), gold (Au), and others [255]. These elements are undesirable in the final products and constitute various challenges ranging from corrosion and erosion of pipelines to catalyst deactivation and/or poisoning during the refining and upgrading process of crude oil [256–258]. More importantly, the metals constitute a major source of environmental pollution. To remove trace metals, hydrogen is added for chemical demetallization in the presence of a catalyst. Hydroprocessing, the chemical demetallization process, is majorly categorized into two namely hydrotreating and hydrocracking. Hydrotreating involves the use of high-pressure hydrogen to remove contaminants (SNO and metals) and to saturate olefins and aromatics to produce a clean fuel [259,260]. The process steps for hydrodesulfurization, hydrodenitrogenation, and hydrodeoxygenation are displayed in **Eqs. (15)** to **(17)**. In similitude to hydrotreating, the hydrocracking process is used to convert low-value gas oils into valuable products via high-pressure removal of heteroatoms and by improving hydrogen-carbon (H/C) ratios [261]. **Fig. 7** illustrates the scheme for the hydrocracking process.
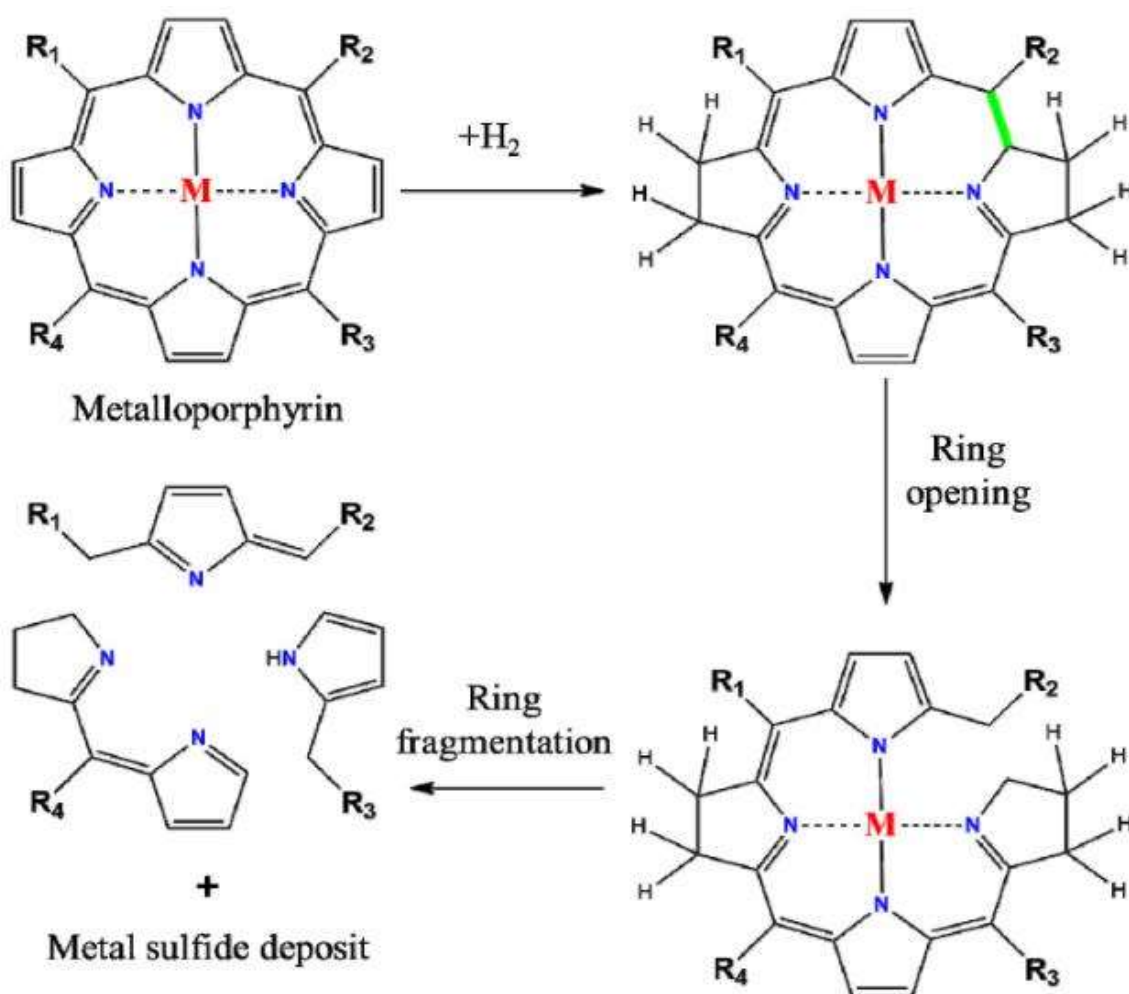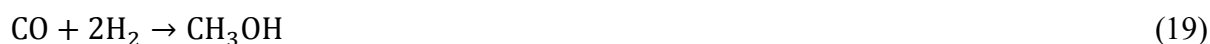
$$C_2H_5SH + H_2 \rightarrow C_2H_6 + H_2S \tag{15}$$

$$R_3N + H_2 \rightarrow 3RH + NH_3 \tag{16}$$

$$R_2O + 2H_2 \rightarrow H_2O + 2RH \tag{17}$$

Fig. 7. Schematic for hydrodemetallization [262].

Due to stringent environmental regulations for the discharge of heavy metals into the environment occasioned by issues of global warming and climate change, there is a high demand for hydrogen for the hydro-processing of crude oil, vegetable oil, and non-edible oils [13]. Moreover, hydrogen is also used for upgrading heavy oils containing high molecular weight compounds and heteroatoms characterized by high viscosity. The addition of hydrogen to the heavy oil stream aids in lowering the oil viscosity, thereby minimizing transportation and oil processing challenges [263,264].

4.5 Conversion of syngas to value-added products

Hydrogen is used to produce value-added chemicals in combination with carbon monoxide (CO) atmospheric pollutants [103,265]. Hitherto, syngas (a combination of CO and H_2) are majorly produced via reforming reactions (steam reforming and dry reforming), pyrolysis, hydrothermal, and thermochemical gasification of coal or natural gas with associated

environmental issues [266–268]. Recently, syngas can be obtained via recycling and renewable technologies. With new developments in carbon capture and sequestration (CCS) techniques, CO can be captured at the source of emission or reduction of captured CO_2 gas [269]. Moreover, hydrogen can be obtained from renewable sources such as wind, solar, and photocatalytic water splitting [270]. The pathways for the conversion of syngas to value-added chemicals are categorized into three namely Fischer-Tropsch synthesis, methanol synthesis, and methanol to hydrocarbon (MTH) synthesis reactions as shown in **Eqs. (18)** to **(20)** [271–273].

$$nCO + (2n+1)H_2 \rightarrow C_nH_{(2n+2)} + nH_2O \tag{18}$$

$$CO + 2H_2 \rightarrow CH_3OH \tag{19}$$

$$CH_3OH \rightarrow CH_2 + H_2O \tag{20}$$

As can be inferred from **Eqs. (18)** to **(20)**, a variety of products is obtained from syngas by activating the $C - O$ bonds and controlling the formation of $C - C$ bonds [271]. This includes long-chain hydrocarbon, light olefins, aromatics, and oxygen-containing compounds. It is noteworthy that the reaction steps involve the use of a catalyst(s) and occur at suitable temperature and pressure conditions [268,273,274]. The refining and product upgrading of the hydrocarbon produced from the reaction is processed into fuel and petrochemical products (see **Eq. (21)**). As compared to fuels derived from petroleum products, Fischer-Tropsch-derived hydrocarbon products are free of heteroatoms and associated impurities [275]. Moreover, they are non-corrosive with a high octane/cetane number and exhibit superior combustion properties [276].

$$-(CH_2-)_n- \xrightarrow{\text{catalyst}} \text{Fuels, lubricants, etc.} \tag{21}$$
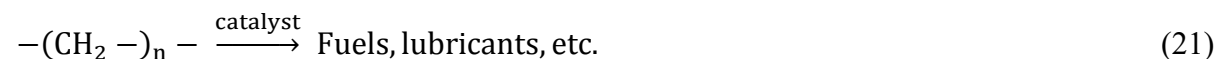
4.6 Metallurgical industry

Welding of metals in the metallurgical industry is commonly carried out using oxy-acetylene flame produced from the oxyfuel combustion process. Despite its efficiency, the use of oxy-acetylene flame generates CO and CO_2 poisoning which poses a danger to human health and the environment. Besides, its application is limited to mild and alloy steels and is less suitable

for thick stainless steels. In recent times, hydrogen has been applied in the metallurgical industry for the production of hot, clean, and stable oxy-hydrogen flames. The flame is generated via an exothermic reaction of oxygen with hydrogen at a high temperature ($\sim 3,000\ °C$). Oxy-hydrogen flames also referred to as knall-gas are used for welding and cutting non-ferrous metals, melting precious metals, and forming laboratory glass. When used for the cutting and welding process, the oxyhydrogen flame is straight, well-focused, and has a high speed of burning with low heat loss [277]. Moreover, the cut and welded spots are smooth with high precision while the byproduct of the process is water which is eco-friendly [278]. Apart from its green nature, oxy-hydrogen flames are of low cost and have proven efficient for cutting thick metals. Nonetheless, extra care is required for handling oxy-hydrogen flames because it is non-luminous under bright light [279].

Additionally, hydrogen is used for the reduction of metals from their ores during metal processing [280]. Conventional metal reduction processes (especially during iron and steel production) are associated with huge energy consumption and contribute large sources of CO_2 emissions into the atmosphere, thereby increasing the carbon footprint [281]. Contrariwise, the use of hydrogen for metal processing reduces greenhouse gas emissions [282]. The process involves the diffusion of hydrogen through the film of the metallic ore to the surface where a chemical (reduction) reaction takes place [283]. The thermodynamic and kinetic property of the reduction process depends on the temperature, pressure, hydrogen flow rate, and mineralogy of the metallic ore [283,284].

4.7 Concluding remarks

Hydrogen utilization technologies encompass a wide spectrum of applications and methodologies that leverage hydrogen as an energy carrier, fuel source, or feedstock across various sectors. These technologies hold substantial promise in mitigating greenhouse gas emissions, fortifying energy security, and facilitating the shift toward a more sustainable, low-carbon energy landscape. Prominent hydrogen utilization methods encompass direct fuel for heat and power, power to gas technology, fuel cells, hydrocarbon fuel upgrade, syngas conversion to value added products and the metallurgical industry.

Based on techno-economic analysis, the refining process is currently the biggest consumer of hydrogen. The consumption of hydrogen during the hydrotreatment process is around 320 – 350 Nm^3/ton of product. This is followed by the use of hydrogen for feedstock in the production

1094	of ammonia and other chemicals. Meanwhile, the use of hydrogen also shows great potential
1095	in fuel-cell electric vehicles with the outlook of replacing conventional internal combustion
1096	engines (ICE). The utilization of hydrogen for transport in fuel cell vehicles has the lowest
1097	consumption of hydrogen based on a recent demonstration in Paris of a Toyota Mirai fuel cell
1098	car which consumed 5.5 kg/1000 km. Nonetheless, to ensure quick integration of hydrogen for
1099	numerous utilizations, the cost of producing hydrogen needs to be lowered, since a large
1100	percentage of hydrogen is produced via the steam reforming process which is quite capital
1101	intensive. A comprehensive overview of these technologies as well as their advantages,
1102	disadvantages and prospects are presented in **Table 5**.

1103

1104 **Table 5.** Summary on the various hydrogen utilization technologies.

Technology	Merits	Demerits	Development stage	Prospects
Direct fuel for heat and power generation.	• High efficiency and versatility. • Reduced air pollution. • Grid balancing.	• High production costs. • Efficiency losses. • Materials Compatibility.	Commercially available	• Expansion of infrastructure. • Enhanced adoption.
Power to gas technology.	• Grid balancing and stability. • Energy independence by harnessing locally available renewable resources. • Integration with renewables.	• High capital costs. • Hydrogen purity may be inefficient. • Scale-up and infrastructure challenges.	Commercially available	• Efficiency improvements. • Electrolyzer design and scalability improvement.
Fuel cells.	• High efficiency • Low emissions levels. • Vast applications.	• Cost. • Sensitivity to impurities.	Commercially available	• Increased adoption • Improvement in applications.
Hydrocarbon fuel refining and upgrading.	• Clean combustion. • Flexibility of fuels.	• Lower efficiency than fuel cells	Commercially available	• Improved efficiency. • Increased adoption.
Syngas conversion to value added products.	• Reduces emissions. • Potential for carbon capture.	• Infrastructure investment.	Commercially available	• Growing demand.

Metallurgical industry.	• Clean energy source. • Reduced carbon footprint. • Improved energy efficiency.	• Infrastructure upgrades. • Hydrogen embrittlement concerns. • Hydrogen transport and storage. • Regulatory compliance.	Emerging technology	• Increased adoption and material design.
				• New applications and better integration.

1105

5   Storage systems for a sustainable economy

1106

1107   H$_2$ storage has been the major obstacle faced in recent times because of the high volatility and

1108   compressive nature of the gas. As an energy carrier, it lacks a safe, efficient, and cost-effective

1109   storage system suitable for various stationery and mobile applications [285]. The choice of its

1110   storage has been a compromise among many criteria which at the present stage of research and

1111   development of related technologies must still be identified. Some of these intriguing criteria

1112   are internally motivated (size, kinetics, charge-discharge, efficiency, safety and, life cycle

1113   environmental impacts) while some are externally motivated (transportation and distribution

1114   infrastructure, connection between the network and final users and, safe handling).

1115   The next subsection of this review will highlight in detail the different storage groups for H$_2$

1116   leveraging on the numerous storage mechanisms discussed in the literature. A systematic and

1117   comprehensive approach is adopted in grouping these storage media as either surface or

1118   subsurface [15]. As can be seen in **Fig. 8**, surface storage technologies are classified into

1119   physical or material based whereas subsurface is further classified into conventional (naturally

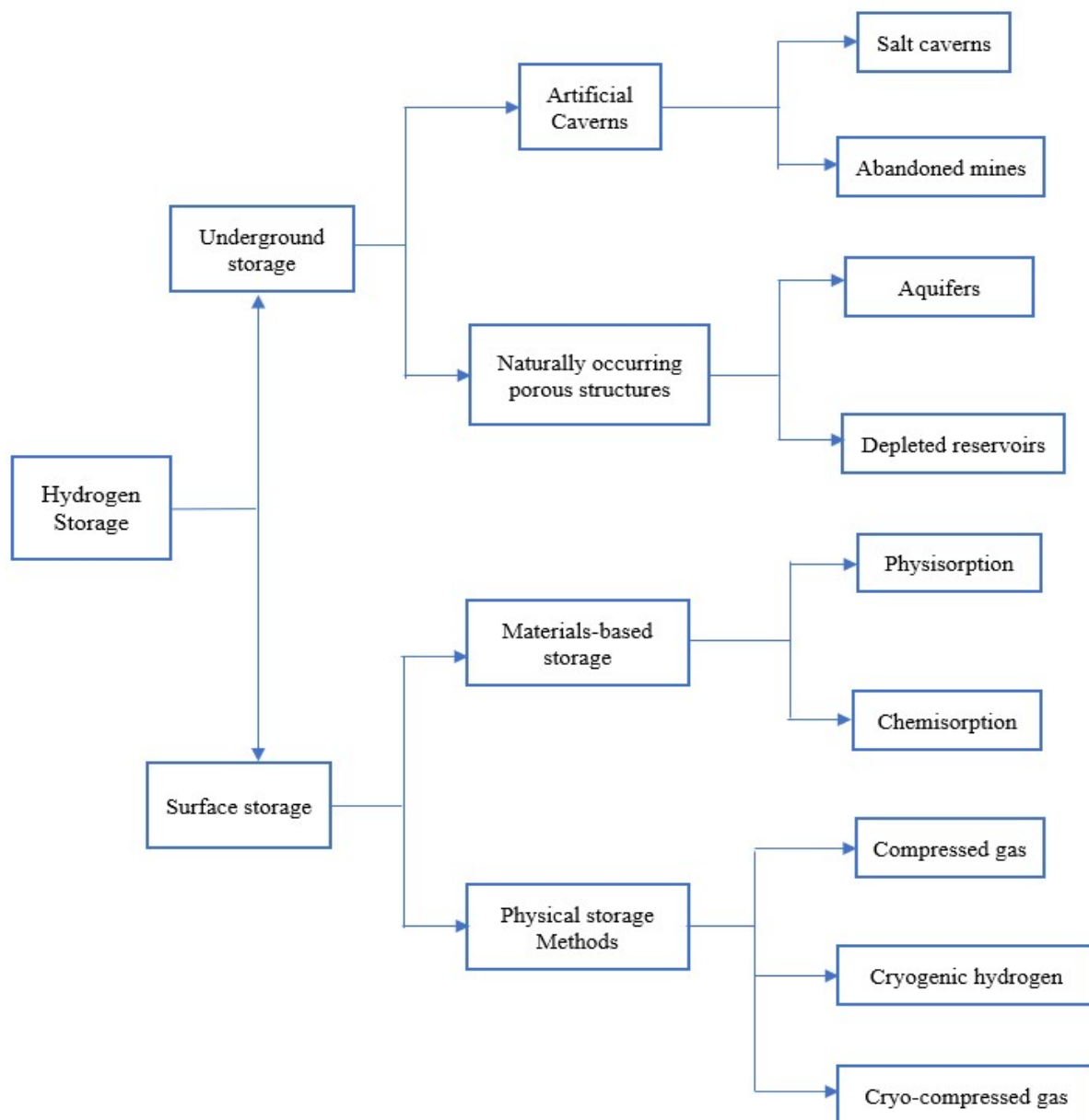1120   occurring porous media) and non-conventional (man-made caverns).

Fig. 8. Classification of hydrogen storage systems. Modified after [15].

1123 **5.1 Surface hydrogen storage**

1124 The drive towards a carbon-free economy gained momentum in 2002 when the US Department

1125 of Energy (DOE) set targets for hydrogen storage systems. These targets, established with five-

1126 year intervals starting in 2005, aimed to achieve goals related to volumetric and gravimetric

1127 capacities, refueling time, costs, cycle life, and minimizing hydrogen loss. In this context,

1128 volumetric capacity refers to the gas quantity held in a specific volume, while gravimetric

1129 capacity indicates the electricity generated by a given fuel weight. Both of these capacities

1130 should ideally be high [1]. When evaluating materials as potential hydrogen storage media,

certain critical properties should be considered to assess their storability. These criteria, among others, should be considered when evaluating H_2 storage materials: (i) high H_2 capacity per unit mass and volume to maximize energy availability, (ii) lightweight for practical use, (iii) cost-effectiveness and availability, (iv) ability to achieve high volumetric and gravimetric densities, (v) ease of activation, (vi) fast sorption kinetics, (vii) low dissociation/decomposition temperature, (viii) suitable thermodynamic properties, (ix) high reversibility, and (x) long-term recycling stability [286–288].

5.1.1 Physical method of storage

Hydrogen's lower energy density per unit of volume (9.9 MJ/m³) compared to fossil fuels necessitates the use of larger storage vessels [289]. This requires specific conditions of high pressure, low temperature, and materials with a strong affinity for hydrogen. The range of physical storage density of hydrogen under different pressure and temperature conditions is depicted in **Fig. 9** [290]. Both liquid and cryo-compressed hydrogen storage necessitates significantly lower temperatures when compared to compressed storage. Consequently, compressed storage closely resembles subsurface storage in terms of temperature requirements. In contrast, hydrogen can be securely stored as a gas under pressure ranging from 50 to 300 bar and at temperatures above 300 K. The three physical methods of hydrogen storage are further analyzed below.
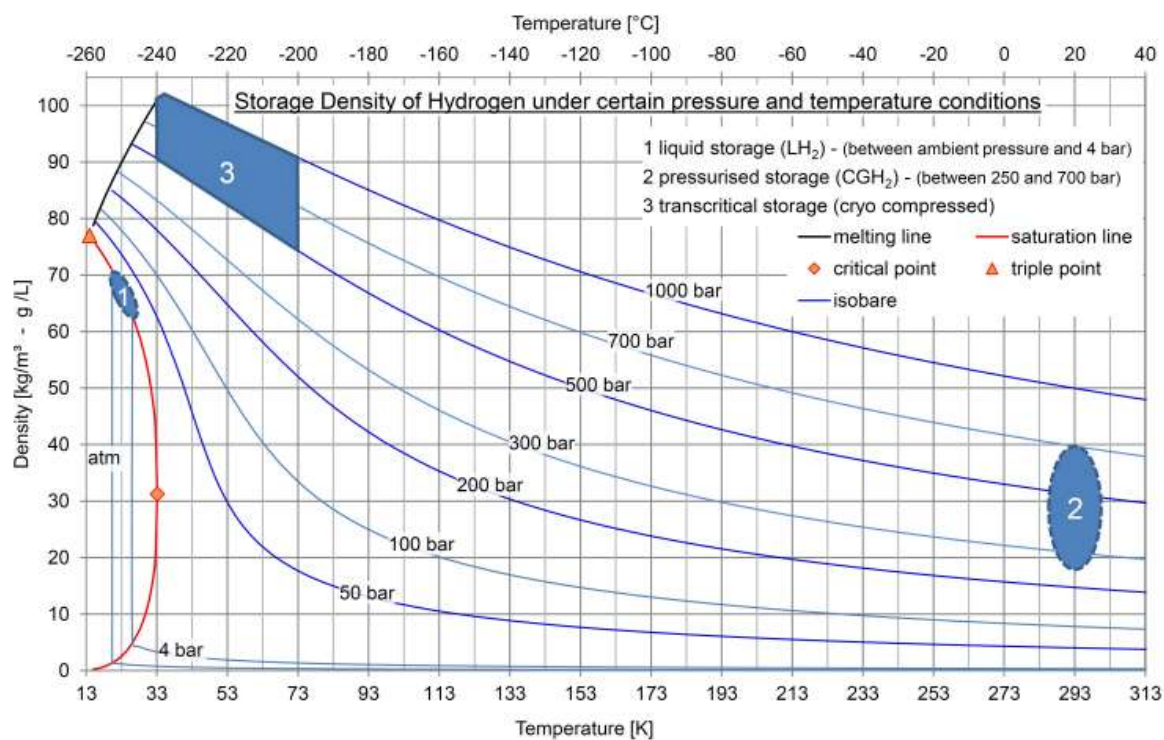
5.1.1.1 Compressed H₂ gas storage

Compressed hydrogen gas storage entails decreasing the gas volume while raising its pressure for storage within a medium. Container-based storage of hydrogen gas is the predominant and widely adopted form of physical storage [10,291]. It accounts for more than 80% of the implemented storage methods [292]. The commonly used storage vessel type is shown in **Fig. 10** with a technical description in **Table 6**. H_2 storage through compression is quite challenging owing to its large disproportionate volume for any given mass (due to its density), compression cost, and energy requirements. As an example, the compression of H_2 gas to 600 bar requires 20.48 MJ/kg of energy, whereas compressing CH_4 gas to the same pressure only requires 1.25 MJ/kg of energy [293]. Compressed H_2 has been widely adopted in the petroleum industry, including in hydrocracking and hydro-processing of crude oil, as well as in producing fertilizers, hydrogen peroxide, and metals such as nickel and iron through processes like syngas fermentation and ammonia production [13].
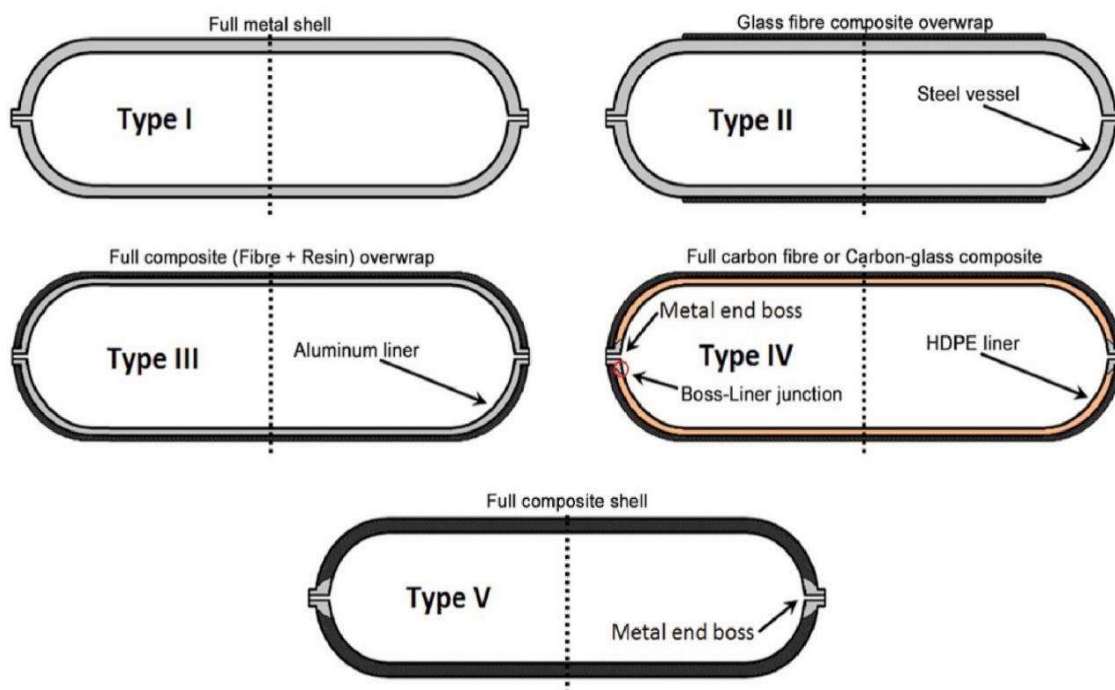

Fig. 10. Commonly used types of H_2 storage vessels [15].

The storage medium and the compressor involved in compressed H_2 storage systems are essential components since they have a substantial impact on the system's reliability, cost, and

46

safety. The efficiency of the storage operation in compressed hydrogen storage systems is influenced by the characteristics of the storage vessels. The classification of the storage vessel, whether it is low pressure (0.95 MPa) or high pressure (≥35 MPa) [294], is determined by the desired storage pressure and the materials chosen for construction. Often, this pressured vessel can be categorized as Type I to Type V [289,294,295] as summarized in **Table 6**. Type IV vessels have successfully met the gravimetric (4.2 wt.%) and volumetric (0.024 kgH_2/L) targets set by the US-DOE for light vehicle fuel requirements [1], which align with the specified gravimetric (4.5 wt.%) and volumetric (0.030 kgH_2/L) targets [163] implying that fully composite and polymer-like designed H_2 vessel is more efficient. Type IV is, however, more expensive compared to other vessel classifications. Type I is metal-based design in terms of composition, therefore making it relatively cheap but at the expense of its weight and low gravimetric density (1 wt.%). Its metallic composition makes it more susceptible to H_2 embrittlement compared to others.

Table 6. Pressure vessel properties and material types. Adapted from Amirthan and Perera [15], Ozaki et al. [294], Moradi and Groth [289], and Elberry et al. [295].

Pressure Vessel Type	Composition (-)	Pressure Limit (MPa)	Density of storage vessel (kg_steel/m³ _steel)	Gravimetric density	Relative cost
Type I (Fully metallic)	Metallic (carbon steel and low alloy steel)	50	1360	~ 1 wt%	Least expensive
Type II (Steel vessel with a glass fiber composite overwrap)	Thick metallic hooped-wrapped composite (aluminum or steel and fiber resin)	N/R	816 to 952	≤ 2.1 wt%	50% more than Type I
Type III (Full composite wrap with metal liner)	Carbon fiber composite and aluminum (liner)	45	340 to 454	≤ 4.2 wt%	Double of Type II
Type IV (Fully composite)	Polymers like HDPE (liner) and carbon fiber or carbon-glass composite	100	Very light	4.4 − 5.7 wt%	Very expensive

| Type V (linerless vessel) | Full composite (fiber-reinforced shell) | Very limited (unfit for large-scale storage) | 20% lighter than Type IV | Under development | Very expensive |

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Currently, automobile industries have leveraged the high-pressure range and reduced weight offered by Type IV vessels for transportation. Two prominent light vehicle models, the Toyota Mirai and Hyundai Nexo, stand out as notable examples of hydrogen-powered fuel cell vehicles. They have storage capacities of 5.6 kg (142.2 L) and 6.3 kg (156 L), respectively, and can drive up to 650 km and 666 km, respectively [15]. Besides the low fueling time, H_2-powered fuel cells are advantageous over batteries since they are lighter, more environmentally friendly, and can be affordable in terms of the cost of fueling systems of the vehicle [15,296]. About 270 USD/kWh is reported for lithium-ion batteries (at a battery charge/discharge rate of 1 C) [297–299] compared to the 15 to 100 USD/kWh for a compressed H_2 storage tank and fuel cell stacks, respectively [298,299].

In conclusion, the success of H_2 integration in the transportation industry will thus be dependent on how well proton-exchange membrane-powered H_2 vehicles perform in terms of overall energy efficiency (41 to 49%) when compared to electric vehicles, which have a 59%–62% range [299]. Nevertheless, the automobile industry will continue to face a significant challenge posed by the low volumetric density of hydrogen. For example, a Toyota Corolla with a fuel tank capacity of 39 liters of conventional 91 octane fuel can offer a driving range similar to that of the Toyota Mirai or Hyundai Nexo, despite their much larger hydrogen fuel tanks of 142.2 and 156 liters, respectively [15]. This underscores the primary constraint of hydrogen fuel storage when compared to conventional fuels. Consequently, the application of compressed hydrogen in heavy-duty vehicles is regarded as a more sustainable approach. Moreover, the extended driving range provided by hydrogen fuel cells, thanks to their faster kinetics compared to battery-powered systems, makes them an appropriate choice for heavy trucks and buses [300].

5.1.1.2 Liquid H_2 storage

Liquid storage (also known as cryogenic) entails the storage of H_2 in liquid form at temperatures below -253 ºC. This storage approach takes advantage of the higher gravimetric density of liquid hydrogen in comparison to compressed hydrogen. For example, with a volumetric density of 71 g/L (at 700 bar), liquid hydrogen storage surpasses the volumetric

density of gaseous hydrogen, which is 42 g/L (at 700 bar) [1]. Thus, H_2 concentration in this form is usually two times higher than in compressed gas, resulting in a higher storage density and a smaller storage space. H_2 storage via liquefaction is achieved due to its low boiling point (20 K) and can be kept in liquid condition using refrigeration, insulation, and vacuum superinsulation [1,301–303]. For instance, the Ingolstadt and Leuna H_2 liquefaction plants successfully used refrigeration to raise hydrogen's temperature to 20 K [304].

Liquefying H_2 involves significant time and energy requirements, leading to a potential energy loss of up to 35-40% during the process. In comparison, compressed H_2 storage experiences a lower energy loss of approximately 10% [1,289]. As a result, the cost of H_2 liquefaction is largely based on the size of production and the location of the plant. Equipment costs account for about 60% of the capital investment, infrastructure costs for 30%, and planning costs for 10% [303]. The aforementioned challenges, along with others, have impeded and restricted the widespread adoption of cryogenic technology.

Although liquid or cryogenic H_2 storage has advanced in terms of gravimetric and volumetric efficiency, it still faces several hurdles that need to be addressed. These include the considerable energy requirement for liquefaction, the cost of storage tanks, heat transfer concerns, and the challenge of H_2 boil-off [289,303]. According to estimates, hydrogen liquefaction costs approximately 1 USD/kg [299]. This emphasizes the fact that the aerospace and automotive industries are the primary drivers of the demand for liquid hydrogen, as they require a higher energy density without incurring additional costs [252,305,306]. In comparison to its other uses, liquid H_2 has been employed more extensively as a propellant alongside liquid O_2 and as a direct fuel in spacecraft [307]. Liquid hydrogen is regarded as a viable substitute for kerosene in aircraft applications [308]. For instance, liquid hydrogen fuel tanks are purposefully designed with a low surface area to volume ratio, in contrast to kerosene tanks that are typically located in the wings of aircraft to mitigate heat transfer. This design choice helps in minimizing heat transfer for efficient operation.

Similar to the case of compressed H_2 storage which depends on vessel types and characteristics, liquid H_2 storage tanks can be spherical or cylindrical. A spherical tank allows for the lowest surface area to volume ratio, followed by cylindrical tanks, and must be installed inside the fuselage to sustain the cryogenic temperatures [309]. The cylindrical tanks for liquid H_2 fuel are favored in aircraft due to their superior gravimetric density. Nevertheless, the lower energy density per unit volume of liquid hydrogen poses a challenge as it occupies valuable space within passenger cabins or cargo areas [310], and, despite its advantage in terms of gravimetric

density over kerosene, the empty weight of liquid hydrogen storage is heavier when compared to kerosene storage in the wings. However, due to the higher energy density of H_2 compared to kerosene, the total weight of an airplane utilizing H_2 as fuel is lower than when using kerosene [310]. Furthermore, to ensure insulation and prevent heat leakage, specialized mitigation techniques such as the implementation of double-walled vessels are utilized.

In general, cryogenic storage tanks are constructed with a double-layered structure to mitigate the risks associated with hydrogen embrittlement. This design comprises an inner pressure vessel made of austenitic stainless steel and an outer protective casing separated by a vacuum [311]. **Fig. 11** illustrates the schematic for a cryogenic storage vessel. The use of cryogenic storage vessels for hydrogen is crucial to prevent ignition-related dangers caused by oxygen condensation. The vessel design should incorporate measures to prevent overpressures, which can be achieved by utilizing an open system equipped with a relief valve. This setup allows the escape of hydrogen in a controlled manner when exposed to a warmer environment, commonly referred to as "boil-off" [312–315]. With an increase in storage capacity, the cost of cryogenic hydrogen storage decreases. For example, estimated costs for capacities of 4300 kg and 100 L are \$167/kg and \$386/kg, respectively [299]. Furthermore, a higher surface area to volume ratio can mitigate the boil-off effect [299,316], as it reduces the proportion of insulating material to stored hydrogen in larger cryogenic tanks [317]. This characteristic holds the potential for liquid H_2 to be suitable for intercontinental transportation via ships [311].
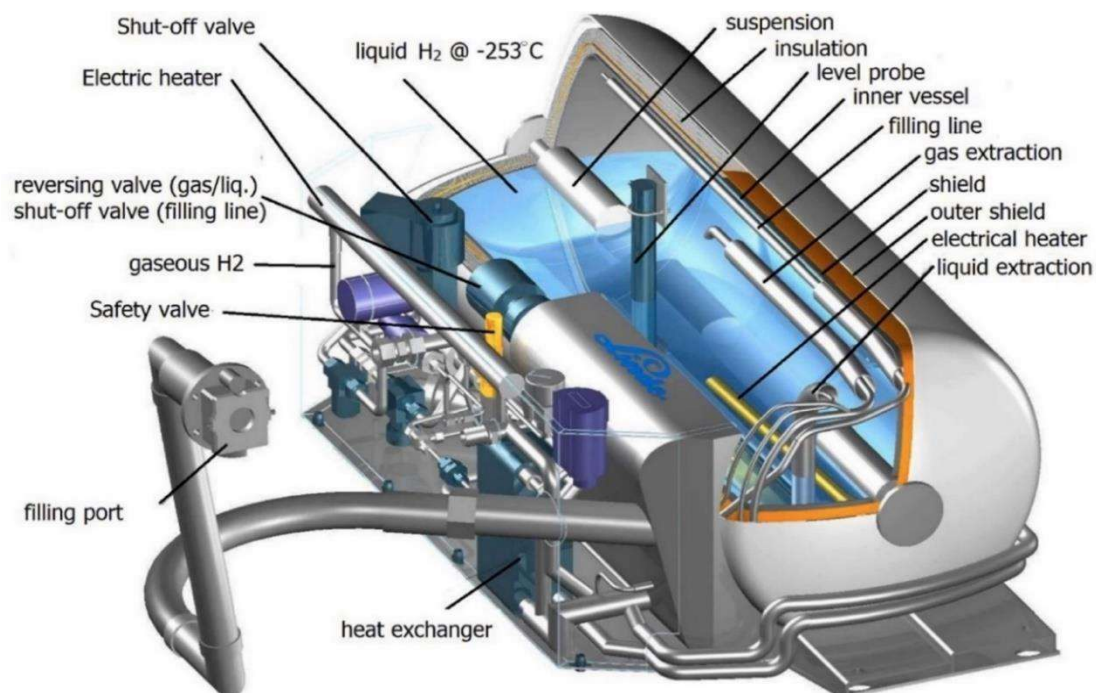

Fig. 11. Liquid (cryogenic) hydrogen storage vessel schematics [318].

1269   It is important to highlights that both liquid hydrogen storage and transportation share the

1270   fundamental requirement of maintaining hydrogen at cryogenic temperatures to keep it in its

1271   liquid state, necessitating specialized, insulated containers or tanks designed to minimize heat

1272   transfer and ensure extremely low temperatures. Safety regulations are rigorously enforced due

1273   to the cryogenic nature and reactivity of liquid hydrogen, thus, with advancements in one area

1274   can often be applicable to the other [319]. For instance, insulation materials and techniques

1275   effective in preserving low temperatures in storage tanks can also be applied to transportation

1276   systems, such as double-walled tanks, vacuum-insulated tanks, vacuum insulation panels,

1277   aerogels, cryogenic insulation foams, and multilayer insulation [176]. Advanced composite

1278   materials, like CFRPs, can serve dual roles in providing insulation and structural support for

1279   both storage tanks and transportation systems [170], offering high strength, low thermal

1280   conductivity, and compatibility with liquid hydrogen [320]. Active cooling systems, like

1281   cryogenic refrigeration cycles, can be employed in both scenarios to counteract heat transfer

1282   and maintain the desired low temperature [176].

1283   However, it should be noted that, liquid hydrogen storage and transportation serve distinct

1284   purposes within the hydrogen supply chain. Storage focuses on preserving hydrogen for future

1285   use or distribution [145], often involving stationary insulated tanks at production sites or

1286   refueling stations or distribution hubs [321]. Conversely, transportation is concerned with

1287   moving the substance from production sites to end-users or storage facilities [322] through the

1288   use of mobile containers, such as cryogenic tanker trucks, rail, cars, or ships typically over

1289   shorter timeframes. To sum up, hydrogen liquid storage and transportation require specialized

1290   equipment and share some handling requirements, yet, their primary objectives, infrastructure,

1291   and duration differs [146].

1292   *5.1.1.3 Cryo-compressed H$_2$ storage*

1293   This storage method involves compressing H$_2$ gas to its supercritical state at -233 °C. In this

1294   state, the gas remains in a non-liquefied form regardless of the compression level. Supercritical

1295   storage offers a higher storage density compared to liquefied storage, with a density of 80 g/L

1296   (approximately 10 g/L more than liquefied storage). It provides several advantages, including

1297   faster refueling, high efficiency during refueling, and enhanced safety due to the presence of a

1298   vacuum enclosure [289]. Though the cryo-compressed H$_2$ storage method holds great potential,

1299   its widespread implementation is currently impeded by the insufficient infrastructure in place.

1300   In a recent study conducted by Aceves et al. [323], it was found that an automotive application

1301   of cryo-compressed storage can utilize a Type III tank configuration. This tank design consists

of an alloy liner wrapped in carbon fiber and protected by a thin outer shell made of stainless steel or aluminum (see **Fig. 12**). To ensure effective insulation, the vacuum space between the inner and outer shells is filled with vacuum superinsulation. This concept, initially proposed by the Lawrence Livermore National Laboratory and pursued by BMW [324], was implemented in the prototype of this storage system. The prototype was installed on a hybrid H_2Toyota Prius and demonstrated impressive results, providing a driving range of approximately 1050 km over two weeks using 10.7 kg of stored H_2, with no noticeable evaporation [323].
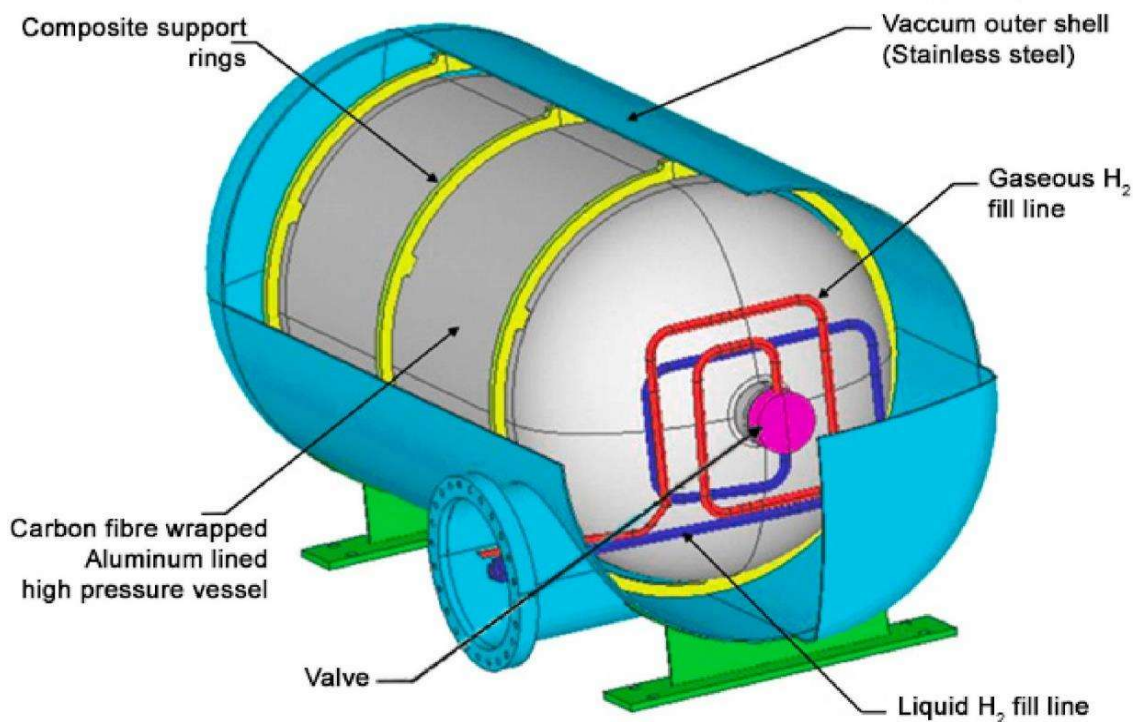

Fig. 12. Type III cryo-compressed tank comprising of a thick aluminum liner (Inner vessel) wrapped with thick carbon fiber composite rings, and an outer jacket of stainless steel. The inner and outer shells are separated by a vacuum space that is filled with vacuum superinsulation [323].

Cryogenic storage tanks are designed to withstand the internal pressure of cryogenic fluids. Unlike liquid hydrogen storage vessels, which are susceptible to the boil-off effect, cryo-compressed H_2 storage vessels are built to operate under high pressure. As a result, the issue of boiling off does not impact cryo-compressed H_2 storage [289]. Furthermore, high-pressure tanks contribute to an extended period of H_2 dormancy. This is because H_2 is released from high-pressure tanks at elevated temperatures, which further enhances its dormancy [325]. Among various physical storage methods, this particular technique boasts the lowest capital

expenses. However, the high cost of liquefaction significantly increases the overall cost, amounting to approximately \$4.8/kg for cryo-compressed H_2, while also exhibiting lower energy efficiency [326]. Moreover, this storage method is more susceptible to heat loss and power consumption compared to the other two types of physical storage [163,327].

5.1.2 Material storage methods

The storage of hydrogen in nanomaterials is an attractive and growing alternative to the use of high-pressure cylinders. Although no material holistically satisfies the requirements (**Table 7**) of the US-DOE for hydrogen storage, recent research developments are demonstrating the increasing potential to meet these targets. Nanomaterials are characterized by their building blocks, which typically consist of a small extension of 1-100 nm in one or more directions in space [328]. The number of directions that exceed 100 nm determines their dimensionality. Nanoparticles, nanodots, and clusters have a dimensionality of 0, while nanowires and nanotubes have a dimensionality of 1; thin films, either as single layers or stacked multilayers, have a dimensionality of 2 [329]. Their unique properties have made them the subject of several experimental and theoretical investigations, particularly focusing on the interaction of these materials with the hydrogen metal.

Table 7: DOE 2025 targets for onboard hydrogen storage for light-duty fuel cell vehicles [330].

Property	Value
Gravimetric hydrogen density	0.055 kg H_2/kg
Volumetric hydrogen density	0.04 kg H_2/L
Minimum delivery temperature	- 40 °C
Maximum delivery temperature	85 °C
Fill time for 5kg of hydrogen	3 – 5 min (1.65 wt.% per minute)
Durability	1500 cycles (1/4 tank to full)

The interaction of hydrogen with nanomaterials can be generally classified into physisorption and chemisorption [321]. In physisorption, molecular interactions between the adsorbate molecules (hydrogen) and the adsorbent are primarily governed by van der Waals forces; whereas, in chemisorption, the dissociation of molecular hydrogen into the atomic form occurs,

after which atomic hydrogen diffuses into the bulk of the material to form a disordered solid solution or a compound [329,331]. Thus, an activation energy barrier must be overcome for chemisorption to take place. Activated carbon, carbon nanotubes and material, metal-organic frameworks (MOFs), zeolites, and clathrates typically fall under the physisorption category, whereas, metal hydrides and complex hydrides are examples of materials in which hydrogen storage by chemisorption occurs.

The structural features of nanomaterials are responsible for the adsorption and desorption behaviors of hydrogen during its storage and release. As proposed by Pasquini et al. [329], **Fig. 13** illustrates the mechanisms of chemisorption-based hydrogen storage in a nanomaterial. It can be observed that the thermodynamic stability of hydrogen sorption depends on the external size/spatial extension of the system (L_{ext}), (in this case, metal hydride formation), the specific change in free energy on the material's outer surface, and at planar defects ($\Delta\gamma^{int}$, $\Delta\gamma^{GB}$), the microstructural length scale (L_{mic}), the surface and interface areas, the stress (σ) and strain (ε) fields in the material, the chemical potential (μ_H), and the enthalpy of formation (ΔH^0). **Fig. 13** also illustrates that the existence of tensile strains facilitates the insertion of hydrogen, thus ensuring better storage stability; whereas, the reverse is the case for compressive strains. Additionally, **Fig. 13** illustrates that the kinetics of hydrogen sorption is also a consequence of L_{ext}, as a small L_{ext} implies a shorter diffusion distance from the surface to the interior regions of the material. This is also crucial for the desorption phase. Pre-existing interfaces such as grain boundaries could act as nucleation sites for accelerated hydrogen storage in the nanomaterial of interest.

Nanomaterials used for hydrogen storage may be in any of the forms of bulk nanocrystalline materials ($L_{ext} \gg 100$ nm), thin films ($L_{ext} < 100$ nm), standing nano-objects such as nanoparticles and nanowires ($L_{ext} < 100$ nm), and composite architectures including supported and embedded nanoobjects [332–335]. Depending on the desired structure, several physical and chemical preparation techniques can be adopted including severe plastic deformation, pulsed laser ablation, ball milling, physical vapor deposition, electron beam lithography, coprecipitation, thermal decomposition, electrochemical deposition, and chemical reduction.
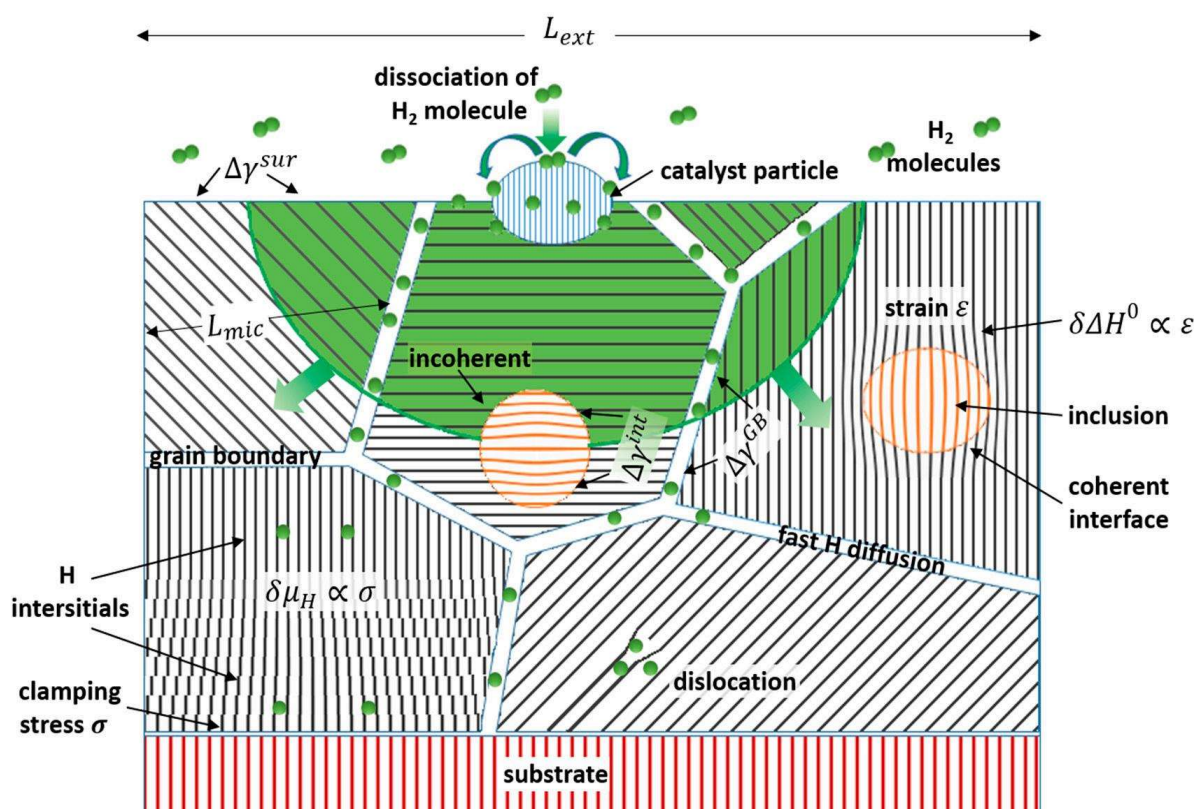
Fig. 13. Crucial geometric and structural characteristics for chemisorption-based hydrogen storage in nanomaterials. The green area represents the expansion of a hydride phase; this involves the dissociation of H_2, which may be facilitated by a catalyst, as well as subsurface penetration and diffusion of H atoms. This process is accelerated along internal interfaces or lines [329].

Fig. 14 illustrates a classification of selected hydrogen storage materials, showing their attainable storage capacities and the required operating conditions. It is evident from **Fig. 14** that there has been notable progress in the development of several nanomaterials for storing hydrogen. While MOFs and activated carbons have exhibited potential for hydrogen storage at very low temperatures (77 K), complex hydrides and nano-confined structures have demonstrated significant enhancements in performance at or near room temperature. Nonetheless, for a material to be practically feasible, scalability for large-scale production, atmospheric stability, cost-effectiveness, and environmental impact, are crucial factors to analyze. A discussion of the recent developments in each class of material is presented next, alongside their suitability as affected by the aforementioned factors.
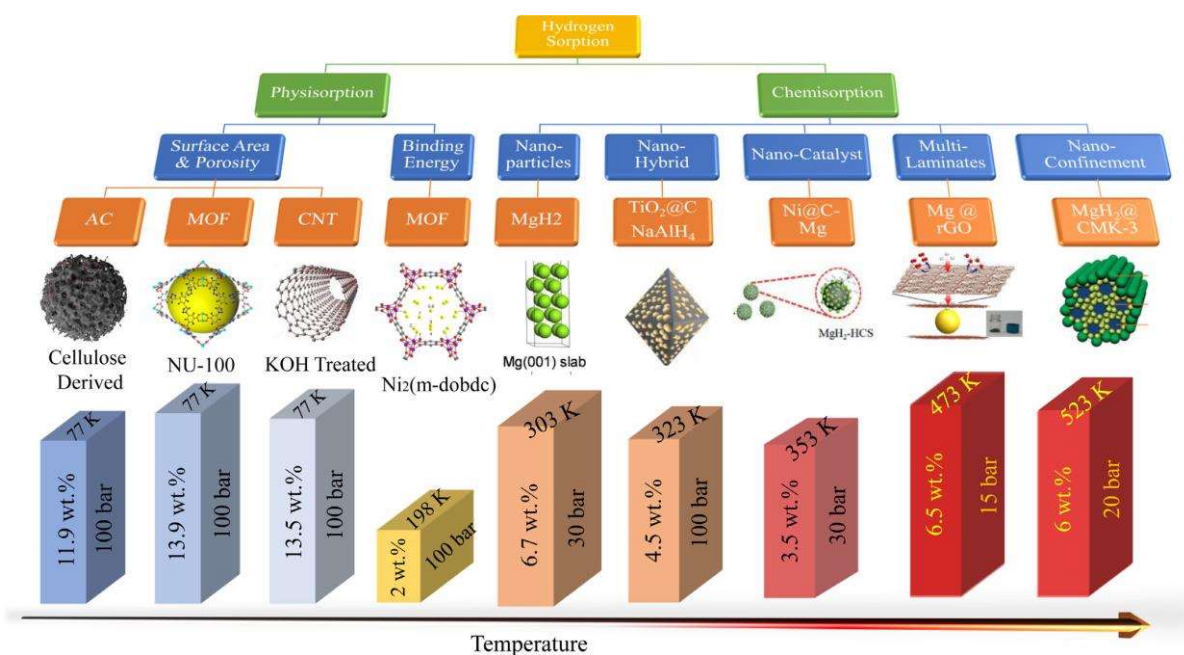
Fig. 14. Hydrogen storage capacities of different materials at increasing temperatures [331]

5.1.2.1 Activated carbon

As a low-cost and lightweight material with high porosity, large surface area, and tunable pore size, activated carbon (ACs) has been investigated as a potential candidate for mainly physisorption-based hydrogen storage. A pioneering study reported storage capacities of up to 5 wt.% at 77k and 20 bar [336]. Newer advancements have employed metal nanoparticle catalysts, including Pt, Pd, V, Co, and Ni to improve the hydrogen storage capacity at room temperature (a consequence of a phenomenon, termed 'hydrogen spillover' [337]); however, this improvement route has often resulted in high-pressure requirements for attaining reasonably high uptake capacities. It has been shown that at 298 K and 100 bar, super-activated carbon doped with Pt demonstrates a hydrogen absorption of 1.2 wt.% [338]. The catalyst facilitates the dissociation of hydrogen molecules on its surface, after which diffusion of hydrogen atoms towards the material and adsorption at the surface sites occur (a mechanism similar to that portrayed in **Fig. 13**). The presence of oxygen functional groups also enhance the effect of the metal catalyst as demonstrated by [339]. Recently, Ramirez-Vidal et al. [340] computationally evaluated the hydrogen sorption characteristics of 6 commercial activated carbons with wide ranging surface areas (996-2216 m^2/g), at temperatures between 77K and 273 K and pressures of up to 15 MPa; as high as 5.8 wt.% H_2 was realised. Their study showed that for ACs with equivalent BET areas, those with higher mesopore volumes and narrower micropores are better suited for hydrogen storage than those with wider micropores and lower

mesopore volumes. This finding is crucial to the optimal preparation and selection of ACs for

effective hydrogen storage.

5.1.2.2 Carbon nanotubes and carbon nanocomposites

Carbon nanotubes (CNTs) have the potential to serve as effective hydrogen storage materials

due to their ability to adsorb large amounts of hydrogen at room temperature [293,341]. These

materials are also highly attractive because of their relative ease of synthesis using renewable

resources, as well as their highly porous structure, chemical stability, and low mass density

[20]. CNTs, which are typically made of a graphene sheet rolled into a cylindrical shape are

categorized as single-walled carbon nanotubes (SWCNTs); whereas, multi-walled carbon

nanotubes (MWCNTs) involve multiple layers of graphene sheets [342]. The fabrication

methods for CNTs and MWCNTs include chemical vapor deposition, laser ablation, and

several other methods highlighted in the literature [343]. SWCNTs tend to exhibit a moderate

capacity for hydrogen adsorption (~2 wt.% at 77 K) [344]. However, as with ACs, surface

doping with metallic nanoparticles has been pursued (primarily for MWCNTs) for the

enhancement of hydrogen storage capacity, but this has only been moderately successful

[345,346]. Despite the computational predictions of high storage capacity (~19 wt.%) in 3D

CNTs [347], experimental investigations that can confirm these uptake capacities are lacking,

but necessary. Newer developments have featured the application of CNT-graphene hybrids

(which can also be classified as carbon nanocomposites). These are 3D carbonaceous

structures, which have been theoretically demonstrated as potential materials for improved

hydrogen storage [348]. In addition to temperature and pressure, the results of molecular

simulations performed on this hybrid material have outlined that the storage efficiency is also

dependent on the interlayer spaces in the graphene structure [349].

Other novel graphene-based nanocomposites have been formulated for improved hydrogen

storage, with promising results obtained. Masjedi-Arani et al. [350] demonstrated that the

hydrogen storage capacity of Cd_2SiO_4 nanoparticles was 1300 mAh/g; however, upon

modification of this nanoparticle with graphene (Cd_2SiO_4/graphene), a storage capacity of 3300

mAh/g was realized. Similar improvements have been reported for Ni/graphene and

$ZnFe_2O_4$/graphene nanocomposites [351,352]. It is worth mentioning that in addition to the

adsorption characteristics, hydrogen desorption was also enhanced. Other CNT-based

nanocomposites such as Ni-Sn alloy/MWCNTs (synthesized by electrochemical deposition)

have shown improved hydrogen storage properties compared to the base materials while

improving the stability (with up to 20 adsorption/desorption cycles achieved) over a relatively

shorter duration [353]. This performance can also be attributed to the multi-constituent structure of carbon nanocomposites and the presence of carbon, which facilitates low-temperature dehydrogenation [293]. The performance of these materials can be further enhanced via nanoconfinement techniques and this holds great potential for reversible hydrogen storage.

5.1.2.3 MOFs

Metal-organic frameworks (MOFs) are porous materials composed of metal ions or metal-containing nodes, primarily d-block transition metals, interconnected by organic linkers or multidentate ligands through coordinate bonds [354]. The metal centers in these MOFs exhibit significant variation in the number of binding sites available for these ligands, and this gives rise to various geometries such as linear, cubic, octahedral, and tetrahedral MOFs. A crucial factor when selecting ligands for the formation of MOFs is that the resulting network must be neutral, as charged networks can reduce porosity and hydrogen storability by accommodating counter ions within the framework [293,355]. MOFs allow for effective modification of the structure and topology of the interior surface where hydrogen is adsorbed, thereby enhancing hydrogen storage (via metal selection and ligand functionalization) [356]. While this design flexibility provided by MOFs is desirable, it also often complicates the process of identifying suitable metals and linkers and optimally selecting their compositions; this is because of the wide parameter space, which has to be searched [357]. Computational methods have been instrumental in accelerating this search via the identification of property performance trends and other useful insights necessary for a tailored design. The grand canonical Monte Carlo (GCMC) is the most widely applied computational method for the evaluation of hydrogen adsorption on MOFs, and is often accredited with good accuracy. Recently, Ahmed et al. [358] conducted a systematic assessment of published databases (both real and hypothetic MOFs) consisting of nearly 500,000 compounds for hydrogen storage. A computational screening was first performed (via GCMC), after which the most promising candidates were assessed experimentally. 3 MOFs, surpassing IRMOF-20 (a record holder for balanced hydrogen capacity) were identified (SNU-70, UMCM-9, and PCN-610/NU-100). It was further demonstrated that a volumetric ceiling at ~40 g H_2/L exists, and is worth considering for MOF formulation. While MOF-5 and MOF-177 have been reported to exceed the DOE gravimetric storage target at 77 K and 100 bar [357], studies such as Garg et al. [359] (where MOF-76(Nd) was synthesized using a solvothermal method) have realized less than 1 wt.% of adsorption/desorption capacity at 20 bar and 77 K; this demonstrates the critical impact of the

storage conditions on the sorption efficiency of MOFs. However, Suresh et al. [360] outlined that the poor performance of some MOFs may also be attributed to poor packing efficiency in the framework. They developed a strategy (via controlled crystal size distributions) for the improvement of the packing efficiency and hydrogen storage efficiency of MOF-5. Better performance was achieved relative to a commercial MOF-5.

5.1.2.4 Zeolites

Zeolites are microporous and crystalline aluminosilica-based materials with good surface area and porosity characteristics for the general storage of non-polar gases [337,361]. Their 3D structures contain tetrahedral primary building units which in turn determine the size of the channels and cages in which hydrogen can be stored [362]. Although it was reported that the performance of zeolites for hydrogen storage has not yielded promising results [331] compared to other nanomaterials, Masika and Mokaya [363] have reported up to 7.3 wt.% hydrogen uptake capacity for zeolite 13X at 20 bar and 77 K; this is comparable to values reported for MOFs [357]. A similar hydrogen uptake (7 wt.%) for different zeolites was reported in the computational study of Sun et al.[364]. However, it should be highlighted that, compared to zeolites, MOFs possess higher surface areas and more tunable characteristics (via metal selection and ligand functionalization) for the enhancement of their hydrogen sorption properties. Besides these attributes (surface area and tunability), the selectivity, ease of regeneration, and stability/cyclability of zeolites are typically not as favorable as MOFs; hence, newer research developments have been directed toward MOFs. A review by Dündar-Tekkaya and Yürüm [365] on the hydrogen storage potential of MCM-41 (a popularly applied zeolite) showed that the maximum reported hydrogen uptake was 2.01% (at 77K and 35 bar). Furthermore, commercial zeolite X has been utilized at atmospheric pressure to achieve up to 2.4 wt.% (at 77 K) in hydrogen uptake [366,367]. Despite this performance, a crucial advantage of applying zeolites as hydrogen sorbents is their relatively simple synthesis method, using a wide range of waste materials. In addition, hydrothermal temperatures of less than 100 °C can be utilised for their formation [368]; thus making for a less energy intensive production of this material.

5.1.2.5 Clathrates

Clathrate hydrates are compounds containing guest molecules within a host framework made up of water molecules connected by hydrogen bonds [369,370]. When water and hydrogen gas are mixed under high pressures and low temperatures, the hydrogen molecules are trapped in polyhedral cages to form the hydrate [371]. The stability of the structure is not only determined

by hydrogen bonding, but also by van der Waals forces, and intermolecular interactions. If the gas molecules are not enclosed within the framework, the hydrate's structure collapses into water. One of the first studies to propose the use of hydrates for hydrogen storage was that of Mao et al. [372], achieving up to 5 wt.% at 200 MPa and 243 K. These extreme and impractical pressure conditions have led the applications of various additives (promoter compounds) that reduce the pressure requirements considerably; some of these include tetrahydrofuran (THF) and tetra-*n*-butylammonium bromide (TBAB) [293,331]. However, there have been considerable disparities regarding the attainable hydrogen uptake capacity at lower and more practical pressure conditions. For example, Lee et al. [373] reported up to 4 wt.% of hydrogen at 12 MPa and 270K; however, Ogata et al. [374] claimed that only 0.26 wt.% can be stored at similar conditions. More recently, Weissman and Masutani [375] reported hydrogen storage capacities of <0.1 wt.% at 11 MPa and 263K using THF and TBAB as promoters (similar performance to the results of Ogata et al. [374]). Liu et al. examined the impact of THF on the hydrogen storage capacity in clathrates (using molecular simulations) and realized a range of 1.6 to 3.8 wt.% as determined by the concentration of THF [376]. They attributed this performance to the preference of hydrogen molecules to occupy the smaller cages of the hydrate; whereas, the larger cages were mainly occupied by 1 THF and 1 hydrogen molecule. Other critical factors such as the feed gas rate and the consequent diffusion rate of hydrogen gas through the liquid phase (mass transfer constraints) critically affect the storage capacity and should be optimally determined to achieve effective hydrogen storage.

5.1.2.6 Metal hydrides and complex metal hydrides

Compared to the previously discussed physisorption-based nanomaterials, the storage of hydrogen in metal hydrides is based on chemisorption (characterized by the existence of hydrogen in the atomic form) [293,377]. The common examples of these materials include metallic (such as Na, Li, K, and Mg) and intermetallic compounds (e.g. $LaNi_5$) [378]. The growing attention by researchers in this class of materials has been spurred by its high storage density, excellent stability, and compactness. Furthermore, Mg and its alloys have received the most attention in metallic hydride synthesis due to its low cost and abundance in the environment [377,379]. However, a critical drawback of these materials (in their pure form) is their poor reversibility. Thus, high temperatures (> 300 ºC) [293] are usually required for the desorption phase (slow desorption kinetics), and this is a result of the strong binding forces between the metal and hydrogen. As documented in [380,381], techniques such as alloying and the formation of hybrids and nanocomposites have been frequently applied to mitigate the slow

desorption kinetics problem. Catalysts such as Ni, Ge, V, graphene, and TiO_2 are typically applied to tackle this challenge. It was demonstrated in the work of Jangir et al. that the hydride decomposition temperature could be decreased from 150 °C to just 50 °C, depending on the amount of the added catalyst [382]. According to the work of Cho et al. the application of MgH_2 on graphene achieved 6.5 wt.% H_2 uptake at 200 °C and 15 bar in 120 min [381]. This uptake is 99.5% of the theoretical value (7.56 wt.%) for Mg nanocrystals. Furthermore, many interstitial sites available in the structure of intermetallic compounds facilitate good hydrogen uptake as mentioned in the work of [378], where LaNi5 was utilized.

In comparison to metal hydrides, complex metal hydrides (CMHs) (e.g. LiAlH4) which are newer nanomaterials are typically formed by the reaction of ionic hydrides (e.g. LiH) with covalent hydrides (e.g. AlH3); this class of CMHs is commonly called alanates [293,383]. The ability of hydrogen to form many combinations with elements such as nitrogen, boron, oxygen, carbon, aluminium, and transition metals for the creation of CMHs is a feature that enables the modification of functional properties for hydrogen storage applications. In addition to this flexibility, the high hydrogen storage efficiency of CMHs has made them a focus of significant research efforts. However, their utilization is also limited by the strong interaction with hydrogen, high desorption temperatures, irreversibility problems, slow kinetics, and complex synthesis procedures. As with metal hydrides, transition metals have been applied for catalytic hydrogen desorption. In addition to transition metals, nanoparticles such as CeF_3 and CeB_6 have been utilized for the improvement of the dehydrogenation of $NaAlH_4$ complex hydrides resulting in observed timeframes as short as 20 min [384]. Partial cation substitution, with smaller size- and/or higher valenced cations such as Mg is also a viable route for decreasing desorption temperatures [378,385]. Moreover, as high as 100 hydrogenation/dehydrogenation cycles have been achieved via the combination of carbon-embedded TiO_2 particles with $NaAlH_4$ [386]. Besides alanates, metal borohydrides, amine hydrides, and metallorganic hydrides are also promising CMHs with immense hydrogen storage potential, as documented in [387–389]. It is evident from this discussion that a majority of research developments in metal hydrides and complex metal hydrides for hydrogen storage have mainly focused on improving the thermodynamics and kinetics of hydrogen desorption, as this appears to be a more serious concern than hydrogen uptake rates, which are typically high (up to 18 wt.% [331]). With this growing research interest, it is expected that the development of low-cost dehydrogenation techniques will increase the potential of utilizing these materials in fuel cell

1574 vehicles. **Table 8** summarizes the key merits and demerits of the discussed materials for
1575 hydrogen storage.

1576   **Table 8.** A summary of key merits and limitations of different hydrogen storage materials.

Material	Advantages	Disadvantages	References
Activated carbon	- Low cost (can be synthesized from readily available and inexpensive materials) - Relatively straightforward synthesis	- Low hydrogen uptake - Low binding energy per hydrogen molecule (with limited binding sites) - Poor selectivity towards hydrogen as other gas may be adsorbed	[293,331,362]
Carbon nanotubes	- High durability and adaptability to harsh environments - Can adsorb hydrogen by physisorption and chemisorption mechanisms – good H_2 uptake - Low-temperature dehydrogenation - Lightweight materials (making them applicable for mobile applications)	- Although they can adsorb hydrogen at ambient conditions, cryogenic conditions are typically required for effective storage. - Their performance is sensitive to defects such as impurities and structural irregularities.	[293,390–392]
Metal-organic frameworks	- Very high surface area and porosity - Excellent structural and chemical stability - Exceptional tunability (can be tailored using a wide combination of metal ions and organic linkers)	- Some MOFs have acute moisture sensitivity (making handling difficult) - Complex synthesis (although several commercially available MOFs exist) - Can be expensive to synthesize	[357,393,394]
Zeolites	- High surface area and porosity - Tunable pore size to match the size of H_2 molecules	- Limited storage capacity - Limited reusability/cyclability - Slow desorption kinetics	[331,365]
Clathrates	- Low operational cost - Low flammability - Environmentally friendly (water is the main by-product	- Slow formation kinetics - Extreme conditions are required in the absence of a promoter - Low storage capacity	[331,371,375]

| | during the dehydrogenation phase)
• Simple and rapid release of hydrogen (no chemical reactions) | • Uncertainty in cyclic performance | |
| Metal and complex metal hydrides | • Good hydrogen uptake capacity
• Relatively mild hydrogen uptake conditions of temperature and pressure
• Long-term stability | • Sensitive to air exposure (violent reaction via oxidation)
• Slow dehydrogenation kinetics coupled with high-temperature desorption)
• Ammonia could be formed as a by-product of amine-based CMH synthesis. | [20,395] |

1577

5.1.3   Concluding remarks on surface storage

1579   In conclusion, surface hydrogen storage technology covers a variety of physical and material-

1580   based methods for storing hydrogen. These techniques play a crucial role in enabling the

1581   widespread adoption of hydrogen as a clean energy carrier. From high-pressure gas cylinders,

1582   cryogenic storage to cryo-compressed storage, as well as physisorption to chemisorption-based

1583   technologies with each offering its own set of advantages and limitations. While high-pressure

1584   and cryogenic storage provide compact solutions suitable for transportation and industrial

1585   applications, materials-based storage methods promise higher gravimetric and volumetric

1586   storage capacities. A comparative summary between the surface storage technologies

1587   highlighting their strength, weakness, opportunities, and threats are presented in **Table 9**.

1588   **Table 9.** Summary on the various surface hydrogen storage technologies.

Technology	Merits	Demerits	Opportunities	Threats
Physical	• Well established and mature technology. • Less prone to contamination allowing for high-purity H_2 storage. • Long operational life when adequately maintained.	• The limited capacity makes them unsuitable for large-scale storage. • Very large tanks can be challenging when space is a huge constraint. • Embrittlement of tank materials over time, potentially causes leaks or failures.	• Physical storage could be utilised to readily store excess renewable energy for later use (Power-to-Gas – P2G). • New developments in materials research could enable improved storage capacity,	• Leaks from storage tanks can impact the environment negatively. • Stringent safety regulations could increase the cost and complexity of tank-based storage.

			safety and efficiency for coatings.	• Emerging technologies like underground storage may offer higher benefits. • Alternative energy carriers (e.g., ammonia) could reduce the demand for tank-based hydrogen storage.
	• Cost-effective compared to alternative storage technologies.	• Potential explosion risk due to hydrogen's flammability if undetected leaks occur.		
Material	• Potential for high volumetric storage capacity, which is crucial for long-range transport applications. • Long term stability and reduced potential for embrittlement. • Greater margin of safety due to reduced leakage potential.	• Large-scale nanomaterial production can be expensive (requiring rare and high-cost materials). • Extreme temperature and pressure conditions for effective and efficient hydrogenation and dehydrogenation. • Contamination may affect thermodynamics and kinetics of adsorption and desorption; and may constitute a safety concern.	• Ongoing advancements in nanomaterials and solid-state storage technologies may lead to breakthroughs in efficiency, cost-effectiveness, and scalability. • Supportive regulations and incentives for clean energy and hydrogen storage technologies can create opportunities for growth.	• Dependency on rare or limited resources may pose challenges for widespread adoption. • Unintended hydrogen release from the variety of promising materials is yet to be intensely explored.

1589

5.2 Subsurface hydrogen storage

Underground gas storage (UGS) is the least expensive way to store significant amounts of gaseous hydrogen. Additionally, it provides significant savings on storage expenses as well as buffer space for potential supply shortages or shifting seasonal demand. This storage method traces its origins back to 1915 when the practice of storing natural gases in geological formations began. The depleted Ontario gas field in Canada marked the first successful underground gas storage (UGS) project [396]. As of January 2010, a total of 642 storage

facilities were explored with most (399) located in the United States and 50 in Canada [397]. Europe had 130 storage facilities, followed by the Commonwealth of Independent States (CIS) countries with 50. Asia and Oceania region were found to have 12 storage facilities while one facility was found in South America: Argentina [398]. Over the years, the number of storage facilities increased from 642 to 672. A closer look at the statistics reveals that depleted hydrocarbon reservoirs accounted for 74.2% (498) of all the facilities, while aquifers represented 11.4% (77), and salt caverns accounted for 14.4% (97). These suggest that the growth of UGS sites is likely to continue, with depleted hydrocarbon reservoirs holding the largest share. Consequently, these findings open up the possibility of exploring more potential sites for H_2 storage through further analysis.

Since the advent of natural gas storage in 1915, the major challenges from the stored gas have been mostly associated with the well integrity as summarized in **Table 10** [399]. For instance, in the Leidy field of Pennsylvania in 1969, the major event that led to the loss of life due to a blowout arose from the casing leak. It was documented that, stored gas migrated via 5 storage wells at 850 m^3/day and later from other 13 wells. As a result, gas gradually accumulated in the storage reservoir within the sandstone at shallower depths, leading to the fracturing of the rock over a wide area spanning several square miles. This phenomenon resulted in significant surface blowouts in both gas and water wells, with gas escaping continuously for a period of up to six weeks. Approximately 56,634 cubic meters per day of gas were vented from the wells, resulting in an estimated total loss of 113.3 million cubic meters of gas. The situation was eventually brought under control by reducing pressures and placing down-hole plugs in the wells [399]. Other aspects of the challenge, such as lack of awareness from the host community (where the facility is to be established based on market structure and transportation) as well as the legal implication relating to (i) ownership of storage rights (ii) the treatment of holdout problems where storage rights are privately owned, (iii) disposition rules for government-owned storage rights and (iv) resource sterilization [397], also existed. However, this has become a general trend as even the current space and awareness of subsurface hydrogen storage also face similar challenges such as techno-economic and societal acceptance, as well as policy and operation license [400,401].

Table 10. Selected reported challenges associated with natural gas storage facilities

Country	Facility location	Product Inventory	Date	Challenge(s)	Operators	Refs.
USA	Montebello, LA California	Gas	1950s – 1980s	• Migration of stored gas via old and poorly completed wells. • Fracture and damage of old well due to higher injection pressure than the original oil field pressure. • Improper well plugging after ceasing injection.	SoCal-Gas	[399,402,403]
	Muhlenberg, Kentucky	Gas	April 1980	• Tubing failure which led to a slipped and dislodged packer causing a gas leak and blowout.	Texas Gas Transmission	[399]
	Fort Morgan, Colorado	Gas	October 2006	• Casing leakage due to complete failure.	Colorado Interstate Gas Company	[399]
	Playa del Rey, California	Gas	April 2003	• The valve in the compressor unit broke which led to the release of gas.	SoCal-Gas	[399]
	Greenlick, Potter County, Pennsylvania	Gas	1988 - 1989	• Casing leak which led to leakage of the stored gas.	Consolidated Natural Gas	[399]
	Castaic Hills and Honor Rancho, California	Gas	1975	• Lateral migration of gas to a shallower formation via faults and cracks. • Leakage flowed to the surface thus oil production was affected in the nearby wells.	SoCal-Gas	[399,402,404]
	Playa del Rey, California	Gas	1940s	• Migration of gas from reservoir structure due to faulting and leakage from wells.	SoCal-Gas	[399,402,403]

	McDonald Island Stockton, California	Gas	October 1993	• Explosion due to gas conditioning failure which led to damage of facility worth 2 million USD.	Pacific Gas and Electric Company	[399,405]
	Downs, Clinton County, Pennsylvania	Gas	July 1975	• Gas leak due to overpressure (during injection) as the Downs storage field initially thought to be connected to Tamarack storage field was not.	Consolidated Gas Supply Corporation	[399]
	Epps, Louisiana	Gas	1980s – 1990s	• A gas leak during injection was observed as the stored gas migrated to an adjacent reservoir within the formation.	Trunkline Gas Company	[406]
Argentina	Lunlunta Carrizal Oilfield, Mendoza	Gas	2003	• Gas migration away from the injection well due to cracks.	Repsol YFP	[407]
Ireland	Kinsale offshore gas field	Gas	1971	• Geochemical studies indicate a gas leak from caprock before operation begins thus leakage of original gas reserves.	Marathon Company	[399,408]
UK	Rough Gas field, Southern North Sea, UK sector	Gas	February 2006	• Cooler unit failure from one of the four glycol dehydration units thus resulted in an explosion and several injuries.	Centrica Company	[399]

1634   As previously discussed in **section 3**, UGS storage facilities are utilized to fulfill peak gas

1635   demand that outpaces supply and long-haul pipeline capacity, and its storage is used to help

1636   market participants manage pipeline imbalance charges as well as the daily and seasonal price

1637   volatility. The essential parameters for natural gas storage are (i) working gas capacity (injected

1638   volume), (ii) cushion gas capacity (base or immobilized native gas), (iii) peak rate (daily peak

1639   flow rate when the reservoir is full), and (iv) efficiency (ratio of working gas to cushion gas)

1640   [409–411]. These parameters are crucial for accessing the general inventory of operations

1641   towards achieving a decarbonized economy by 2050 [412,413]. Particular attention should be

1642   given to working gas (in this case hydrogen) and the cushion gas type (such as CH_4, CO_2, N_2,

and even H_2 in some cases [16,414–417]) to alleviate the water inflow issues, pressure maintenance and increase in the volume of the working gas during withdrawal. A simple schematic between these two gases (working and cushion gas) is shown in **Fig. 15** during geological storage and withdrawal.
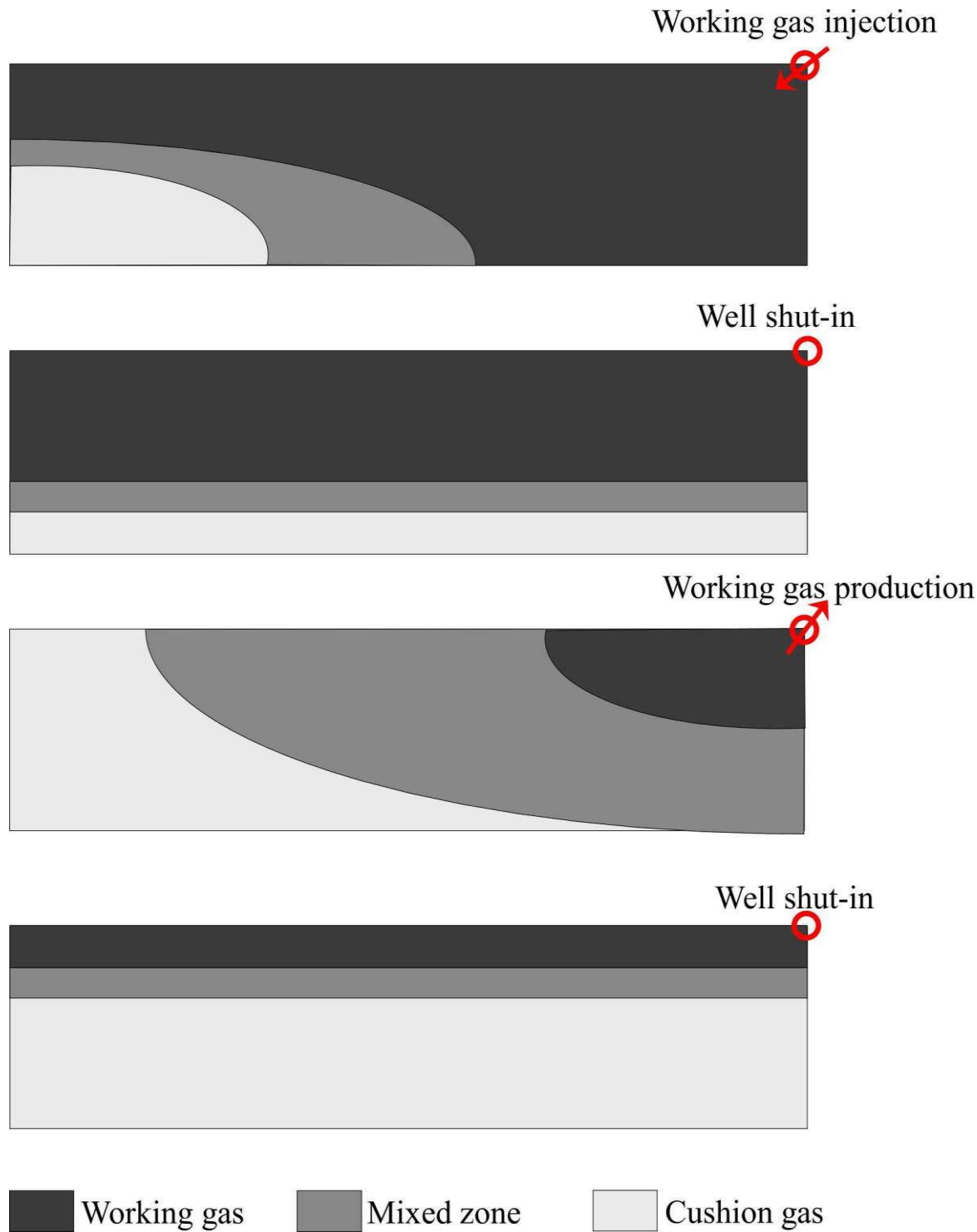

Fig. 15. Schematic diagram of working gas, cushion gas, and mixing zone during injection and withdrawal operations in geological formations [18].

Recently, underground hydrogen storage (UHS) studies have gained wide interest from various groups (such as universities, research institutions, and industry operators) to achieve safe and successful operations. Specifically, laboratory and field case studies have confirmed UHS feasibility based on safety [418]; capacity [418,419]; geochemical [420–422], physiochemical [423–425], biochemical, or microbial [218,426–429], and economics [430–432]. Despite their acceptance and potential, key factors for UHS application are required before implementation. For instance, the field and type of storage systems largely depend on some concerns, such as in situ reactions [426], large amounts of water extraction during H_2 production [433,434], undesirable hydrodynamic effects [435,436], leakage [218,426], environmental issues [437], site suitability and capacity [423] and costs [430] as it generally influences storage and withdrawal capacity, injectivity, gas immobilization, and efficient gas containment [409]. The selection of an appropriate location for UHS involves careful consideration of geological characteristics and cost factors. In the subsequent section, we thoroughly analyze these potential sites. Furthermore, **Fig. 16** offers a visual depiction of the sustainable carbon-free hydrogen energy generation and supply chain.
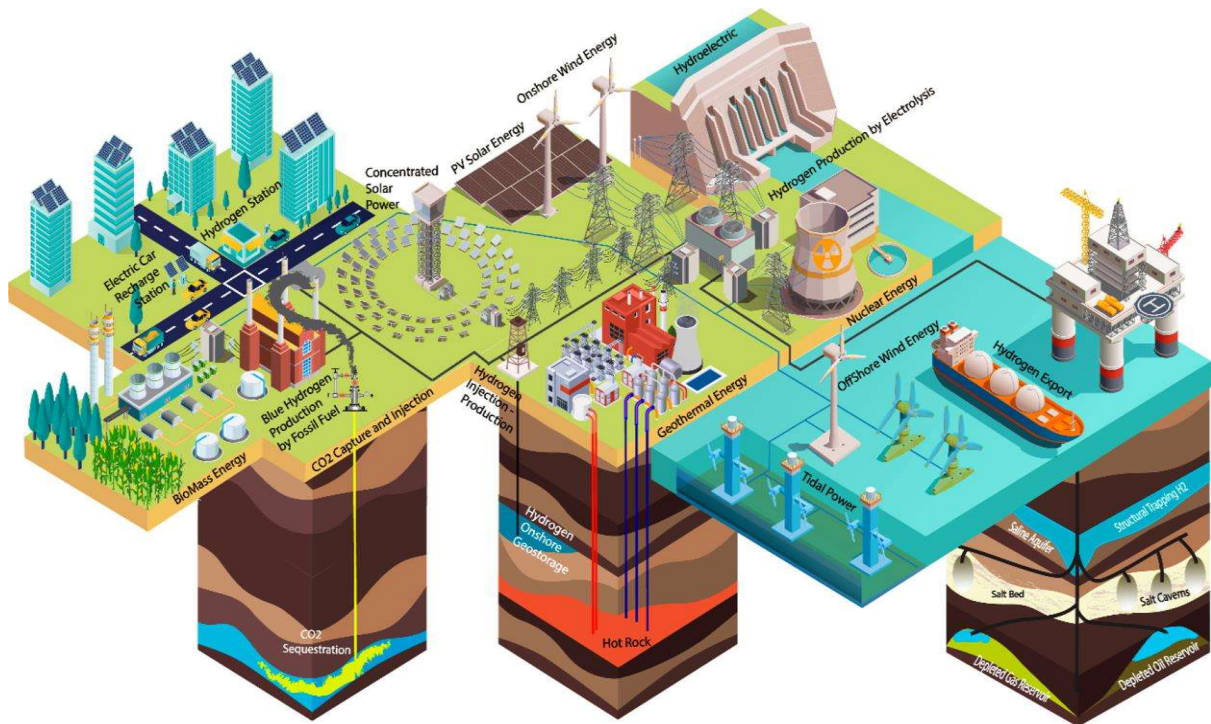

Fig. 16. Graphical schematic for a sustainable carbon-free economy in geological formation [438].

5.2.1 Salt Caverns

Gas storage caverns can be created either within salt domes or in layered salt deposits. Salt domes are large and uniform structures that can support operations at depths of about 6,000

feet. However, at depths exceeding 6000 feet, salt deformation can still occur despite the presence of well-designed caverns, primarily due to the combined effects of high pressure and temperature [439]. Bedded salts, which consist of thinner layers located at shallower depths compared to salt domes, often exhibit alternating layers of salt halite (NaCl) and non-soluble beds such as dolomite, anhydrite, and shale [440,441]. Caverns constructed within bedded salt formations for UHS tend to be thinner and less stable due to the increased heterogeneity of the rock [439,441]. **Fig. 17** is a simplified salt cavern schematic depicting the hanging and foot walls as a function of depth. The safety of caverns in terms of geo-mechanics and thicknesses, particularly regarding the hanging and foot walls, must be strategically determined. This involves carefully assessing the structural integrity and stability of the cavern, considering factors such as the diameter of the cavern and the thicknesses of the surrounding walls. To ensure stability, it is recommended that the hanging wall's minimum thickness be around 75% of the cavern diameter, while the footwall should have a minimum thickness of approximately 20% of the cavern diameter [442]. Additionally, capsule-shaped caverns are considered more stable and less prone to stress risks compared to elliptical or cylindrical shapes, especially when operating at depths of 1200 meters and under an overburden pressure of 270 bar [442].
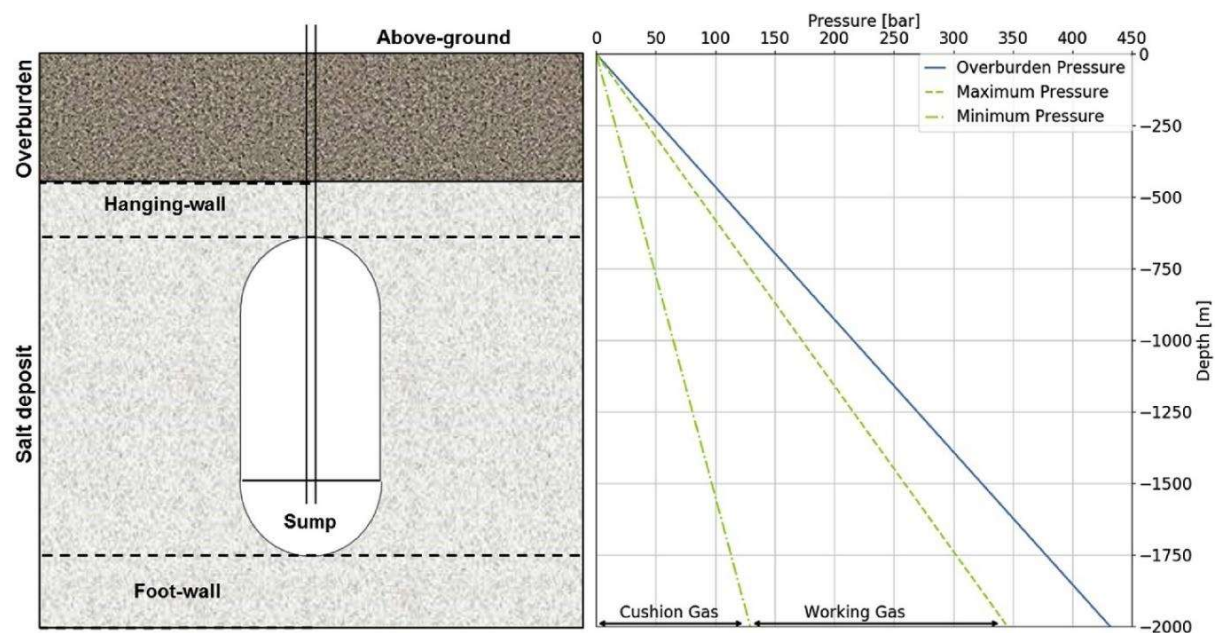

Fig. 17. Simplified salt cavern schematic with estimated pressure limits as a function of depth [443].

Salt caverns are created using a controlled solution mining technique, which eliminates the need for expensive tunneling or shaft-sinking operations. This technique takes advantage of the high solubility of rock salts, such as halite or NaCl, in water. By injecting water into the salt formations and then extracting the dissolved salt water, caverns can be formed without the need

for extensive excavation work or associated costs [418,430]. As can be seen in **Fig. 18**, this process involves three stages: leaching, debrining, and filling [444]. The salt surrounding these caverns possesses high impermeability, making it virtually leakproof. The primary potential avenue for gas loss is through any wells that may have leaks, although the overall gas loss through such leaks is estimated to be less than 1% [431]. Cavern storage can be classified into two common practices: wet storage and dry storage. In wet storage, the necessary pressure within the cavern is maintained by using brine, a concentrated solution of salt in water. On the other hand, dry storage relies on a cushion gas to provide the required pressure. However, dry storage is generally considered more expensive due to the additional cost of supplying and maintaining cushion gas [445].
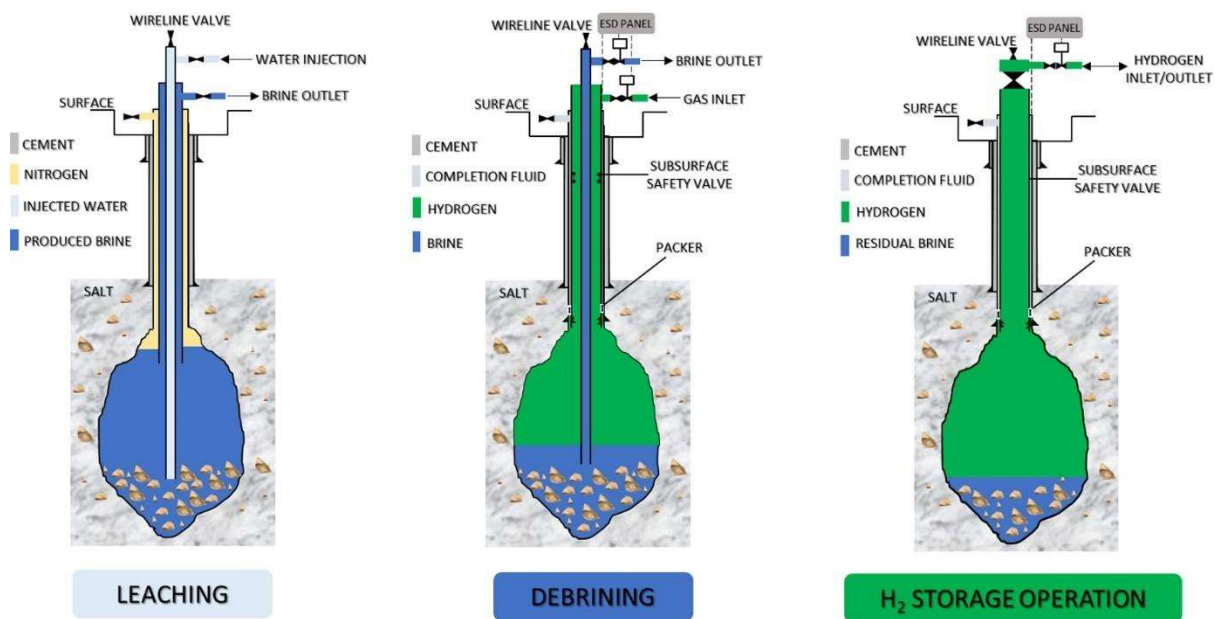

Fig. 18. Salt cavern construction stages: Leaching phase (left), debrining phase (middle), and filling phase (right) [446]. The *leaching phase* involves the injection of water via an access well into the salt formation for it to dissolve. Then pipes are cast in place with cement to make the process leakproof. After ascertaining the integrity of the cavern, the *debrining phase* begins by displacing the brine through cushion gas injection into the cavern. In the *filling phas*e, the brine is displaced with hydrogen gas.

The suitability of a salt formation for UHS can be evaluated by considering various factors including the depth, thickness, geographical size, and the presence of insoluble impurities such as anhydride or sedimentary interbeds like dolostone, limestone, or shale. These factors play a crucial role in determining the feasibility and stability of the salt formation for UHS

applications. In addition, visco-plastic properties of the caprocks can be used as a reference for effective seals during design as the requirement of the project informs the technicality of the caverns. For instance, when designing a cavern within a thin-bedded formation, it is important to consider the diverse lithologies present within the bedded salt formation. These variations in lithology can result in different properties that influence factors such as creep rates, deformation, and slip between the bedding planes. Thus, understanding and accounting for these distinct sets of properties is crucial for ensuring the stability and integrity of the cavern within the thin-bedded formation [441]. Consequently, the operating pressure of the cavern must be carefully determined, taking into account several factors: (i) the fracturing pressure of the weakest lithology present in the bedded salt formation, (ii) the minimum pressure required to prevent roof creep and ensure stability, and (iii) a pressure that remains below the maximum threshold that could potentially induce slip along the bedding planes [439,447].

Geological conditions like tightness, resistivity, mechanical properties, and in-situ chemical reactions make salt caverns a practical H_2 storage option [398]. Furthermore, their excellent mechanical stability during the injection and withdrawal process is not affected, thus – caverns are a good candidate for medium and short-term storage. Microbial activities are also less since a highly saline environment restricts the consumption of H_2 by microbes [433]. One of the main challenges associated with salt caverns, similar to domes, is their depth. However, there is an advantage to having a lower depth for the caverns. A shallower depth requires a smaller volume of cushion gas to maintain the desired pressure, which ultimately helps to reduce the operational costs associated with pressure maintenance [431]. However, about 30% of the total volume is recommended for cushion [448]. Other challenges such as managing the water quantity needed for leaching the cavern, ensuring proper disposal of brine, and determining the optimal number of wells for injection and withdrawal stages may arise during implementation [398,448,449].

Numerous UHS feasibility projects for example, in Europe under the HyUnder project [418,450], as well as in other countries [451] indicate caverns as an appropriate option for storing different substances of specific gases under high pressures. From the literature, the practical utilization of salt caverns for H_2 storage is primarily observed in specific regions such as the USA (e.g., Clemens Dome, Moss Bluff, and Spindletop), the UK (e.g., Teesside and Yorkshire), and Germany (e.g., Kiel town) [451]. This establishes them as the sole proven underground hydrogen storage technology thus far, although ongoing research suggests that other technologies are likely suitable as well. Many pilot projects are currently underway or

1747 planned across Europe to demonstrate hydrogen storage in salt caverns. These initiatives

1748 include HyCAVmobil by EWE, HyPSTER by Storengy, HyGeo by Teréga, H_2 @Epe by

1749 Uniper, the Green Hydrogen Hub by Gas Storage Denmark, HYPOS by VNG, and HyStock

1750 by Gasunie as highlighted in the report of Cihlar et al. [452].

5.2.2 Depleted hydrocarbon reservoirs

1752 A hydrocarbon reservoir, consisting of oil and gas, is a geological trap that has undergone

1753 multiple diagenetic formation stages (including source rock formation, migration, and

1754 maturation) to become a suitable storage medium for hydrocarbons (**Fig. 19**). Typically, these

1755 traps are sealed by an impermeable caprock (such as shale) and often supported by an

1756 underlying (bottom) or surrounding (edges) aquifer [453,454]. Depleted hydrocarbon storage

1757 options are advantageous over aquifer-type reservoirs because the pre-existing reservoir gas

1758 gives room for lower cushion gas and they do not require any integrity test for caprock

1759 efficiency [453,454]. However, in some cases, secondary integrity tests are conducted on the

1760 caprock if the structural integrity is compromised [448]. These types of reservoirs are often

1761 ideal for gas storage because they have a wide geographical distribution and huge storage

1762 capacity with a less ambiguous site evaluation. They also require less capital and operational

1763 cost as the existing infrastructure can be easily repurposed to operate with H_2 [454,455] (refer

1764 to previous discussion in **section 3**). Moreover, H_2 storage in these types of reservoirs is safe

1765 as demonstrated in recent modeling and numerical simulation studies
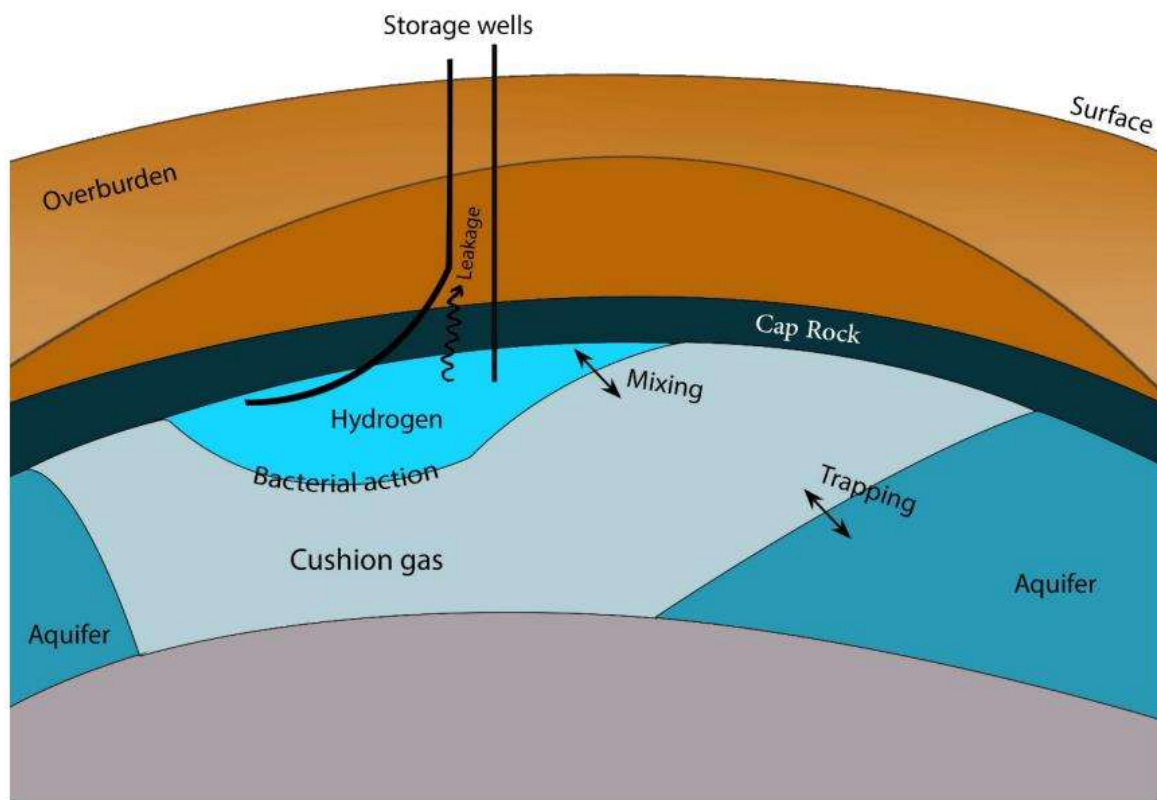
1766 [417,422,461,426,456,457,457,458,458–460].

Fig. 19. A simple schematic for a depleted hydrocarbon reservoir. At the storage site, the vertical scale is exaggerated for visualization purposes. The actual total depth typically ranges from 1 to 3 kilometers, whereas the lateral extent of the H_2 plume can span several kilometers. The height of the storage formation itself is typically a few tens of meters [462].

The experience gained in countries utilizing depleted oil and gas reservoirs for gas storage can be utilized in the selection process of suitable fields for conversion. This selection involves a thorough analysis of geological data and the physical parameters of the pre-selected structures [463,464]. Geological data such as the shape and dimensions of the structure, aquifer size, gas-water contact, reservoir properties, cap rock properties, and native fluid compositions are important factors in identifying suitable candidates for gas storage. Similarly, physical data of the storage rock including the (i) porosity, which should be high, (ii) the permeability, (iii) the water saturation, which should be as low as possible (iv) the drive mechanism (i.e., the ability of the aquifer to move within the reservoir rock when filled or emptied) are of great significance for gas storage [465]. The drive mechanisms could be depletion or water drive. In depletion drive reservoirs, the gas-water contact remains stable, resulting in high performance and fewer issues. However, in water-drive reservoirs, the gas-water contact moves upward during production, requiring additional pressure to push the water back during gas injection. This leads to reduced reservoir performance due to water production and the need for higher pressure to displace the water [465].

When the above conditions and factors from natural gas storage sites are known, H_2 storage in such a depleted gas field is potentially feasible. However, the difference in the thermophysical properties of these gases (i.e., CH_4 in the UGS and H_2 in the UHS, refer also to **Table 1**) shows that caution must be exercised when converting a natural gas storage site for H_2. For instance, the high reactivity, less viscosity, and smaller molecular size of H_2 increase the risk of H_2 loss via diffusion (vertical leakage) and dispersion (lateral migration) [413]. Furthermore, it is challenging to reduce the residually trapped H_2 during the storage cycle [422,436], as only a portion of the injected H_2 is recovered, with approximately 25-75% of the injected H_2 being lost as cushion gas, based on the reservoir and storage conditions [466]. However, H_2 losses via dissolution in brine are merely 0.4%, with an estimated overall loss from cyclic processes of 1% (per cycle) [435]. Cushion gas requirement is however higher than salt caverns but lower than aquifer storage sites [431,448].

After conducting appropriate investigations, only a limited number of existing gas storage sites have been evaluated for hydrogen storage capabilities. This is demonstrated in Austria's underground sun storage project where about 20% mixing of H_2 with 80% CH_4 was pilot tested in a natural gas storage site in the Molasse Basin [464]. The storage layer has an approximate thickness of 800 m and exhibits characteristics such as 22% porosity, 22% irreducible water saturation, and a temperature of 40°C. The field displays homogeneous characteristics, featuring a well-sealed shale layer without any aquifer connection, rendering it an attractive location for a pilot project [464]. Furthermore, the Diadema-led project by Hychico in Argentina is another field case where an existing Glauconitic sandstone reservoir with the following characteristics (thickness = 815 m, porosity = 25%, permeability = 500 mD, water saturation = 55% and temperature = 55 °C) is to be used for H_2 storage [467].

Several factors however come into play in converting a depleted oil reservoir to a UHS site because of the in-situ residual oil present in the form of an oil droplet, oil film, and clusters [468]. This is apparent because H_2 injection might trigger in-situ chemical reactions and subsequently convert the H_2 into methane gas. Again, the interaction between the H_2-oil interface could lower the H_2 purity and even result in H_2 loss due to dissolution. In addition, H_2 can react with formation minerals (via precipitation and dissolution) such as sandstones, carbonates, anhydrite, and calcites, which not only leads to the loss of stored H_2 but also degrade the overall reservoir properties and storage sites' performance [422,469,470]. Microbial activities such as methanogenesis, acetogenesis, and sulfate-reducing bacteria as

reviewed by Thaysen et al. [429] have been found to affect these types of reservoirs with temperature and salinity being the most controlling parameters.

Compared to caverns, porous media like depleted reservoirs and aquifers are well suited for base load requirements as the flow of fluid via the pores encounters significant resistance. However, this storage option has limitations in terms of the number of cycles per year and a modest rate of gas injection [471], unlike caverns which can accommodate multiple cycles [398]. Furthermore, the flow rate must also be maintained optimally, as an incorrect rate can lead to H_2 loss. For example, a slow injection flow rate can result in H_2 plume disintegration due to dissolution or diffusion into the surrounding formation, while a fast rate will lead to vertical gas rising and lateral spreading, or viscous fingering, which causes the injected gas to go beyond the intended boundary, increasing H_2-water contact and causing permanent loss [15]. Generally, at different injection rates, the interplay between gravitational forces and viscous forces or their combined effect gives rise to three distinct displacement regimes namely (i) a flow regime where gravity dominates, (ii) a flow regime characterized by the lateral spreading of gas, and (iii) a flow regime dominated by the propagation of lateral fingers [472]. This concept of varying injection rates can be seen in **Fig. 20**.
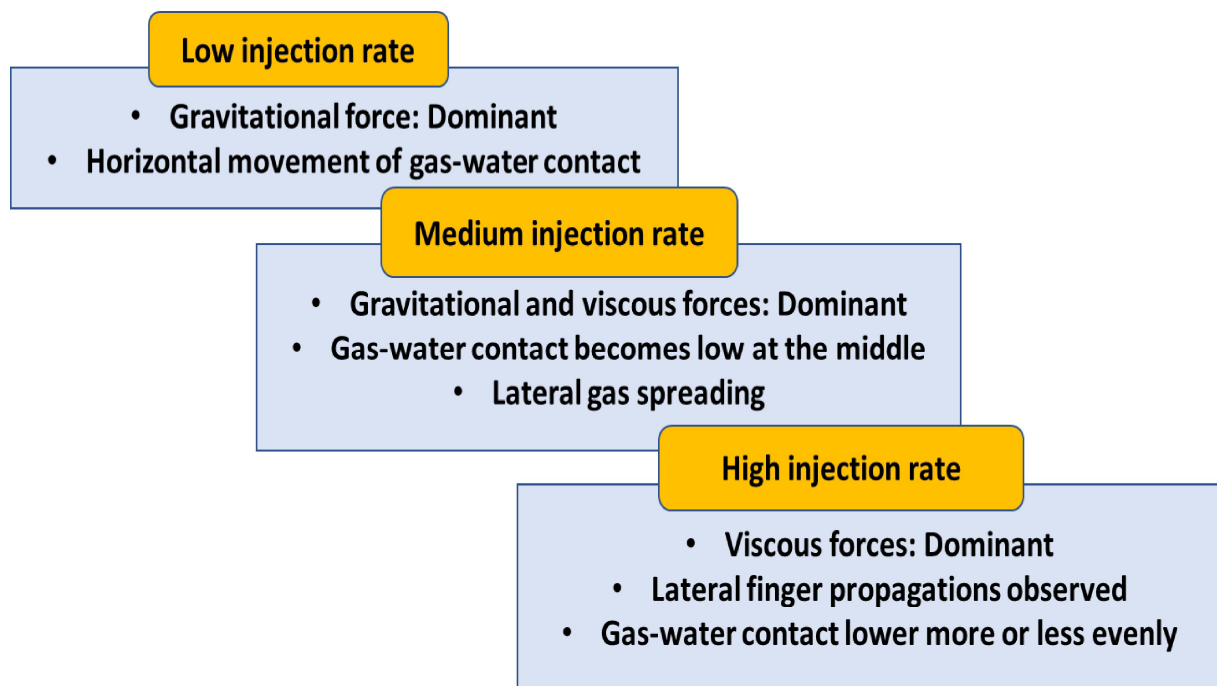
Low injection rate
- **Gravitational force: Dominant**
- **Horizontal movement of gas-water contact**

Medium injection rate
- **Gravitational and viscous forces: Dominant**
- **Gas-water contact becomes low at the middle**
- **Lateral gas spreading**

High injection rate
- **Viscous forces: Dominant**
- **Lateral finger propagations observed**
- **Gas-water contact lower more or less evenly**

Fig. 20. Varying injection rates based on flow regimes [472].

5.2.3 Aquifer

Aquifers are rock formations that contain porous and permeable spaces filled with fresh or saline water. These formations are found thousands of feet below the surface in sedimentary

basins worldwide. Aquifers provide an alternative for UHS in areas where salt caverns or depleted hydrocarbon deposits are not accessible [398]. There are two primary categories of aquifers suitable for this purpose: confined and unconfined aquifers. Confined aquifers involve the storage of hydrogen in underground water-bearing formations naturally enclosed by both overlying and underlying geological layers [473]. These geological layers act as a natural barrier, effectively preventing the migration of gas and ensuring the secure containment of hydrogen. On the other hand, unconfined aquifers pertain to groundwater systems where there is no impermeable sealing layer at the top, allowing groundwater to be in direct contact with the surface [474]. In the case of unconfined aquifers, the absence of a natural sealing layer can potentially lead to the upward movement of hydrogen through the groundwater and its release into the atmosphere. This could result in reduced efficiency and safety when it comes to hydrogen storage.

Gas storage in deep aquifers is similar to that in depleted deposits, however, information such as the structure (for storage capacity evaluation) and rock properties needed to characterize such subsurface aquifers, are rarely available as compared to depleted reservoirs [401,475]. The optimal configuration for a trapping system typically involves an anticlinal structure paired with an impermeable cap-rock layer. This cap-rock, which can consist of rock salt, clay stone, anhydrite, or similar materials, serves as an effective barrier to prevent the migration of fluids and gases. Geologic traps, formed as a result of plate tectonics and other geological processes, are also considered advantageous for trapping resources [423,473] with appropriate surrounding hydrostatic and threshold pressure [431]. A schematic description of this type of storage reservoir is shown in **Fig. 21.**

In the course of H_2 injection into the already-filled aquifer, a hydrodynamic force-displacement due to a change in density is induced between the gas and the liquid which often leads to either a downward or side displacement. As a result, the pressure in the porous media is increased due to the H_2 addition into the same aquifer volume without any withdrawal of the liquid originally present. This results in a gas/brine interface between the liquid and gas (H_2 plus cushion) which thus leads to a simultaneous percentage of liquid production in the production streams [450,476,477]. Storage gas volume, however, is a function of the reservoir porosity, temperature, and pressure as both injection and withdrawal pressures change systematically [398]. Recent simulation studies have reported a maximum recovery of up to 78% [433] in aquifers compared to 80% of the total injected H_2 after continuous operation for depleted hydrocarbon reservoirs [457,478] implying that porous medium can be advantageous for

seasonal H_2 storage as recovery generally improves with storage cycles. In terms of practicality, pure hydrogen storage in aquifers has not yet been tested on the ground. However, the geological storage of hydrogen, alongside other gases such as CO_2, CH_4, CO, and N_2, has been documented in projects conducted in locations like Ketzin in Germany, Lobodice in the Czech Republic, and Beynes in France [449].
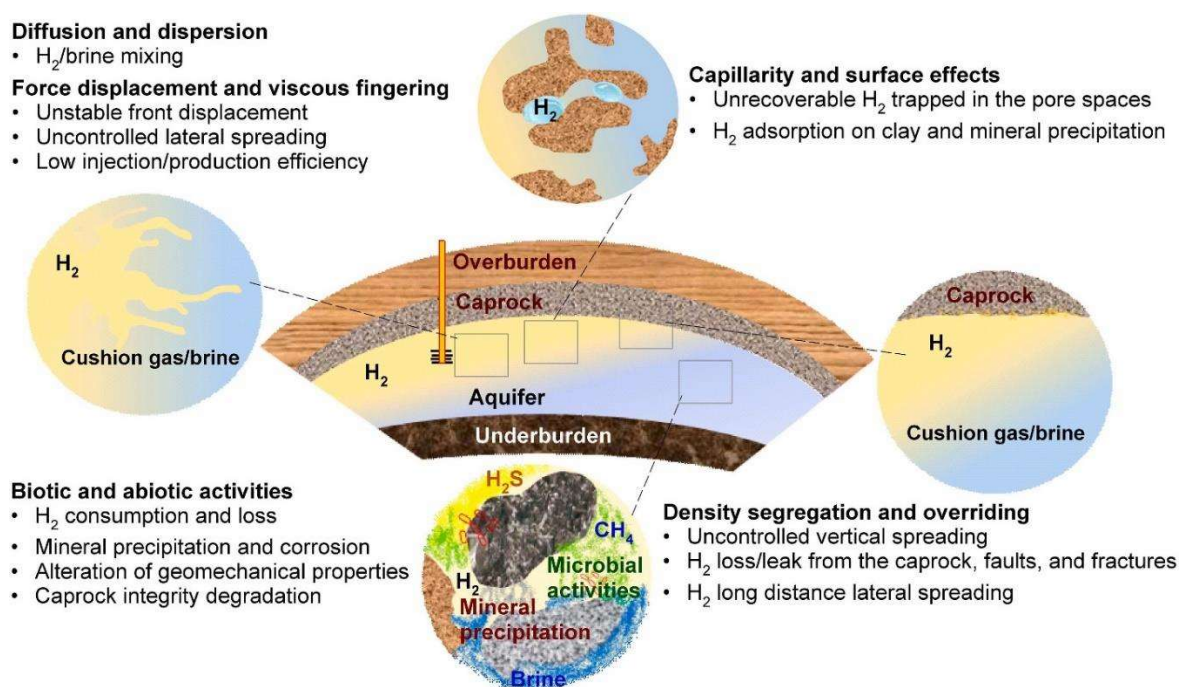

Fig. 21. Hydrogen storage schematics in saline aquifers depicting the primary fluid flow, transport mechanisms, and potential technical challenges [475].

Cushion gas for aquifer UHS is often greater than depleted reservoirs since no naturally occurring gas is present to offset the total volume needed [431]. In comparison with other options for cushion gas, as high as 80% is reported for aquifer-type storage thus, making it more expensive than depleted storage options. As can be seen in **Fig. 21**, aquifer storage options are majorly affected by (i) diffusion and dispersion, (ii) force-displacement and fingering, (iii) biotic and abiotic, (iv) capillarity and surface effects, and (v) density override and segregation [475]. While some of these challenges are common to porous media (aquifer and depleted reservoirs), the case of the aquifer is more demanding due to the huge uncertainty related to the unknown geology. Thus, detailed and laborious information (particularly on its integrity) must be properly captured, as well as the fluid pressure range to understudy the injection and production flow rates, before implementation for H_2 storage.

5.2.4 Hard rock caverns and abandoned mines

In comparison to salt caverns, which are built in sedimentary formations, the case of hard rock caverns involves the mining of caverns out of igneous or metamorphic rocks, which is followed by a liner (for encasement) and a concrete layer for efficient sealing [431]. The concrete layer needs to be engineered to effectively distribute the pressure load from the liner to the surrounding rock while also ensuring a uniform and smooth surface for the lining. Its design should incorporate structural strength to withstand the forces exerted on it and provide stability to the overall system. Simultaneously, it should offer a seamless and even surface that promotes the efficient functioning of the lining, ensuring its integrity and optimal performance. In addition, the lining must be gas-tight (leakproof) and chemically resistant to in-situ fluids. The liners in this design are typically made of stainless steel or polypropylene plastic. These liners serve a crucial purpose: they should not bear any load but instead need to withstand the gas pressure exerted on the concrete, which is then transmitted to the rock face. It is essential for the liners to exhibit resistance to both elastic and plastic deformation, ensuring their integrity and preventing any significant structural changes caused by the gas pressure [431,479].

Based on the geology of these types of rocks (i.e., igneous or metamorphic rocks), hard rock caverns will require higher pressure for effective H_2 storage. Similarly, their structural stability makes them require minimum cushion gas for pressure maintenance (as with the case of salt caverns), and since they are carefully crafted and lined, hard rock caverns have no risk of impurities. Salt and hard rock caverns are comparable in terms of operational standards such as multiple cyclicities (per year) and suitability for peak load requirements compared to porous medium (characterized for seasonal storage). Albeit, hard rock caverns are more expensive in terms of construction and design compared to salt caverns. An example of hard rock caverns is that in Sweden (Skallen project) (**Fig. 22**) which was used for natural gas storage [418,480,481]. It has also received storage applications for waste management purposes [418]. So far, no H_2 storage has been reported in a hard rock cavern; however, SSAB and LKAB (a Swedish company), are preparing a site as part of the HYBRIT green steel project [462].
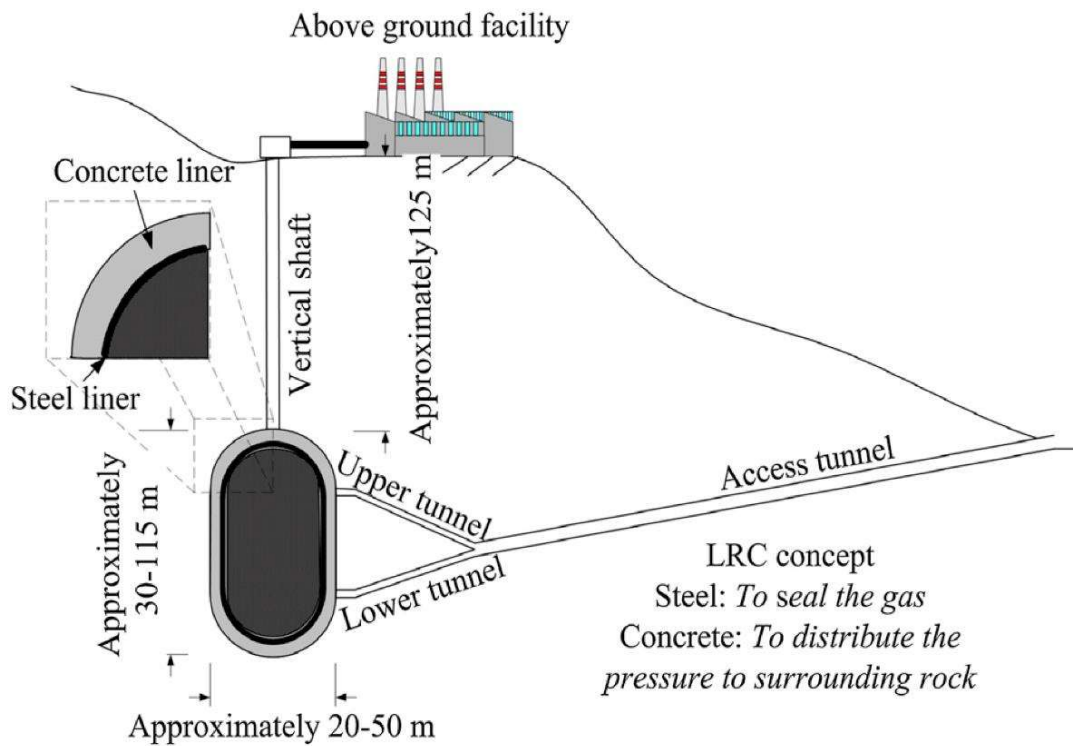
Above ground facility

Concrete liner

Steel liner

Vertical shaft

Approximately 125 m

Approximately 30-115 m

Upper tunnel

Lower tunnel

Access tunnel

LRC concept
Steel: *To seal the gas*
Concrete: *To distribute the*
pressure to surrounding rock

Approximately 20-50 m

Fig. 22. A diagram illustrating the application of lined rock caverns for hydrogen storage [480].

Abandon mines, which were previously used for the extraction of natural resources such as coal, can also be, converted into gas storage facilities. These storage media are advantageous in that, the associated cost of mining to create caverns, and liner installation as seen in hard rock mediums is not applicable. To store gas in this type of medium, the mining and geological history is vital as it helps to plan the required pressure and operational strategy for storage [16]. However, these facilities are not specifically engineered for the cyclic storage process, and the integrity of the surrounding rocks remains uncertain, thus, H_2 storage will require a careful and detailed survey before practical application. Literature studies have confirmed its feasibility for natural gas storage such as in Germany (the potash mine in Bernsdorf) [482], Belgium (the abandoned coal mines of Peronnes and Anderlus) [482], and in the United States (the abandoned coal mine of Leyden) [483]. Recently, experimental studies on H_2/brine/rocks systems have also indicated the potential for H_2 adsorption on coal seams [484–487]. We also note the application of other volcanic rock types such as basaltic rock for H_2 storage medium via H_2/brine/rock experimental investigation [488,489].

5.2.5 Concluding remarks on subsurface storage

In summary, UHS plays a pivotal role in the advancement and utilization of hydrogen as a sustainable energy solution. Porous mediums (such as aquifers or depleted hydrocarbon

reservoirs) are best considered as seasonal storage options compared to caverns (salts and rock) and mines which are best known for their multiple cyclicities. Moreover, UHS technologies offer numerous advantages over surface storage methods. These advantages encompass improved safety measures against external threats like terrorist attacks, military actions, and fires. UHS also enables higher storage pressures, greater energy density, reduced investment costs, and necessitates a smaller surface area compared to surface storage techniques [490]. However, to ensure the efficiency of UHS for practical implementation, establishing a general criterion becomes significant. These criteria will primarily depend on the careful selection of geological sites, taking into account various interconnected factors [462] such as (i) technical; e.g., geology, depth, areal extent, thickness, caprock strength, porosity and permeability, storage capacity, reservoir pressure, (ii) economics; e.g., labor, demand and supply distance, infrastructure availability, and the initial cost of investment, (iii) HSE factor; e.g., regional risks, environmental and public perception, and legal restriction and (iv) social criteria; e.g., societal acceptance and job creation.

The costs associated with developing underground hydrogen storage facilities can vary significantly due to a combination of factors, some of which are influenced by human decisions and actions, while others are inherent to the specific project and geological conditions [452]. Several key cost components include cushion gas, site exploration and development, compressors, and the construction of surface and subsurface infrastructure. These costs are not uniform across all storage projects, as they depend on various factors such as the size and capacity of the storage facility, the operating conditions, including pressure and temperature requirements, and the frequency of injection and withdrawal cycles. Consequently, it is challenging to provide a one-size-fits-all estimate for hydrogen storage costs, given the diverse range of variables involved in each project. For a concise overview of these hydrogen storage technologies, **Table 11** presents a comparative assessment based on different performance criteria and indicators whereas **Table 12** reports the current operating UHS projects around the world.

Table 11. Comparative assessment of subsurface formations for hydrogen storage.

Performance Indicators	Salt caverns	Depleted oil and gas reservoirs	Aquifer reservoirs	Rock caverns
Geographic availability and description	Formation of cavities through the dissolution of salt rock by injecting water from the surface into the salt [431].	Permeable carbonate or sandstone rocks [419,491].	Permeable and porous rock formations (typically carbonate or sandstone rocks) containing saline or fresh H_2O [431,482].	Homogenous and massive rocks such as gneiss and granite that are characterized by low permeability, exceptional mechanical strength, and minimal textural or structural weaknesses [482,492].
Suitability factors and consideration	Salt domes are most preferred over bedded salt structures [452].	Microbial activity, reservoir rock properties, fluid composition and operational parameters [452].	Caprock integrity for new developments, microbial activity, reservoir rock type, fluid composition, and operational considerations [452].	Metamorphic or igneous rock; low steel price [452].
Expenditure during new development	Exploration and assessment of geological characteristics, well infrastructure, cushion gas, and compression requirements [452].	Well infrastructure, cushion gas, compression [452].	Exploration and determination of geology, well infrastructure, cushion gas, and compression [452].	Cavern blasting, steel lining, cushion gas, and compression cost [452].
Usability	Frequent [493].	Seasonal [493].	Seasonal [493].	Frequent [490].
Safety	Very good [482].	Weak [482].	Weak [482].	Weak [482].

Geological tightness	Very good [482,494].	Very good [482,494].	Fair [482,494].	Fair [482].
Storage capability	High [452].	Site-specific [452].	Site-specific [452].	High [452].
Development cost (relative)	Low [452].	Low [452].	Low [452].	High [452].
Operation cost (relative)	Medium [452].	Low [452].	Low [452].	Medium [452].
Injection rate	Medium [482].	High [482].	Medium [482].	Medium [482].
Withdrawal rate	Medium [482].	High [482].	Medium [482].	Medium [482].
Working gas	High [452].	Medium [452].	Low [452].	High [452].
Cushion gas	Low [493,494].	Medium [493,494].	High [493,494].	Low [494].
Leakage risks	Low [493].	High [493].	High [493].	Low [493].
Cycles per year	High [452].	Low [452].	Low [452].	High [452].
Cyclic cost	Low [493].	Medium [493].	Medium [493].	Low [493].
Depth (ft)	984 – 5905 [452].	984 – 8858 [452].	1312 – 7545 [452].	3280 [452].
Operating pressure (psi)	507 – 3045 [452].	217 – 4133 [452].	435 – 4568 [452].	290 – 2900 [452].
Storage suitability	Proven [443,446].	Pure H_2 under investigation; proven for H_2–CH_4 mixtures [452].	Still under investigation; proven for town gas with up to 60% H_2 [452].	First H_2 storage is still in the development phase [452].
Technical readiness level	8 [452].	3-6 [452].	3 [452].	5-6 [452].
Chemical conversion rate	Low [493].	Medium [493].	High [493].	Low [493].
Research prospects	Precision and optimization of the production and injection time [452].	Impacts of in situ bacterial reactions and native natural gas [452].	Rocks' impermeability and in situ bacterial reactions [452].	Compatibility of H_2 with lining materials [452].

1975

1976 Finally, to harness the advantages of these technologies for hydrogen storage, particularly in
1977 porous media, it is crucial to take into account key insights derived from underground gas
1978 storage (UGS) and carbon capture and storage (CCS) methods. These include [453]:

1979 • Conducting a geological study of the selected structure and its cap rock to assess their
1980   suitability for H_2 storage.
1981 • Investigating the behavior of the structure during the production phase, particularly for
1982   depleted or partially depleted oil and gas reservoirs.
1983 • Employing dynamic simulation and mathematical modeling techniques to understand the
1984   reservoir's behavior based on various drive mechanisms.
1985 • Determining the performance of the reservoir when it is filled to the original pressure and
1986   beyond, by assuming different dynamic pressure values at the wellhead.
1987 • Evaluating reservoir performance in relation to the number and type of wells (vertical or
1988   horizontal) and their completion methods (such as completion with gravel pack or large-
1989   diameter tubing).

1990 By considering these aspects, a comprehensive understanding of underground hydrogen
1991 storage (UHS) operations can be achieved, allowing for the effective assessment of storage
1992 integrity.

Table 12. Geological details of operating UHS projects around the world.

	Field name	Country	Average depth (m)	Storage capacity (m³)	Operating conditions	Gas composition	Year of commission	Operators	Refs.
Salt caverns	Teesside	UK	400	210,000	653 psi	95% H$_2$ + 4% CO$_2$	Sabic Petrochemicals	1972	[449,490]
	Clemens	USA	1,000	580,000	1015-1987 psi	95% H$_2$	Conoco-Phillips	1983	[449,490]
	Moss Bluff	USA	1,200	566,000	797-2024 psi	95% H$_2$	Linde PLC	2007	[449,490]
	Spindletop	USA	1340	906,000	986-2929 psi	95% H$_2$	Air Liquide USA	2014	[451,490]
	Kiel	Germany	1330	32,000	1160 -1450 psi	60% H$_2$	Not reported	1971	[449,490]
Depleted oil and gas reservoir	Underground Sun Storage	Austria	1200	115,444	1131 psi/30 – 80 °C	10 – 20% H$_2$ + 80% CH$_4$	RAG Austria	2014 - 2021	[464]
	Diadema	Argentina	600	Not reported	145 psi/55 °C	10% H$_2$	Hychico	2015	[449,451]
Aquifer	Lobodice	Czech Republic	400-500	Not reported	1305 psi/34 °C	50% H$_2$ + 22% CH$_4$ + 12% CO$_2$	RWE	2007	[451]
	Beynes	France	430	3.3 × 10^8	Not reported	16 to 50% H$_2$ + Natural Gas	Storengy	1983	[451]
	Ketzin	Germany	200 - 250	1.3 × 10^8	Not reported	Town gas – up to 62% H$_2$ + Natural gas	Formerly VNG	1964 - 2000	[451]

1993
1994

1995

6 Conclusions and Perspectives

The hydrogen economy refers to a vision and concept where hydrogen is utilized as a primary energy carrier, replacing or supplementing fossil fuels in various sectors of the economy. It involves the production, transportation (and/or distribution), utilization, and storage of hydrogen as a clean and sustainable energy source. Based on our investigation and comprehensive review of the literature, the following conclusion can be drawn:

1. The push for a hydrogen economy holds significant potential for addressing the challenges of greenhouse gas emissions, towards transition into a free carbon ecosystem. Specifically, it offers benefits in promoting clean energy, energy storage, decentralized energy production, energy source diversification, industrial application, energy conversion, and renewable energy integration.

2. Various methods of hydrogen production, such as electrolysis, reforming, and biomass conversion, offer diverse pathways for obtaining clean hydrogen. However, electrolysis (via water splitting) is currently gaining more attention for hydrogen production.

3. Hydrogen transportation options, including pipelines, compressed gas cylinders, liquid hydrogen carriers, and more recently, hydrogen blending with natural gas provide flexibility and scalability for widespread adoption. The current attention is gradually shifting towards hydrogen blending with an effort to increase the fraction of H_2 (which is currently at 20 to 50%) in the existing natural gas stream for transmission and distribution.

4. The utilization of hydrogen in fuel cells for power generation and in various sectors like transportation, industry, and residential applications demonstrates its versatility and potential for decarbonization.

5. Storage technologies, such as underground storage in salt caverns and depleted reservoirs, as well as surface (materials and physical-based) storage, enable efficient and reliable utilization of hydrogen. Furthermore, large-scale hydrogen storage (via geological formations) offers several benefits in terms of energy system flexibility, grid balancing, seasonal energy requirement, transportation fuel, and energy security.

6. The integration of hydrogen into existing energy systems requires the development of infrastructure, policy frameworks, and supportive regulations to ensure its safe and efficient deployment.

7. Further research is needed to address technical challenges, enhance system integration, and optimize the overall hydrogen value chain to maximize its environmental and economic

benefits. This can be achieved by a collaborative effort between industry, academia, and government as they all play a crucial impact in achieving a sustainable carbon-free society.

8. Finally, a detailed economic analysis that takes into consideration all the essential components of hydrogen economy is recommended for future research.

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Authors contribution

Nasiru S. Muhammed: Conceptualization, Investigation, Data curation, Writing – original draft. **Afeez O. Gbadamosi:** Conceptualization, Investigation, Data curation, Writing – original draft. **Emmanuel I. Epelle:** Investigation, Data curation, Writing – original draft **Abdulrasheed A. Abdulrahman:** Investigation, Data curation, Writing – original draft **Bashirul Haq:** Supervision, Writing – review & editing. **Shirish Patil:** Project administration, Formal analysis, Supervision. **Dhafer Al Shehri:** Resources, Formal analysis. **Muhammad Shahzad Kamal:** Writing – review & editing.

Conflict of Interest

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

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