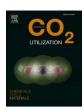
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Review article

# Blue hydrogen production from natural gas reservoirs: A review of application and feasibility

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#### ABSTRACT

Recently, interest in developing  $H_2$  strategies with carbon capture and storage (CCS) technologies has surged. Considering that, this paper reviews recent literature on blue  $H_2$ , a potential low-carbon, short-term solution during the  $H_2$  transition period. Three key aspects were the focus of this paper. First, it presents the processes used for blue  $H_2$  production. Second, it presents a detailed comparison between blue  $H_2$  and natural gas as fuels and energy carriers. The third aspect focuses on  $CO_2$  sequestration in depleted natural gas reservoirs, an essential step for implementing blue  $H_2$ . Globally,  $\sim 75\%$  of  $H_2$  is produced using steam methane reforming, which requires CCS to obtain blue  $H_2$ . Currently, blue  $H_2$  needs to compete with other advancing technologies such as green  $H_2$ , solar power, battery storage, etc. Compared to natural gas and liquefied natural gas, blue  $H_2$  gas results in lower  $CO_2$  emissions since CCS is applied. However, transporting liquefied and compressed blue  $H_2$  entails higher energy, economic, and environmental costs. CCS must be appropriately implemented to produce blue  $H_2$  successfully. Due to their established capacity to trap hydrocarbons over geologic time scales, depleted natural gas reservoirs are regarded as a viable option for CCS. Such a conclusion is supported by several simulation studies and field projects in many countries. Additionally, there is much field experience and knowledge on the injection and production performance of natural gas reservoirs. Therefore, using the existing site infrastructure, converting these formations into storage reservoirs is undemanding.

#### 1. Introduction

#### 1.1. Background

Currently, most of the global energy supply is being provided by conventional fossil fuels. In 2020,  $\sim$ 80.7% of this supply was met by oil, natural gas, and coal [1], as shown in Fig. 1. Despite their abundance [2], our dependence on fossil fuels needs to be reduced due to their environmental and health impacts. The use of fossil fuels leads to consequences such as increased greenhouse gas (GHG) emissions and accelerated climate change [3,4]. Furthermore, the extraction and processing of fossil fuels impose other environmental impacts, such as water

pollution, which results from fracking fluids, oil spills, coal sludge, coal mine drainage, etc. [5,6]. In addition, the production and combustion of fossil fuels result in the release of particulate matter and gases (e.g., sulfur dioxide and nitrogen oxides), causing respiratory illnesses and increasing mortality rates [7,8].

Over the coming decades, the best candidates expected to replace the depleting fossil fuels are renewable energy sources, specifically biomass, wind, solar, geothermal, hydro, and ocean renewable energy [9,10]. However, due to the variability and uncertainty of power supply from renewables (e.g., wind and solar), hydrogen has emerged as a critical player that can be utilized both as a fuel and energy carrier [11,12]. Since the start of the 1990 s, hydrogen generation has been studied as a

Abbreviations: ACE, Acetogens; AMU, Atomic mass unit; ASU, Air separation unit; ATR, Autothermal reforming; CCS, Carbon capture and storage; CG, Cushion gas; CNG, Compressed natural gas; DMC, Dimethyl carbonate; DME, Dimethyl ether; EGR, Enhanced gas recovery; F-T, Fischer-Tropsch; GHG, Greenhouse gas; GWC, Gas-water contact; IEA, International Energy Agency; IRENA, International Renewable Energy Agency; IPCC, Intergovernmental Panel on Climate Change; LCA, Life cycle assessment; LNG, Liquefied natural gas; LOHC, Liquid organic hydrogen carriers; MeOH, Methanol; MET, Methanogens; MMV, Monitoring, measurement, and verification; NG, Natural gas; NGD, Natural gas decomposition; PSA, Pressure swing adsorption; PV, Photovoltaic; SMR, Steam methane reforming; SMR-CL, Steam methane reforming with CO<sub>2</sub> looping; SRM, Sulfate-reducing microorganisms; UHS, Underground H<sub>2</sub> storage; WG, Working gas; WGS, Water-gas shift.

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method for decarbonizing the mix of energy sources [13,14].

Hydrogen is demanded by energy-consuming sectors like industry, transport, commercial, residential, etc. [15]. Besides, hydrogen is greatly demanded as a feedstock for many industrial processes such as the production of ammonia and methanol, glass making, food processing, petroleum refining, and metal treatment [16,17]. According to the International Energy Agency (IEA), the global hydrogen demand in 2018 was estimated at  $\sim 73.9$  Mt H<sub>2</sub>/yr., mainly for the refining industry (38.2 Mt  $H_2/yr$ .) and the ammonia industry (31.5 Mt  $H_2/yr$ .) [18]. The countries with the highest demand for hydrogen include China, the United States, the European Union, India, Japan, and South Korea. These countries have implemented hydrogen strategies and invested in the development and deployment of fuel cell technologies. Other countries, such as Chile, Morocco, and Namibia, are emerging as exporters of low-carbon hydrogen. On the other hand, countries like Australia, Oman, Saudi Arabia, and the United Arab Emirates, which are traditionally exporters of fossil fuels, are exploring the use of clean hydrogen to diversify their economies [19].

Recently, several countries have announced major H2 plans and funds supporting the development, implementation, and dissemination of hydrogen technologies. For example, the Australian government has allocated more than 100 million AUD to support H<sub>2</sub> research and pilot projects. China has launched a Ten Cities program for H2 transport and announced that Wuhan is expected to be the first Chinese Hydrogen City by 2025. China has also set targets for the deployment of fuel cell electric vehicles and refueling stations on a large scale. France has allocated 100 million euros in support of deployment of hydrogen technologies. Additionally, France has set 2023 and 2028 targets for low-carbon H2 in industry and transport sectors. Germany has endorsed a 10-year national program with 1.4 billion euros for fuel cell and H2 technologies. The UK has launched two projects each of 20 million GBP, aimed at the development of technologies for low-carbon H2 supply and storage. Besides, other countries that have important H2 plans include Austria, Belgium, Brazil, India, Italy, Japan, The Netherlands, New Zealand, Norway, Saudi Arabia, South Africa, South Korea, and the United States [18].

Currently, fossil fuels (e.g., natural gas, coal, and oil) are the most cost-effective sources of H<sub>2</sub>, with costs ranging between US \$1 and US \$3

per 1 kg  $H_2$  [20]. In addition to  $H_2$  generation from fossil fuels,  $H_2$  can also be generated depending on other energy and/or material sources and processes, such as biomass [21], solar thermal energy [22], solar photovoltaic (PV) [23], hydro [24], wind [25], nuclear electric [26], and nuclear heat [27], as shown in Fig. 2.

Nowadays, the different types of  $H_2$  are codified according to the production technology and source used, which are referred to as the " $H_2$  colors". An important type of  $H_2$  is blue  $H_2$ , produced mainly from natural gas through different processes resulting in  $CO_2$  as a by-product. However, as this  $CO_2$  is trapped and stored, the resulting  $H_2$  is referred to as blue  $H_2$  [28–30]. Blue  $H_2$  could play an essential decarbonizing role, especially in the coming few decades.

#### 1.2. Review methodology

This review paper presents major aspects of blue H<sub>2</sub> production, which employs carbon capture and storage (CCS) technologies to minimize the CO<sub>2</sub> emissions associated with conventional H<sub>2</sub> production. In addition, the review gives significant insights into blue H2 as a potential energy carrier. The steps that were followed to conduct this review are detailed in Table 1. Initially, we selected the research topic due to the importance of reducing global CO2 emissions and the importance of H2 production for many industrial sectors. After determining our research interest, we defined four research questions, which are presented in Subsection 1.3. We then identified keywords to use in searching for relevant sources, which are listed in Table 1. These keywords pertain to topics such as the different types of hydrogen, hydrogen production processes, hydrogen and natural gas storage and transportation, subsurface gas storage, CO2 sequestration in natural gas reservoirs, and CO2 utilization and conversion processes. We conducted our search using search engines and databases such as Google Scholar, ScienceDirect, OnePetro (SPE Publications), ACS Publications, and Springer. Our search mainly focused on finding relevant journal articles (research articles and review papers), but we also included books, conference proceedings, and reports from reputable organizations such as the International Energy Agency (IEA), International Renewable Energy Agency (IRENA), and the European Commission when necessary.

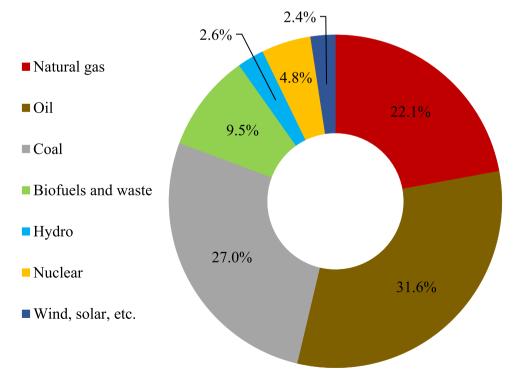
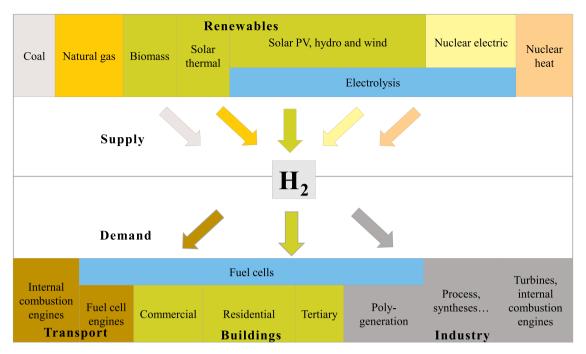


Fig. 1. Distribution of global supply of primary energy by fuel type in 2020. Estimated by [1].



**Fig. 2.** H<sub>2</sub> supply and demand. Modified from [15].

Table 1
Key steps of the review process.

Step	Details
1. Defining the review questions	Defining review questions that have not been thoroughly investigated in currently published review papers. The review questions should be specific, relevant, feasible, and researchable.
2. Stating the main keywords	Several keywords were used to reach the relevant articles, including: "hydrogen color codes," "blue hydrogen," "steam methane reforming," "autothermal reforming of methane," "hydrogen storage," "natural gas storage," "liquefied hydrogen," "liquefied natural gas," "gas transportation," "compressed gas," "underground gas storage," "oil and gas reservoirs," "gas storage in aquifers," "gas storage in salt caverns," "carbon capture and storage," "CO <sub>2</sub> leakage," and "methane desorption by CO <sub>2</sub> ," "CO <sub>2</sub> utilization," and "CO <sub>2</sub> conversion."
3. Searching for relevant sources	The keywords were entered into the search boxes of the databases/search engines: ScienceDirect OnePetro (SPE Publications) ACS Publications Springer Google Scholar Boolean operators (i.e., AND, OR, and NOT) were used to narrow down or expand the search results.
4. Reviewing the sources	Reading the abstract Reading the results, discussion, and conclusions Summarizing the relevant data and findings Referring to new references
<ol> <li>Analyzing, interpreting the findings, and writing</li> </ol>	Linking the summarized data and findings Identifying trends and patterns Identifying contradictions and conflicts Identifying gaps Drawing conclusions Preparing tables and figures Writing the introduction, body, and conclusions

Citing the used references

We screened the sources obtained through our search by carefully examining the titles, keywords, and abstracts to exclude those that were not relevant to our review paper. We then read and summarized the acceptable sources, focusing on the results, discussion, and conclusions. We analyzed, interpreted, and correlated the findings and identified trends, patterns, contradictions, conflicts, and gaps. We used this information to draw conclusions and write the entire paper, citing a total of 242 sources.

### 1.3. Review scope, objectives, and structure

The primary purpose of this review paper is to assess the feasibility of producing  $H_2$  from natural gas and injecting the resulting  $CO_2$  into natural gas reservoirs. The paper aims to address four main research questions, including:

- 1. To what extent is blue H<sub>2</sub> feasible to be produced from natural gas reservoirs as a fuel/energy carrier?
- 2. What are the comparative costs,  $CO_2$  emissions, and embodied energy of natural gas, liquefied natural gas (LNG), blue  $H_2$ , and grey  $H_2$  based on recent research in the literature?
- 3. Is it viable to utilize depleted natural gas reservoirs as a potential site for CCS?
- 4. What are the impacts of CO<sub>2</sub> sequestration on depleted natural gas reservoirs?

This review is organized into seven sections. Section 1 has introduced the review. Section 2 presents a brief overview of the chemical element hydrogen. Furthermore, Section 2 presents the different colors of hydrogen, focusing on the blue hydrogen definition and the processes used for hydrogen production. Sections 3 and 4 provide a detailed comparison between hydrogen (blue and grey) and natural gas, considering four main aspects: (1) transportation and storage, (2) environment, (3) economics, and (4) energy efficiency. The other sections of the review concentrate on CO<sub>2</sub> sequestration and utilization, an essential part of the blue hydrogen concept. These sections involve CO<sub>2</sub> sequestration into depleted natural gas reservoirs (Section 5), environmental effects of CO<sub>2</sub> sequestration (Section 6), field application of CO<sub>2</sub>

sequestration in natural gas reservoirs (Section 7), and  $CO_2$  utilization pathways (Section 8). Finally, the conclusions are stated in Section 9.

# 2. Blue hydrogen

#### 2.1. Overview of the chemical element hydrogen

On earth, less than 1% of hydrogen exists in the form of a stable diatomic molecule ( $H_2$ ) [31], which is a preferred form of hydrogen that can function as a fuel and energy carrier. Most hydrogen is found chemically bound in water and organic compounds (e.g., hydrocarbons) [32]. Table 2 provides the main physicochemical properties of hydrogen [33–35]. Because of its physical properties (i.e., small molecular size, low viscosity, low molecular weight, and high diffusivity),  $H_2$  is highly susceptible to leakage from transportation and storage facilities [36,37], posing an environmental and safety risk.

For decades, hydrogen has been used as a fuel and as an industrial substance in many applications and processes, such as fuel desulfurization [38,39], production of ammonia fertilizers [40,41], methanol synthesis [42,43], heat treating of steel [44], welding [45], glass production [46], semiconductor industry [47], buoyancy lifting [48], polymer production [49], leak detection [50], and cryogenic applications [51]. This widespread hydrogen use indicates that hydrogen will continue to be produced to satisfy the various industrial markets, at least for decades to come [52]. Therefore, developing technologies to decarbonize hydrogen production is becoming critically important.

There are several methods that are used for hydrogen storage. The application of these methods is highly governed by the physical state of hydrogen under given pressure and temperature conditions, which can be determined from the hydrogen phase diagram (Fig. 3). At low temperatures, hydrogen exists in the form of solid  $\rm H_{2}$ , exhibiting a density of 70.6 kg/m³ at 11.15 K ( $\rm -262~^{\circ}C$ ). At higher temperatures, hydrogen exists as  $\rm H_{2}$  gas having a low density, which, for instance, equals 0.08989 kg/m³ at 273.15 K (0  $^{\circ}C$ ) and 1 bar. The phase diagram of hydrogen also shows a region of liquid  $\rm H_{2}$ , determined by the solid line, the triple point (21.2 K), and the critical point (32 K) [31].

At atmospheric pressure and ambient temperature, 1 kg of hydrogen occupies a volume of 11 m<sup>3</sup>. Subsequently, hydrogen storage involves reducing the large volume of hydrogen gas to reach a relatively high hydrogen density and hence a high storage capacity. To achieve that, three approaches are followed: (1) applying work to compress the gas; (2) decreasing the gas temperature below the critical point; or (3) reducing the molecular repulsion forces by interacting hydrogen with other materials. It is noteworthy that for a storage method to be efficient, hydrogen uptake and release should be reversible. As such, covalent hydrogen—carbon compounds are usually excluded as materials for the

**Table 2** Physicochemical properties of H<sub>2</sub>.

Property	Value
Density at 25 °C and 1.01325 bar	$0.089 \text{ kg/m}^3$
Molecular weight	2.016 g/mol
Viscosity at 25 °C and 1.01325 bar	$0.89 \times 10^{-5}$ Pa.s
Diffusion coefficient in air	$6.1 \times 10^{-5} \text{ m}^2/\text{s}$
Boiling point	-252.95 °C
Critical pressure	13.0 bar
Critical temperature	-239.95 °C
Lower heating value	120 MJ/kg
Autoignition temperature	585 °C
Lower flammability limit	4% (by volume in air)
Upper flammability limit	75% (by volume in air)
Adiabatic flame temperature in air	2044.85 °C
Minimum spark ignition energy	0.02 mJ
Diffusion in pure water at 25 °C and 1.01325 bar	$5.13 \times 10^{-9} \text{ m}^2/\text{s}$
Research octane number	> 130
Thermal conductivity	$182.0 \times 10^{-3}$ W/m K
Quenching distance	0.64 mm

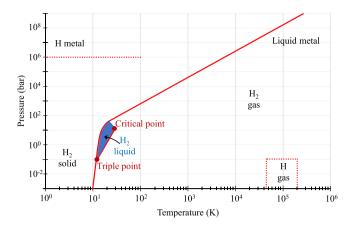


Fig. 3. Phase diagram of hydrogen. Based on [31,53].

storage of hydrogen. This is because releasing hydrogen from these compounds requires heating above 800  $^{\circ}$ C or oxidizing the carbon [31]. The various hydrogen storage methods are explained later in Subsection 3.1.

Since hydrogen is not readily available in sufficient quantities for energy applications, in the form of  $H_2$ , it is usually classified as an energy carrier rather than an energy source [54,55]. Generally, the energy sources (i.e., the primary energy forms) exist in the natural environment as two sources. The first source is of a finite quantity, such as uranium, peat, conventional fossil fuels (e.g., coal and natural gas), and unconventional fossil fuels (e.g., crude oil from oil sands). The second source is renewable (or relatively renewable), and it includes wind, biomass fuels, solar energy, geothermal heat, falling water, tides, etc., given that the rate of use is below the rate of growth. Often, these energy resources need to be processed prior to use in their raw forms. On the other hand, the energy carriers (or the energy currencies) are the energy forms that are transported and used. These include, for example,  $H_2$ , gasoline, heat, and electricity, which normally do not exist in the environment in their processed form [56–58].

### 2.2. Definition of blue hydrogen

Blue  $H_2$  is defined as the type of  $H_2$  that is produced from fossil fuels (normally from natural gas). However, in this case, the resulting  $CO_2$  emissions are captured and stored in subsurface formations (e.g., natural gas reservoirs) using CCS techniques, aiming for maximum elimination of greenhouse gas (GHG) emissions [59,60]. Table 3 compares the main  $H_2$  colors, including blue  $H_2$ .

**Table 3**Comparison between the different types (i.e., colors) of H<sub>2</sub>.

Туре	Primary source/process	CO <sub>2</sub> emissions	References
Brown H <sub>2</sub>	Fossil fuels (coal)	CO <sub>2</sub> is released into	[61]
Gray H <sub>2</sub>	Fossil fuels (mostly from natural gas)	the environment CO <sub>2</sub> is released into the environment	[62]
Blue H <sub>2</sub>	Fossil fuels (mostly from natural gas)	CCS techniques are applied (minimal	[63]
	natural gas)	CO <sub>2</sub> emissions)	
Green H <sub>2</sub>	Water electrolysis using electricity produced from renewable energy	Zero or minimal CO <sub>2</sub> emissions	[64]
Turquoise H <sub>2</sub>	Fossil fuels (mostly from natural gas)	Solid carbon production	[65]
Gold H <sub>2</sub>	Biologically produced from residual hydrocarbons using microbes in the subsurface (i.	Carbon neutral	[66,67]
Purple H <sub>2</sub>	e., depleted oil reservoirs) Water electrolysis based on nuclear energy	Zero or minimal CO <sub>2</sub> emissions	[68]

#### 2.3. Processes of blue hydrogen production

While several processes are used to produce  $H_2$ , in general, the production of blue  $H_2$  involves two main processes, as discussed below.

#### 2.3.1. Steam methane reforming process

The steam methane reforming (SMR) process (shown in Fig. 4) is the most widely used method of  $H_2$  production [69]. Globally, it is estimated that 75% of all  $H_2$  supply comes from SMR technology [70,71]. Initially, natural gas (i.e., methane) is desulfurized to remove any small amounts of sulfur that could poison the catalyst used in the SMR process [72]. Then, desulfurized methane is fed into the SMR unit in the presence of a nickel-based catalyst at a pressure of 35 bar and under high temperature (697–827 °C). In addition, it is required to heat the SMR boiler to produce the steam needed in the process. This heat is created by the combustion of gases produced from a pressure swing adsorption (PSA) unit. At this stage, CH<sub>4</sub> reacts with the steam, leading to the production of CO and  $H_2$  [73,74], as indicated in the following equation [75,76]:

$$CH_4 + H_2O \rightarrow CO + 3H_2, \Delta H_{298K}^{\circ} = +241kJ/mol$$
 (1)

After that, at  $\sim 197\text{-}247~^\circ\text{C}$  and 347–547  $^\circ\text{C}$ , CO undergoes another reforming process to convert it to CO<sub>2</sub> using water–gas shift (WGS) reactors, resulting in additional H<sub>2</sub> production, as shown in the following equation [76,77]:

$$CO + H_2O \rightarrow CO_2 + H_2, \Delta H_{298K}^{\circ} = -41.1 \text{kJ/mol}$$
 (2)

Finally, the impurities and produced  $CO_2$  are captured from the offgas by different methods (e.g., the PSA process), depending on the amount of  $CO_2$ , to enhance the quality of produced  $H_2$  [76].

#### 2.3.2. Autothermal reforming of methane

The autothermal reforming (ATR) process aims for  $H_2$  production while saving significant amounts of energy for the reaction compared with the SMR endothermic reaction. However, an air separation unit

(ASU) is required to provide a pure oxygen supply for the ATR process [79], as shown in Fig. 5. Initially, in the presence of a nickel catalyst, steam, natural gas, and oxygen (obtained from the ASU) react in an adiabatic ATR reactor to form syngas, which contains steam, H<sub>2</sub>, CO, CO<sub>2</sub>, and some trace gases. The heat generated due to partial oxidation is simultaneously used for the endothermic reforming reaction without the need for additional fuel. After that, the resulting syngas is cooled and fed to the WGS unit, where steam reacts with CO over a Fe and Cr catalyst to produce H<sub>2</sub> and CO<sub>2</sub>. Then, CO<sub>2</sub> is separated from the H<sub>2</sub>-rich gas using an amine-based syngas purification unit. Additionally, the H<sub>2</sub>-rich gas is further purified from any unconverted CO, Ar, and other impurities, using a PSA unit, up to 99.9% H<sub>2</sub> [78,79]. It is important to note that the ATR process is also applied when specific H<sub>2</sub>:CO ratios are required by certain industries [80].

#### 2.4. Challenges related to blue hydrogen

Blue  $H_2$  production is challenged by the competition from green  $H_2$ . Currently, blue  $H_2$  is viewed as a low-carbon, short-term solution during the  $H_2$  transition period. However, green  $H_2$  is being perceived as a better prospect for a zero-carbon sustainable energy future [81,82]. As such, the blue  $H_2$  market is expected to shrink due to green competition. Furthermore, blue  $H_2$  needs to compete with other advancing technologies such as solar power, battery storage, and thermal energy storage.

Another main challenge for blue  $\rm H_2$  production is its dependence on multi-step energy-intensive processes such as SMR and ATR [83,84]. Several steps in the SMR and ATR require a heat/energy supply that is usually provided by the combustion of external fuel. This heat/energy is needed for steps such as steam formation, air separation, pre-reforming, pre-heating, oxidation, steam reforming, autothermal reacting, syngas purification, pressure swing adsorption, etc. [76,78,85]. To overcome this challenge, it is important to optimize and improve each step within the methane-based  $\rm H_2$  production processes. For instance, Qiu et al. [86] suggested the use of concentrated solar heat to drive the endothermic

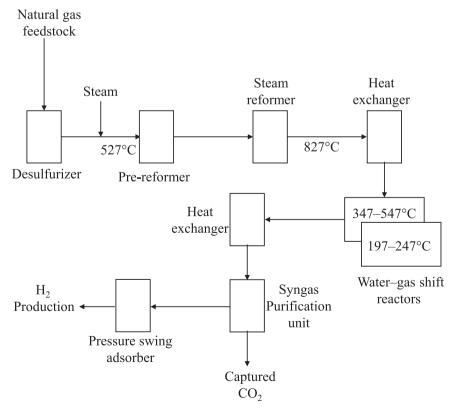


Fig. 4. Conventional SMR process for H<sub>2</sub> production with carbon capture. Based on [76,78].

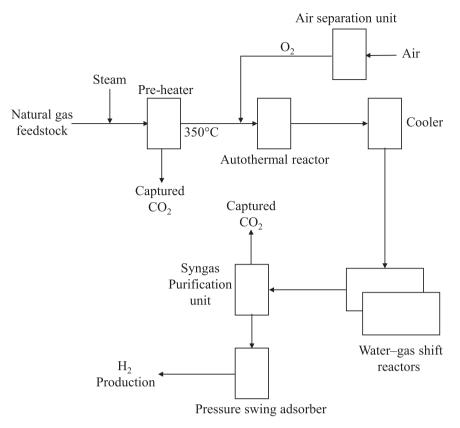


Fig. 5. ATR process for H<sub>2</sub> production with carbon capture. Based on [78,79].

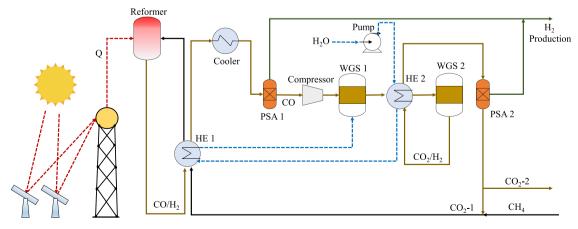
reactions of methane reforming in a process referred to as solar-driven SMR with  $CO_2$  looping (SMR-CL), as shown in Fig. 6. According to Qiu et al. [86], the SMR-CL system could reduce  $CO_2$  emissions by 34.98% compared with conventional SMR. In addition, this system could save the fuel consumed for combustion, achieving energy efficiency of 67.13%, which is relatively high when compared with other systems used to produce  $H_2$ , such as solar photovoltaic, photoelectrolysis, and methanol-reforming systems.

Another challenge for blue H<sub>2</sub> production is that it requires CCS technologies, adding costs to the production process. These costs are related to the use of booster compressors, transportation facilities, offshore/onshore injection facilities (e.g., injection wells), monitoring equipment on the storage sites, etc., which all entail capital, operating,

and maintenance costs [87,88]. This challenge is aggravated by the lack of studies that estimate the cost of CCS due to the large number of factors affecting the process, such as the capture technology, fuel prices, fuel type,  ${\rm CO_2}$  concentration in the exhaust gases, transportation methods and distance, etc.

## 3. Blue hydrogen vs. natural gas

To reduce global greenhouse gas emissions, many countries have taken serious steps to transition toward clean sources of energy. Blue  $\rm H_2$ , being a clean energy source, has a promising future in the next 30 years to reduce the current climate change problems and hence contribute toward sustainable energy. This has encouraged researchers and policy



 $HE = Heat\ exchanger,\ PSA = Pressure\ swing\ adsorber,\ WGS = Water-gas\ shift\ reactorber$ 

Fig. 6. Solar-driven SMR-CL for  $H_2$  production. Modified from Qiu et al. [86].

makers to evaluate further the feasibility of blue  $H_2$  production from various aspects, but detailed analysis is still needed. In this section, a comparative assessment of blue  $H_2$  versus natural gas is presented based on the current information in the literature. This comparison is performed by focusing on four main aspects: (1) transportation and storage; (2) environment; (3) economics; and (4) energy efficiency.

#### 3.1. Transportation and storage

Efficient, reliable transportation and storage technologies are essential for a successful, safe production market. This subsection discusses the techniques that are used for transporting and storing natural gas and  $H_2$ , which are generally common between both fluids.

#### 3.1.1. Transportation and surface storage

Natural gas can be transported (and/or stored) through several technologies, which involve: (1) Pipelines; (2) LNG; (3) compressed natural gas (CNG); and (4) gas to products. Pipelines are known as the most convenient way to transport natural gas. The system in this technology consists of a complex network of pipelines designed to transport the gas from its origin to various locations. These pipelines can range from 0.152 to 1.422 m in diameter and can operate at pressures higher than 49.3 bar. According to Rajnauth et al. [89], pipeline transportation suits better for short distances that are less than 3000 km [89]. The LNG technology involves the conversion of natural gas into a liquefied form at a low temperature (-160 °C). In LNG technology, the volume of the gas is shrunk to 1/600 of its volume in standard conditions, making it easier for transportation over large distances (> 4023.3 km) [90]. The CNG technology is useful for offshore gas reserves that cannot be produced due to the unavailability of pipelines or the high cost of LNG. The process of CNG involves compressing the natural gas to high pressures (higher than 137.9 bar) and low temperatures [89]. Economides et al. [90] performed a technical and economic evaluation of CNG and LNG. The results indicated that for short distances up to 4023.3 km, the cost of transporting natural gas as CNG is low at \$0.93-2.23 per MMBTU compared to \$1.5-2.5 per MMTBU of LNG. However, LNG is cheaper for distances greater than 4023.3 km.

Gas-to-product technology involves several modes, which are: (1) gas to wire; (2) gas to liquids; (3) gas to solids. According to Rajnauth et al. [89], in the gas-to-wire mode, natural gas is converted to electricity at the upstream source and transported by cable to markets. This technique is costly due to the DC transmission and requires a reserve of 0.283 bcm to 28.316 bcm of gas for transportation. In the gas-to-liquid mode, natural gas is converted to liquid hydrocarbons, such as methanol or synthetic crude oil, which are then transported. Although this technique is useful for transportation over large distances, its efficiency is not as high as LNG.

In the gas-to-solid mode, natural gas is converted into natural gas hydrates under higher pressure and low temperature by mixing natural gas with water [89]. Gas hydrates are often found in vast quantities in challenging environments, such as deep-sea beds and permafrost regions. The extraction of gas hydrates from these environments in commercial quantities is problematic due to several factors. In many cases, gas hydrates are contained in low-permeability sediments (e.g., claystone), resulting in low gas accumulation and low rates of gas production. Moreover, the gas enrichment factor decreases with depth. Therefore, with increasing depth, the amount of gas contained in the same volume of gas hydrate decreases [91]. Nevertheless, these challenges are not applicable to gas hydrates produced in a controlled environment, such a production facility. In this case, artificially produced gas hydrates can be transported to the market using special carriers such as LPG ships and towed barges. Compared to the cost of pipelines or LNG, researchers found that the cost of gas hydrates is much cheaper (e.g.,  $\sim 25\%$  less than the LNG cost), which makes the technique competitive to other transportation methods. The only concern with this method is the dissociation of natural gas where heat is required.

However, this can be solved by using the waste heat generated from power plants to dissociate the hydrates [89,92].

Moving to H<sub>2</sub>, there are several techniques that are used for H<sub>2</sub> storage:

- 1. Compressed  $H_2$ :  $H_2$  is stored in high-pressure gas steel cylinders with an operating pressure of 200 bar. In order to maximize the storage capacity, newly designed lightweight cylinders have been constructed. These cylinders can withstand a much higher operating pressure (800 bar), aiming to further improve the volumetric density of  $H_2$  to half of its value in the liquid state. Although this technique is commercially available, it can lead to some safety issues because of the high pressure required to achieve the high volumetric density [31,93].
- 2. Liquid storage form: an alternative and safer technique to enhance the  $\rm H_2$  energy density is through liquefaction. The advantage of this technique is that required  $\rm H_2$  densities can be achieved at atmospheric pressure. In this technique, several components are needed, including compressors, heat exchangers, and valves. The liquefaction process starts by compressing the gas, then cooling it at the heat exchanger, and finally passing it through a valve where it undergoes an enthalpic Joule-Thomson expansion to produce liquefied  $\rm H_2$ . Additionally, the liquefaction process involves cooling  $\rm H_2$  at cryogenic temperatures that are as low as  $-253\,^{\circ}{\rm C}$  using some gases such as helium and neon. Although the liquefaction process is found to be effective in achieving higher  $\rm H_2$  energy density than compressed gaseous  $\rm H_2$ , there are some significant disadvantages, such as  $\rm H_2$  evaporation and leakage [93].
- 3. *Cryo*-compressed H<sub>2</sub> *storage*: H<sub>2</sub> in this process is stored at a low cryogenic temperature in a vessel that can withstand a pressure ranging between 253.3 and 354.6 bar. The advantage of this process is the increased H<sub>2</sub> energy density, which in turn leads to greater storage capacity. In addition, this process is a safe way for H<sub>2</sub> storage due to the double-layer design and outer vacuum enclosure that protects it from mechanical and chemical intervention. However, the downside of this technique is the unavailability of cryo-compressed tanks in the market. Also, its dormancy time is short (7 days), compared to that of liquid H<sub>2</sub>, which is 7 times longer [93].
- 4. *Solid storage form:* hydrogen can be stored in solids where it is absorbed reversibly by solid compounds under certain temperature and pressure conditions. It can also be produced irreversibly by hydrolyzing some compounds, such as alkali metal borohydrides. The process of hydrogen absorption involves first dissociating it on the solid surface, and then its atoms diffuse into the solid host creating what is known as hydrides [94]. Storing hydrogen in the form of metal hydrides is relatively expensive. In a study reported by Amos [95], the capital cost of this form of hydrogen storage varies widely from \$820/kg to \$22,000/kg H<sub>2</sub>, which is mostly attributed to the cost of the material or alloy used in the storage. In certain scenarios, such as with very small hydride units, the capital cost can escalate to substantial levels, reaching \$60,000/kg H<sub>2</sub>.

Table 4 provides more details on the main methods of  $H_2$  storage, which include the gravimetric  $(\rho_m)$  and volumetric  $(\rho_v)$  densities.

In addition to  $H_2$  generation and storage,  $H_2$  transportation is critical in implementing  $H_2$  fuel in several industrial applications. The most widely used transportation methods are the following:

1. Compressed gas containers: medium amounts of  $H_2$  can be transported by trucks containing compressed gas containers or tubes under pressures of commonly 200–700 bar. For large  $H_2$  quantities, these gas containers and tubes are attached to compressed  $H_2$  gas tube trailers that are locked inside a protective frame. The maximum amount of  $H_2$  to be transported depends on the weight of these compressed cylinders or tubes. Therefore, in order to transport

**Table 4**Main hydrogen storage methods [96].

Method of storage	$\rho_{m}$ (mass%)	$ ho_{ m v}$ (kg H $_2$ / ${ m m}^3$ )	T (°C)	P (bar)	Remarks
Liquid H <sub>2</sub> (cryogenic tanks)	Varies (depending on size)	70.8	-252	1	<ul> <li>Liquid H<sub>2</sub></li> <li>Continuous loss of a few percent per day at room temperature (25 °C)</li> </ul>
High-pressure gas cylinders	13	< 40	Room	Up to	- Compressed H <sub>2</sub> gas
			temperature	800	- Light-weight composite cylinders are used (the material's tensile strength $= 2000 \text{ MPa}$ )
Adsorption of molecular hydrogen	pprox 2	20	-80	100	- Physisorption of $H_2$ on materials having very large specific surface area such as carbon - Totally reversible
Adsorption of atomic hydrogen on interstitial sites in host metals	pprox 2	150	Room temperature	1	- Totally reversible in metallic hydrides at room temperature
Complex compounds	< 18	150	> 100	1	- Complex compounds ( $[BH_4]$ or $[AlH4]$ ) - Desorption at high temperatures and adsorption at high pressures
Metals and complexes together with $H_2O$	< 40	> 150	Room temperature	1	- Chemical oxidation of metals with ${\rm H}_2{\rm O}$ and release of hydrogen - Not directly reversible

greater amounts of  $H_2$ , lighter tank materials need to be manufactured [93].

- 2. Cryogenic liquid tankers: H<sub>2</sub> in liquid form can be transported by trucks. In contrast to compressed gas containers, a higher quantity of H<sub>2</sub> can be loaded into the liquid truck as the H<sub>2</sub> density in the liquid state is higher than that in the gaseous state [93].
- 3. *Pipelines*: H<sub>2</sub> can be transported by pipelines that are linked to compressors, city stations, and storage facilities. It is to be noted that the role of the compressor station is to use the transmission system to provide the energy required to ensure continued gas flow according to the pressure and temperature needs. Pipeline transportation has many advantages, such as being cost-effective for large power plants, environmentally friendly, and having a long enough pipeline lifetime for several decades. However, there are some disadvantages of this technique, such as the possibility of gas leakage and the need to compress the H<sub>2</sub> to high pressures to improve its delivery speeds [93].
- 4. Blending with natural gas: one of the latest techniques of transporting H<sub>2</sub> is by blending it with natural gas in pipelines. This technique includes several separation and purification processes to separate H<sub>2</sub> from H<sub>2</sub>-natural gas blends. Additionally, it is found that blending

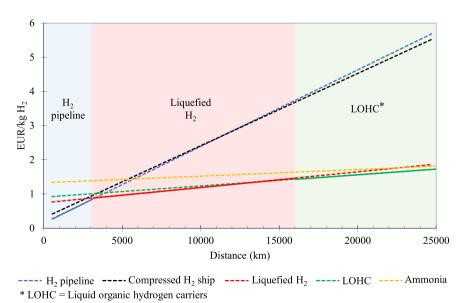
natural gas with H<sub>2</sub> reduces pipeline friction leading to an increased flowrate [93].

The cost of an  $\rm H_2$  transportation method is highly affected by the distance between the production and delivery points. As shown in Fig. 7, for distances that are below 3000 km, pipeline transportation of compressed  $\rm H_2$  gas seems to be the cheapest method, whereas for distances ranging from 3000 km to 16,000 km, transportation of liquefied  $\rm H_2$  exhibits the lowest cost. For distances beyond 16,000 km, hydrogen transportation in the form of LOHC (liquid organic hydrogen carriers) and ammonia (NH<sub>3</sub>) appears to be the cheapest.

#### 3.1.2. Subsurface storage

Both natural gas and H<sub>2</sub> can be stored in similar underground formations, which mainly include salt caverns, mines, natural aquifers, and depleted gas and oil reservoirs, as shown in Fig. 8.

Salt caverns are formed by injecting freshwater down into mineral deposits (e.g., nahcolite/trona, magnesium chlorides, potash, borates, and sodium sulfates) in a process referred to as solution mining. These caverns can be used to store chemicals, H<sub>2</sub>, and hydrocarbons (e.g., natural gas) [98,99]. Depending on their depth, salt caverns can



**Fig. 7.** Effect of transportation distance between a single production and delivery point on the hydrogen delivery costs, considering a scenario of 1 Mt H<sub>2</sub> per year and low electricity cost.

Source [97].

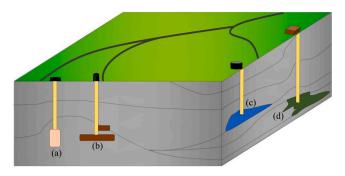


Fig. 8. Main subsurface storage formations for natural gas and  $H_2$ : (a) salt caverns, (b) mines, (c) aquifers, and (d) depleted gas and oil fields.

withstand pressures up to 250 bar with storage volumes that exceed  $1\times 10^6$  m³ [100]. Salt caverns are considered ideal sites for subsurface fluid storage for three main reasons. First, these formations are formed of salt, which has a crystalline structure, leading to a very low matrix permeability [101,102]. Second, salt caverns have favorable rheological characteristics involving the self-sealing of fractures [103,104]. Third, salt formations exhibit visco-plastic behavior, redistributing stress peaks around the cavern [105–107].

There are many success cases of  $H_2$  storage in salt caverns. Commissioned in 1972, three salt caverns at the Teesside location in the United Kingdom have been successfully used to store  $H_2$  at a depth of  $\sim$  350 m. Each cavern provided a storage volume of  $\sim$  7  $\times$  10<sup>4</sup> m³ [108, 109]. Other locations that have been used to store  $H_2$  include Clemens Dome (since 1983) and Moss Bluff (since 2007), both of which are in the United States. The salt caverns located at Clemens Dome and Moss Bluff provided an  $H_2$  storage capacity of  $\sim$  5.80  $\times$  10<sup>5</sup> m³ and 5.66  $\times$  10<sup>5</sup> m³, respectively [100,109].

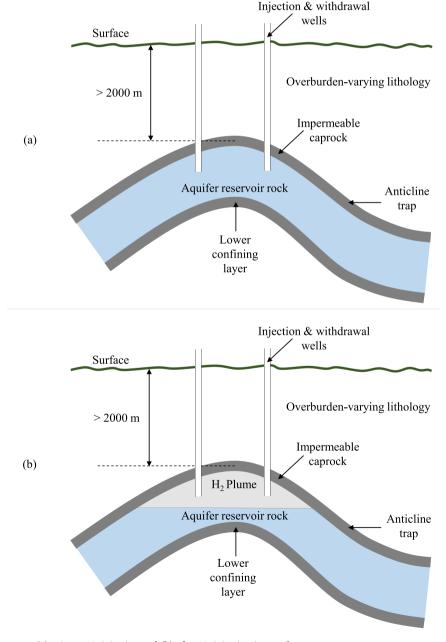
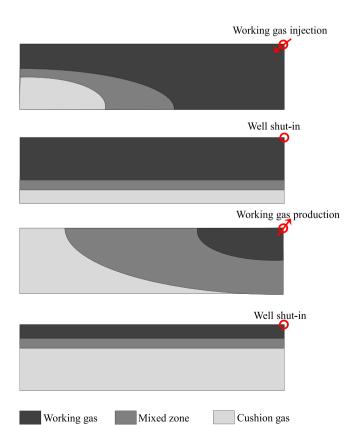


Fig. 9. Aquifer  $H_2$  storage structure: (a) prior to  $H_2$  injection and (b) after  $H_2$  injection is complete. Modified from [120].

Aquifers, which are subsurface formations containing fresh or saline water in their pores [110,111], represent a suitable option for subsurface storage of  $\rm H_2$  [36,112] and natural gas [109,113]. Many natural aquifers are characterized by appropriate size, structure, porosity, and permeability, allowing for feasible gas storage and recovery [114,115]. However, for an aquifer to be used for subsurface gas storage, it needs to have an impermeable caprock to prevent the vertical migration of gas, which may result from the buoyancy effect [116,117]. Additionally, the aquifer needs to be bounded by an anticline structure hindering the horizontal migration of gas, which helps form a significant gas plume around the well (i.e., at the upper part of the aquifer) [118,119], as shown in Fig. 9.

When a gas is injected into an aquifer for storage purposes, the gas moves upwards, whereas the liquid (i.e., water) present in the aquifer is displaced downwards and sideways due to the density difference between the fluids. This, in turn, increases the pressure in the aquifer due to adding gas without water withdrawal. Subsequently, the gas-liquid interface moves with the continuous injection of gas. However, when the stored gas is withdrawn, the gas-liquid interface changes, which can lead to disadvantageous simultaneous water production with gas [121, 122]. To alleviate the water inflow issues, maintain the aguifer pressure, and increase the volume of the working gas (e.g., H<sub>2</sub> and CH<sub>4</sub>), which can be produced from storage aquifers, the concept of cushion gas (CG) has been introduced. Several gases can be used as CG, including CO2 and N2. Typically, CG is injected prior to the working gas (WG), which prevents water from coning during the withdrawal stage [118, 123-125]. Fig. 10 shows a schematic diagram of subsurface WG storage with CG.

Despite its advantages in increasing the amount of recovered WG from subsurface formations, CG application could be challenged by two main issues. First, elevated reservoir pressure resulting from injecting CG of relatively low compressibility can cause formation damage. However, the use of supercritical fluids (e.g., supercritical CO<sub>2</sub>) as CG



**Fig. 10.** Schematic diagram of subsurface WG storage with CG. Modified from [126].

can tackle this issue due to the significant change in their density near the critical conditions [126,127]. Second, mixing between CG and WG may occur. Therefore, it is recommended to apply CG-assisted gas storage in formations of large thickness and dip angle [126]. Furthermore, several studies showed that when  $CO_2$  is used as CG in the supercritical state, its mixing with  $CH_4$  as WG is controlled due to the significant variations in their physical properties [128–131].

Compared to aquifers, depleted gas reservoirs are more fitting for gas storage, especially when the WG is natural gas. This is mainly because the residual gas in the reservoir can be used as CG. Generally, in aquifers, around 80% of CG is needed allowing for only 20% of WG. However, in gas reservoirs, only 50-60% of CG is required [109,132]. Moving to depleted oil reservoirs, these formations are highly reliable for gas storage due to their proven ability to trap hydrocarbons over geologic time scales, which indicates their caprock integrity [133–135]. However, when the WG to be stored in a depleted oil reservoir is H<sub>2</sub>, an extensive investigation should be conducted prior to H2 injection because many chemical (or biochemical) reactions can occur in the reservoir, which can lead to the irreversible conversion of H2 into other forms. Examples of these reactions include methanogenesis, biological souring, acetogenesis, and abiotic calcite dissolution [136–140]. Fig. 11 presents the main reactions that take place in carbonate hydrocarbon reservoirs used for H2 storage.

Fig. 12 compares the technical and economic aspects of underground  $\rm H_2$  storage (UHS) in different geological formations. Based on the comparison, salt caverns present the best UHS performance, providing the highest production rate and accounting for minimal losses that occur due to leakage and chemical conversion of  $\rm H_2$ . Furthermore, salt caverns exhibit the lowest seismic risks. Nevertheless, depleted gas reservoirs provide a larger storage capacity as one reservoir can be equivalent to several salt caverns. Additionally, there is an abundance of knowledge and field experience regarding the production from gas reservoirs, which facilitates the conversion of depleted gas reservoirs from production to storage reservoirs, utilizing the current site infrastructure.

#### 3.2. Environment

Fossil fuels represent the main energy source for most countries, leading to several negative environmental impacts, including global warming, air quality deterioration, acid rain, and oil spills [142,143]. According to an analysis conducted by Martins et al. [144], many European countries heavily depend on fossil fuels. Fossil fuel energy consumption is 60%, corresponding to 24 of 29 European countries [144]. This number shows the dependency on fossil fuels and how it can lead to several environmental problems in the long term. Crow et al. [145] built a deterministic model of GHG emissions from US gas fields with time. It is estimated that with future drilling activities, GHG emissions will show a rising trend with time. Additionally, for long-lived gas fields, it is estimated that 40–50% of GHG emissions are emitted during the last third of the production cycle [145].

Due to the high dependency of the world on natural gas, the industry has been working on several ways to optimize the production of natural gas to get a minimal impact on the environment while looking for other resources of energy. One of these ways is to produce blue  $\rm H_2$  while capturing and storing CO<sub>2</sub> [146]. There are several blue  $\rm H_2$  production technologies that lead to different amounts of greenhouse gas emissions. Oni et al. [78] conducted a complete techno-economic and GHG emission analysis using the life cycle assessment (LCA) framework of three blue  $\rm H_2$  production technologies: (1) SMR + CCS; (2) ATR + CCS; and (3) natural gas decomposition (NGD) + CCS. The results indicated that ATR + CCS yields the lowest GHG emissions, which is followed by NGD + CCS and then SMR + CCS [78].

Although most of the researchers highlighted the benefits of blue  $H_2$  technology in reducing overall GHG emissions, a limited number of researchers claim that the GHG emissions of blue  $H_2$  are higher than that of natural gas. For instance, Howarth and Jacobson [70] reported in

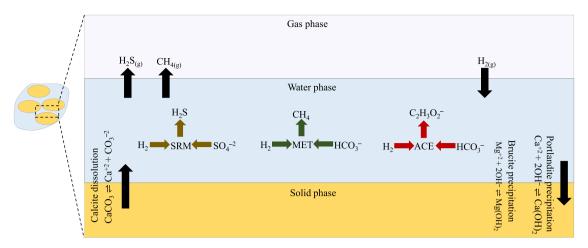


Fig. 11. Possible reactions occurring in depleted carbonate hydrocarbon reservoirs due to H<sub>2</sub> injection in the presence of SRM (sulfate-reducing microorganisms), MET (methanogens), and ACE (acetogens).

Adapted from [139].

	Salt caverns	Saline aquifer	Depleted gas reservoir
Capacity	Average	High	High
Discharge rate	High	Low	Average
Initial costs	High	Average	Average
Cyclic costs	Low	Average	Average
Seismic risks	Low	High	Average
Chemical conversion of H <sub>2</sub>	Low	High	High
Cushion gas need	Low	High	High
Leakage risks	Low	High	High
	Less preferred performance		Preferred performance

Fig. 12. Characteristics of UHS in different geological formations. Based on [141].

their paper that the footprint of blue  $H_2$  is more than 20% higher than that of burning natural gas and 60% greater than that of burning diesel oil. This particularly occurs in these cases where some of the  $CO_2$  equivalent emissions from blue  $H_2$  are being emitted during the extraction and processing of natural gas that is used as a feedstock in the SMR process, which may limit the benefit of using blue  $H_2$  powered by natural gas compared to using the energy produced from natural gas directly. Nevertheless, Howarth and Jacobson [70] indicated that the GHG emissions associated with blue  $H_2$  production could be reduced by using renewable energy to drive the SMR process, such as hydro, wind, and solar.

The paper reported by Howarth and Jacobson [70] has caused a lot of interest and has sparked discussions among scientists and engineers, who doubted the accuracy of the paper's findings due to its method and assumptions. For instance, Romano et al. [147] argued that the oversimplified method and assumptions used by Howarth and Jacobson [70]

overestimate the CO<sub>2</sub> emissions and natural gas consumption during blue hydrogen production. Romano et al. [147] also argued that the assumed CH4 leakage rate in the previous paper was on the high end of the estimated emissions for current natural gas production in the US and therefore cannot be used to represent all-natural gas and blue hydrogen value chains worldwide. They also stated that the  ${\rm CO}_2$  emissions associated with blue H2 production should be based on data from blue H2 plants that use current technology and are aimed at reducing CO2 emissions, which would achieve significantly higher CO<sub>2</sub> capture rates. In response to Romano et al. [147], Howarth and Jacobson [148] argued that Romano et al.'s discussion of CH<sub>4</sub> emissions is flawed because it is based on outdated and non-peer-reviewed sources, including a cartoon on a webpage from the oil and gas industry. Howarth and Jacobson [148] also maintain that their own analysis, which is based on data from real-world operations, is more reliable than the theoretical estimates used by Romano et al. [147]. Howarth and Jacobson [148] conclude

that blue hydrogen cannot be part of a decarbonized energy future due to its high greenhouse gas emissions, particularly from CH<sub>4</sub>.

According to Oni et al. [78], Howarth and Jacobson's analysis only looked at current SMR facilities with low carbon capture rates and assumed higher fugitive CH<sub>4</sub> emissions. Furthermore, Howarth and Jacobson did not consider that the CH<sub>4</sub> regulations are being developed to reduce fugitive CH<sub>4</sub> emissions. For instance, Canada has implemented regulations that have reduced emissions by 9% from 2014 to 2019, and in 2020 they set a goal to reduce fugitive CH<sub>4</sub> emissions by 45% by 2025. Besides, the global warming potential (GWP) considered in Howarth and Jacobson's study is debatable, resulting in higher GHG emissions for blue H<sub>2</sub>. According to Diab et al. [29], the significant difference in the results of Howarth and Jacobson's study and other studies is mainly due to the assumptions used in modeling, such as the choice of GWP time horizon (e.g., 100-year GP versus a 20-year GWP) and the selection of CH<sub>4</sub> leakage and carbon capture percentages.

#### 3.3. Economics

Blue  $H_2$  economy is a medium- and short-term prospect for most of the natural gas exporting countries. This is because blue  $H_2$  production is acceptable from an environmental point of view. As indicated previously, there are different techniques for blue  $H_2$  production, and each leads to a different cost. Gaudernack and Lynum [149] compared in their paper the cost of producing blue  $H_2$  through SMR followed by  $CO_2$  sequestration to that of high-temperature pyrolysis, and they found that the SMR process is around 25% more expensive than the pyrolysis process.

According to Hjeij et al. [150], there are several factors that impact blue  $\rm H_2$  cost, such as the capital cost, fossil fuel prices, effects of the system, and CCS costs. In 2020, the cost of blue  $\rm H_2$  production was estimated to be 1.3–2.9 \$/kg of  $\rm H_2$ , assuming a fossil fuel and electricity consumption of 1–10/ MMBtu and 25–60/MWh, respectively. Furthermore, the energy transition toward blue  $\rm H_2$  production seems appropriate to natural gas exporting countries from an economic perspective since they can utilize carbon capture for their existing fossil fuel power stations and other  $\rm H_2$  production facilities that are based on fossil fuels. Additionally, they also have the capital needed for these investments from the natural gas income as well as their well-established relationship with the energy-importing countries.

As reported by Hjeij et al. [150], CCS equipment increases the cost of blue H<sub>2</sub>, which results from the additional capital and operating costs. Nevertheless, the interest in blue H2 production increases with the increasing demand for clean H2 resources in the European and Asian markets, making it a great investment opportunity. The global annual demand for  $H_2$  throughout the past years has increased from  $20 \times 10^6$ tons in 1975 to  $70 \times 10^6$  tons in 2018. Furthermore,  $H_2$  production capacity is expected to increase in the upcoming years in which the current funding for H<sub>2</sub> production projects is more than \$70 billion. In their study, Hjeij et al. [150] reported a framework developed for evaluating the competitiveness of natural gas exporting countries as H<sub>2</sub> exporters according to several indicators, including economic potential. Based on the results from the study, Canada, Australia, the USA, Russia, and Norway showed high competitiveness. However, Qatar's competitiveness as an H2 exporter compared to other natural gas-exporting countries (NGECs) was low (economic potential = 3). This is due to the high dependency on producing and exporting natural gas itself, which is expected to increase with time. In addition, Qatar exhibited relatively lower scores in terms of industrial knowledge and experience in H<sub>2</sub> production and exporting. Therefore, one of the ways to stabilize Qatar's revenue and that of other NGECs is by economic diversification through H2 exports.

Ali Khan et al. [151] presented in their study a spatial techno-economic framework for assessing blue H<sub>2</sub> production using the SMR process with CCS using Australia as a case study. Ali Khan et al. [151] explored the option of converting the gas emissions captured at

SMR hubs by converting them into formic acid through  $CO_2$  electroreduction. This technique was found to be effective in reducing the cost of blue  $H_2$  by 4–9% as well as  $CO_2$  storage dependency. Additional cost reduction can be obtained through  $CO_2$  electrolyzer capacity scaling to convert a greater amount of captured GHG emissions.

Lau [152] presented a comparison between the cost of producing 1 Mscf of blue natural gas to that of blue  $H_2$  with the same heat content. It was found that the cost of blue natural gas is lower than that of blue  $H_2$ , which in turn is less than the cost of green  $H_2$ .

#### 3.4. Energy efficiency

Regarding the energy efficiency of blue  $H_2$  compared to other fossil fuels,  $H_2$  has certain characteristics that are more beneficial. For instance,  $H_2$  has the highest specific energy content than all conventional fuels. Its energy yield is 120 MJ/kg corresponding to  $\sim 2.75$  times higher than other hydrocarbon fuels. For instance, gasoline and natural gas have an energy content of 44 MJ/kg and 48.6 MJ/kg, respectively. Additionally,  $H_2$  has a density that is lower than that of gasoline, enabling it to store 2.6 more energy per mass [153–155].

 $\rm H_2$  is highly flammable in comparison with other fuels. Its flammability limits range between 4% and 75% versus 5.3–15% for methane. Therefore, the operability of  $\rm H_2$  engines will be more effective on lean mixtures than gasoline engines. Additionally, it has low ignition energy (0.02 MJ) and high ignition temperature. However, compared to other fuels,  $\rm H_2$  has very high diffusivity. This property can lead to potential risk issues, as indicated by most researchers. Nevertheless, the advantage of this point is that if an  $\rm H_2$  leak occurs, it will dissipate quickly, which will therefore lead to less unsafe conditions.

Lastly, a main disadvantage of  $H_2$  is that it has a low gas density of 0.09 kg/ $N_A$ m<sup>3</sup>, which can pose two main problems when used as a transportation fuel in internal combustion engines. First, a large volume is required to drive the vehicle's energy. Second, the energy density of the mixture of  $H_2$  and air leads to low power output [156].

# 4. Comprehensive evaluation of cost, CO<sub>2</sub> emissions, and embodied energy of natural gas and hydrogen

As indicated previously, deciding between blue  $\rm H_2$  and natural gas (NG) as a better fuel or energy carrier is not a straightforward decision, due to the multiple factors that contribute to the decision-making process aiming to achieve acceptable environmental and economic objectives. In this section, a concluding comparison between blue  $\rm H_2$  and NG is presented, considering three main parameters: cost,  $\rm CO_2$  emissions, and embodied energy. The comparison also includes grey  $\rm H_2$ , providing more insights facilitating the process of decision-making. In this comparison, we depended on the most commonly used values reported in the literature as well as the average values in some cases.

#### 4.1. Cost

Fig. 13 presents the cost of NG, LNG, grey  $H_2$  (gaseous and liquid), and blue  $H_2$  (gaseous and liquid). Obviously, among the different fuels, NG gas exhibits the lowest cost (2.48 \$/GJ fuel). This was followed by the cost of gaseous grey  $H_2$  (7.83 \$/GJ fuel), LNG (9.48 \$/GJ fuel), and gaseous blue  $H_2$  (12.49 \$/GJ fuel). Fig. 13 also demonstrates that producing the fuels in their liquid form significantly increases their production cost, with the highest cost being that of liquid blue  $H_2$ , which is 32.46 \$/GJ fuel. Such a high cost of liquid fuels can be attributed to the expensive liquefication process for both LNG and liquid  $H_2$ . This process is generally similar for NG and  $H_2$ , as both gases are cooled and compressed to reach their liquefaction temperature. However, there are some differences in the specifics of the liquefaction technology used, which leads to different liquefication costs. For example, NG is usually cooled to -162 °C (-260°F) using refrigerants such as propane ( $C_3H_8$ ) or ethane ( $C_2H_6$ ) [157], while  $H_2$  is typically cooled to -253 °C

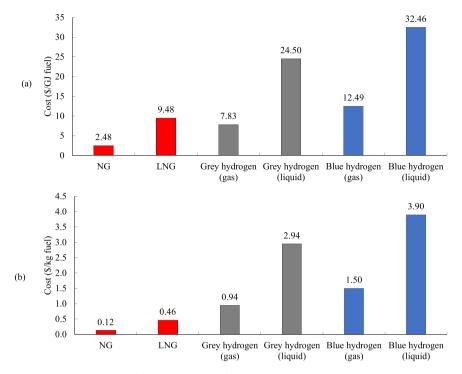


Fig. 13. Cost of NG, LNG, and H<sub>2</sub> (grey and blue): (a) in \$/GJ fuel and (b) in \$/kg fuel. The values were evaluated based on data sourced from [155,162-166].

 $(-423^{\circ}\text{F})$  using specialized cryogenic refrigerants such as helium (He) or neon (Ne), which are be expensive and require special handling and storage [158,159]. Furthermore, unlike  $H_2$ , NG often contains impurities such as water, sulfur compounds, and other hydrocarbons that must be removed before the gas can be liquefied, adding costs to NG liquefication. Moreover,  $H_2$  has a wide range of flammable concentrations (4–75%) in air and requires less energy to ignite than natural gas, making it more prone to ignition, so it requires special handling and

storage to prevent accidents or leaks. This can add to the complexity and cost of  $H_2$  liquefaction [160,161].

## 4.2. CO<sub>2</sub> emissions

Fig. 14 shows the  $CO_2$  emissions resulting from the production and utilization of NG, LNG, grey  $H_2$  (gaseous and liquid), and blue  $H_2$  (gaseous and liquid). In this regard, blue  $H_2$  (gas) results in the lowest

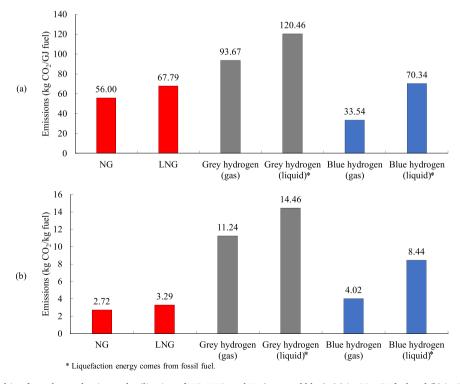


Fig. 14. CO<sub>2</sub> emissions resulting from the production and utilization of NG, LNG, and H<sub>2</sub> (grey and blue): (a) in CO<sub>2</sub>/GJ fuel and (b) in CO<sub>2</sub>/kg fuel. The values were evaluated based on data sourced from [155,163,165,167,168].

amount of CO<sub>2</sub> emissions, with a value of 33.54 kg CO<sub>2</sub>/GJ fuel, hence posing the lowest environmental impact when used for energy applications. This was followed by NG emitting 56.00 kg CO<sub>2</sub> per GJ fuel. However, when liquefaction is involved in the production, CO<sub>2</sub> emissions increase since liquefaction energy comes from fossil fuels. As a result, the emission values of LNG and liquid blue H<sub>2</sub> are comparable, at 67.79 and 70.34 kg CO<sub>2</sub>/GJ fuel, respectively. Nevertheless, if the liquefaction energy comes from renewables, then it is expected that the CO<sub>2</sub> emissions will be considerably less. Regarding grey H<sub>2</sub>, it unsurprisingly results in the highest CO<sub>2</sub> emissions in both of its forms due to the lack of CCS during the SMR process, with grey H<sub>2</sub> (gas) producing 93.67 kg CO<sub>2</sub>/GJ fuel and grey H<sub>2</sub> (liquid) producing 120.46 kg CO<sub>2</sub>/GJ fuel.

#### 4.3. Embodied energy

Fig. 15 presents the energy embodied in NG, LNG, and H<sub>2</sub> (grey and blue), which includes the energy stored in the materials as well as the energy consumed during the production processes. For instance, during H<sub>2</sub> production, energy is consumed through the NG combustion process. Moreover, electricity is required to drive some processes within the SMR plant. Regarding embodied energy, NG requires the lowest amount of embodied energy (~1.0003 MJ input/MJ fuel), which is followed by LNG with a value of (1.06 MJ input/MJ fuel). However, for grey H<sub>2</sub>, the required amount of embodied energy is remarkably higher due to the SMR process, with grey H2 (gas) requiring 1.37 MJ input/MJ fuel and grey H<sub>2</sub> (liquid) requiring 1.67 MJ input/MJ fuel. Compared to grey H<sub>2</sub>, the energy embodied in blue H<sub>2</sub> is higher in each state due to the CCS process, which was 1.56 and 1.86 MJ input/MJ fuel for blue H<sub>2</sub> (gas) and blue H<sub>2</sub> (liquid), respectively. It is worth mentioning that a significant amount of energy is embodied during the liquefaction of NG and H2. On average, a total energy of 36 MJ is required to liquefy 1 kg of H<sub>2</sub> [169], compared to that of 2.9 MJ/kg required for NG liquefaction [170].

#### 5. CO<sub>2</sub> sequestration into depleted natural gas reservoirs

Considering that the oil and gas industry emits a huge amount of  $\mathrm{CO}_2$  into the atmosphere, leading to serious environmental problems, countries have been looking for ways and technologies to mitigate these climate issues.  $\mathrm{CO}_2$  sequestration is considered one of the effective ways to slow down  $\mathrm{CO}_2$  emissions and mitigate global warming and climate change. Given its significance, researchers have studied this topic from both experimental and simulation aspects.

# 5.1. Impact of $CO_2$ sequestration on the rock, fluid, and petrophysical properties

The concept of  $CO_2$  injection was discussed in several papers. However, it was not examined widely through field application. One of the reasons for that is the concern of the injected  $CO_2$  getting mixed with the existing natural gas, which may change the reservoir properties. In a study reported by Oldenburg [171], it was indicated that the technique is achievable, and it was proved through some operational strategies that the mix between  $CO_2$  and methane can be controlled to some extent. Oldenburg [171] performed numerical simulation using the  $CO_2$ -CH4 gas mixtures during the  $CO_2$  injection process. It was found that the density and viscosity variation between the two gases favors limited mixing between both gases [171].

The injection of  $CO_2$  influences the gas phase behavior to be encountered in gas reservoirs [172]. Generally, gas reservoir fluids are classified into three categories including dry gas, wet gas, and retrograde gas (condensate gas). This classification depends mainly on the hydrocarbon composition and reservoir temperature and pressure, governing the gas phase behavior in the reservoir during the depletion process [173]. As Fig. 16 (a) indicates, a dry-gas reservoir is one that consistently produces a single composition of gas in the reservoir, wellbore, and lease-separation facility throughout the field's production life, where no production of associated liquids occurs as the reservoir gets depleted [174,175].

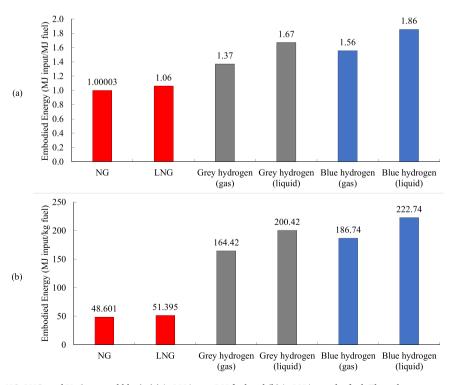
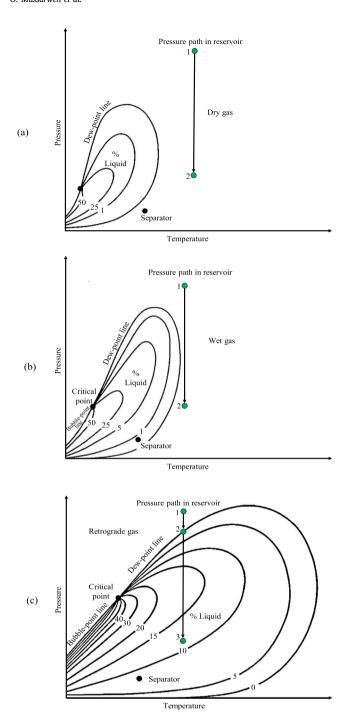


Fig. 15. Energy embodied in NG, LNG, and H<sub>2</sub> (grey and blue): (a) in MJ input/MJ fuel and (b) in MJ input/kg fuel. The values were evaluated based on data sourced from [155,167,169,170].



**Fig. 16.** Phase diagram of a typical: (a) dry gas with isothermal pressure reduction path (1-2), (b) wet gas with isothermal pressure reduction path (1-2), and (c) retrograde gas with isothermal pressure reduction path (1-2-3).

Modified from [177].

In a wet-gas reservoir (Fig. 16 (b)), the reservoir temperature is above the cricondentherm. As the reservoir gets depleted, the gas mixture inside the reservoir stays in the gas phase. However, when the temperature-pressure trajectory of the gas cuts into the two-phase region, condensate drops out either inside the borehole or at the surface facilities [175,176]. In a retrograde gas reservoir, the reservoir temperature is below the cricondentherm but above the critical temperature. As Fig. 16 (c) indicates, at first (point 1), the retrograde fluid is totally gas in the reservoir. As the reservoir pressure drops, reaching

point 2, the retrograde gas undergoes a dew point, then any additional drop in pressure leads to liquid condensation from the gas, forming a free liquid within the reservoir. This liquid usually does not flow and can hardly be produced using conventional gas production technologies [175,177].

In a study conducted by Sobers et al. [172], PVT analysis showed that CO<sub>2</sub> has a wetting effect on dry gas reservoirs and a drying influence on retrograde and wet gas reservoirs. The drying effect means that at given temperature and pressure values, there is a reduction in the volume of the liquid as CO<sub>2</sub> concentration increases. Fig. 17 shows an example of this effect for CO2-retrograde gas mixtures. It is to be noted that in retrograde gas systems, the composition of CO2-natural gas mixtures varies according to the abandonment pressure of the reservoir since the decrease in pressure causes condensation of the heavy ends of gas, leading to the production of the lighter components. Sobers et al. [172] also reported a decrease in natural gas compressibility due to the addition of CO<sub>2</sub>. Furthermore, the formation volume factor (B<sub>g</sub>) of a dry reservoir decreases with the addition of CO<sub>2</sub>, implying an increase in the quantity of CO<sub>2</sub> that can be sequestered in the reservoir (i.e., a lower B<sub>g</sub> implies a larger surface volume of CO2 expected to occupy the pore spaces of the reservoir). However, in the presence of liquid, CO<sub>2</sub> storage capacity is lower because liquids are much less compressible than gases. Therefore, it is recommended to vaporize the retrograde condensate within the reservoir, aiming to enhance gas recovery and maximize CO2 storage capacity. In a study conducted by Adisoemarta [178], it was experimentally shown that adding CO2 to hydrocarbon gas mixtures reduces the compressibility factor (or the z-factor) of gas, indicating a larger storage capacity of CO2 compared to the hydrocarbon gas. Moreover, it was shown that CO2 has a vaporizing effect on the retrograde condensate to various extents depending on the depletion pressure and temperature, as given in Fig. 18.

The effect of CO<sub>2</sub> sequestration on the mechanical properties of rock was also investigated by researchers. For example, Fatima et al. [179] studied the impact of CO2 sequestration on the mechanical strength and petrophysical parameters of the rock. This was accomplished using CMG-GEM simulation where CO<sub>2</sub> was injected into the reservoir model generated by the software for 25 years, and then the impacts following the injection were modeled for the next 200 years. Base case results showed that during the first few years of injection, CO2 rises upward, creating a structural trap. After 16 years of injection, CO<sub>2</sub> begins to be produced from a production well, and after 100 years of injection, it begins dissolving in the formation water, leading to acidic solutions (-HCO<sub>3</sub>) that dissolve minerals except for quartz. This also leads to a further precipitation of calcite and dolomite. Furthermore, sensitivity cases showed that a reservoir with reduced vertical permeability is more favorable for CO2 storage than the base case reservoir due to the decreased rate of vertical migration of CO<sub>2</sub> toward a production well. However, a reduction in the porosity by 40% reduces the CO<sub>2</sub> trapping capability of the formation, causing a 79% increase in the produced CO<sub>2</sub> from the production well. Lastly, the increase in the water salinity increases the density of brine, causing the CO<sub>2</sub> phase to rise more quickly and reach the production well [179].

# 5.2. Optimum location, well type, injection rate, and reservoir selection for highest gas recovery

To obtain the highest gas recovery and maximum  $CO_2$  Storage, researchers have studied the impact of well and reservoir characteristics during the process of  $CO_2$  sequestration. Narinesingh and Alexander [180] studied the impact of different well placements on gas recovery using CMG-GEM numerical reservoir simulation coupled with WIN-PROP. In that study, the well locations were varied while keeping the injection pressure constant at 137.9 bar for 1000 years to model the flow of  $CO_2$  plume into the reservoir. The results showed that 60% of the injected  $CO_2$  remained in the reservoir, which can be attributed to the small amount of  $CO_2$  injected into a relatively large reservoir at low

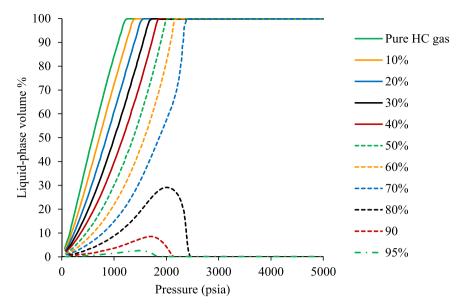
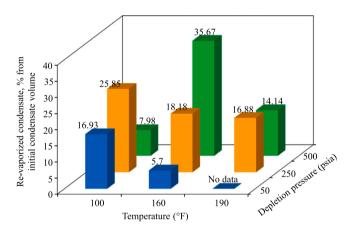


Fig. 17. Effect of CO<sub>2</sub> concentration (0–95%) and pressure on the liquid-phase volume percent of a retrograde gas at 150 °F. Modified from [172].



**Fig. 18.** Re-vaporized condensate as a function of temperature and depletion pressure. Modified from [178].

injection pressure, leading to  $\sim$ 7% of condensate recovery. Additionally, placing a horizontal well between two producers was found to be the optimal placement, with 37.1% condensate recovery and 77.9%  $\rm CO_2$  storage, followed by a vertical well with 37.8% condensate recovery and 70.8%  $\rm CO_2$  storage [180].

Korelskiy et al. [181] modeled the stress-strain changes of the rock during  $\mathrm{CO}_2$  injection. They also indicated the conditions that lead to faults reactivation and caprock failure, which assists in potential reservoir selection for long-term  $\mathrm{CO}_2$  storage. Korelskiy et al. [181] performed their study through geomechanical modeling using the finite element method, where the pore pressure changes were studied in 5 steps of increasing injection volume and reservoir pressure. The results indicated that the maximum displacement of the reservoir top is 0.53 m. Additionally, rock failure and reactivation of faults begins in step 3 for the carbonate rock due to its high stiffness, which is impacted by the significant changes in stress during the early stages of injection [181].

The flow of  $\mathrm{CO}_2$  into the reservoir was modeled by other researchers who investigated its influence on the water pressure system of retrograde gas condensate reservoirs. For instance, Matkivskyi et al. [182] modeled the effect of increasing the rate of  $\mathrm{CO}_2$  injection at gas-water contact (GWC) on the rate of gas production from producing wells. In

the numerical simulation tool, the CO<sub>2</sub> injectors were placed at the boundary of GWC. It was found that as the CO<sub>2</sub> injection rate increases, the gas production rate increases while the water production rate decreases. This is due to the solubility of CO<sub>2</sub> in water, which slows down the water production rate [182]. Cao et al. [183] investigated the impact of reservoir temperature, injection rate, and impurity concentration on the performance of CCS and gas recovery. They used TOUGH2MP software to simulate the impact of CO<sub>2</sub> sequestration on a depleted gas reservoir with a 5-spot well pattern of injectors. Results from numerical simulation indicated that CO<sub>2</sub> storage decreases with the increase in the level of CO<sub>2</sub> impurity. Moreover, CO<sub>2</sub> storage is more favorable in a low temperature reservoir of 75 °C compared to 114 °C. Regarding the impact of CO<sub>2</sub> injection rate on the storage capacity, the results showed that the capacity decreases with the increase in the injection rate [183].

# 5.3. CH<sub>4</sub> desorption by CO<sub>2</sub>

The desorption of CH<sub>4</sub> under the influence of CO<sub>2</sub> is an important aspect of CO<sub>2</sub>-based enhanced gas recovery (EGR) and CO<sub>2</sub> sequestration in natural gas reservoirs. Generally, gases (e.g., CH<sub>4</sub>) are adsorbed on solid adsorbents (e.g., rock surfaces) by physisorption and/or chemisorption. Physisorption involves mainly the deposition of the adsorbate on the solid surface where van der Waals interactions (and/or induced dipoles) attract the adsorbate molecules to the surface. On the other hand, chemisorption is characterized by the formation of chemical

**Table 5**Comparison between chemisorption and physisorption.

Property	Chemisorption	Physisorption	References
Bonding forces	Chemical bonding	Van der Waals	[185]
	(involving electron	interactions (and/or	
	transfer or sharing)	induced dipoles)	
Heat of	High (> 1.5 times the	Low (1.0 – 1.5 times	[188]
adsorption	evaporation heat)	the evaporation heat)	
Formation of multilayers	Monolayer only	Monolayer or multilayer	[188]
Change in the adsorbed molecule	A significant change in the chemisorbed molecule due to reaction or dissociation	Negligible change (the physisorbed molecule mostly preserves its identity)	[189]
Reversibility	Chemisorption is irreversible due to strong chemical bonding	Highly reversible	[190]

bonds between the molecules of the gas and the adsorbent surface where electron transfer or sharing occurs [184,185]. Table 5 provides a comparison between chemisorption and physisorption. Several factors affect the gas adsorption capacity of reservoir rock, such as sorption affinity, reservoir temperature, reservoir pressure, pore size, specific surface area, and the presence of organic minerals, which have higher CH<sub>4</sub> adsorption capacity than inorganic minerals [186,187].

According to Cui et al. [191], the size of the adsorbate gas molecule is an important parameter affecting the selective adsorption of gases on adsorption sites. The  $CO_2$  molecule has a kinetic diameter  $(\sigma_k)$  of 0.33 nm, while CH<sub>4</sub> has a  $\sigma_k$  of 0.38 nm. Therefore, the adsorptive capacity of rock for CO2 is larger than that for CH4 since molecules of smaller  $\sigma_k$  can enter a larger number of pores of smaller diameter, which are located on the rock surface. Subsequently, CO<sub>2</sub> can be used to desorb pre-adsorbed CH<sub>4</sub> in gas reservoirs. Eliebid et al. [192] experimentally studied the competitive adsorption between CO2 and CH4 on carbonate rock, considering the pressure and temperature effects. The results showed that the adsorption of  $CO_2$  on Pink Desert limestone is  $\sim 4$  times the adsorption of CH<sub>4</sub> on the same rock at a temperature of 100 °C and pressures ranging from 0 to 50 bar. Such a value of CO2 adsorption indicates that carbonate reservoirs are good candidates for CO<sub>2</sub> capturing. It was also shown that the increase in temperature significantly reduces CH<sub>4</sub> adsorption, was measured at 50 °C, 100 °C, and 150 °C. Furthermore, desorption experiments showed that CO2 could desorb CH4 from carbonate surfaces. For example, adding 10% CO2 to CH4 gas desorbed 7 mg/g CH<sub>4</sub> at 100 °C and 50 bar. Therefore, these effects can be exploited in EGR by injecting hot CO2 into natural gas reservoirs.

In another study, Eliebid et al. [193] reported that Indiana limestone exhibited a significantly higher adsorption capacity for  $\mathrm{CO}_2$  compared to  $\mathrm{CH}_4$ . The high  $\mathrm{CO}_2$  adsorption was attributed to the high affinity of calcite surfaces for  $\mathrm{CO}_2$  since the measured Gibbs free energy was low and negative. Regarding sandstone rock, Eliebid et al. [194] reported that Kentucky sandstone also has a significantly higher  $\mathrm{CO}_2$  adsorption compared to  $\mathrm{CH}_4$ , indicating a higher tendency of  $\mathrm{CO}_2$  to be captured by the rock. However, increasing the temperature up to  $100\,^{\circ}\mathrm{C}$  promoted a less adsorption uptake of gases, including  $100\%\,^{\circ}\mathrm{CH}_4$ ,  $90\%\,^{\circ}\mathrm{CH}_4/10\%\,^{\circ}\mathrm{CO}_2$ , and  $100\%\,^{\circ}\mathrm{CO}_2$ . Nevertheless, increasing the temperature to  $150\,^{\circ}\mathrm{C}$  caused the opposite effect, meaning higher  $\mathrm{CO}_2$  uptake when it is injected at high temperatures into depleted natural gas reservoirs.

# 5.4. Potential CO2 leakage

Despite the positive effects of CO<sub>2</sub> sequestration in reducing CO<sub>2</sub> emissions into the atmosphere, researchers have shown some concerns related to the potential leakage of CO2. In research reported by Nygaard et al. [195], the risk of CO<sub>2</sub> leakage due to thermal and pressure changes introduced from CO<sub>2</sub> injection was evaluated. In their research, Nygaard et al. [195] used a 3D simulation model to study the cement-casing and cement-formation interactions with the temperature of 95 wells from the Wabamun area, Alberta. Based on the simulation results, it was found that only 4 wells out of a selected subset of 27 wells would require workover due to impacted wellbore integrity due to CO2 leakage. Additionally, Zulqarnain et al. [196] performed a risk-based approach to categorize the wells that have leakage potential in a depleted gas field in Sothern Louisiana. They used Petrel and CMG-GEM to simulate four sensitivity cases of CO2 leakage potential. Based on the model findings, it was found that uncased wells have the highest CO2 leakage potential, followed by cased-uncemented and cased-cemented. In addition, a leaky well at 328 ft distance from the injector has the highest leakage volume compared to other wells far away, and hence the maximum injector-leaky well distance leads to minimum CO2 leakage potential. Lastly, three sensitivity cases were also generated on the storage boundary type (closed, semi-closed, and open boundary) to simulate the pressure build-up rate. The simulation results indicated that in a closed system boundary, the pressure increase is the highest and the CO<sub>2</sub> plume spread is the lowest, leading to the highest leakage volume [196].

Lastly, in order to ensure a safe  $CO_2$  sequestration project, appropriate  $CO_2$  storage site selection, along with a plan for monitoring, measurement, and verification (MMV), is required. This was studied by Tiwari et al. [197,198], who used a 3D reservoir simulation model coupled with geomechanical models on a field located in Malaysia to understand storage capacity, integrity, and  $CO_2$  migration path. In the end, an MMV plan was recommended. Their study results suggested that no changes in the porosity or the mineralogy are expected throughout the injection period, and hence there will not be any impact on the storage capacity. Regarding the MMV plan, it is recommended to install fiber optic system in the injectors, and perform surface seismic acquisition for full field monitoring and time-lapsed evaluations of  $CO_2$  plume migration [197,198].

#### 6. Environmental effects of CO2 sequestration

Fossil fuels are a major contributor to the increase in  $\mathrm{CO}_2$  emissions into the atmosphere, leading to climate change and global warming [199]. According to the climate analysis indicator tool, in Qatar, for example, around 96% of greenhouse gases come from the energy sector, which consists mainly of oil and natural gas production [200]. Additionally, the Intergovernmental Panel on Climate Change (IPCC) estimated that the 8000 largest point sources are responsible for 40% of the total  $\mathrm{CO}_2$  emissions associated with human activities [201].

To address and solve this issue, countries have taken several mitigation measures to reduce  $CO_2$  emissions by 50–90% and to stabilize the greenhouse gas emissions at 450–550 ppm  $CO_2(e)$  in the atmosphere by 2050, following the Paris agreement on climate change [202]. These mitigation measures include: (1) using carbon capture and storage technologies by capturing  $CO_2$  emissions from power plants; (2) compressing and liquefying  $CO_2$ ; and (3) injecting  $CO_2$  into depleted gas reservoirs [203].

 ${\rm CO_2}$  sequestration is identified as one of the most effective techniques for reducing the  ${\rm CO_2}$  emissions causing global warming. For instance, it is estimated that atmospheric  ${\rm CO_2}$  emissions can be reduced by around 80–90% when CCS technologies are implemented in a power plant [204,205]. It is also estimated that the appropriate application of CCS technologies can reduce the concentrations of  ${\rm CO_2}$  to 450 ppm by the year 2100 [206,207].

Additionally, researchers have conducted several life-cycle assessment (LCA) studies to estimate the amount of  $\mathrm{CO}_2$  reduction resulting from CCS. Pehnt and Henkel [208] found that GHG emissions are reduced during all process stages with CCS. Odeh and Cockerill [209] performed LCA on three fossil fuel power plants, and they estimated GHG emissions with and without CCS. Their results showed that with 90% carbon capture efficiency, GHG emissions are reduced by 75–84%. These results align with the LCA study performed by Singh et al. [210] on a natural gas power plant.

While  $CO_2$  sequestration proved its effectiveness in reducing the global warming, however, there are some concerns related to the potential impacts of the accidental leakage of  $CO_2$  on the environment. (Amir Rashidi et al. [211] modeled the environmental consequence of  $CO_2$  leakage on the marine environment in offshore Sarawak. They used 3D-hydrodynamic modeling to understand the ambient flow conditions at which  $CO_2$  seepage will occur, leading to variations in the pH of seawater. The scenario of  $CO_2$  leakage was modeled using plugged and abandoned well at leakage rates derived from the literature. The simulation results indicated that  $CO_2$  would dissolve at the seabed only and would not escape into the atmosphere. Additionally, the pH reduction to less than 6.5 is estimated to be within the variations of seawater acidity and within a 200 m radius from the leakage source.

Despite the positive effects of CCS, researchers have shown some concerns related to the environmental impact. For instance, in their paper, Wilberforce et al. [212] reported several challenges associated with this  $CO_2$  sequestration. One of the key environmental issues is the accidental release of  $CO_2$  from the pipelines causing groundwater to be

acidic and leading to water pollution that would need further treatment due to the increased salinity. Furthermore, injecting  $\mathrm{CO}_2$  into the soil increases its pH, leading to heavy metal mobilization. An additional impact of  $\mathrm{CO}_2$  sequestration was highlighted by Shukla et al. [213] as they indicated that a high  $\mathrm{CO}_2$  injection rate could increase the stress pressure reducing the rock strength and leading to additional fractures.

# 7. Field application of CO<sub>2</sub> sequestration in natural gas reservoirs

#### 7.1. Conditions for field application of CO<sub>2</sub> sequestration

For efficient  $CO_2$  sequestration, geologic formations need to: (1) have a large storage capacity to accumulate large quantities of  $CO_2$ ; (2) have a seal rock preventing the vertical escape of  $CO_2$  to the earth's surface; (3) have sufficient permeability to allow large injection rates of  $CO_2$  through a feasible number of wells; (4) be sufficiently deep ( $\geq$  800 m) to maintain the stored  $CO_2$  under pressure ensuring a dense supercritical state; and (5) be free of leakage conduits (e.g., fractures, faults, and improperly sealed wells) extending from the injection formation to the land surface [214–217].

The entrapment of gases (e.g., CH<sub>4</sub>) in natural gas reservoirs indicates the capability of these reservoirs for long-term gas containment. With proven gas-production records, many natural gas reservoirs were shown to have not only the required volume but also adequate integrity against gas escape [218–220]. Furthermore, depleted gas reservoirs have a larger  $\text{CO}_2$  storage capacity than depleted oil reservoirs, considering the same initial hydrocarbon pore volume. The first reason for that is the higher ultimate gas recovery factor ( $\sim$  65% of the initial-gas-in-place) in gas reservoirs versus that of  $\sim$  35% of the initial-oil-in-place in oil reservoirs. The second reason is the high compressibility of gas, which is nearly 30 times more than that of oil or water. Additionally,  $\text{CO}_2$  behaves as a supercritical critical fluid at typical gas reservoir conditions (i.e., above 31 °C and 73.8 bar). As such, the injection of  $\text{CO}_2$  into deep depleted gas fields could be deemed a reasonable strategy for geologic  $\text{CO}_2$  storage [221].

# 7.2. Case studies

Recently, many simulation studies and pilot projects have been implemented focusing on  $\mathrm{CO}_2$  storage in natural gas fields. In this subsection, studies and pilot projects aiming to evaluate the potential of natural gas fields for  $\mathrm{CO}_2$  storage are discussed.

#### 7.2.1. Austria

In a study conducted by Polak and Grimstad [222],  $CO_2$  storage potential in Austria's Atzbach-Schwanenstadt gas field was estimated based on the volume of produced gas. The field had an available storage pore volume corresponding to  $14.5 \times 10^6$  tonnes of  $CO_2$ . Since low reservoir permeability affects the distribution of the injected  $CO_2$ , the actual storage capacity was expected to be less than the available pore volume. The simulation study considered  $CO_2$  storage until reaching the initial reservoir pressure (160 bars) to avoid reservoir seal fracturing. The source of  $CO_2$  was estimated to provide the field with  $3 \times 10^5$  tonnes of  $CO_2$  per year. Reservoir simulation showed that  $8.2 \times 10^6$  tonnes of  $CO_2$  could be stored over an injection period of 30 years. Long-term simulation of the storage process indicated that only 10% of the stored  $CO_2$  would dissolve in the immobile water of the reservoir over a period of 1500 years.

### 7.2.2. The Netherlands

Van der Meer et al. [223] studied the feasibility of a  $CO_2$  sequestration project in the K12-B field, which is located in the North Sea, the Netherlands. The project aimed to sequester the  $CO_2$  separated during natural gas production. The  $CO_2$  content in the produced gas was estimated at 13%. Reservoir simulation showed that  $\sim 60$  tonnes of  $CO_2$  per

day could be injected into the K12-B site. A pilot test was also conducted, which included the injection of CO<sub>2</sub> at a rate of 2350 kg per hour ( $\sim$  56.4 tonnes per day). A total of 9000 tonnes of CO<sub>2</sub> were successfully injected over the test period, leading to an increase of 9 bars in the bottom hole pressure (BHP). The environmental impact of the project was deemed positive. In a feasibility study reported by Leeuwenburgh et al. [224], two gas fields in the Netherlands were investigated for CO<sub>2</sub> sequestration. The study demonstrated that at the end of the life of the investigated fields, the amount of sequestered CO<sub>2</sub> is estimated to be 0.4  $\times$  106 tonnes of CO<sub>2</sub>. However, the study revealed that the associated CO<sub>2</sub>-EGR is limited to 1%.

#### 7.2.3. United Kingdom

Hughes [225] reported that depleted offshore gas fields in the UK have the capacity to store  $3.8 \times 10^9$  tonnes of CO<sub>2</sub>. Particularly,  $1 \times 10^9$  tonnes and  $2.8 \times 10^9$  tonnes can be stored in the East Irish Sea fields and the Southern North Sea (SNS) fields, respectively.

#### 7.2.4. South Africa

In a study conducted by Saffou et al. [226], a numerical model was developed to evaluate the potential for  $CO_2$  sequestration in a near-depleted gas field located in the Bredasdorp Basin, South Africa. The model considered compartment C1, which is part of a sandstone reservoir. The study showed that  $7.78 \times 10^7$  MSCF of  $CO_2$  could be stored over an injection period of 71 years with no leakage or integrity issues. It was also shown that  $6.79 \times 10^7$  MSCF of natural gas could be recovered during a 10-year period of  $CO_2$ -EGR.

#### 7.2.5. Nigeria

In another study, Ojo and Tse [227] estimated the  $\rm CO_2$  storage potential of three depleted sandstone gas reservoirs situated in a field in the Niger Delta, Nigeria. The reservoirs are located at around 3319 m to 3539 m below the surface, which allows for maintaining the  $\rm CO_2$  in a supercritical fluid state. The geological assessment demonstrated that the reservoirs have the potential to store a total of 147 MM tonnes of  $\rm CO_2$ .

# 7.2.6. United States

In a study reported by Godec et al. [228], reservoir simulation was conducted to estimate the EGR and  $\text{CO}_2$  storage capacity in the Marcellus gas shales, USA. It was shown that the shale formation has a storage capacity of  $55 \times 10^9$  tonnes of  $\text{CO}_2$ , which can lead to additional  $\text{CH}_4$  recovery of  $12 \times 10^{12} \, \text{m}^3$ , based on an incremental gas recovery of 7% due to  $\text{CO}_2$  injection.

# 7.2.7. Germany

Kühn et al. [229] reported that a pilot project is being planned to inject  $100 \times 10^3$  tonnes of  $CO_2$  into the Altensalzwedel block, which is a subfield of the near-depleted Altmark natural gas field in Germany. A simulation study conducted by Ganzer et al. [230] demonstrated that the Altmark gas field is highly suitable for  $CO_2$  injection. At injection rates of 143 tonnes per day and 200 tonnes per day (over 8 years of injection), the injected amounts of  $CO_2$  will be retained in the formation without reaching the production wells. Additionally, the distribution of  $CO_2$  in the formation will depend on heterogeneities in the formation properties, such as litho-facies type, porosity, and permeability.

#### 7.2.8. Other projects

An important project of  $CO_2$  sequestration is the Otway Project, Victoria, Australia. The project involved the injection of a fluid mixture into the Waarre Formation ( $\sim$ 2 km below the surface), which consisted of around 75 mol%  $CO_2$  and 21 mol%  $CH_4$ , with the remaining content including nitrogen, condensate, and wet hydrocarbons. The total amount of the injected fluid was 65,445 tonnes [231,232]. The Otway Project demonstrated that  $CO_2$  could be safely injected into depleted gas reservoirs. Moreover, the project developed methods for monitoring and

verification of  $CO_2$  storage, which can be used in future commercial-scale projects of  $CO_2$  sequestration [233]. Similar to the Otway Project, the feasibility of the  $CO_2$  sequestration in other gas fields has been evaluated, such as the DF1–1 gas field (South China Sea) [234] and the P18–4 gas field (offshore the Netherlands) [235], which exhibited a potential  $CO_2$  storage capacity of 8 Mt and 1 Gt, respectively [236]. In another study, Aimard et al. [237] reported that a plan was developed to store  $150 \times 10^3$  tonnes of compressed  $CO_2$  in a depleted gas reservoir of the carbonate type, which is located in the southwest of France, over a 2-year injection period.

#### 8. CO<sub>2</sub> utilization pathways

 $CO_2$  utilization assists in mitigating climate change by removing atmospheric  $CO_2$  and storing it for a long term in a stable form or by reducing the amounts of  $CO_2$  released into the atmosphere. Typically,  $CO_2$  utilization pathways are categorized into two main categories: (1) pathways of direct  $CO_2$  utilization and (2) pathways of  $CO_2$  conversion to energy and chemical products [238,239]. Table 6 demonstrates some applications and industries that involve  $CO_2$  utilization.

As shown in Table 6, there are several vital industrial applications where  $CO_2$  is utilized. For example,  $CO_2$  is favorable for use in cooling systems due to its unique thermophysical properties, such as the high heat transfer coefficient [240], nonflammability [241], and low viscosity, which results in high circulation rate [242]. Moreover,  $CO_2$  is widely used as a chemical feedstock to synthesize chemical intermediates (e.g., solvents, urea, Fischer-Tropsch (F-T) products) and fuels (e.g., methanol and ethanol). However, the consumption of these fuels could emit  $CO_2$  again.

#### 9. Conclusions

The development of  $H_2$  strategies has gained considerable pace in recent years. In that context, this paper reviewed recent literature concerning blue  $H_2$ , a potential low-carbon, short-term solution during the  $H_2$  transition period. This paper focused on three main aspects. First, it discussed the production of blue  $H_2$  and the challenges it faces in the rapidly developing energy harvesting and conversion fields. Second, the paper presented a detailed comparison between blue  $H_2$  and NG, aiming to inform the strategy developers of their advantages and disadvantages as fuels and energy carriers. Third, this paper discussed  $CO_2$  sequestration, an essential part of implementing blue  $H_2$ .

Blue  $H_2$  production from NG involves two main processes: SMR and ATR. These are the same processes used to produce conventional  $H_2$ , but the main difference here is that blue  $H_2$  production is complemented with CCS. SMR is more commonly used than ATR, often implemented in cases where specific  $H_2$ :CO ratios are desired for some industries. Despite its potential, blue  $H_2$  as an energy carrier is expected to be challenged by other advancing technologies such as solar power, battery storage, and thermal energy storage.

NG and blue  $\rm H_2$  employ common methods of transportation (e.g., compressed gas containers, cryogenic liquid tankers, Pipelines, etc.). However,  $\rm H_2$  transportation is more expensive because more energy is needed for its compression, liquefaction, and regasification compared to NG. Regarding the production cost, blue  $\rm H_2$  gas is relatively more expensive than NG due to the cost of the SMR and CCS processes. In terms of  $\rm CO_2$  emissions, blue  $\rm H_2$  gas poses a lower environmental impact compared to NG and LNG. However, in its liquid form, blue  $\rm H_2$  has comparable  $\rm CO_2$  emissions to that of LNG, which in turn is higher than that of NG.

Successful blue  $H_2$  production necessitates a proper implementation of CCS. Recently, numerous research studies have been conducted on  $CO_2$  sequestration in depleted NG reservoirs. These formations are considered a viable option for subsurface  $CO_2$  storage due to their proven ability to trap hydrocarbons over geologic time scales, which many simulation studies and field projects in many countries have

**Table 6** Applications and industries involving CO<sub>2</sub> utilization [238].

${\rm CO_2}$ utilization route	Direct/indirect use	${\rm CO}_2$ utilization process
Physical routes ( $CO_2$ remains in molecular form, which is used pure or in mixtures)	Direct use (CO <sub>2</sub> goes directly into the product)	Dry ice Refrigerant Carbonated beverage Fire extinguisher Solvent
	Indirect use (CO <sub>2</sub> boosts a process generating a product)	Welding medium Power via supercritical CO <sub>2</sub> -cycle Additional oil via enhanced oil recovery
		(EOR) Additional gas via enhanced gas recovery (EGR)
Chemical routes (CO <sub>2</sub> molecules crack and convert to other products)	Direct use (CO <sub>2</sub> goes directly into the product)	Calcium carbonate Cetylsalicylic acid Polypropylene carbonate Salicylic acid Urea
	Indirect use (CO <sub>2</sub> boosts a process generating a product)	Cyclic carbonate Formic acid Synthetic gas Polyurethane Fischer-Tropsch (F-T) products
		Algae fuel Dimethyl carbonate (DMC) Dimethyl ether (DME) Methanol (MeOH)

demonstrated. Furthermore, the conversion of depleted NG reservoirs into storage sites is relatively undemanding due to an abundance of field experience regarding the injection and production performance of these formations, utilizing the current site infrastructure.

In conclusion, the demand for  $H_2$  as an energy carrier and feedstock for many essential industrial applications is continuously increasing. Currently, most  $H_2$  is produced from natural gas through the SMR and ATR processes. Therefore, it is apparent that the need for improving the blue  $H_2$  technology is inevitable to meet the high  $H_2$  demand, at least over the coming decades. Based on the current literature, this can be achieved through optimizing the SMR and ATR processes and CCS operations and improving  $H_2$  compression and liquefaction technologies. Finally, it is important to incorporate renewables to reduce the energy consumed at the different stages of blue  $H_2$  production.

# **Declaration of Competing Interest**

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

# **Data Availability**

Data will be made available on request.

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